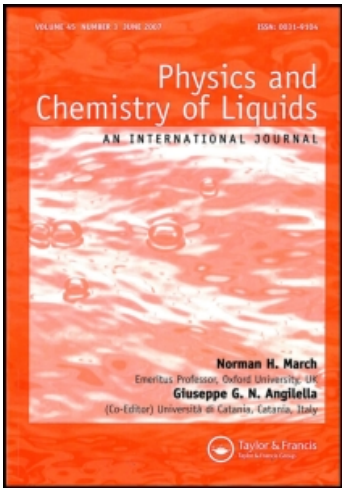

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Refractive indices and molar refractions for isomeric chlorobutanes with isomeric butanols

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Experimental refractive indices and molar refractions for the binary mixtures of each of the isomers of chlorobutane with each of the isomers of butanol are given at 298.15 K. From these data the refractive index deviations were calculated. The experimental refractive index results were compared with those predicted by several mixing rules: Lorentz–Lorenz, Gladstone–Dale, Arago–Biot, Wiener and Heller.

Keywords: Refractive index; Chlorobutane; Butanol

1. Introduction

The study of different thermophysical properties of a liquid mixture provides complementary information about the mixing process and about interactions between their pure components. Following previous studies on thermodynamic and transport properties of binary mixtures involving chlorobutane and butanol isomers [1–8], we report here refractive indices and molar refractions for the binary mixtures of each of the isomers of chlorobutane with each of the isomers of butanol at 298.15 K. A survey of the literature shows that there is one reference for 1-chlorobutane with 2-butanol at 313.15 K (at $\lambda = 435.8$ nm, mercury arc) [9] and another for 1-chlorobutane with 1-butanol at 298.15 K [10]. Our results show that the refractive index behaviour is strongly influenced by the butanol structure.

Finally, we have used several mixing rules for the prediction of the refractive indices, these mixing rules were: Lorentz–Lorenz [11,12], Gladstone–Dale [13], Arago–Biot [14], Wiener [15] and Heller [16].

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2. Experimental

The liquids used were: 1-butanol (>99.8%), 2-methyl-1-propanol, 2-methyl-2-propanol (>99.5%), 1-chlorobutane, 2-chlorobutane, 2-methyl-2-chloropropane, and 2-butanol (>99%) provided by Aldrich as well as 2-methyl-1-chloropropane (>99%) obtained from Fluka. The liquids were used without further purification, although the isomeric butanols were dried with activated molecular sieve type 0.3 nm from Merck.

The refractive indices were measured using a high-precision automatic refractometer Abbemat-HP DR Kernchen. The sample to be measured was placed on the polished surface of a prism made of synthetic sapphire. A cone-shaped yellow light beam of 589.3 nm sodium D wavelength illuminated the sample from below, under different angles of reflection. Using reflected light has the advantage that the colour and cloudiness of the sample do not significantly influence the result. The temperatures of the sample and of the prism were controlled within ± 0.002 K by a built-in Peltier device, a second Peltier thermostat is used to keep the temperature of internal refractometer components constant, in order to increase the precision and accuracy of the measurements. The apparatus was calibrated with twice deionized distilled water. The reproducibility of the measurements is $\pm 1 \times 10^{-6}$ and the corresponding accuracy is $\pm 2 \times 10^{-5}$.

The comparison between measured refractive indices and literature values [17] for the pure compounds are given in table 1.

The densities, ρ, employed to calculate the molar refractions were measured using an Anton Paar DMA-58 vibrating tube densimeter in which temperature is controlled automatically within ± 0.01 K. The apparatus was calibrated with twice deionized distilled water and dry air. The precision of density measurements is $\pm 5 \times 10^{-6}$ g cm^{-3} and the accuracy of these measurements after proper calibration is $\pm 1 \times 10^{-5}$ g cm^{-3}. Densities of the pure compounds at 298.15 K are shown in table 2.

The mixtures were prepared by weight using a Mettler H20T balance. The maximum estimated error in the mole fraction is $\pm 1 \times 10^{-4}$.

3. Results and discussion

The refractive indices of the studied binary mixtures at the temperature of 298.15 K are gathered in table 2. From refractive indices of the mixtures, n_D, and their corresponding densities the molar refractions, R, can be calculated by using the

Table 1. Experimental and literature refractive indices, n_D, of the pure compounds and their experimental densities, ρ.

Compound	ρ/g cm^{-3}	$n_{D,\text{exptl}}$	$n_{D,\text{lit}}$
1-Chlorobutane	0.88047	1.399511	1.4001
2-Chlorobutane	0.86743	1.394061	1.3941
2-Methyl-1-chloropropane	0.87105	1.395410	1.3951
2-Methyl-2-chloropropane	0.83678	1.382275	1.3828
1-Butanol	0.80575	1.397336	1.3973
2-Butanol	0.80241	1.395164	1.3949
2-Methyl-1-propanol	0.79784	1.393762	1.3939
2-Methyl-2-propanol	0.78100	1.384904	1.3851

Table 2. Refractive indices, n_D, refractive index deviations, Δn_D, and molar refractions, R, of the binary mixtures.

ϕ_1	n_D	$R/cm^3\,mol^{-1}$	Δn_D	ϕ_1	n_D	$R/cm^3\,mol^{-1}$	Δn_D
			1-Chlorobutane + 1-butanol				
0.0726	1.397548	22.384	0.000054	0.6399	1.398331	24.177	−0.000397
0.1093	1.397641	22.492	0.000067	0.7316	1.398460	24.494	−0.000467
0.2182	1.397865	22.819	0.000054	0.8166	1.398644	24.793	−0.000468
0.3301	1.398021	23.164	−0.000033	0.8330	1.398695	24.852	−0.000453
0.4254	1.398122	23.467	−0.000139	0.9645	1.399256	25.326	−0.000178
0.5290	1.398218	23.805	−0.000269				
			1-Chlorobutane + 2-butanol				
0.0665	1.395207	22.349	−0.000246	0.6311	1.396171	24.146	−0.001736
0.1193	1.395217	22.507	−0.000466	0.7303	1.396680	24.488	−0.001659
0.2236	1.395261	22.827	−0.000875	0.8127	1.397251	24.779	−0.001446
0.3330	1.395357	23.170	−0.001255	0.9084	1.398131	25.121	−0.000982
0.4319	1.395535	23.487	−0.001506	0.9488	1.398636	25.267	−0.000653
0.5325	1.395799	23.816	−0.001680				
			1-Chlorobutane + 2-methyl-1-propanol				
0.0683	1.394108	22.407	−0.000046	0.6329	1.396603	24.171	−0.000797
0.1244	1.394390	22.572	−0.000087	0.7274	1.397087	24.490	−0.000857
0.1978	1.394731	22.792	−0.000168	0.8192	1.397697	24.811	−0.000774
0.3306	1.395279	23.198	−0.000383	0.9095	1.398401	25.128	−0.000590
0.4327	1.395704	23.519	−0.000546	0.9468	1.398781	25.262	−0.000424
0.5376	1.396154	23.856	−0.000699				
			1-Chlorobutane + 2-methyl-2-propanol				
0.0685	1.385132	22.456	−0.000773	0.6213	1.391381	24.186	−0.002598
0.1059	1.385313	22.568	−0.001138	0.7171	1.393013	24.502	−0.002365
0.2209	1.386171	22.919	−0.001960	0.8203	1.395001	24.849	−0.001886
0.3278	1.387274	23.250	−0.002419	0.8998	1.396741	25.116	−0.001307
0.4268	1.388520	23.561	−0.002618	0.9554	1.398162	25.304	−0.000698
0.5244	1.389888	23.872	−0.002676				
			2-Chlorobutane + 1-butanol				
0.0672	1.397230	22.370	0.000114	0.6379	1.395174	24.205	−0.000073
0.1173	1.397144	22.520	0.000192	0.7397	1.394720	24.564	−0.000193
0.2387	1.396824	22.889	0.000270	0.8179	1.394416	24.848	−0.000241
0.2995	1.396620	23.080	0.000265	0.9015	1.394156	25.158	−0.000228
0.4405	1.396061	23.535	0.000168	0.9439	1.394077	25.318	−0.000168
0.5364	1.395639	23.855	0.000060				
			2-Chlorobutane + 2-butanol				
0.0510	1.394924	22.304	−0.000184	0.6449	1.392995	24.230	−0.001458
0.1018	1.394713	22.456	−0.000339	0.7390	1.392945	24.563	−0.001404
0.2348	1.394152	22.867	−0.000753	0.8154	1.393016	24.839	−0.001249
0.3389	1.393740	23.199	−0.001050	0.9111	1.393308	25.193	−0.000851
0.4398	1.393432	23.530	−0.001247	0.9434	1.393501	25.314	−0.000622
0.5371	1.393160	23.856	−0.001412				
			2-Chlorobutane + 2-methyl-1-propanol				
0.0663	1.393832	22.406	0.000050	0.6351	1.393488	24.219	−0.000464
0.1216	1.393858	22.571	0.000060	0.7265	1.393437	24.536	−0.000542
0.2261	1.393847	22.889	0.000017	0.8228	1.393445	24.876	−0.000563
0.3359	1.393772	23.233	−0.000090	0.8943	1.393547	25.135	−0.000482
0.4342	1.393682	23.549	−0.000210	0.9481	1.393710	25.334	−0.000335
0.5469	1.393565	23.921	−0.000361				
			2-Chlorobutane + 2-methyl-2-propanol				
0.0659	1.384798	22.448	−0.000709	0.6281	1.388205	24.245	−0.002451
0.1156	1.384821	22.600	−0.001142	0.7638	1.389847	24.709	−0.002051

(*continued*)

Table 2. Continued.

ϕ_1	n_D	R/cm^3 mol^{-1}	Δn_D	ϕ_1	n_D	R/cm^3 mol^{-1}	Δn_D
0.2245	1.385098	22.938	−0.001861	0.8100	1.390479	24.869	−0.001842
0.3258	1.385594	23.257	−0.002294	0.9106	1.392050	25.217	−0.001192
0.3963	1.386052	23.481	−0.002481	0.9540	1.392948	25.372	−0.000692
0.5297	1.387197	23.917	−0.002558				
			2-Methyl-1-chloropropane + 1-butanol				
0.0666	1.397321	22.369	0.000113	0.6302	1.395881	24.170	−0.000241
0.1200	1.397269	22.528	0.000164	0.7299	1.395579	24.521	−0.000351
0.2249	1.397090	22.849	0.000187	0.8185	1.395341	24.839	−0.000419
0.3352	1.396821	23.195	0.000131	0.9066	1.395220	25.162	−0.000370
0.4445	1.396500	23.547	0.000020	0.9452	1.395241	25.305	−0.000274
0.5407	1.396186	23.866	−0.000109				
			2-Methyl-1-chloropropane + 2-butanol				
0.0623	1.395025	22.342	−0.000154	0.6294	1.393665	24.168	−0.001654
0.1179	1.394787	22.508	−0.000406	0.7299	1.393724	24.524	−0.001620
0.2282	1.394408	22.848	−0.000812	0.8169	1.393955	24.840	−0.001410
0.3354	1.394096	23.188	−0.001151	0.8983	1.394318	25.136	−0.001067
0.4303	1.393870	23.495	−0.001400	0.9489	1.394721	25.320	−0.000676
0.5417	1.393708	23.866	−0.001589				
			2-Methyl-1-chloropropane + 2-methyl-1-propanol				
0.0656	1.393895	22.398	0.000025	0.6350	1.394208	24.201	−0.000600
0.1142	1.393965	22.541	0.000015	0.7268	1.394261	24.519	−0.000699
0.2241	1.394073	22.874	−0.000058	0.8118	1.394387	24.822	−0.000713
0.3355	1.394142	23.222	−0.000173	0.8948	1.394650	25.124	−0.000587
0.4352	1.394150	23.539	−0.000329	0.9495	1.394939	25.322	−0.000388
0.5265	1.394164	23.837	−0.000466				
			2-Methyl-1-chloropropane + 2-methyl-2-propanol				
0.0670	1.384882	22.451	−0.000726	0.6228	1.388895	24.209	−0.002552
0.1178	1.384962	22.606	−0.001180	0.7215	1.390179	24.546	−0.002305
0.2250	1.385336	22.936	−0.001932	0.8157	1.391556	24.873	−0.001918
0.3186	1.385910	23.227	−0.002341	0.9113	1.393196	25.202	−0.001282
0.4169	1.386686	23.536	−0.002598	0.9543	1.394118	25.348	−0.000812
0.5353	1.387879	23.918	−0.002649				
			2-Methyl-2-chloropropane + 1-butanol				
0.0612	1.396687	22.358	0.000273	0.6394	1.388405	24.318	0.000699
0.1230	1.395994	22.550	0.000510	0.7366	1.386751	24.689	0.000508
0.2330	1.394619	22.900	0.000792	0.8326	1.385081	25.067	0.000284
0.3417	1.393122	23.260	0.000932	0.9097	1.383763	25.382	0.000129
0.4394	1.391666	23.596	0.000948	0.9558	1.382995	25.574	0.000054
0.5441	1.390000	23.967	0.000858				
			2-Methyl-2-chloropropane + 2-butanol				
0.0626	1.394320	22.347	−0.000037	0.6319	1.386185	24.291	−0.000835
0.1231	1.393475	22.537	−0.000102	0.7379	1.384816	24.696	−0.000837
0.2330	1.391923	22.895	−0.000238	0.8235	1.383791	25.035	−0.000758
0.3432	1.390264	23.261	−0.000476	0.8952	1.383003	25.323	−0.000623
0.4370	1.388868	23.583	−0.000664	0.9546	1.382489	25.568	−0.000371
0.5533	1.387229	23.999	−0.000803				
			2-Methyl-2-chloropropane + 2-methyl-1-propanol				
0.0586	1.393266	22.388	0.000177	0.6359	1.386679	24.329	0.000221
0.1748	1.392144	22.750	0.000390	0.7210	1.385565	24.647	0.000085
0.2363	1.391504	22.949	0.000457	0.8229	1.384249	25.040	−0.000060
0.3420	1.390320	23.298	0.000486	0.9068	1.383219	25.375	−0.000126
0.4353	1.389215	23.616	0.000453	0.9636	1.382598	25.608	−0.000095
0.5405	1.387891	23.984	0.000337				

(*continued*)

Table 2. Continued.

ϕ_1	n_D	R/cm^3 mol^{-1}	Δn_D	ϕ_1	n_D	R/cm^3 mol^{-1}	Δn_D
2-Methyl-2-chloropropane + 2-methyl-2-propanol							
0.0617	1.384161	22.444	−0.000581	0.6305	1.381020	24.358	−0.002226
0.1102	1.383650	22.599	−0.000964	0.7239	1.381011	24.699	−0.001990
0.2342	1.382564	22.998	−0.001724	0.8129	1.381113	25.031	−0.001654
0.3355	1.381917	23.333	−0.002105	0.9087	1.381429	25.394	−0.001086
0.4396	1.381426	23.685	−0.002322	0.9450	1.381645	25.535	−0.000775
0.5336	1.381155	24.012	−0.002346				

Lorentz–Lorenz relation:

$$R = \frac{n_D^2 - 1}{n_D^2 + 2} V_m \tag{1}$$

where V_m is the molar volume of the mixture. The molar refractions for the studied mixtures are given in table 2.

Fialkov and Fenerly [18] and Fialkov [19] stated that the refractive index is an additive property of the pure components when the composition is expressed in volume fraction. Following these suggestions we have calculated the refractive index deviation, Δn_D, by means of the following equation:

$$\Delta n_D = n_D - \phi_1 n_{D,1} - \phi_2 n_{D,2} \tag{2}$$

where n_D is the refractive index of the mixture, ϕ_i and $n_{D,i}$ are the volume fraction referred to the unmixed state and the refractive index of component i respectively. The values of refractive index deviations are also collected in table 2 and they are graphically represented in figures 1–4.

The refractive index deviations were correlated by means of a Redlich–Kister type equation in ϕ_i [20]:

$$\Delta n_D = \phi_1 \phi_2 \sum_{p=0}^{r} A_p (\phi_1 - \phi_2)^p \tag{3}$$

where A_p are adjustable parameters determined by the method of least-squares. The values of these parameters are given in table 3 together with the standard deviations.

Our experimental data for 1-chlorobutane with 1-butanol show an average absolute deviation of 0.0002 with respect to those of Chen *et al.* [10].

For a given isomer of chlorobutane, Δn_D increases in the sequence: 2-methyl-2-propanol < 2-butanol < 2-methyl-1-propanol < 1-butanol, while if we consider a given isomer of butanol Δn_D increases in the sequence: 1-chloropropane < 2-methyl-1-chloropropane < 2-chloropropane < 2-methyl-2-chloropropane. Refractive index deviations for the mixtures containing 1-butanol or 2-methyl-1-propanol show sigmoid curves except for the mixture 2-methyl-2-chloropropane that presents positive Δn_D values

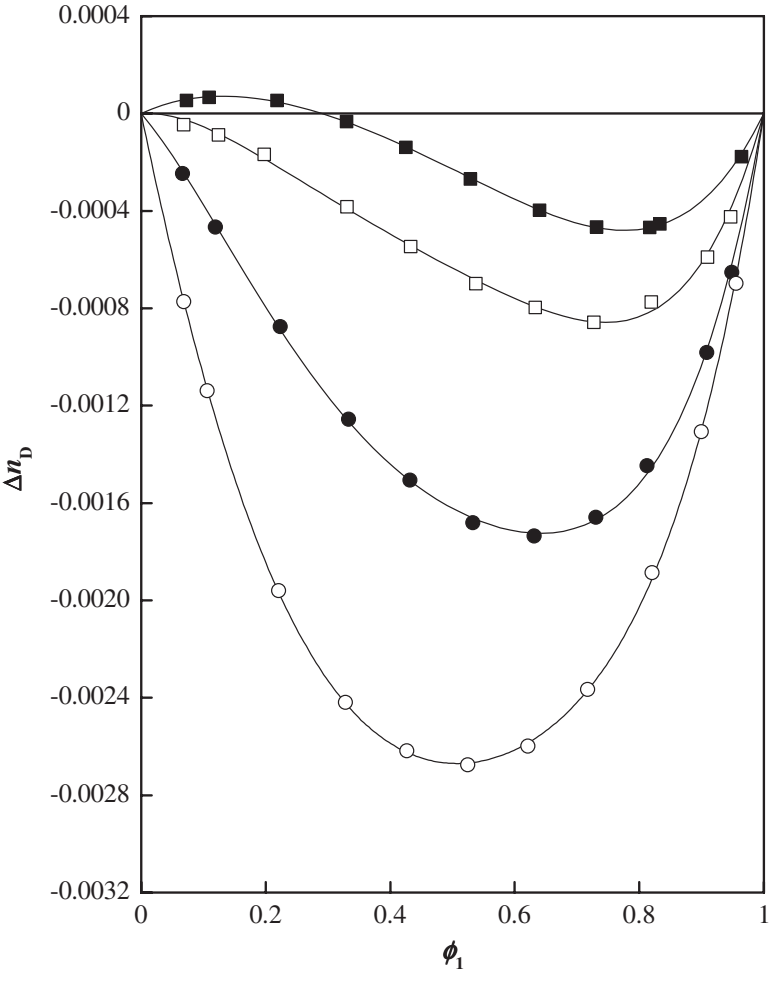

Figure 1. Refractive index deviation, Δn_D, for 1-chlorobutane (1) + isomeric butanols (2) as a function of volume fraction ϕ_1: 1-butanol (■); 2-butanol (●); 2-methyl-1-propanol (□); 2-methyl-2-propanol (○).

over the whole composition range. On the other hand, for the mixtures containing 2-butanol or 2-methyl-2-propanol Δn_D values are negative although the mixture 2-methyl-2-chloropropane with 2-butanol which shows slightly positive Δn_D values at very high volume fractions of butanol. As the experimental results show, we can remark the strong influence of the isomeric butanol in the refractive index deviation.

Δn_D values for these mixtures show a similar trend, although with opposite sign, as their corresponding excess molar volumes [1–3,7]. According to Brocos *et al.* [21], refractive index deviations can be interpreted as a sign-reversed measure of the deviation of reduced free volume, and hence of changes in intermolecular interactions. For these systems, and as also confirmed by our previous studies on different thermophysical properties [1–8], the main molecular interactions are the breaking of the multimeric structure of the butanols during the mixing process.

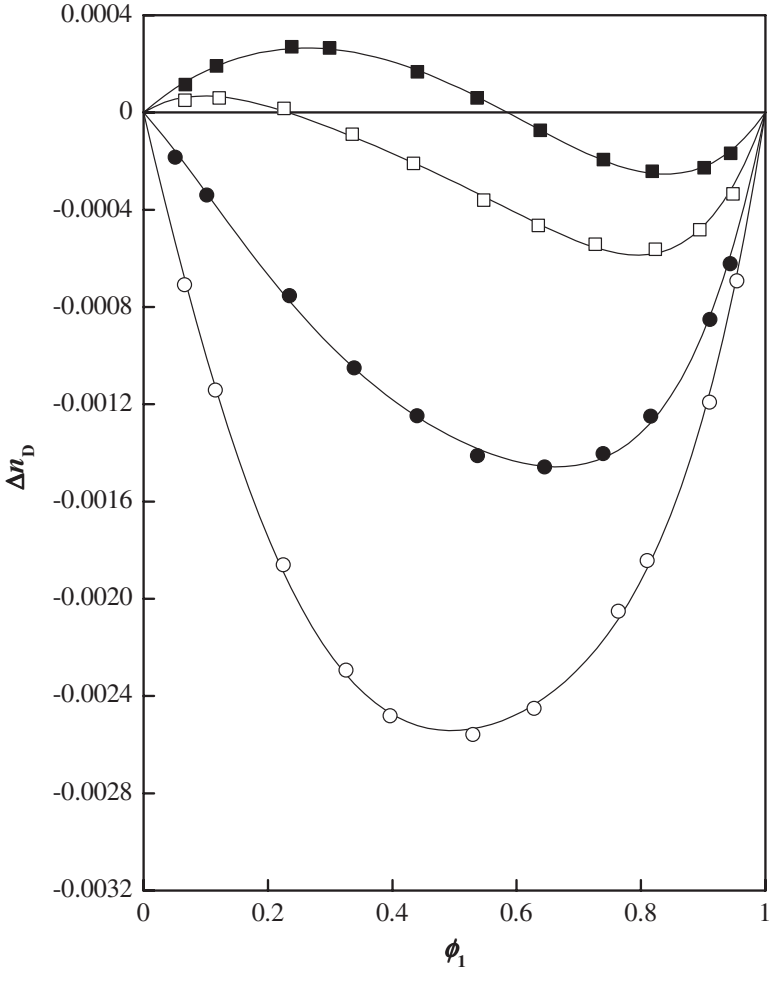

Figure 2. Refractive index deviation, Δn_D, for 2-chlorobutane (1) + isomeric butanols (2) as a function of volume fraction ϕ_1: 1-butanol (■); 2-butanol (●); 2-methyl-1-propanol (□); 2-methyl-2-propanol (○).

4. Refractive index mixing rules

The refractive index of a liquid mixture can be predicted from its density together with the refractive indices and densities of the pure components by using a mixing rule.

In this work the experimental refractive index were compared with those predicted by several mixing rules.

$$\text{Lorentz–Lorenz:} \quad \left(\frac{n_D^2 - 1}{n_D^2 + 2}\right) = \frac{w_1\rho}{\rho_1}\left(\frac{n_{D,1}^2 - 1}{n_{D,1}^2 + 2}\right) + \frac{w_2\rho}{\rho_2}\left(\frac{n_{D,2}^2 - 1}{n_{D,2}^2 + 2}\right) \tag{4}$$

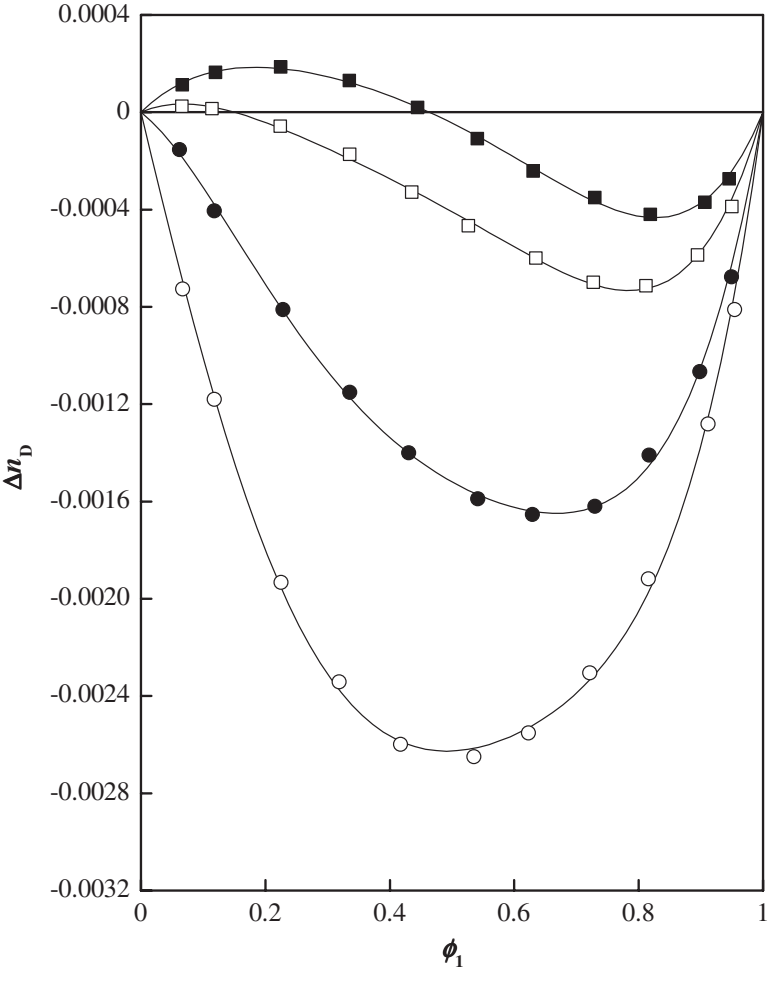

Figure 3. Refractive index deviation, Δn_D, for 2-methyl-1-chloropropane (1) + isomeric butanols (2) as a function of volume fraction ϕ_1: 1-butanol (■); 2-butanol (●); 2-methyl-1-propanol (□); 2-methyl-2-propanol (○).

$$\text{Gladstone–Dale:} \quad (n_D - 1) = \frac{w_1\rho}{\rho_1}(n_{D,1} - 1) + \frac{w_2\rho}{\rho_2}(n_{D,2} - 1) \tag{5}$$

$$\text{Arago–Biot:} \quad n_D = \frac{w_1\rho}{\rho_1}n_{D,1} + \frac{w_2\rho}{\rho_2}n_{D,2} \tag{6}$$

$$\text{Wiener:} \quad \left(\frac{n_D^2 - n_{D,1}^2}{n_D^2 + 2n_{D,1}^2}\right) = \frac{w_2\rho}{\rho_2}\left(\frac{n_{D,2}^2 - n_{D,1}^2}{n_{D,2}^2 + 2n_{D,1}^2}\right) \tag{7}$$

$$\text{Heller:} \quad \left(\frac{n_D^2 - n_{D,1}^2}{n_D^2 + 2n_{D,1}^2}\right) = \frac{3}{2}\frac{w_2\rho}{\rho_2}\left(\frac{m^2 - 1}{m^2 + 2}\right); \quad m = n_{D,2}/n_{D,1} \tag{8}$$

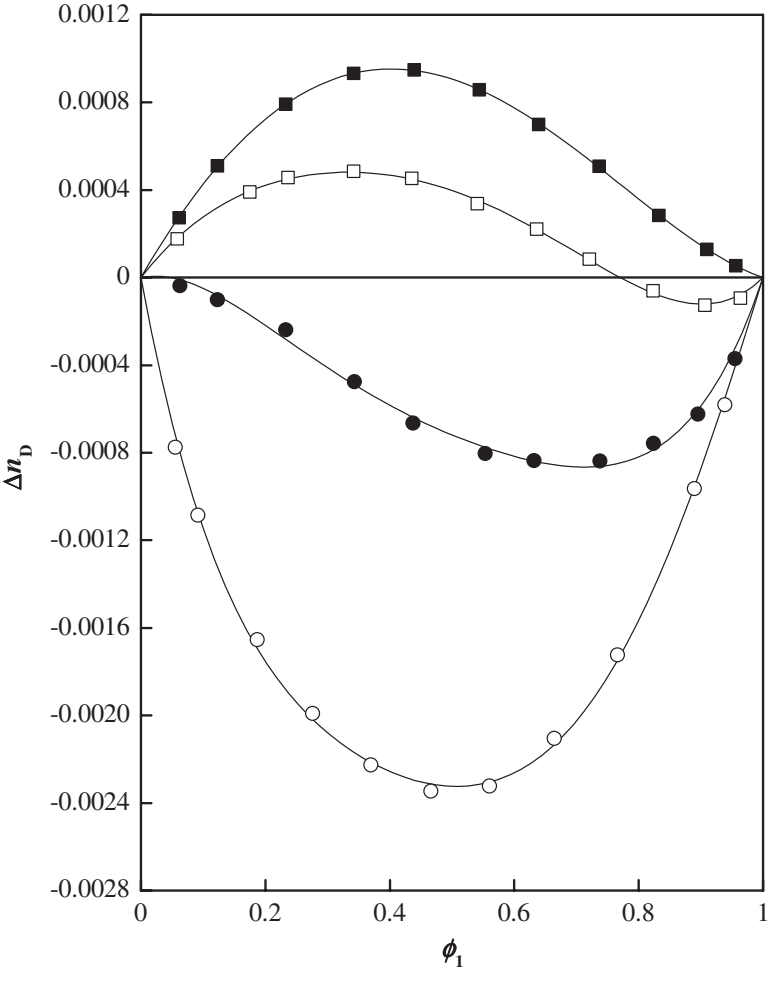

Figure 4. Refractive index deviation, Δn_D, for 2-methyl-2-chloropropane (1) + isomeric butanols (2) as a function of volume fraction ϕ_1: 1-butanol (■); 2-butanol (●); 2-methyl-1-propanol (□); 2-methyl-2-propanol (○).

in these equations ρ is the density of the liquid mixture and w_i and ρ_i are the weight fraction and density of the component i respectively.

The root mean square deviations (RMSD) between experimental and predicted n_D values are given in table 4, the RMSD is defined as:

$$\mathrm{RMSD} = \left(\frac{1}{N} \sum_i \left(n_{D,\,\mathrm{exptl}} - n_{D,\,\mathrm{pred}} \right)^2 \right)^{1/2} \tag{9}$$

where N is the number of experimental data.

A close similarity in the predictions was observed between Wiener and Heller equations as well as between Lorentz–Lorenz and Gladstone–Dale equations. The best

Table 3. Parameters, A_i, and standard deviations, σ, of the Redlich–Kister equation.

System	A_0	A_1	A_2	A_3	σ
1-Chlorobutane +					
1-butanol	−0.000913	−0.002518	−0.001109	−0.000707	0.000029
2-butanol	−0.006487	−0.002754	−0.002074	−0.002845	0.000077
2-methyl-1-propanol	−0.002539	−0.002685	−0.001831	−0.001877	0.000044
2-methyl-2-propanol	−0.010678	−0.000200	−0.003929	−0.002096	0.000086
2-Chlorobutane +					
1-butanol	0.000439	−0.002371	−0.001149	−0.000581	0.000033
2-butanol	−0.005356	−0.002571	−0.002342	−0.002268	0.000072
2-methyl-1-propanol	−0.001144	−0.002488	−0.001687	−0.001931	0.000037
2-methyl-2-propanol	−0.010169	0.000123	−0.003614	−0.002937	0.000204
2-Methyl-1-chloropropane +					
1-butanol	−0.000178	−0.002560	−0.001616	−0.001731	0.000037
2-butanol	−0.006078	−0.002871	−0.002279	−0.003524	0.000083
2-methyl-1-propanol	−0.001635	−0.002842	−0.002167	−0.002074	0.000042
2-methyl-2-propanol	−0.010504	0.000216	−0.004238	−0.004253	0.000099
2-Methyl-2-chloropropane +					
1-butanol	0.003626	−0.001846	−0.000680	−0.000142	0.000067
2-butanol	−0.002875	−0.002352	−0.001023	−0.002159	0.000064
2-methyl-1-propanol	0.001591	−0.001962	−0.001130	−0.001260	0.000044
2-methyl-2-propanol	−0.009293	0.000199	−0.002984	−0.003285	0.000115

Table 4. Root mean square deviations for the Lorentz–Lorentz (LL), Gladstone–Dale (DL), Arago–Biot (AB), Wiener (W) and Heller (H) equations.

System	LL	GD	AB	W	H
1-Chlorobutane +					
1-butanol	0.00729	0.00646	0.02210	0.00032	0.00032
2-butanol	0.00802	0.00722	0.02225	0.00131	0.00131
2-methyl-1-propanol	0.00811	0.00722	0.02402	0.00065	0.00065
2-methyl-2-propanol	0.01046	0.00950	0.02822	0.00229	0.00229
2-Chlorobutane +					
1-butanol	0.00577	0.00509	0.01810	0.00021	0.00021
2-butanol	0.0059	0.00594	0.01843	0.00104	0.00104
2-methyl-1-propanol	0.00671	0.00596	0.02042	0.00035	0.00035
2-methyl-2-propanol	0.00895	0.00815	0.02403	0.00204	0.00204
2-Methyl-1-chloropropane +					
1-butanol	0.00618	0.00546	0.01906	0.00024	0.00024
2-butanol	0.00702	0.00633	0.01942	0.00119	0.00119
2-methyl-1-propanol	0.00716	0.00637	0.02155	0.00047	0.00047
2-methyl-2-propanol	0.00947	0.00862	0.02540	0.00217	0.00217
2-Methyl-2-chloropropane +					
1-butanol	0.00255	0.00220	0.00955	0.00073	0.00073
2-butanol	0.00343	0.00310	0.00981	0.00050	0.00050
2-methyl-1-propanol	0.00354	0.00311	0.01192	0.00039	0.00040
2-methyl-2-propanol	0.00606	0.00557	0.01564	0.00170	0.00170

predictions are obtained with Wiener and Heller mixing rules, these equations are better than all the others, while the predictions with Arago–Biot are very unsatisfactory.

The worst results with all the mixing rules are presented for the mixtures containing 2-methyl-2-propanol.

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