Density, Surface Tension, and Refractive Index of Octane + 1-Alkanol Mixtures at $T = 298.15$ K

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Surface tension and refractive index for binary mixtures of {octane + ethanol, + 1-propanol, + 1-butanol, + 1-pentanol, + 1-hexanol, + 1-heptanol, and + 1-octanol}, and density for {octane + ethanol} have been measured at $T = 298.15$ K for the whole composition range. From the experimental data we extract excess molar volumes, surface tension deviations, and changes of refractive index. All these experimental data are compared among them and with previous results of the binary mixtures hexane + 1-alkanol and dibutyl ether + 1-alkanol. Also we compare our present results with other published data when available.

Introduction

The accurate measurement of experimental data of physical properties in mixtures of organic liquids is required for a full understanding of their thermodynamic behavior. In particular, the alkane + 1-alkanol mixtures are used as additives to petrol and in rectification processes for binary azeotropes. Compared with other physical properties such as density or viscosity, very little attention has been focused on the measurement of surface tension of liquid mixtures, although this is a very important property for many physical, chemical, and biological processes. In this work, we present measurements on surface tension, density, and refractive index of $\{C_8H_{18} + C_MH_{2M+1}OH\}$ covering the whole range of mole fractions at $T = 298.15$ K and atmospheric pressure. The number of carbons (M) in the 1-alkanol ranges from two to eight, which allows the study of the influence of the 1-alkanol chain length with respect to the physical quantities measured. The density of some of these binary mixtures was previously measured in our laboratory and already published.[1−4] Those data are reproduced in this paper and used for different calculations.

From the measured density, surface tension, and refractive index we extract, respectively, the excess molar volumes, surface tension deviations, and changes of refractive index for all seven mixtures. The resulting data were correlated using a Redlich−Kister[5] type equation, and the obtained fitting parameters are presented. These results were compared with those of {hexane + 1-alkanol}[6] and {dibutyl ether + 1-alkanol}[7] mixtures, previously published by us, and with other literature data.

Experimental Section

The mass purities and the source of the chemicals employed were as follows: octane (Fluka, ≥99.5); ethanol (Panreac, ≥99.8); 1-propanol (Sigma-Aldrich, ≥99.5); 1-butanol (Fluka, >99.5); 1-pentanol (Fluka, >99.0); 1-hexanol (Fluka, ≥99.0); 1-heptanol (Sigma, ≥99.0); 1-octanol (Fluka, >99.5). The liquids were degassed by ultrasound and dried over molecular sieves (Sigma type 0.4 nm) and otherwise used as supplied.

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The surface tensions of the pure liquids and their corresponding mixtures were measured with a thermostated (within ±0.05 K) drop volume tensiometer LAUDA TVT1 with a real sensitivity of 10^{-3} mN·m^{-1}. Detailed instrument design and experimental procedure have been described elsewhere;[8] only note that we need to know the density of the mixtures to calculate the surface tension from the drop volume measured by the tensiometer. Density was measured with an Anton-Paar DMA 60/602 densimeter, thermostated at $T = (298.15 \pm 0.01)$ K in a Haake F3 circulating-water bath. The sensitivity of the densimeter is around 10^{-5} g·cm^{-3}. Immediately prior to each series of measurements, double distilled water and heptane (Fluka, mole fraction ≥ 99.5) were used to calibrate the densimeter. The refractive index was measured with a thermostated (within ±0.01 K) automatic refractometer Atago RX-1000 that uses the D-line of Na, with reproducibility in the refractive index data of 10^{-4}. Finally, all mixtures were prepared by mass using a Mettler AT 201 balance with a sensitivity of 10^{-5} g. The precision of the mole fraction calculated is then estimated to be better than 10^{-4}.

Results and Discussion

The measured density, refractive index, and surface tension of the pure liquids at $T = 298.15$ K are listed in Table 1 together with literature values. For the surface tension we include two values from two different sources, when available, due to the lack of agreement among the data sources used. The agreement, within the experimental uncertainties, between both sets of data indicates that the compounds used were pure and that our experimental equipment has good accuracy.

The only density we have measured for this work corresponds to the binary mixture {(x)octane + (1 − x)-ethanol}, because the other mixtures studied were previously measured by us and are already published: {octane + butanol, + hexanol, and + octanol},[1] {+ propanol},[2] {+ pentanol},[3] and {+ heptanol}.[4] Those data were used in this paper to obtain the surface tension. In Table 2 we present the measured density of {(x)octane + (1 − x)-ethanol} for the whole composition range.

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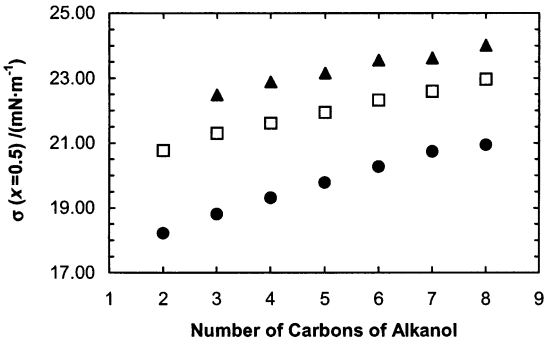

Figure 1. Surface tension for (●) hexane,[6] (▲) octane, and (□) dibutyl ether[7] + 1-alkanol mixtures at the equimolar composition versus the number of carbons of the 1-alkanol.

Table 1. Physical Properties of the Pure Components at $T = 298.15$ K

	ρ/g·cm^{-3}		n_D		σ/mN·m^{-1}	
substance	exptl	lit.	exptl	lit.	exptl	lit.
octane	0.698 50	0.698 54[9]	1.3951	1.39512[9]	21.17	21.18[10] 21.14[11]
ethanol	0.785 15	0.785 09[12]	1.3593	1.35941[12]	21.74	21.68[10] 21.97[11]
1-propanol	0.799 62	0.799 60[10]	1.3830	1.3830[13]	23.39	23.11[10] 23.32[11]
1-butanol	0.805 76	0.805 75[10]	1.3973	1.3973[12]	24.02	23.38[10] 24.93[11]
1-pentanol	0.810 83	0.810 80[10]	1.4080	1.4080[10]	24.97	24.74[10] 25.36[11]
1-hexanol	0.815 15	0.815 34[10] 0.815 15[14]	1.4159	1.4157[10] 1.4161[14]	25.73	25.81[11]
1-heptanol	0.818 75	0.818 6[12]	1.4224	1.4225[12]	26.47	26.5[12]
1-octanol	0.821 63	0.821 57[10]	1.4274	1.4276[10,12]	27.13	26.94[10] 27.10[11]

Table 2. Density of {(x)Octane + $(1 - x)$Ethanol} at $T = 298.15$ K

x	ρ/g·cm^{-3}	x	ρ/g·cm^{-3}
0.0131	0.781 56	0.3985	0.727 44
0.0523	0.771 81	0.4536	0.725 27
0.0788	0.766 05	0.5175	0.721 13
0.1433	0.758 82	0.5832	0.716 94
0.1881	0.754 27	0.6709	0.713 14
0.2228	0.747 52	0.7226	0.708 80
0.2616	0.742 94	0.8201	0.706 53
0.3011	0.738 29	0.8707	0.702 91
0.3725	0.734 06		

Table 3 lists the surface tensions for the seven binary mixtures measured. In Figure 1 we plot the equimolar surface tensions of {0.5octane + (0.5)1-alkanol} against the number of carbon atoms of 1-alkanols; also we plot the equimolar surface tensions of those mixtures with hexane + 1-alkanol[6] and dibutyl ether + 1-alkanol[7] for comparison. As can be noted, the surface tension increases as the chain length of the 1-alkanol does for the three systems, and the shape and increasing rate are also similar for all. Thus, we conclude that the cohesion forces among the molecules depend only on the length of the carbon chain in the different molecules and not on the distribution of the functional groups.

The refractive indexes for the seven measured mixtures are presented in Table 4. In Figure 2 we plot the equimolar refractive indexes of all mixtures as well as those for hexane + 1-alkanol[6] and dibutyl ether (DBE) + 1-alkanol[7] for comparison. As we can observe from Figure 2, the refractive index increases with the increasing of the chain length of the 1-alkanol, as also happens for the mixtures containing hexane and DBE (which also contains eight carbon atoms). The values for the mixtures with hexane

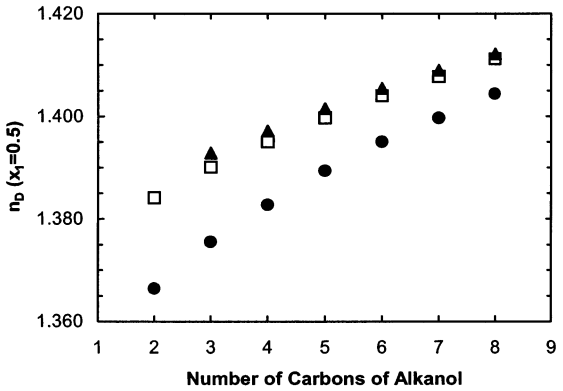

Figure 2. Refractive index for (●) hexane,[6] (▲) octane, and (□) dibutyl ether[7] + 1-alkanol mixtures at the equimolar composition versus the number of carbons of the 1-alkanol.

Table 3. Measured Surface Tensions of {(x)Octane + $(1 - x)$1-Alkanol} at $T = 298.15$ K

x	σ/mN·m^{-1}	x	σ/mN·m^{-1}	x	σ/mN·m^{-1}
		Octane + Ethanol			
0.0686	21.37	0.4898	20.77	0.7650	20.89
0.1132	21.20	0.5401	20.78	0.8041	20.92
0.1721	21.06	0.5865	20.79	0.8191	20.94
0.2867	20.88	0.6299	20.80	0.9095	21.01
0.3571	20.82	0.6457	20.81	0.9212	21.05
0.4544	20.77	0.6690	20.84		
		Octane + 1-Propanol			
0.1230	22.38	0.4493	21.36	0.7592	21.17
0.1859	22.05	0.5154	21.30	0.7884	21.17
0.2349	21.81	0.5218	21.29	0.8449	21.16
0.2900	21.66	0.6224	21.22	0.8918	21.16
0.3454	21.53	0.6308	21.21	0.9245	21.16
0.3547	21.51	0.6938	21.21		
		Octane + 1-Butanol			
0.0453	23.65	0.3526	21.99	0.6918	21.36
0.1130	23.15	0.4467	21.73	0.7770	21.29
0.1636	22.84	0.4857	21.65	0.8167	21.28
0.1788	22.74	0.5747	21.49	0.8650	21.24
0.2956	22.20	0.5886	21.49	0.9376	21.21
0.3281	22.08	0.6326	21.42		
		Octane + 1-Pentanol			
0.1150	23.88	0.4927	21.96	0.7932	21.46
0.1459	23.60	0.5188	21.91	0.8340	21.41
0.1629	23.50	0.5951	21.75	0.8869	21.35
0.2727	22.80	0.6550	21.65	0.9457	21.27
0.3271	22.54	0.6713	21.63		
0.4586	22.06	0.7676	21.50		
		Octane + 1-Hexanol			
0.0515	25.19	0.4619	22.48	0.7478	21.65
0.0944	24.79	0.4987	22.32	0.7794	21.58
0.1600	24.22	0.5049	22.30	0.8504	21.45
0.1837	24.03	0.5984	22.02	0.8950	21.38
0.2785	23.38	0.6717	21.83	0.9590	21.25
0.3309	23.08	0.7019	21.74		
		Octane + 1-Heptanol			
0.0995	25.35	0.4567	22.79	0.7830	21.68
0.1699	24.68	0.4918	22.65	0.7981	21.63
0.1767	24.63	0.5587	22.38	0.8409	21.53
0.2957	23.72	0.5956	22.24	0.9111	21.38
0.3286	23.52	0.6564	22.04	0.9580	21.28
0.3530	23.37	0.6626	22.01		
		Octane + 1-Octanol			
0.0641	26.39	0.3554	23.84	0.6897	22.12
0.1200	25.83	0.4526	23.23	0.7547	21.88
0.1378	25.59	0.4950	23.00	0.8114	21.70
0.1501	25.50	0.5413	22.76	0.8617	21.55
0.2923	24.29	0.6269	22.38	0.8832	21.51
0.3168	24.11	0.6566	22.24		

increase faster than those with octane, and both increase faster than that with DBE. The equimolar value of the

Table 4. Refractive Indexes {(*x*)Octane + (1 − *x*)1-Alkanol} at *T* = 298.15 K

x	n_D	*x*	n_D	*x*	n_D
		Octane + Ethanol			
0.0686	1.3641	0.3571	1.3792	0.6457	1.3880
0.1132	1.3670	0.4544	1.3827	0.6690	1.3885
0.1721	1.3705	0.4898	1.3839	0.7650	1.3906
0.1834	1.3711	0.5401	1.3853	0.8191	1.3917
0.2867	1.3762	0.5865	1.3866	0.9095	1.3934
0.3500	1.3789	0.6299	1.3877	0.9212	1.3939
		Octane + 1-Propanol			
0.0237	1.3835	0.3547	1.3885	0.6938	1.3921
0.1230	1.3852	0.4493	1.3896	0.7592	1.3927
0.1859	1.3862	0.5154	1.3903	0.7884	1.3930
0.2349	1.3869	0.5218	1.3904	0.8449	1.3935
0.2900	1.3877	0.6224	1.3914	0.8918	1.3940
0.3454	1.3884	0.6308	1.3915	0.9245	1.3943
		Octane + 1-Butanol			
0.0453	1.3970	0.3526	1.3955	0.6918	1.3948
0.1130	1.3966	0.4467	1.3952	0.7770	1.3948
0.1636	1.3964	0.4857	1.3951	0.8167	1.3948
0.1788	1.3963	0.5747	1.3949	0.8650	1.3948
0.2956	1.3957	0.6326	1.3948	0.9376	1.3949
		Octane + 1-Pentanol			
0.0189	1.4075	0.4586	1.4003	0.7676	1.3968
0.1150	1.4058	0.4927	1.3999	0.7932	1.3966
0.1459	1.4052	0.5188	1.3996	0.8340	1.3963
0.1629	1.4049	0.5951	1.3986	0.8869	1.3958
0.2727	1.4030	0.6550	1.3979	0.9457	1.3955
0.3271	1.4022	0.6713	1.3977		
		Octane + 1-Hexanol			
0.0515	1.4147	0.4619	1.4049	0.7478	1.3992
0.0944	1.4136	0.4987	1.4040	0.7794	1.3987
0.1600	1.4119	0.5049	1.4039	0.8504	1.3974
0.1837	1.4113	0.5984	1.4020	0.8950	1.3966
0.2785	1.4091	0.6717	1.4007	0.9590	1.3956
0.3311	1.4079	0.7019	1.4000		
		Octane + 1-Heptanol			
0.0616	1.4205	0.3530	1.4119	0.6629	1.4034
0.0995	1.4194	0.4567	1.4091	0.7830	1.4002
0.1699	1.4174	0.4918	1.4081	0.7981	1.3999
0.1767	1.4170	0.5587	1.4062	0.8409	1.3988
0.2957	1.4136	0.5956	1.4051	0.9111	1.3971
0.3286	1.4126	0.6564	1.4036		
		Octane + 1-Octanol			
0.0641	1.4257	0.4950	1.4114	0.8114	1.4012
0.1378	1.4231	0.5413	1.4100	0.8617	1.3995
0.2923	1.4180	0.6269	1.4072	0.8832	1.3989
0.3168	1.4172	0.6566	1.4062	0.9635	1.3961
0.3554	1.4159	0.6897	1.4051		
0.4526	1.4128	0.7547	1.4030		

mixtures with octane and DBE approaches as the carbon chain of the 1-alkanol increases. It seems that n_D is sensitive not only to the length of the carbon chain but also to the distribution of the functional groups in the molecules.

The density, surface tension, and refractive index were fitted to a polynomial equation,

$$Q = Q_2 + \sum_{i=0}^{n} A_i x^{i+1} \qquad (1)$$

where $Q = \rho$, σ, or n_D; $Q_2 = \rho_2$, σ_2, or n_{D2}, the value of the corresponding magnitude for the pure 1-alkanol (given in Table 1); A_i are the fitting parameters, and x is the mole fraction of the octane. The obtained A_i values of the different magnitudes for all measured mixtures are plot in Table 5. In eq 1 we have included a constraint to ensure that the value of Q for the pure octane ($x = 1$) is that given in Table 1.

From the measured density, surface tension, and refractive index data for the binary mixtures we have calculated,

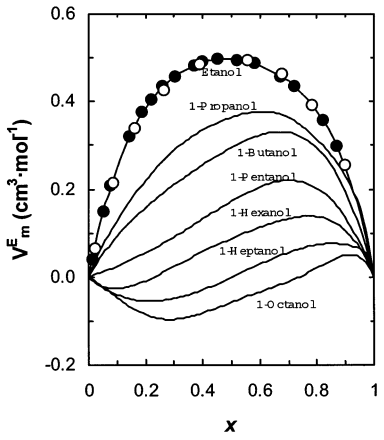

Figure 3. Experimental excess molar volumes V_m^E at $T = 298.15$ K: ●, {(*x*)octane + (1 − *x*)ethanol}; ○, {(*x*)octane + (1 − *x*)ethanol} from ref 16; {(*x*)octane + (1 − *x*)1-propanol} from ref 2; {(*x*)hexane + (1 − *x*)1-butanol} from ref 1; {(*x*)hexane + (1 − *x*)1-pentanol} from ref 3; {(*x*)hexane + (1 − *x*)1-hexanol} from ref 1; {(*x*)hexane + (1 − *x*)1-heptanol} from ref 4; {(*x*)hexane + (1 − *x*)1-octanol} from ref 1.

Table 5. Parameters A_i and A_k of Eqs 1 and 2 and Standard Deviations *s*

	eq	A_0	A_1	A_2	A_3	*s*
			Octane + Ethanol			
ρ/g·cm^{-3}	1	−0.2278	0.3127	−0.2605	0.0890	0.0014
V_m^E/cm^3·mol^{-1}	2	1.9732	−0.0589	1.1792	0.0665	0.0032
σ/mN·m^{-1}	1	−5.3201	10.4214	−9.0506	3.3808	0.026
$\delta\sigma$/mN·m^{-1}	2	−2.7144	1.3608	−1.2146		0.014
n_D	1	0.0746	−0.0609	0.0221		0.00008
Δn_D	2	0.0278	−0.011			0.00008
			Octane + 1-Propanol			
σ/mN·m^{-1}	1	−10.1214	18.5392	−15.9499	5.3137	0.013
$\delta\sigma$/mN·m^{-1}	2	−3.8874	2.7263	−1.5478		0.011
n_D	1	0.0196	−0.0135	0.0060		0.00002
Δn_D	2	0.0044	−0.0030			0.00002
			Octane + 1-Butanol			
σ/mN·m^{-1}	1	−8.6237	9.4627	−3.6872		0.010
$\delta\sigma$/mN·m^{-1}	2	−3.9066	1.8816	−0.2023		0.008
n_D	1	−0.0062	0.0027	0.0014		0.00004
Δn_D	2	−0.0048	−0.0008			0.00004
			Octane + 1-Pentanol			
σ/mN·m^{-1}	1	−10.9247	12.3626	−5.2361		0.010
$\delta\sigma$/mN·m^{-1}	2	−4.4988	2.6096			0.011
n_D	1	−0.0202	0.0074			0.00005
Δn_D	2	−0.0074	−0.0002			0.00005
			Octane + 1-Hexanol			
σ/mN·m^{-1}	1	−10.9640	10.1879	−3.7823		0.007
$\delta\sigma$/mN·m^{-1}	2	−4.5068	1.9171			0.007
n_D	1	−0.0264	0.0056			0.00010
Δn_D	2	−0.0057	−0.0011			0.00007
			Octane + 1-Heptanol			
σ/mN·m^{-1}	1	−11.9625	10.2390	−3.5749		0.019
$\delta\sigma$/mN·m^{-1}	2	−4.8626	1.9661			0.013
n_D	1	−0.0311	0.0038			0.00009
Δn_D	2	−0.0038	−0.0010			0.00006
			Octane + 1-Octanol			
σ/mN·m^{-1}	1	−11.9058	8.3933	−2.4730	0.0271	0.021
$\delta\sigma$/mN·m^{-1}	2	−4.7334	1.3790			0.015
n_D	1	−0.0323				0.00010

respectively, the corresponding excess molar volumes, V_m^E (this only for {(*x*)octane + (1 − *x*)ethanol}), surface tension deviations, $\delta\sigma$, and changes in the refractive index, Δn_D, using the usual expressions.[7] These results were fitted to a Redlich−Kister type equation[5] of the form

$$Q^E = x(1-x)\sum_{k=0}^{N} A_k(2x-1)^k$$

where Q^E represents V_m^E; $\delta\sigma$, or Δn_D; x is the mole fraction

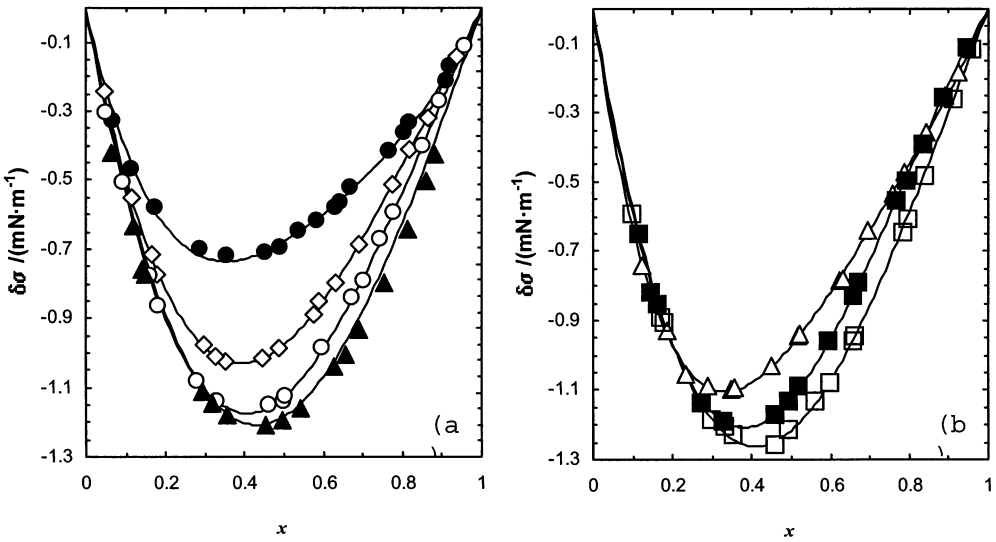

Figure 4. Surface tension deviations at $T = 298.15$ K: (a) ●, {(x)hexane + (1 − x)ethanol}; ◇, {(x)hexane + (1 − x)1-butanol}; ○, {(x)-hexane + (1 − x)1-hexanol}; ▲, {(x)hexane + (1 − x)1-octanol}; (b) △, {(x)hexane + (1 − x)1-propanol}; ■, {(x)hexane + (1 − x)1-pentanol}; □, {(x)hexane + (1 − x)1-heptanol}.

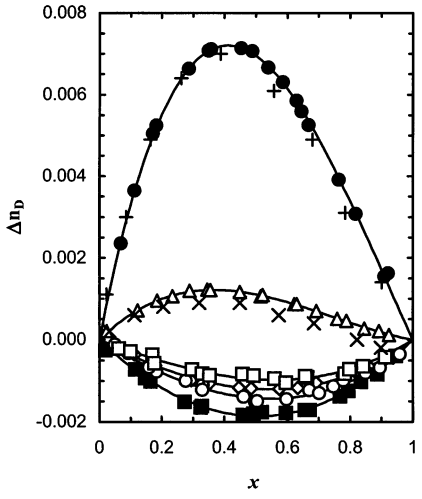

Figure 5. Changes of refractive index: ●, {(x)hexane + (1 − x)-ethanol}; +, {(x)hexane + (1 − x)ethanol} from ref 16; △, {(x)-hexane + (1 − x)1-propanol}; ×, {(1 − x)hexane + (1 − x)1-propanol} from ref 16; ◇, {(x)hexane + (1 − x)1-butanol}; ■, {(x)hexane + (1 − x)1-pentanol}; ○, {(x)hexane + (1 − x)1-hexanol}; □, {(x)hexane + (1 − x)1-heptanol} at $T = 298.15$ K.

of octane; A_k are parameters obtained by the unweighted least-squares method; and their number was determined using an f-test.[15] The parameters A_i are also shown in Table 5. Figure 3 shows the excess molar volumes presented in this work besides the other ones obtained from the literature.[1−4] The results show that the excess molar volumes of {(x)octane + (1 − x)1-alkanol} decrease as the chain length of 1-alkanol increases. In the case of {(x)octane + (1 − x)ethanol}, V_m^E's are positive and the maximum is slightly skewed toward low mole fractions of octane. This curve is very similar to that previously published for the same mixture,[16] which is included in Figure 3.

Figure 4 shows the surface tension deviations, $\delta\sigma$, for the seven binary mixtures measured. For clarity we have divided the figure in two: in Figure 4a we plot the mixtures with alkanols containing an even number of carbons, and in Figure 4b we plot those alkanols with an odd number of carbons. To our knowledge it is the first time than $\delta\sigma$ data for the studied mixtures are published, and so we cannot compare them with any previous data. From this figure we observe that $\delta\sigma$ decreases with the length of the alkanol for both even and odd number of carbons independently. But if we observe all of them, that decrease does not happen. The cohesion forces between the molecules of octane and alkanol get weaker as the length of the alkanol increases, but this effect is more important for those alkanol molecules with an odd number of atoms.

Figure 5 presents the obtained changes in the refractive index on mixing, Δn_D. All curves are negative except those for the mixtures containing ethanol and 1-propanol, which are positive. The value of Δn_D decreases when the 1-alkanol length increases from ethanol to 1-pentanol, and for longer 1-alkanols the absolute value of Δn_D gets lower and is null for {octane + 1-octanol} (not presented in Figure 5). If we compare our measured Δn_D with data previously published in the literature for {octane + ethanol}[16] and {octane + 1-propanol},[16] we observe in Figure 5 that they are similar in absolute value and curve shape.

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