Analysis of thermophysical properties of binary systems containing ethyl acetate and 1-propanol or 1-butanol

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Abstract

The aim of this research is the experimental determination of density, viscosity, refractive index and speed of sound for binary mixtures of the ester ethyl acetate and alcohols. Experimental measurements were carried out for two systems with 1-propanol and 1-butanol at atmospheric pressure and in a temperature range 288.15 - 323.15 K. Results of experimental measurements were further used to determine excess molar volumes, viscosity deviations, refractive index deviations, and excess isentropic compressibility for each investigated mixture. These calculated data were correlated using the empirical Redlich-Kister equation. Positive values of the excess molar volume and isentropic compressibility appear for both analysed systems, while values of viscosity and refractive index deviations are negative. The structure and specific characteristics of different molecules in considered mixtures and determined non-ideal behaviour allow the insight into the possible type of interactions in the mixture, interstitial accommodation and structural effects.

Keywords: density; viscosity; refractive index; speed of sound; deviation properties; molecular interactions.

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1. INTRODUCTION

Density, refractive index, viscosity and speed of sound represent thermophysical properties of a fluid that characterize each pure substance in the liquid state, as well as their mixtures. Knowledge of these properties is very important for design and operation of separation processes and equipment in the chemical industry, calculation of heat exchangers, fluid transport, as well as for the development of new thermodynamic models [1].

Theoretical significance of the investigation of thermodynamic as well as transport properties of liquid mixtures is reflected in extension of the database with new experimental values. Various substances and their mixtures are used in the industry and for many of them literary values of thermodynamic properties and related quantities do not exist. On the other hand, density and the excess molar volume, primarily, but also other deviation properties, are the best indicators of molecular interactions, structural changes and packing effects in mixtures.

Part of our research with ester and alcohol mixtures [2-4] is related to ethyl acetate, ethyl ester of acetic acid. It is widely used in the industry, primarily as a solvent and diluent, being favoured because of its low cost, low toxicity, and agreeable sweet odour. Since it finds use in different industries, from cosmetic and pharmaceutical, industry of paints and coatings, to printing and even food industry for decaffeination of coffee/tea, its worldwide production was already estimated to 1.3 Mt in 2004 with a significant increase in the following years [5].

Ethyl acetate is highly miscible with all organic compounds, such as glycols, ketones, alcohols, and esters. It is most commonly used in mixtures with alcohols, and a combination with 20 % ethanol found application as an excellent solvent for cellulose acetate.

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This paper presents experimental determination of density, refractive index, viscosity and speed of sound for binary mixtures ethyl acetate + 1-propanol and ethyl acetate + 1-butanol. The experiments were performed in a temperature interval of 288.15 - 323.15 K with a 5 K temperature step and at atmospheric pressure. The values obtained by measuring these properties were further used to determine the excess molar volume V^E, viscosity deviation $\Delta\eta$, deviation in the refractive index Δn_D and the excess isentropic compressibility k_s^E. The Redlich-Kister polynomial was used to show the analytical dependence of the experimental data on the mixture composition.

2. EXPERIMENTAL

Information about the purity and manufacturers of used chemicals is presented in Table 1. All chemicals were degassed before preparation of the mixtures and used without further purification.

Table 1. Sample information

Chemical name	CAS number	Source	Initial mass fraction purity
ethyl acetate	141-78-6	Sigma Aldrich, USA	0.998
1-propanol	71-23-8	Merck KGaA, USA	0.995
1-butanol	71-36-3	Merck KGaA, USA	0.995

For the investigated pure components, a comparison with literature sources [6-130] for density, viscosity, refractive index and speed of sound at atmospheric pressure is provided graphically (Figure 1 for ethyl acetate, Figure 2 for 1-propanol and Figure 3 for 1-butanol). The average absolute percentage deviations of experimental from literature data for density, viscosity, refractive index and speed of sound for all three investigated pure substances are 0.06, 2.72, 0.04 and 0.13 %, respectively.

Experimental data at atmospheric pressure were obtained using the Anton Paar (Austria) devices, DMA 5000 densimeter, Stabinger SVM 3000 viscometer and RXA 156 refractometer. The working procedure on each apparatus is described in detail in our previous works [131,132].

Mixtures were prepared gravimetrically on a Mettler AG 204 balance (Mettler Toledo, USA) with the precision of $1\cdot10^{-7}$ kg and the standard uncertainty in mole fraction of $1\cdot10^{-4}$. The combined expanded uncertainty with a 0.95 level of confidence, for measured properties, is within 0.5 kg m^{-3} for density, 0.25 % for viscosity, $7\cdot10^{-4}$ for refractive index and 0.61 m s^{-1} for speed of sound. For mixing properties, the estimated uncertainties are $6\cdot10^{-8}$ m^3 mol^{-1} for excess molar volume, 0.46 % for viscosity deviation, $8.6\cdot10^{-4}$ for refractive index deviation and $0.96\cdot10^{-12}$ Pa^{-1} for excess isentropic compressibility.

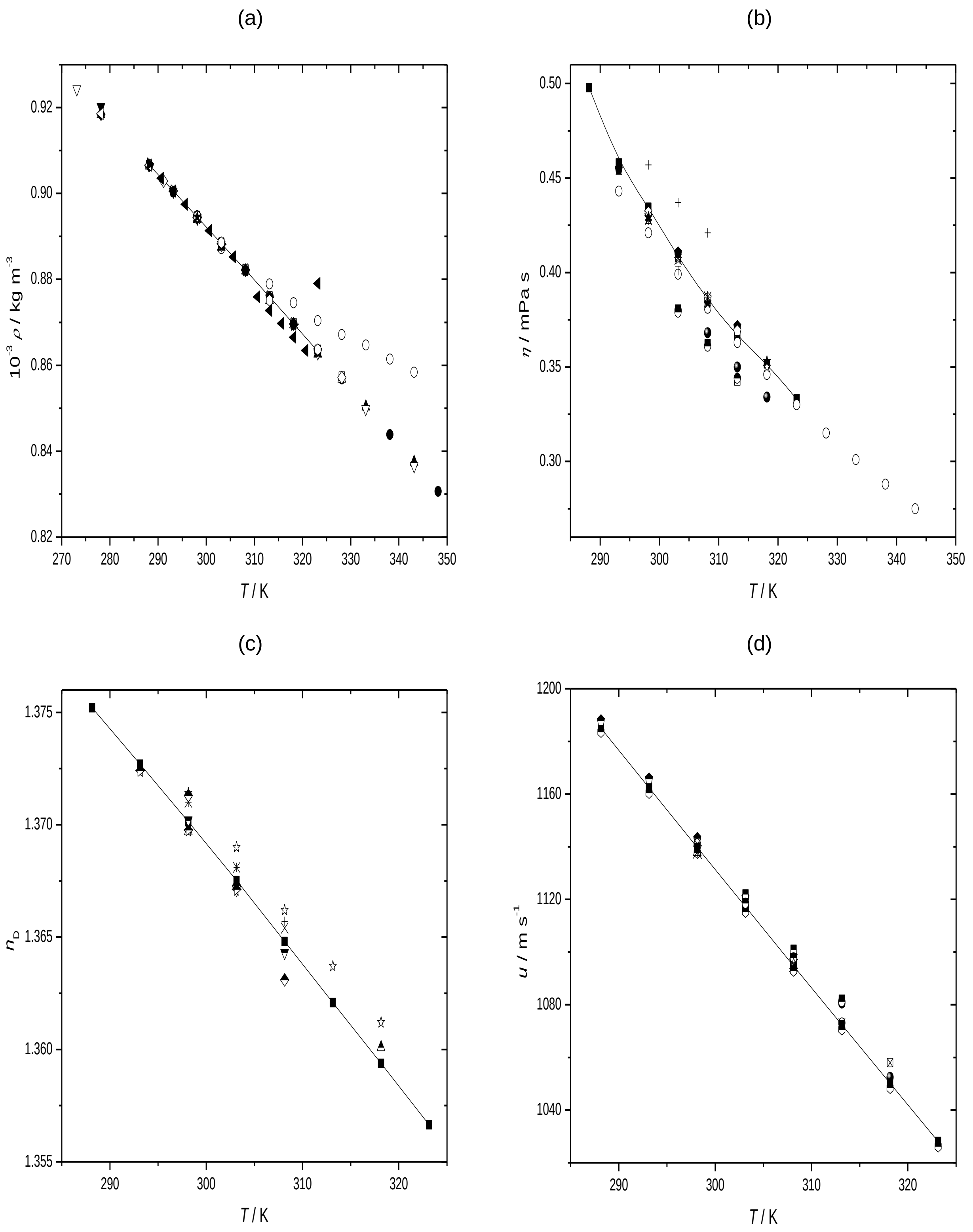

Figure 1. Comparison of (a) density, (b) viscosity, (c) refractive index and (d) speed of sound for ethyl acetate covering the entire investigated temperature range: (■) *this work,* (□) [6], (●) [7], (○) [8], (▲) [9], (△) [10], (▼) [11], (▽) [12], (◆) [13], (◇) [14], (◀) [15], (◁) [16], (▶) [17], (▷) [18], (⬢) [19], (⬡) [20], (★) [21], (☆) [22], (⬟) [23], (⬠) [24], (◐) [25], (+) [26], (×) [27], (✳) [28], (−) [29], (|) [30], (⬒) [31], (◓) [32], (▲) [33], (▼) [34], (◆) [35], (◀) [36], (▶) [37], (⬢) [38], (★) [39], (⬟) [40], (⊠) [41], (⊗) [42], (⋇) [43]. *The lines are a guide to the eye.*

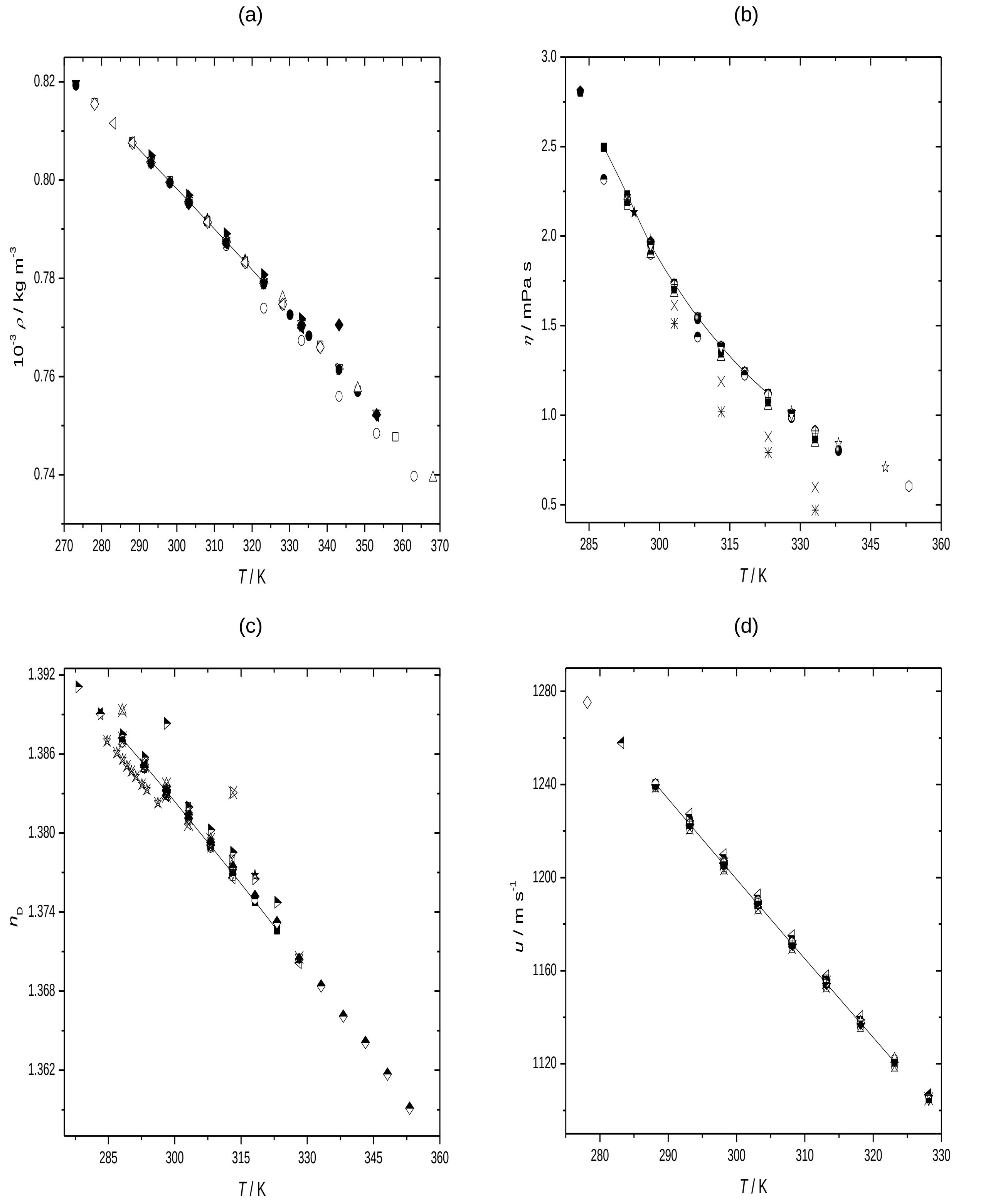

Figure 2. Comparison of (a) density, (b) viscosity, (c) refractive index and (d) speed of sound for 1-propanol over the entire investigated temperature range: (■) *this work*, (□) [44], (●) [45], (○) [46], (▲) [47], (△) [48], (▼) [49], (▽) [50], (◆) [51], (◇) [52], (◀) [53], (◁) [54], (▶) [55], (▷) [56], (⬢) [57], (⬡) [58], (★) [59], (☆) [60], (⬟) [61], (⬠) [62], (◕) [63], (+) [64], (×) [65], (✳) [66], (−) [67], (|) [68], (⬒) [69], (◓) [70], (▲) [71], (▼) [72], (⬘) [73], (◀) [74], (▶) [75], (⬢) [76], (★) [77], (⬟) [78], (☒) [79], (⊠) [80], (⨷) [81], (⨷) [82], (⨷) [83], (⨷) [84], (⨷) [85], (⊠) [86], (⨷) [87], (⊠) [88], (⬓) [89], (◒) [90], (▲) [91], (▼) [92], (⬙) [93], (◀) [94], (▶) [95], (⬢) [96], (★) [97], (⬟) [98], (⊞) [99], (⊕) [100]. *The lines are a guide to the eye.*

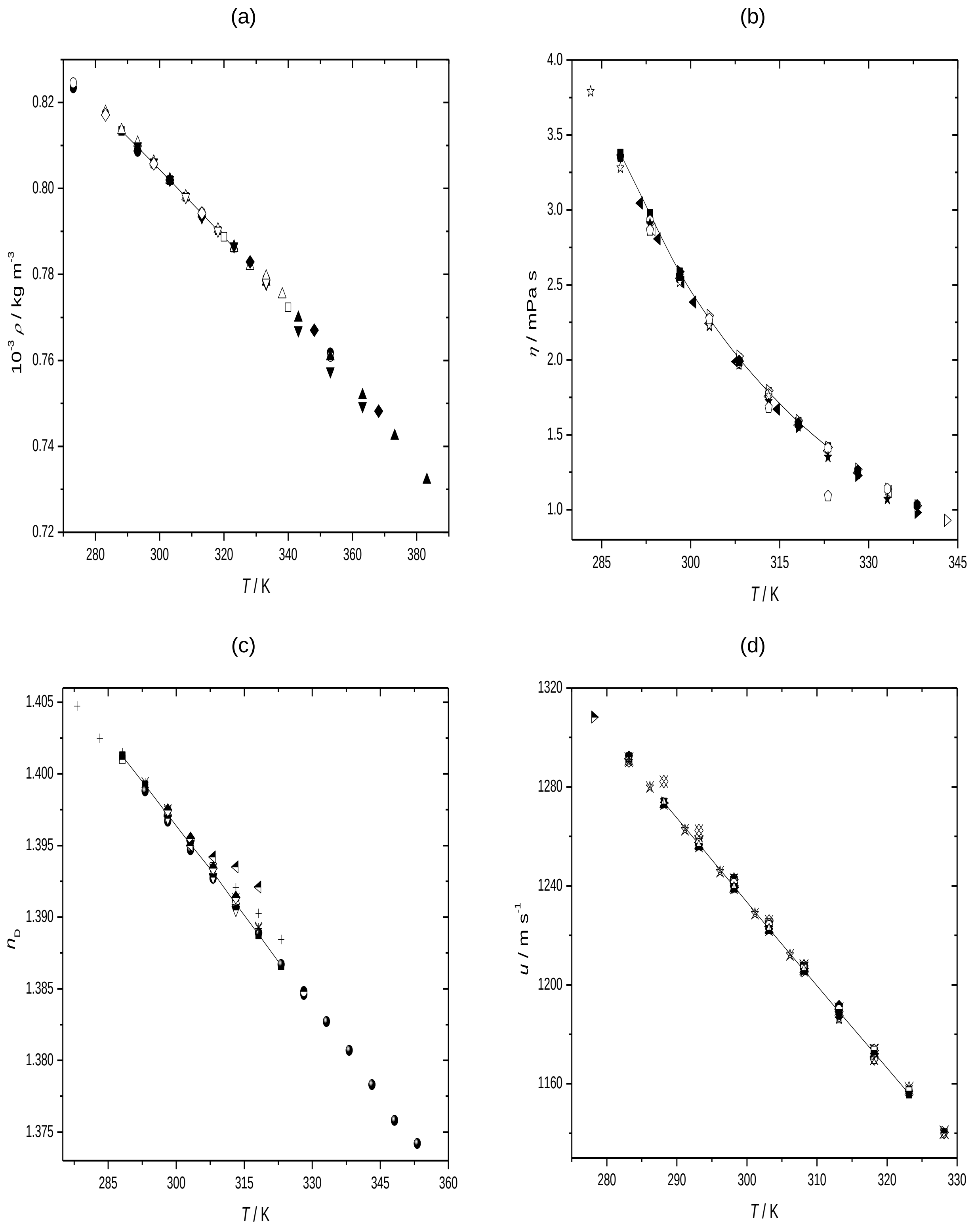

Figure 3. Comparison of (a) density, (b) viscosity, (c) refractive index and (d) speed of sound for 1-butanol over the entire investigated temperature range: (■) *this work*, (□) [101], (●) [102], (○) [103], (▲) [104], (△) [105], (▼) [46], (▽) [47], (◆) [48], (◇) [106], (◀) [107], (◁) [59], (▶) [63], (▷) [108], (⬢) [109], (⬡) [64], (★) [69], (☆) [110], (⬟) [111], (⬠) [112], (●) [73], (+) [75], (×) [113], (✳) [77], (−) [78], (|) [114], (⬒) [80], (◒) [82], (▲) [85], (▼) [115], (◆) [86], (◀) [116], (▶) [117], (⬢) [118], (★) [119], (⬟) [120], (⊠) [121], (⊠) [90], (⊠) [91], (⊠) [122], (⊠) [123], (⊠) [124], (⊠) [125], (⊠) [126], (⊠) [127], (⊠) [128], (⬓) [129], (◓) [100], (▲) [130]. *The lines are a guide to the eye.*

3. RESULTS AND DISCUSSION

Measured data of densities ρ, viscosities η, refractive indices n_D and the speed of sound u for binary systems ethyl acetate + 1-propanol and ethyl acetate + 1-butanol are given in Table 2, at atmospheric pressure and at eight temperatures, T = 288.15, 293.15, 298.15, 303.15, 308.15, 313.15, 318.15 and 323.15 K.

Table 2. Values of density, excess molar volume, dynamic viscosity, viscosity deviations, refractive index, refractive index deviations, speed of sound, isentropic compressibility, and excess isentropic compressibility for binary mixtures ethyl acetate (1) + alcohol (2), at T = (288.15 - 323.15) K and p = 0.1 MPa.[a]

x_1	$10^{-3}\rho$ / kg m^{-3}	$10^6 V^E$ / m^3 mol^{-1}	η /mPa s	$\Delta\eta$ /mPa s	n_D	Δn_D	u / m s^{-1}	$10^{12} k_S$ / Pa^{-1}	$10^{12} k_S^E$ / Pa^{-1}
				Ethyl acetate + 1-propanol					
				288.15 K					
0.0000	0.80769	--	2.4965	--	1.38715	--	1240.24	804.90	--
0.1002	0.81941	0.0794	1.6983	-0.5979	1.38531	-0.0006	1229.54	807.26	4.31
0.1999	0.83060	0.1368	1.2725	-0.8244	1.38364	-0.0011	1219.87	809.06	8.05
0.3000	0.84134	0.1801	0.99828	-0.8986	1.38218	-0.0014	1211.38	809.97	10.90
0.4000	0.85168	0.2029	0.83090	-0.8661	1.38079	-0.0016	1204.56	809.22	12.09
0.5000	0.86164	0.2079	0.72182	-0.7753	1.37956	-0.0016	1198.86	807.49	12.30
0.6000	0.87120	0.2027	0.64708	-0.6502	1.37843	-0.0016	1194.46	804.53	11.29
0.7000	0.88047	0.1771	0.58664	-0.5108	1.37750	-0.0013	1191.44	800.10	8.80
0.7999	0.88935	0.1440	0.53914	-0.3586	1.37654	-0.0011	1189.84	794.24	4.88
0.8999	0.89812	0.0795	0.51591	-0.1820	1.37580	-0.0006	1187.60	789.45	2.04
1.0000	0.90664	--	0.49779	--	1.37521	--	1185.00	785.47	--
				293.15 K					
0.0000	0.80371	--	2.2297	--	1.38518	--	1222.92	831.96	--
0.1002	0.81514	0.0847	1.5415	-0.5107	1.38322	-0.0007	1211.60	835.70	4.75
0.1999	0.82606	0.1455	1.1579	-0.7176	1.38148	-0.0012	1201.29	838.86	8.92
0.3000	0.83656	0.1913	0.91661	-0.7815	1.37992	-0.0015	1192.27	840.92	11.99
0.4000	0.84666	0.2168	0.76437	-0.7566	1.37848	-0.0017	1184.88	841.29	13.37
0.5000	0.85640	0.2229	0.66965	-0.6741	1.37718	-0.0018	1178.69	840.47	13.56
0.6000	0.86576	0.2175	0.59705	-0.5695	1.37600	-0.0017	1173.58	838.64	12.74
0.7000	0.87486	0.1890	0.54048	-0.4489	1.37502	-0.0014	1170.43	834.39	9.50
0.7999	0.88357	0.1538	0.49959	-0.3128	1.37401	-0.0012	1168.14	829.41	5.53
0.8999	0.89219	0.0854	0.47346	-0.1617	1.37325	-0.0007	1165.37	825.31	2.43
1.0000	0.90058	--	0.45784	--	1.37269	--	1162.36	821.86	--
				298.15 K					
0.0000	0.79971	--	1.9533	--	1.38316	--	1205.62	860.30	--
0.1002	0.81082	0.0912	1.3697	-0.4314	1.38109	-0.0008	1193.13	866.36	6.02
0.1999	0.82148	0.1563	1.0424	-0.6073	1.37926	-0.0013	1182.41	870.70	10.32
0.3000	0.83172	0.2050	0.83595	-0.6617	1.37765	-0.0016	1172.86	874.04	13.61
0.4000	0.84159	0.2320	0.70702	-0.6387	1.37610	-0.0018	1164.85	875.70	15.24
0.5000	0.85113	0.2388	0.62035	-0.5735	1.37474	-0.0019	1157.95	876.25	15.74
0.6000	0.86028	0.2332	0.55714	-0.4849	1.37352	-0.0018	1152.67	874.88	14.33
0.7000	0.86919	0.2039	0.50625	-0.3839	1.37248	-0.0016	1149.08	871.33	10.74
0.7999	0.87775	0.1647	0.47202	-0.2664	1.37143	-0.0013	1146.39	866.89	6.25
0.8999	0.88622	0.0921	0.44821	-0.1383	1.37066	-0.0008	1143.11	863.54	2.86
1.0000	0.89448	--	0.43445	--	1.37014	--	1139.68	860.72	--
				303.15 K					
0.0000	0.79567	--	1.7352	--	1.38110	--	1188.47	889.80	--
0.1002	0.80649	0.0973	1.2355	-0.3667	1.37893	-0.0008	1175.07	898.00	6.98
0.1999	0.81686	0.1671	0.95591	-0.5140	1.37700	-0.0014	1163.38	904.50	12.26
0.3000	0.82685	0.2193	0.77737	-0.5597	1.37530	-0.0017	1153.53	908.90	15.44
0.4000	0.83649	0.2476	0.65671	-0.5476	1.37373	-0.0019	1144.68	912.37	17.69
0.5000	0.84580	0.2561	0.58431	-0.4873	1.37229	-0.0020	1137.60	913.59	17.70

x_1	$10^{-3}\rho$ / kg m^{-3}	$10^6 V^E$ / m^3 mol^{-1}	η /mPa s	$\Delta\eta$ /mPa s	n_D	Δn_D	u / m s^{-1}	$10^{12} k_S$ / Pa^{-1}	$10^{12} k_S^E$ / Pa^{-1}
0.6000	0.85475	0.2502	0.52762	-0.4113	1.37102	-0.0019	1131.87	913.20	16.08
0.7000	0.86349	0.2190	0.48278	-0.3234	1.36991	-0.0017	1127.55	910.90	12.57
0.7999	0.87189	0.1759	0.45342	-0.2202	1.36884	-0.0014	1124.76	906.61	7.05
0.8999	0.88021	0.0988	0.43204	-0.1089	1.36808	-0.0008	1120.94	904.17	3.40
1.0000	0.88834	--	0.40807	--	1.36752	--	1117.14	902.00	--
				308.15 K					
0.0000	0.79160	--	1.5473	--	1.37902	--	1171.43	920.59	--
0.1002	0.80211	0.1034	1.1170	-0.3139	1.37674	-0.0009	1157.14	931.10	7.97
0.1999	0.81220	0.1783	0.87305	-0.4420	1.37473	-0.0015	1144.72	939.59	13.92
0.3000	0.82193	0.2343	0.71623	-0.4825	1.37293	-0.0018	1133.95	946.19	17.99
0.4000	0.83134	0.2641	0.60833	-0.4742	1.37132	-0.0020	1124.87	950.64	19.90
0.5000	0.84043	0.2738	0.54452	-0.4218	1.36979	-0.0021	1117.37	953.03	19.74
0.6000	0.84918	0.2676	0.49441	-0.3557	1.36848	-0.0020	1111.17	953.76	17.94
0.7000	0.85773	0.2345	0.45376	-0.2802	1.36733	-0.0017	1106.38	952.44	14.08
0.7999	0.86598	0.1873	0.42735	-0.1905	1.36622	-0.0014	1103.19	948.84	7.94
0.8999	0.87415	0.1058	0.40783	-0.0938	1.36539	-0.0008	1098.88	947.36	3.92
1.0000	0.88215	--	0.38533	--	1.36481	--	1094.68	945.98	--
				313.15 K					
0.0000	0.78748	--	1.3848	--	1.37693	--	1154.49	952.75	--
0.1002	0.79769	0.1098	1.0112	-0.2716	1.37453	-0.0009	1139.30	965.81	9.04
0.1999	0.80750	0.1898	0.79714	-0.3841	1.37243	-0.0015	1126.17	976.45	15.68
0.3000	0.81697	0.2494	0.65715	-0.4221	1.37055	-0.0019	1114.77	984.98	20.20
0.4000	0.82615	0.2809	0.55940	-0.4180	1.36887	-0.0021	1105.16	991.04	22.25
0.5000	0.83500	0.2927	0.50290	-0.3727	1.36728	-0.0022	1097.20	994.81	22.01
0.6000	0.84356	0.2854	0.45764	-0.3161	1.36590	-0.0021	1090.53	996.80	19.99
0.7000	0.85193	0.2508	0.41859	-0.2533	1.36471	-0.0018	1085.29	996.57	15.75
0.7999	0.86002	0.1993	0.39554	-0.1746	1.36358	-0.0015	1081.71	993.73	8.91
0.8999	0.86803	0.1131	0.37646	-0.0919	1.36270	-0.0009	1076.91	993.35	4.52
1.0000	0.87592	--	0.36641	--	1.36209	--	1072.33	992.84	--
				318.15K					
0.0000	0.78331	--	1.2413	--	1.37481	--	1137.61	986.46	--
0.1002	0.79321	0.1164	0.91805	-0.2341	1.37230	-0.0010	1121.54	1002.26	10.16
0.1999	0.80274	0.2018	0.73132	-0.3321	1.37012	-0.0016	1107.69	1015.28	17.57
0.3000	0.81195	0.2650	0.60971	-0.3647	1.36812	-0.0021	1095.69	1025.87	22.53
0.4000	0.82090	0.2986	0.52280	-0.3626	1.36640	-0.0022	1085.52	1033.80	24.83
0.5000	0.82954	0.3111	0.47386	-0.3226	1.36474	-0.0024	1077.09	1039.11	24.51
0.6000	0.83789	0.3038	0.43432	-0.2732	1.36333	-0.0022	1069.99	1042.44	22.21
0.7000	0.84607	0.2676	0.39866	-0.2199	1.36207	-0.0019	1064.32	1043.39	17.53
0.7999	0.85401	0.2119	0.37874	-0.1509	1.36092	-0.0015	1060.33	1041.49	10.01
0.8999	0.86187	0.1206	0.36147	-0.0792	1.36001	-0.0009	1055.05	1042.34	5.23
1.0000	0.86963	--	0.35164	--	1.35938	--	1050.13	1042.75	--
				323.15K					
0.0000	0.77910	--	1.1194	--	1.37265	--	1120.79	1021.79	--
0.1002	0.78869	0.1231	0.84193	-0.1987	1.37004	-0.0010	1103.84	1040.60	11.36
0.1999	0.79794	0.2136	0.67575	-0.2865	1.36777	-0.0017	1089.26	1056.25	19.60
0.3000	0.80688	0.2809	0.56917	-0.3144	1.36569	-0.0022	1076.65	1069.15	25.06
0.4000	0.81559	0.3167	0.48717	-0.3177	1.36394	-0.0023	1065.97	1079.04	27.51
0.5000	0.82401	0.3307	0.44575	-0.2805	1.36219	-0.0025	1057.08	1086.06	27.10
0.6000	0.83216	0.3228	0.41000	-0.2377	1.36074	-0.0023	1049.56	1090.88	24.48
0.7000	0.84016	0.2843	0.37552	-0.1935	1.35944	-0.0020	1043.42	1093.25	19.41
0.7999	0.84794	0.2244	0.35548	-0.1350	1.35825	-0.0016	1039.02	1092.41	11.15
0.8999	0.85566	0.1282	0.34310	-0.0688	1.35731	-0.0009	1033.26	1094.66	5.97
1.0000	0.86329	--	0.33316	--	1.35664	--	1027.99	1096.14	--

x_1	$10^{-3}\rho$ / kg m^{-3}	$10^6 V^E$ / m^3 mol^{-1}	η /mPa s	$\Delta\eta$ /mPa s	n_D	Δn_D	u / m s^{-1}	$10^{12} k_S$ / Pa^{-1}	$10^{12} k_S^E$ / Pa^{-1}
				Ethyl acetate + 1-butanol					
				288.15 K					
0.0000	0.81338	--	3.3752	--	1.40124	--	1273.65	757.89	--
0.1001	0.82235	0.1031	2.3174	-0.7698	1.39811	-0.0005	1258.95	767.23	6.58
0.2000	0.83160	0.1569	1.6252	-1.1745	1.39510	-0.0009	1245.70	774.92	11.52
0.3000	0.84076	0.2074	1.2758	-1.2362	1.39218	-0.0012	1233.57	781.63	15.47
0.3999	0.85003	0.2302	1.0300	-1.1945	1.38943	-0.0014	1223.12	786.37	17.45
0.5000	0.85936	0.2357	0.86497	-1.0715	1.38679	-0.0014	1214.08	789.46	17.78
0.5999	0.86863	0.2321	0.74109	-0.9079	1.38427	-0.0014	1206.15	791.34	16.91
0.6998	0.87795	0.2098	0.64317	-0.7184	1.38180	-0.0012	1199.97	791.02	13.83
0.7999	0.88734	0.1704	0.57729	-0.4963	1.37945	-0.0010	1194.91	789.29	9.34
0.8999	0.89690	0.0995	0.52888	-0.2569	1.37722	-0.0006	1190.12	787.18	4.47
1.0000	0.90664	--	0.49779	--	1.37521	--	1185.00	785.47	--
				293.15 K					
0.0000	0.80960	--	2.9734	--	1.39921	--	1256.85	781.92	--
0.1001	0.81831	0.1073	2.0834	-0.6382	1.39599	-0.0006	1241.46	792.89	6.98
0.2000	0.82730	0.1657	1.4777	-0.9926	1.39292	-0.0010	1227.58	802.11	12.20
0.3000	0.83622	0.2186	1.1689	-1.0498	1.38994	-0.0013	1214.59	810.62	16.72
0.3999	0.84526	0.2424	0.94949	-1.0179	1.38715	-0.0015	1203.63	816.62	18.73
0.5000	0.85436	0.2484	0.80304	-0.9126	1.38447	-0.0015	1194.08	820.91	19.02
0.5999	0.86340	0.2449	0.68318	-0.7811	1.38190	-0.0014	1185.94	823.50	17.62
0.6998	0.87251	0.2210	0.59416	-0.6189	1.37936	-0.0013	1179.30	824.10	14.23
0.7999	0.88169	0.1789	0.53307	-0.4281	1.37694	-0.0011	1173.44	823.69	9.82
0.8999	0.89104	0.1049	0.48685	-0.2228	1.37470	-0.0007	1168.26	822.29	4.43
1.0000	0.90058	--	0.45784	--	1.37269	--	1162.36	821.86	--
				298.15 K					
0.0000	0.80578	--	2.5832	--	1.39714	--	1239.80	807.38	--
0.1001	0.81424	0.1122	1.8389	-0.5292	1.39386	-0.0006	1223.70	820.16	7.44
0.2000	0.82298	0.1740	1.3210	-0.8325	1.39071	-0.0010	1208.88	831.47	13.42
0.3000	0.83165	0.2298	1.0589	-0.8797	1.38766	-0.0014	1195.62	841.15	17.77
0.3999	0.84045	0.2555	0.86421	-0.8597	1.38482	-0.0015	1184.13	848.57	19.86
0.5000	0.84931	0.2620	0.73814	-0.7707	1.38208	-0.0016	1174.09	854.14	20.09
0.5999	0.85813	0.2578	0.63307	-0.6611	1.37946	-0.0015	1165.19	858.32	18.95
0.6998	0.86702	0.2326	0.55469	-0.5248	1.37685	-0.0014	1158.06	860.02	15.31
0.7999	0.87599	0.1879	0.50115	-0.3633	1.37439	-0.0011	1151.94	860.28	10.23
0.8999	0.88513	0.1108	0.46057	-0.1890	1.37210	-0.0007	1145.83	860.50	5.12
1.0000	0.89448	--	0.43445	--	1.37014	--	1139.68	860.72	--
				303.15 K					
0.0000	0.80194	--	2.2744	--	1.39509	--	1222.63	834.19	--
0.1001	0.81014	0.1173	1.6473	-0.4403	1.39171	-0.0006	1205.79	848.98	8.00
0.2000	0.81862	0.1830	1.2138	-0.6873	1.38847	-0.0011	1190.60	861.76	14.00
0.3000	0.82705	0.2412	0.96613	-0.7484	1.38537	-0.0014	1176.78	873.13	18.60
0.3999	0.83561	0.2685	0.79925	-0.7288	1.38248	-0.0016	1164.75	882.13	20.82
0.5000	0.84424	0.2749	0.68979	-0.6514	1.37969	-0.0016	1154.21	889.13	21.03
0.5999	0.85283	0.2709	0.59621	-0.5586	1.37701	-0.0015	1144.83	894.65	19.79
0.6998	0.86150	0.2445	0.52812	-0.4402	1.37434	-0.0015	1137.20	897.58	15.94
0.7999	0.87026	0.1969	0.47972	-0.3018	1.37182	-0.0012	1130.55	899.03	10.60
0.8999	0.87919	0.1167	0.44185	-0.1530	1.36949	-0.0008	1123.77	900.67	5.46
1.0000	0.88834	--	0.40807	--	1.36752	--	1117.14	902.00	--
				308.15 K					
0.0000	0.79807	--	2.0095	--	1.39301	--	1205.84	861.75	--
0.1001	0.80600	0.1221	1.4805	-0.3664	1.38951	-0.0007	1188.24	878.73	8.55

x_1	$10^{-3}\rho$ / kg m^{-3}	$10^6 V^E$ / m^3 mol^{-1}	η /mPa s	$\Delta\eta$ /mPa s	n_D	Δn_D	u / m s^{-1}	$10^{12} k_S$ / Pa^{-1}	$10^{12} k_S^E$ / Pa^{-1}
0.2000	0.81423	0.1916	1.1116	-0.5731	1.38623	-0.0011	1172.44	893.45	14.86
0.3000	0.82241	0.2526	0.88584	-0.6364	1.38307	-0.0015	1158.04	906.70	19.69
0.3999	0.83073	0.2812	0.73828	-0.6217	1.38012	-0.0016	1145.48	917.41	21.98
0.5000	0.83912	0.2891	0.64009	-0.5573	1.37727	-0.0016	1134.44	926.00	22.14
0.5999	0.84748	0.2846	0.55644	-0.4787	1.37452	-0.0016	1124.56	933.05	20.77
0.6998	0.85593	0.2564	0.49432	-0.3786	1.37179	-0.0015	1116.46	937.29	16.60
0.7999	0.86448	0.2062	0.44982	-0.2605	1.36923	-0.0012	1109.25	940.13	11.01
0.8999	0.87319	0.1226	0.41469	-0.1332	1.36685	-0.0008	1101.82	943.34	5.79
1.0000	0.88215	--	0.38533	--	1.36481	--	1094.68	945.98	--
				313.15 K					
0.0000	0.79416	--	1.7845	--	1.39092	--	1189.12	890.52	--
0.1001	0.80183	0.1269	1.3360	-0.3065	1.38730	-0.0007	1170.80	909.82	9.06
0.2000	0.80979	0.2007	1.0124	-0.4885	1.38398	-0.0012	1154.37	926.69	15.71
0.3000	0.81773	0.2639	0.81234	-0.5467	1.38075	-0.0015	1139.39	941.99	20.77
0.3999	0.82581	0.2940	0.67892	-0.5385	1.37773	-0.0017	1126.33	954.53	23.09
0.5000	0.83396	0.3024	0.59207	-0.4834	1.37483	-0.0017	1114.79	964.87	23.18
0.5999	0.84209	0.2978	0.51562	-0.4182	1.37201	-0.0016	1104.38	973.65	21.75
0.6998	0.85032	0.2680	0.45774	-0.3344	1.36923	-0.0015	1095.82	979.35	17.22
0.7999	0.85865	0.2151	0.41922	-0.2309	1.36661	-0.0012	1088.02	983.81	11.44
0.8999	0.86715	0.1283	0.38501	-0.1234	1.36418	-0.0008	1079.97	988.74	6.14
1.0000	0.87592	--	0.36641	--	1.36209	--	1072.33	992.84	--
				318.15 K					
0.0000	0.79020	--	1.5862	--	1.38879	--	1172.50	920.53	--
0.1001	0.79761	0.1314	1.2080	-0.2546	1.38507	-0.0008	1153.45	942.35	9.59
0.2000	0.80532	0.2090	0.92572	-0.4136	1.38170	-0.0012	1136.39	961.56	16.59
0.3000	0.81300	0.2752	0.74962	-0.4662	1.37841	-0.0016	1120.86	979.05	21.85
0.3999	0.82084	0.3067	0.63513	-0.4574	1.37532	-0.0017	1107.24	993.70	24.30
0.5000	0.82876	0.3158	0.55648	-0.4124	1.37235	-0.0017	1095.26	1005.86	24.22
0.5999	0.83666	0.3113	0.48668	-0.3589	1.36948	-0.0017	1084.29	1016.63	22.78
0.6998	0.84466	0.2800	0.43495	-0.2873	1.36664	-0.0016	1075.21	1024.07	18.01
0.7999	0.85277	0.2246	0.40140	-0.1973	1.36401	-0.0013	1066.88	1030.23	11.94
0.8999	0.86107	0.1339	0.37021	-0.1050	1.36152	-0.0008	1058.24	1037.04	6.52
1.0000	0.86963	--	0.35164	--	1.35938	--	1050.13	1042.75	--
				323.15 K					
0.0000	0.78620	--	1.4161	--	1.38662	--	1155.92	951.95	--
0.1001	0.79335	0.1358	1.1131	-0.1946	1.38278	-0.0008	1136.16	976.47	10.08
0.2000	0.80080	0.2174	0.85172	-0.3478	1.37939	-0.0012	1118.49	998.19	17.40
0.3000	0.80823	0.2870	0.69549	-0.3957	1.37605	-0.0016	1102.43	1018.04	22.83
0.3999	0.81583	0.3191	0.58819	-0.3948	1.37291	-0.0017	1088.23	1035.04	25.43
0.5000	0.82351	0.3292	0.52211	-0.3525	1.36987	-0.0018	1075.70	1049.42	25.38
0.5999	0.83117	0.3247	0.46138	-0.3051	1.36696	-0.0017	1064.30	1062.14	23.69
0.6998	0.83895	0.2917	0.40961	-0.2486	1.36406	-0.0016	1054.71	1071.51	18.65
0.7999	0.84685	0.2336	0.38433	-0.1655	1.36141	-0.0012	1045.85	1079.58	12.29
0.8999	0.85493	0.1394	0.35357	-0.0880	1.35888	-0.0008	1036.59	1088.57	6.86
1.0000	0.86329	--	0.33316	--	1.35664	--	1027.99	1096.14	--

[a]Standard uncertainties u for each variables are $u(T)$ = 0.01 K; $u(p)$ = 5 %; $u(x_1)$ = 0.0001, and the combined expanded uncertainties U_c are $U_c(\rho)$ = 0.5 kg m^{-3}; $U_c(V^E)$ = 6·10^{-8} m^3 mol^{-1}; $U_r(\eta)$ = 0.25 %; $U_r(\Delta\eta)$ = 0.46 %; $U_c(n_D)$ = 7·10^{-4}, $U_c(\Delta n_D)$ = 8.6·10^{-4}, $U_c(u)$ = 0.61 m s^{-1}; $U_c(k_S)$ = 0.78·10^{-12} Pa^{-1} and $U_c(k_S^E)$ = 0.96·10^{-12} Pa^{-1} with 0.95 level of confidence ($k \approx 2$).

Along with directly measured data, the excess properties are also presented in Table 2. Using the acquired experimental data, excess molar volumes V^E, viscosity deviations $\Delta\eta$, deviations in refractive index Δn_D and excess isentropic compressibility k_S^E are calculated by the equations, respectively:

$$V^{E}=\sum_{i=1}^{2}x_iM_i\left[\left(\frac{1}{\rho}\right)-\left(\frac{1}{\rho_i}\right)\right] \tag{1}$$

$$\Delta\eta=\eta-\sum_{i=1}^{2}x_i\eta_i \tag{2}$$

$$\Delta n_D=n_D-\sum_{i=1}^{2}x_in_{Di} \tag{3}$$

$$k_S^E=k_S-\sum_{i=1}^{2}x_ik_{Si} \tag{4}$$

where M_i is the molecular weight of the component i, x_i is its mole fraction, ρ, η and n_D are measured data for density, viscosity and refractive index for the mixture, while ρ_i, η_i and n_{Di} refer to these properties for the pure component i. Isentropic compressibility of the mixture k_S and isentropic compressibility for the pure substances k_{Si} are calculated from the experimental data for density and speed of sound by the equation:

$$k_{S(i)}=1/(\rho_{(i)}u_{(i)}^2) \tag{5}$$

Excess and deviation functions were correlated with the Redlich-Kister (RK) equation [133]:

$$Y=x_ix_j\sum_{p=0}^{k}A_p\left(2x_i-1\right)^p \tag{6}$$

where Y represents V^E, $\Delta\eta$, Δn_D or k_S^E while A_p and k+1 are fitting parameters and their number.

The objective function used to optimize RK parameters is:

$$OF=\frac{1}{m}\sum_{i=1}^{m}\left(\frac{Y_{i,exp}-Y_{i,calc}}{Y_{i,exp}}\right)^2\rightarrow\min \tag{7}$$

where the subscripts (exp) and (calc) correspond to the experimental and calculated values of excess molar volume, viscosity deviation, refractive index deviation or excess isentropic compressibility, while m is the number of experimental data points.

The values of fitting parameters for all investigated properties are presented in Table 3. The number of parameters is determined using the F-test, while the quality of correlation was estimated by the value of the root-mean-square deviations (rmsd) σ, calculated by the equation:

$$\sigma=\left(\sum_{i=1}^{m}\left(Y_{exp}-Y_{calc}\right)_i^2/(m-n)\right)^{1/2} \tag{8}$$

where n is the number of fitting parameters.

Table 3. Redlich–Kister parameters for excess molar volume V^E, viscosity deviation $\Delta\eta$, deviation in refractive indices Δn_D and excess isentropic compressibility k_S^E and the corresponding rmsd values $10^{-6}\sigma(V^E)$ / m^3 mol^{-1}, $\sigma(\Delta\eta)$ / mPa s, $\sigma(\Delta n_D)$ and $10^{-12}\sigma(k_S^E)$ / Pa^{-1} for investigated binary systems.

Function	T / K	A_0	A_1	A_2	A_3	σ
			Ethyl acetate + 1-propanol			
10^6 V^E / m^3 mol^{-1}	288.15	0.8443	-0.0291	0.0990		0.0027
	293.15	0.9007	0.0098	0.0636		0.0027
	298.15	0.9656	0.0121	0.0731		0.0025
	303.15	1.0300	0.0134	0.0878		0.0023
	308.15	1.0997	0.0182	0.0960		0.0021
	313.15	1.1715	0.0253	0.1045		0.0019
	318.15	1.2466	0.0296	0.1073		0.0019
	323.15	1.3231	0.0408	0.1170		0.0017

Function	T / K	A_0	A_1	A_2	A_3	σ
$\Delta\eta$ / mPa s	288.15	-3.1013	2.1730	-1.6281	0.7514	0.0174
	293.15	-2.6965	1.8776	-1.4558	0.6429	0.0129
	298.15	-2.2941	1.5596	-1.2212	0.5876	0.0106
	303.15	-1.9493	1.3065	-0.9584	0.6242	0.0073
	308.15	-1.6872	1.1196	-0.8035	0.5309	0.0063
	313.15	-1.4908	0.9480	-0.7309	0.3621	0.0063
	318.15	-1.2904	0.8082	-0.6329	0.3381	0.0054
	323.15	-1.1221	0.6645	-0.5444	0.3427	0.0059
Δn_D	288.15	-0.0065	0.0003	-0.0007		0.00004
	293.15	-0.0070	0.0001	-0.0012		0.00004
	298.15	-0.0075	-0.0001	-0.0016		0.00004
	303.15	-0.0080	-0.0001	-0.0020		0.00008
	308.15	-0.0084	0.0002	-0.0016		0.00003
	313.15	-0.0088	0.0003	-0.0017		0.00003
	318.15	-0.0093	0.0004	-0.0019		0.00003
	323.15	-0.0096	0.0005	-0.0017		0.00004
10^{12} k_S^E / Pa^{-1}	288.15	49.76	-9.40	-21.24	-11.46	0.29
	293.15	54.68	-14.02	-23.07	-3.14	0.33
	298.15	62.89	-12.14	-30.23	-25.71	0.46
	303.15	70.93	-16.10	-22.61	-15.66	0.44
	308.15	79.50	-20.91	-20.83	-11.57	0.53
	313.15	88.73	-24.09	-21.38	-11.64	0.61
	318.15	99.08	-26.84	-23.16	-14.91	0.66
	323.15	108.99	-30.58	-20.06	-12.15	0.81
Ethyl acetate + 1-butanol						
10^6 V^E / m^3 mol^{-1}	288.15	0.9513	0.0144	0.2613		0.0046
	293.15	1.0019	0.0153	0.2741		0.0044
	298.15	1.0558	0.0167	0.2802		0.0045
	303.15	1.1140	0.0130	0.2733		0.0082
	308.15	1.1641	0.0221	0.2966		0.0045
	313.15	1.2176	0.0238	0.3035		0.0044
	318.15	1.2707	0.0284	0.3185		0.0046
	323.15	1.3247	0.0314	0.3224		0.0047
$\Delta\eta$ / mPa s	288.15	-4.2910	2.9467	-2.2126	0.9801	0.0196
	293.15	-3.6726	2.4309	-1.8739	0.9149	0.0167
	298.15	-3.1072	2.0423	-1.5186	0.6811	0.0151
	303.15	-2.6357	1.7526	-1.1513	0.5305	0.0092
	308.15	-2.2534	1.4730	-0.9722	0.4266	0.0088
	313.15	-1.9600	1.2451	-0.8213	0.2158	0.0084
	318.15	-1.6735	1.0168	-0.6724	0.2331	0.0092
	323.15	-1.4407	0.9356	-0.4308	-0.0014	0.0137
Δn_D	288.15	-0.0057	0.0004	-0.0007	-0.0014	0.00001
	293.15	-0.0059	0.0003	-0.0013	-0.0016	0.00002
	298.15	-0.0062	0.0004	-0.0018	-0.0027	0.00002
	303.15	-0.0065	0.0002	-0.0021	-0.0022	0.00002
	308.15	-0.0066	0.0005	-0.0025	-0.0019	0.00002
	313.15	-0.0067	0.0004	-0.0027	-0.0012	0.00003
	318.15	-0.0070	0.0004	-0.0026	-0.0013	0.00003
	323.15	-0.0070	0.0002	-0.0025	0.00006	0.00004
10^{12} k_S^E / Pa^{-1}	288.15	71.50	-11.50	-17.33		0.28
	293.15	76.08	-13.64	-18.15		0.23
	298.15	80.46	-16.06	-16.95		0.27
	303.15	84.20	-16.57	-16.16		0.28
	308.15	88.66	-18.00	-15.52		0.32
	313.15	92.68	-20.81	-14.86		0.37
	318.15	97.22	-23.06	-14.81		0.40
	323.15	101.53	-24.90	-17.15		0.43

Certain amount of data for investigated systems can already be found in the literature [134-142] although not at all temperatures and for all properties presented in this manuscript. For the system with 1-propanol available are data for density at 298.15 – 323.15 K and viscosity at 298.15 – 308.15 K, while for the system with 1-butanol beside the density at 298.15 – 313.15 K and viscosity at 298.15 and 303.15 K, data for refractive index can be also found but only at 298.15 K.

Comparison is graphically presented only for the temperature of 298.15 K (Figs 4 and 5) for all directly measured properties or calculated deviation properties that are published in literature. Since measurements in papers [137] and [142] are only performed at 303.15 K, the reported data are not used for the graphical comparison.

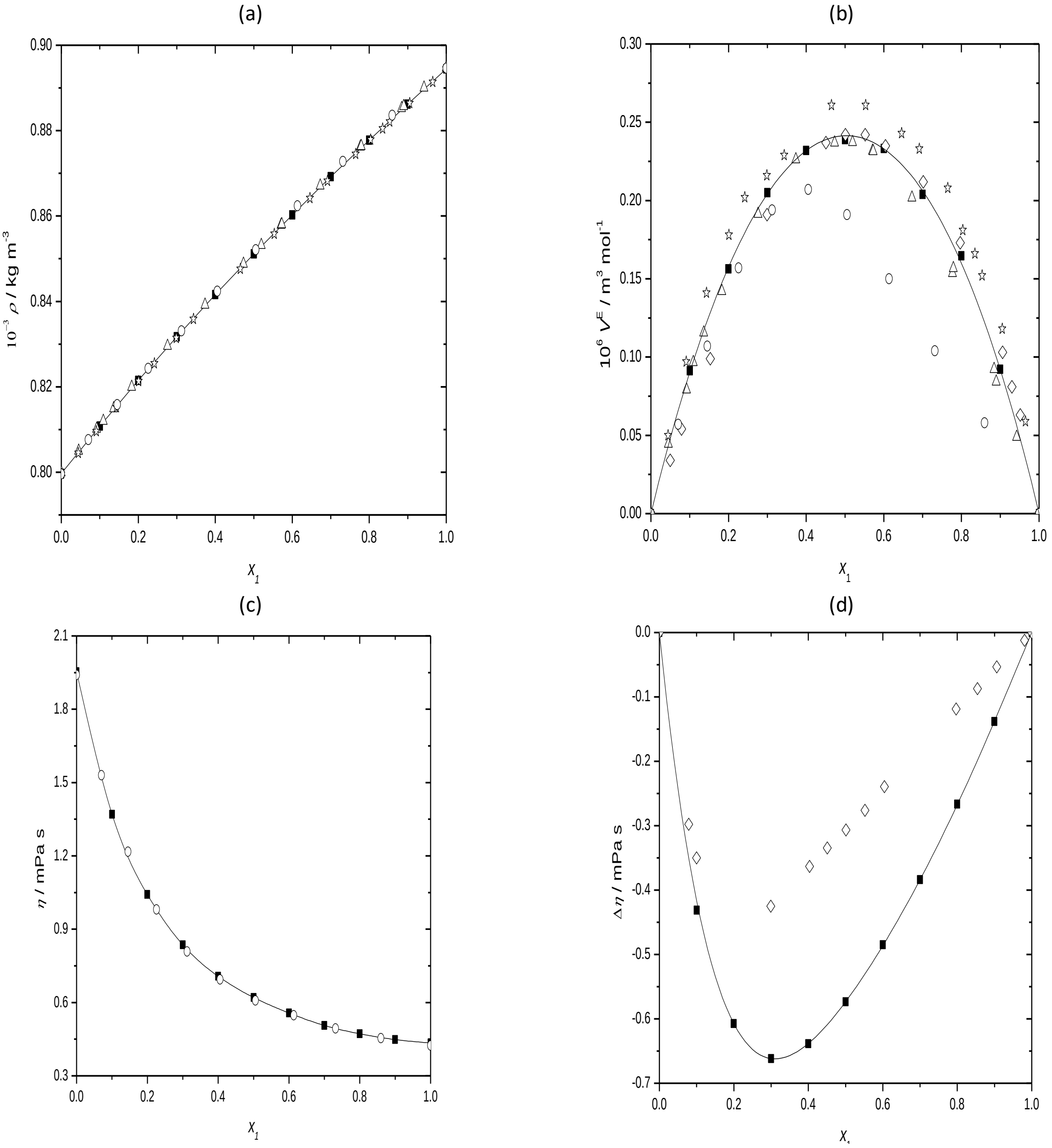

Figure 4. Comparison of (a) density, (b) excess molar volume, (c) viscosity and (d) viscosity deviation for binary mixtures ethyl acetate + 1-propanol as a function of the ethyl acetate molar fraction x_1 at 298.15 K: (■) this work, (△) [20], (☆) [134], (○) [135], (◇) [136]. For density and viscosity lines are a guide to the eye, while for the excess molar volume and viscosity deviation lines are obtained by the RK correlation.

In the case of both binary systems, curves that fit the experimental data and literature values for excess molar volume and viscosity deviation have rather similar shapes with maximum deviations of $9\cdot10^{-8}$ m^3 mol^{-1} for V^E and

0.3 mPa s for $\Delta\eta$. For directly measured properties ρ, η and n_D, the experimental and literature values are also following the same trend and the agreement is satisfactory for both systems with average deviations of 0.6 kg m^{-3} for density, 0.03 mPa s for viscosity and $2\cdot10^{-3}$ for refractive index.

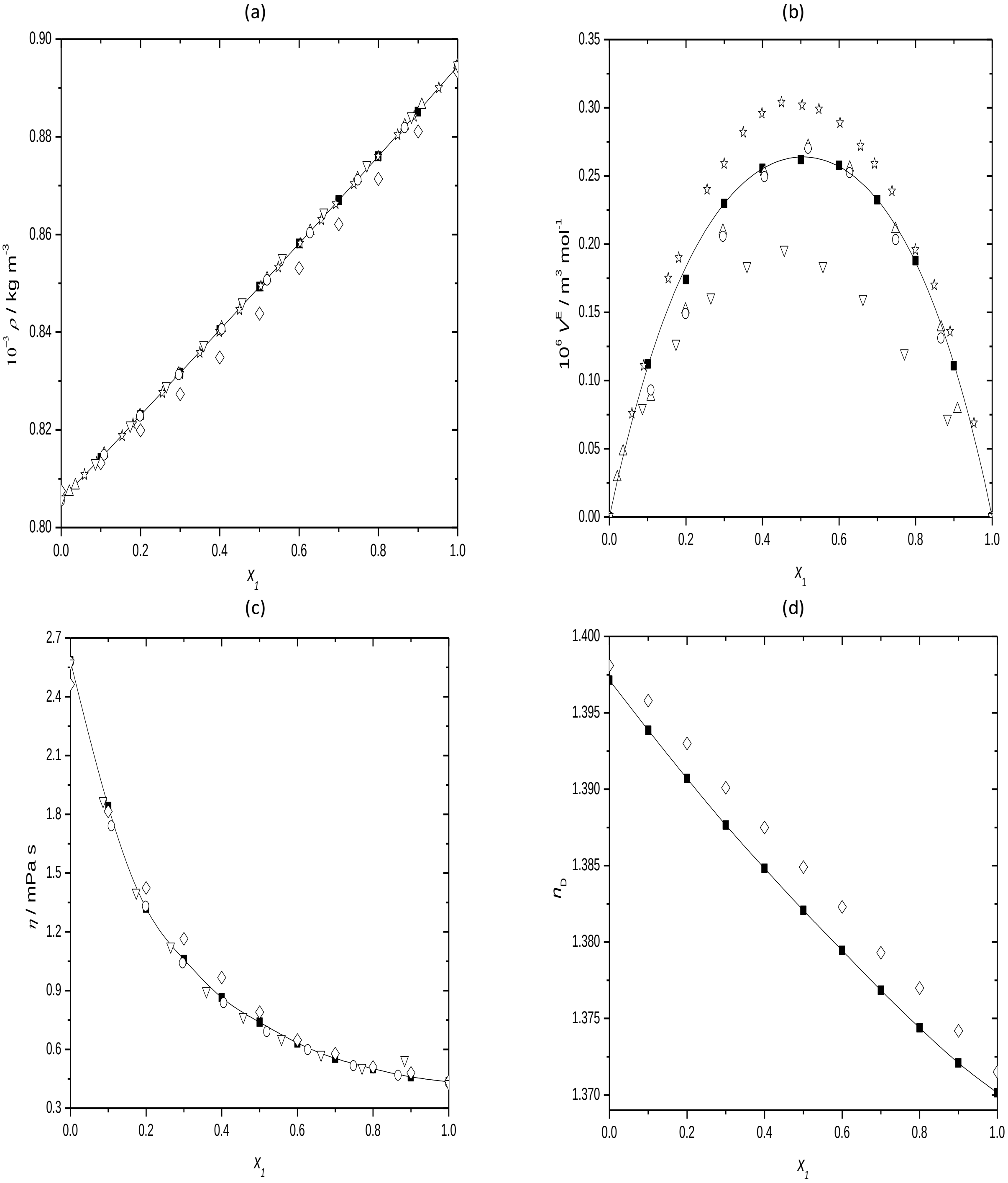

Figure 5. Comparison of (a) density, (b) excess molar volume, (c) viscosity and (d) refractive index for binary mixtures ethyl acetate + 1-butanol as a function of the ethyl acetate molar fraction x_1 at 298.15 K: (■) *this work,* (△) [138], (☆) [139], (○) [140], (◇) [141], (▽) [135]. *For density, viscosity and refractive index lines are a guide to the eye, while for the excess molar volume line is obtained by the RK correlation.*

By inspecting the data presented in Table 2, it can be concluded that the density for the ethyl acetate + 1-propanol/1-butanol systems increases with the increase in the molar fraction of acetate in the mixtures. Also, with the increase in temperature, density of the mixture decreases.

The excess molar volumes V^E of these two systems in the temperature range from 288.15 to 323.15 K are presented in Figure 6, where the lines show the V^E-x_1 dependencies obtained by the RK polynomial by correlating the V^E values experimentally obtained in this work (symbols). It can be seen that the excess molar volume has positive values for both systems in the whole concentration range for all temperatures and that they increase with the increase in temperature, which means that at lower temperatures the deviation from the ideal behaviour is smaller.

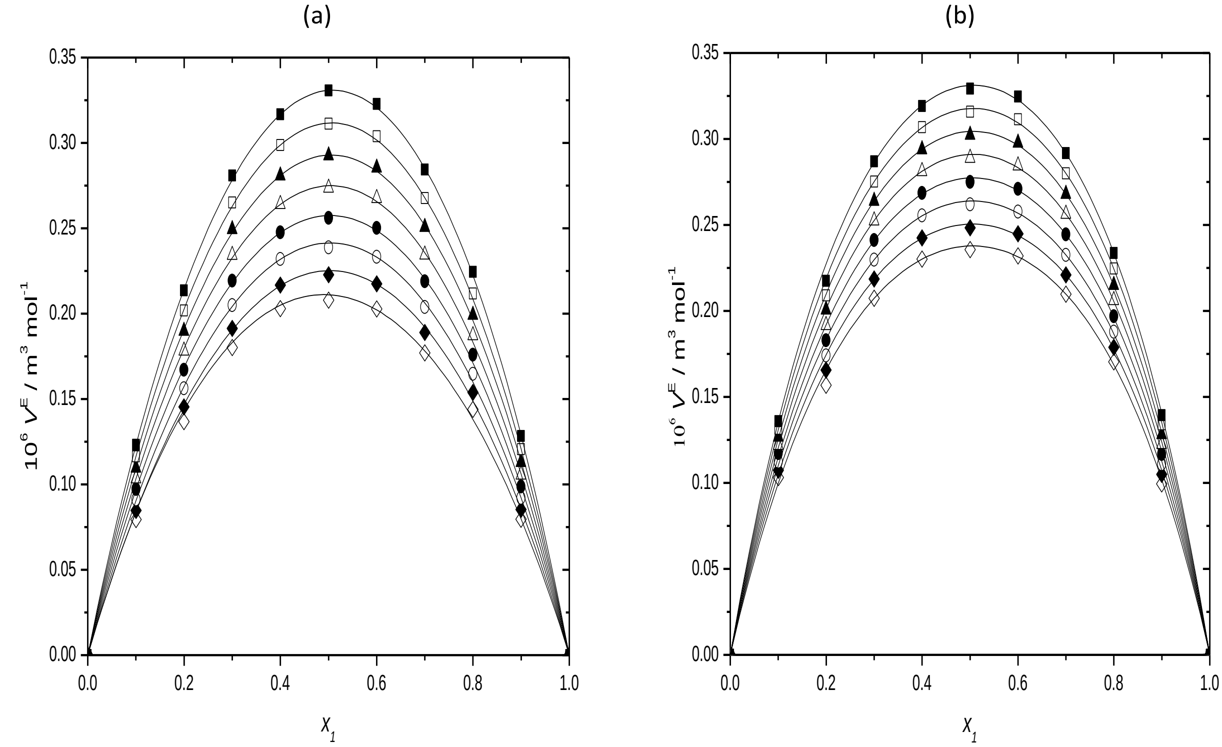

Figure 6. Experimental values of excess molar volume V^E as a function of the ethyl acetate molar fraction x_1 for the systems: (a) ethyl acetate (1) + 1-propanol (2); (b) ethyl acetate (1) + 1-butanol (2) at following temperatures: (◇) *288.15 K*, (◆) 293.15 K, (○) 298.15 K, (●) 303.15 K, (△) 308.15 K, (▲) 313.15 K, (□) 318.15 K, (■) 323.15 K, (—) *RK equation.*

Results of excess and deviation properties provide insights into the positive or negative deviation from the ideal mixture, indicating which interactions are prevalent in the systems under study. When mixing pure components deviations from the ideal mixture can be smaller or larger depending on the formation of different types of intermolecular bonds and interactions and the packing ability.

Dipole-dipole and H-bond interactions between hetero molecules in the mixture lead to negative excess molar volumes, as well as structural effects such as favourable interstitial accommodation and efficient packing. On the other hand, disruption of dipole-dipole interactions and intermolecular hydrogen bonds between molecules of the same substance when put in a mixture, as well as steric hindrance, result in positive values of V^E [143].

Alcohols are organic compounds with a highly polar hydroxyl group, which enables interconnection of molecules with strong hydrogen bonds and construction of associated liquids. In mixtures with another organic component, alcohol molecules tend to dissociate from the aggregates and form hydrogen bonds with molecules of another kind. The degree of dissociation from the aggregate depends on the affinity towards the proton of a functional group of another molecule, a proton acceptor.

Ethyl acetate is also a polar compound, which acts as H-bond acceptor, and therefore there is a possibility of intermolecular interactions in mixtures with alcohols. Depending on the strength of these two types of interactions, disruption of bonds between molecules of alcohol and formation of new ones between molecules in the mixture, positive or negative deviation from ideal behaviour can be noticed.

Another possibility to interact are dipol-dipol interactions. Since the dipole moment of ethyl acetate is higher than the ones for investigated alcohols, it can be concluded that these interactions are stronger between molecules of acetate than those with alcohol molecules. This will have a positive influence on the excess molar volume as a result.

The viscosity of both investigated systems decreases with the increase in the molar content of acetate as well as the temperature. The deviation in viscosity is negative for both systems throughout the temperature and concentration ranges and the curves are characterized by an asymmetric behaviour (Fig. 7).

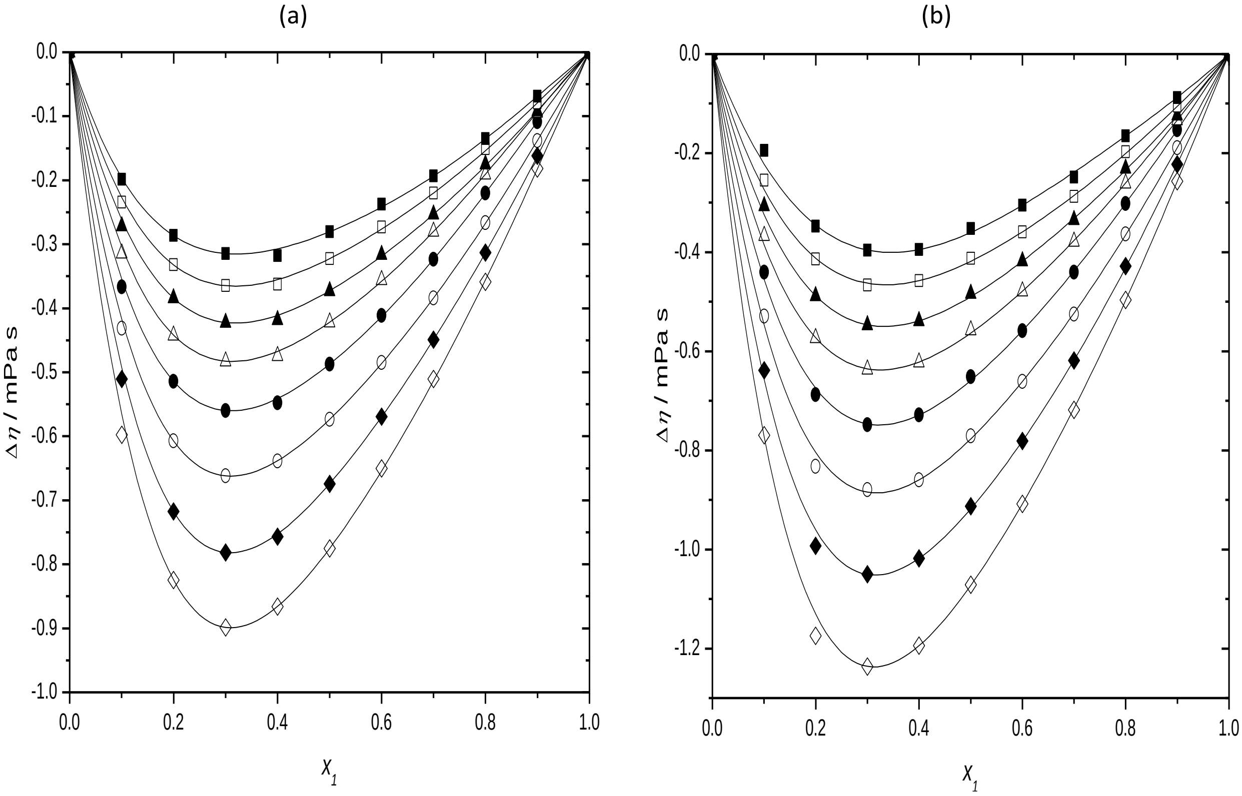

Figure 7. Experimental values of viscosity deviation Δη as a function of the ethyl acetate molar fraction x_1 for the systems: (a) ethyl acetate (1) + 1-propanol (2); (b) ethyl acetate (1) + 1-butanol (2) at following temperatures: (◇) 288.15 K, (◆) 293.15 K, (○) 298.15 K, (●) 303.15 K, (△) 308.15 K, (▲) 313.15 K, (□) 318.15 K, (■) 323.15 K, (—) *RK equation.*

The additional effect that contributes to the non-ideal behaviour is related to the molecular structure, possible interstitial accommodation or steric hindrances. These effects have a larger influence on viscosity deviation, making this property more suitable for this discussion. In the present research, negative values of Δ*η* are obtained, meaning that viscosities of the real investigated mixtures are lower than those for the ideal ones. This is the consequence of poor packing and steric hindrances, which will also contribute to positive values of the excess molar volume.

These conclusions are supported by the literature dealing with ethyl acetate + alcohol mixtures [20,135]. Nikam e*t al.* [135] showed how the chain length influences the excess molar volume data by investigating densities of ethyl acetate mixtures with methanol, ethanol, 1-propanol and 1-butanol. It is concluded that the negative V^E values for the system with methanol are due to geometrical fitting of the small methanol molecule with molecules of ethyl acetate. Positive V^E values for all other systems are a consequence of weak hetero-association effects since proton donating ability decreases with the increase in chain length of *n*-alkanols.

In Table 2 it can be noticed that the refractive index decreases with increasing the content of ethyl acetate and temperature. Deviations in refractive index are negative for both systems over the entire concentration as well as the temperature range (Fig. 8). Negative refractive index deviations generally agree with positive excess molar volume values.

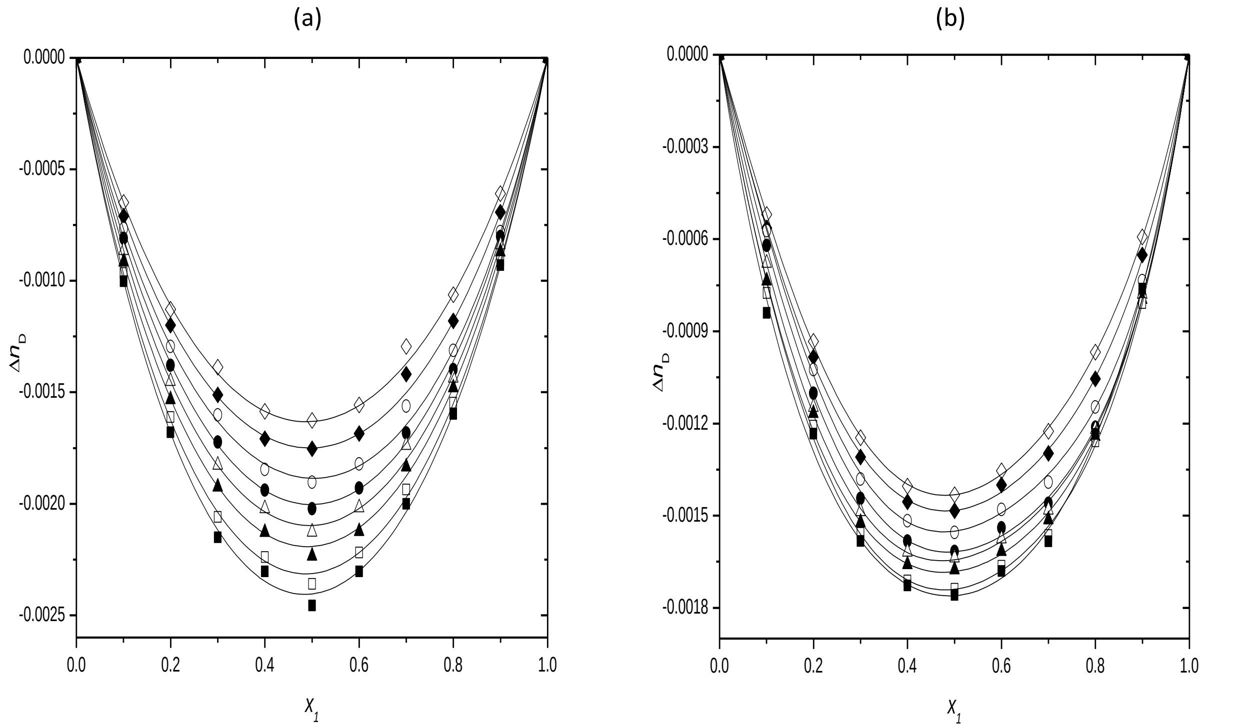

Figure 8. Experimental values of deviation in refractive index Δn_D as a function of the ethyl acetate molar fraction x_1 for the systems: (a) ethyl acetate (1) + 1-propanol (2); (b) ethyl acetate (1) + 1-butanol (2) at following temperatures: (◇) 288.15 K, (◆) 293.15 K, (○) 298.15 K, (●) 303.15 K, (△) 308.15 K, (▲) 313.15 K, (□) 318.15 K, (■) 323.15 K, (—) *RK equation.*

The excess isentropic compressibility is determined as positive throughout the concentration and temperature ranges (Fig. 9). Positive deviations are higher, and the temperature influence is more pronounced for the ethyl acetate + 1-propanol system. Breaking of interactions and the corresponding disruption of the molecular order in pure components lead to positive k_S^E values [144], similarly as data of excess molar volume were discussed above.

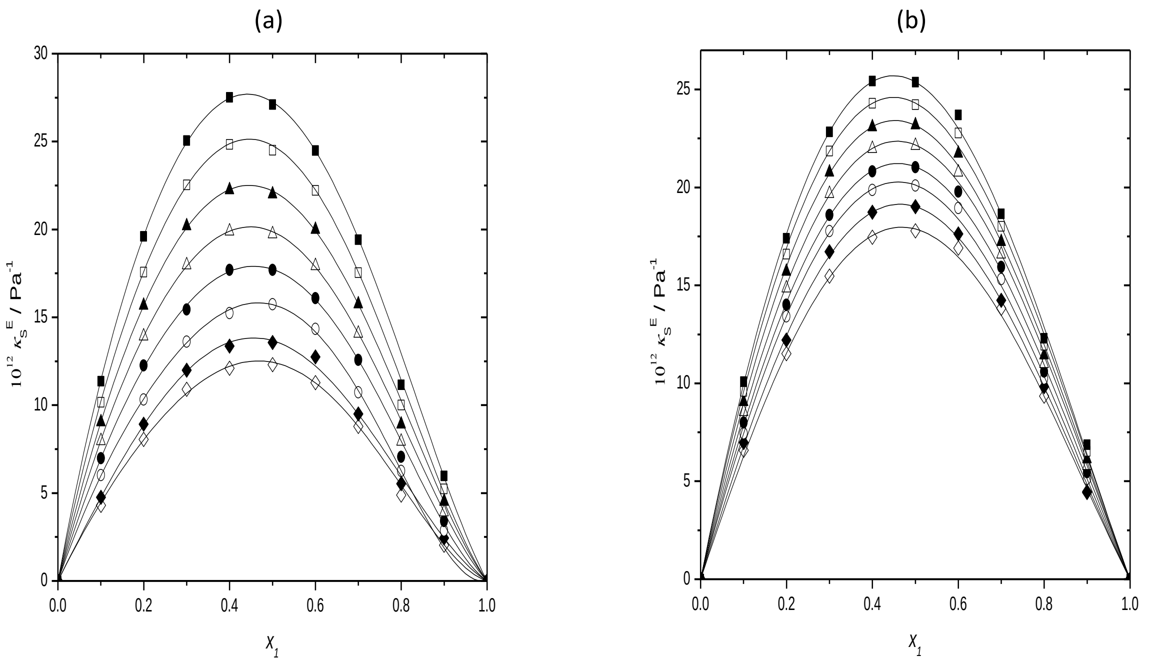

Figure 9. Experimental values of excess isentropic compressibility k_S^E as a function of the ethyl acetate molar fraction x_1 for the systems: (a) ethyl acetate (1) + 1-propanol (2); (b) ethyl acetate (1) + 1-butanol (2) at following temperatures: (◇) 288.15 K, (◆) 293.15 K, (○) 298.15 K, (●) 303.15 K, (△) 308.15 K, (▲) 313.15 K, (□) 318.15 K, (■) 323.15 K, (—) RK equation.

4. CONCLUSION

Thermodynamic and transport properties of binary mixtures containing industrially and ecologically important components (acetate and alcohol), have been investigated at atmospheric pressure and in a wide temperature range. Results for the excess molar volume and other deviation properties, calculated from experimentally measured ρ, η, n_D and u values, were correlated using the Redlich-Kister polynomial, where the optimal number of polynomial parameters is determined by the statistical F-test. In the case of investigated ethyl acetate + 1-propanol/1-butanol systems, positive values of V^E indicate that the effects of breaking the hydrogen bonds between alcohol molecules and dipole-dipole interactions between acetate molecules, which induce volume expansion, quantitatively overcame volume contraction caused by the interstitial accommodation of the ethyl acetate molecule into alcohol multimers and formation of new bonds between different molecules. The overall result is a less effective molecule packing in these mixtures than in pure components.

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SAŽETAK

Analiza termofizičkih svojstava binarnih sistema koji sadrže etil acetat i 1-propanol ili 1-butanol

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(Naučni rad)

Cilj ovog istraživanja je eksperimentalno određivanje gustine, viskoznosti, indeksa refrakcije i brzine zvuka binarnih smeša estra etil acetata i alkohola. Eksperimentalna merenja urađena su za dva sistema sa 1-propanolom i 1-butanolom na atmosferskom pritisku i u temperaturnom opsegu 288.15 - 323.15 K. Rezultati eksperimentalnih merenja dalje su korišćeni za određivanje dopunske molarne zapremine V^E, promene viskoznosti $\Delta\eta$, promene indeksa refrakcije Δn_D i promene izentropske kompresibilnosti k_S^E za svaku ispitivanu smešu. Ovi izračunati podaci korelisani su pomoću empirijske Redlih-Kisterove (Redlich-Kister) jednačine. Pozitivne vrednosti dopunske molarne zapremine i promene izentropske kompresibilnosti javljaju se kod oba analizirana sistema, dok su vrednosti promene viskoznosti i indeksa refrakcije negativne. Struktura i specifične karakteristike različitih molekula u razmatranim smešama i određeno neidealno ponašanje omogućavaju uvid u moguću vrstu interakcija u smeši, mogućnosti međumolekulskog pakovanja i strukturne efekte.

Ključne reči: gustina; viskoznost; indeks refrakcije; brzina zvuka; dopunske veličine; molekulske interakcije.