



Density, speed of sound, refractive index and relative permittivity of methanol, propan-1-ol or pentan-1-ol + benzylamine liquid mixtures. Application of the Kirkwood-Fröhlich model



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ARTICLE INFO

Article history:

Received 30 August 2021

Received in revised form 16 January 2022

Accepted 17 January 2022

Available online 21 January 2022

Keywords:

Alkan-1-ol

Benzylamine

Permittivity

Refractive index

Density

Speed of sound

ABSTRACT

Densities (ρ), speeds of sound (c), relative permittivities at 1 MHz (ϵ_r) and refractive indices at the sodium D-line (n_D) at $T = (293.15\text{ K to }303.15\text{ K})$ and $p = 0.1\text{ MPa}$ are reported for binary liquid mixtures alkan-1-ol + benzylamine. Methanol, propan-1-ol and pentan-1-ol are the alkan-1-ols studied in this work. The values of the excess molar volume (V_m^E), excess isentropic compressibility (κ_S^E), excess speed of sound (c^E), excess refractive index (n_D^E), excess relative permittivity (ϵ_r^E) and its temperature derivative ($(\partial\epsilon_r^E/\partial T)_p$) are calculated, and they are adjusted to Redlich-Kister polynomials. The V_m^E values are negative, indicating a predominance of the solvation between unlike molecules and structural effects. ϵ_r^E values indicate a positive contribution from the creation of (alkan-1-ol)-benzylamine interactions, and the positive value for the methanol mixture emphasises the importance of solvation. Calculations on excess molar refractions point out to weaker dispersive interactions than in the ideal mixture, which may be explained by the mentioned solvation effects. The Kirkwood-Fröhlich model has been applied to the mixtures, and the Kirkwood correlation factors suggest an important relative weight, especially in the methanol system, of linear-like molecules in the solutions, which is in accordance with the positive contribution of the formed multimers to ϵ_r^E due to their good effective response to the electric field.

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1. Introduction

We are carrying out a systematic thermophysical characterization of binary mixtures containing alkan-1-ols and amines [1–16] with the aim to understand and model the interactions and structure of these liquid systems. In a series of previous works, we have provided volumetric, dielectric and refractive properties of systems containing cyclohexanamine [13,14], hexan-1-amine [10], *N*-propylpropan-1-amine [11], *N,N*-diethylethanamine [12] and aniline [15]. More recently, we have reported excess molar enthalpies of cyclohexanamine systems [16]. Now, we turn our attention to alkan-1-ol + benzylamine liquid mixtures. Among the numerous industrial uses of benzylamine, it is receiving attention in the field of carbon capture, utilization and storage, which is well-known to be a common research topic nowadays. Amine solutions seem to be very suitable for this type of application, and aqueous solutions of benzylamine could be candidates in this field. Recently, heats of

absorption of CO_2 and heat capacities have been experimentally determined for this type of solutions [17]. From the theoretical perspective, it is interesting to investigate the effect of replacing aniline by benzylamine in systems with a given alkan-1-ol. While aniline and benzylamine differ only in a $-\text{CH}_2-$ group between the amine group and the aromatic ring that is present in the latter, their macroscopic properties are very different. In pure aniline there exist very strong dipolar interactions, while in pure benzylamine such interactions are weaker. This can be clearly shown by comparing the upper critical solution temperatures (UCST) of mixtures with alkanes. For instance, UCST(aniline + heptane) = 343.11 K [18], and UCST(benzylamine + decane) = 280.09 K [19].

In this article, we report experimental densities (ρ), speeds of sound (c), relative permittivities at 1 MHz (ϵ_r) and refractive indices at the sodium D-line (n_D) for methanol, propan-1-ol and pentan-1-ol + benzylamine liquid mixtures at pressure $p = 0.1\text{ MPa}$ and in the temperature range $T = (293.15\text{ to }303.15)\text{ K}$, and we calculate the Kirkwood correlation factors in the framework of the one-fluid model version of the Kirkwood-

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Fröhlich theory [20,21] in order to gain insight into the correlations between the molecular dipoles in the studied solutions.

2. Experimental

2.1. Materials

Pure liquids were used without further purification. Information about their source and purity is displayed in Table 1. Table 2 lists the measured properties of the pure compounds, whose agreement with literature values is analysed below, and their dipole moments.

2.2. Apparatus and procedure

The concentration of the liquid mixtures was calculated from mass measurements. The masses were determined by weighing using a Sartorius MSU125p analytical balance and correcting for buoyancy effects, with a standard uncertainty of $5 \cdot 10^{-5}$ g. The corresponding standard uncertainty of the mole fraction is 0.0005. Some cautions were taken: i) To minimize interaction with air, pure liquids were stored with 4 Å molecular sieves, except for methanol, which interacted with the sieves; ii) the density of the pure compounds was measured over time to check their stability, staying constant within the uncertainty of the measurements; iii) the measurement cells were appropriately filled and closed to avoid partial evaporation.

Pt-100 resistances, calibrated using the triple point of water and the melting point of Ga, were used to measure the temperature of the samples, with a standard uncertainty of 0.01 K for ρ and c , and 0.02 K for ε_r and n_D .

A densimeter and sound analyser from Anton Paar, model DSA 5000, was employed to determine densities (by the vibrant tube method) and speeds of sound (using ultrasonic pulses at 3 MHz centre frequency) of liquid samples, with a temperature stability of 0.001 K. More details about the technique and the calibration and test of the apparatus can be found elsewhere [15,22,23].

The refractive property n_D was determined using an automatic refractometer Bellingham + Stanley RFM970. The temperature is regulated with Peltier modules with a stability of 0.02 K. The apparatus was calibrated with 2,2,4-trimethylpentane and toluene at $T = (293.15 \text{ to } 303.15)$ K, with reference values recommended by Marsh [24]. For 2,2,4-trimethylpentane, the values are 1.39145 (293.15 K), 1.38898 (298.15 °C) and 1.38650 (303.15 K); for toluene, 1.49693 (293.15 K), 1.49413 (298.15 K) and 1.49126 (303.15 K).

The experimental device to determine ε_r and its calibration has been described in detail elsewhere [25]. The test of the technique and the uncertainty assessment were aided by the comparison with literature values of many pure liquids at $T = (288.15 \text{ to } 333.15)$ K [12]. The procedure involves precise impedance measurements of a parallel-plate capacitor (Agilent 16452A, $\approx 4.8 \text{ cm}^3$) with an impedance analyser (Agilent 4294A). The temper-

ature of the cell is regulated by a thermostatic bath (LAUDA RE304) with a stability of 0.02 K.

2.3. Uncertainty of the measurements

The uncertainty assessed from measurements of different samples of the same liquid (but always prepared from the same source liquids), carried out by us, using the measurement procedures and instruments described above, under the same conditions, in the same location and repeated over a short period of time will be called *repeatability*. This term will be therefore referred to our measurements only. It will also include the uncertainty associated to calibration constants and/or reference values where appropriate, but it will not consider the purity of the source liquids. For the measurements reported in this work, the expanded uncertainties (with a coverage factor of 2, approximately 95% confidence level) associated with the repeatability were found to be: 0.0001 for the mole fraction, $0.00005 \text{ g} \cdot \text{cm}^{-3}$ for ρ , $0.8 \text{ m} \cdot \text{s}^{-1}$ for c , 0.00008 for n_D , and $0.001\varepsilon_r$ (0.1 % relative expanded uncertainty) for ε_r . These values were used to define the number of digits reported for each of the properties.

However, the total uncertainty associated with these quantities should ideally include other factors, such of the purity of the compounds, which is very hard to quantify. For the mole fraction of the mixtures, the total expanded uncertainty has been roughly estimated as 0.0010. For the other properties mentioned, this total uncertainty has been quantified by the *reproducibility*, which we will take as the uncertainty evaluated from measurements of the same liquid carried out by different authors –and therefore, different source liquids, principles, methods, instruments, locations, conditions and/or periods of time-. For the case of alkan-1-ols, the agreement of the measured properties by different authors is generally better than for benzylamine. For the latter case, we include in Table 2 more literature values in order to analyse the differences and assess a reasonable estimate of the total uncertainty. Some of them have been considered as outliers (the ρ value from reference [26], the ρ and c values from reference [27,28], and the n_D values from reference [29]) due to their unusually large differences to the corresponding average values. For the relative permittivity of benzylamine, there are not enough data to perform a significant analysis (to the best of our knowledge, only one reference is available [85]), so they have been ignored in the uncertainty assessment for ε_r . With the above considerations, the expanded uncertainties (with a coverage factor of 2) associated with the reproducibility have been estimated as: $0.0006 \text{ g} \cdot \text{cm}^{-3}$ for ρ , $1.3 \text{ m} \cdot \text{s}^{-1}$ for c , 0.0003 for n_D , and $0.01\varepsilon_r$ (1 % relative expanded uncertainty) for ε_r .

The uncertainties of the excess functions (defined below) are given in Tables 3–7. They are estimated from repeatability considerations and/or comparison with reference values of test systems, such as cyclohexane + benzene for the excess molar volume, excess isentropic compressibility and excess speed of sound [30–32]. The estimation of the uncertainty of these func-

Table 1
Sample description.

Chemical name	CAS Number	Source	Purification method	Purity ^a	Water content ^b
methanol	67-56-1	Sigma-Aldrich	none	0.999	$2 \cdot 10^{-5}$
propan-1-ol	71-23-8	Fluka	none	0.999	$1 \cdot 10^{-3}$
pentan-1-ol	71-41-0	Sigma-Aldrich	none	0.999	$3 \cdot 10^{-4}$
benzylamine	100-46-9	Fluka	none	0.998	$6.8 \cdot 10^{-4}$

^a In mole fraction. By gas chromatography. Initial purity provided by the supplier.

^b In mass fraction. By Karl-Fischer titration.

Table 2

Thermophysical properties of the pure liquids used in this work at temperature T and pressure $p = 0.1$ MPa: dipole moment (μ), density (ρ^*), speed of sound (c^*), isobaric thermal expansion coefficient (α_p^*), isentropic compressibility (κ_S^*), molar isobaric heat capacity (C_{pm}^*), isothermal compressibility (κ_T^*), refractive index at the sodium D-line (n_D^*) and relative permittivity at frequency $f = 1$ MHz (ϵ_r^*).^a

Property	T/K	methanol	propan-1-ol	pentan-1-ol	benzylamine
μ/D $\rho^*/\text{g}\cdot\text{cm}^{-3}$	293.15	1.664 [55]	1.629 [55]	1.598 [55]	1.38 [56]
		0.79191	0.80352	0.81454	0.98238
		0.7916 [57]	0.80361 [59]	0.81468 [60]	0.983 [61]
		0.791400 [58]			0.98366 [29]
	298.15	0.78720	0.79951	0.81087	0.97809
		0.7869 [62]	0.79960 [59]	0.81103 [60]	0.978 [61]
		0.786884 [63]			0.981 [26]
	303.15	0.78259	0.79547	0.80724	0.978337 [64]
		0.782158 [63]	0.79561 [59]	0.81737 [60]	0.97935 [29]
					0.9789 [29]
$c^*/\text{m}\cdot\text{s}^{-1}$	293.15	1119.1	1222.5	1292.4	1580.1
		1119 [68]	1223 [69]	1292 [68]	1580.59 [29]
					1579.7 [61]
		1102.3	1205.1	1275.3	1559.9
	298.15	1101.9 [70]	1206 [69]	1276 [68]	1560.1 [61]
					1561.06 [29]
		1086.6	1188.6	1259.0	1539.9
	303.15	1186.37 [71]	1189 [69]	1259 [68]	1540.2 [61]
					1541.20 [29]
					1541.2 [66]
					1538.2 [27,28]
$\alpha_p^*/10^{-3}\text{K}^{-1}$	298.15	1.184	1.007	0.900	0.887
		1.196 [49]	1.004 [49]	0.905 [49]	0.882 [29]
	293.15	1008.3	832.7	735.0	407.7
					406.93 [29]
		1045.5	861.3	758.3	420.2
		1028 [49]	849 [49]	758 [72]	419.01 [29]
	303.15	1082.2	889.8	781.5	433.1
					431.79 [29]
$C_{pm}^*/\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$	298.15	81.92 [73]	146.88 [73]	207.45 [74]	206.74 [64]
		1253.2	1016.0	884.9	544.5
	298.15	1248 [49]	1026 [49]	884 [49]	
		1.32852	1.38521	1.40989	1.54364
	293.15	1.32859 [75]	1.38512 [76]	1.40986 [68]	1.54380 [77]
					1.544 [61]
					1.5427 [29]
n_D^*	298.15	1.32639	1.38305	1.40794	1.54132
		1.32652 [79]	1.38307 [68]	1.40789 [68]	1.5413 [80]
					1.542 [61]
					1.5387 [29]
	303.15	1.32431	1.38103	1.40597	1.53893
		1.32457 [81]	1.38104 [68]	1.40592 [82]	1.539 [61]
		1.32410 [68]			1.5393 [78]
	293.15	33.576	21.224	15.745	5.019
		33.61 [83]	21.15 [84]	15.63 [83]	5.18 [85]
		32.624	20.547	15.161	4.945
		32.62 [83]	20.42 [84]	15.08 [86]	5.20 [85]
ϵ_r^*	303.15	31.684	19.875	14.580	4.871
		31.66 [83]	19.75 [84]	14.44 [83]	

^a Standard uncertainties, u : $u(T) = 0.01$ K for ρ^* and c^* measurements; $u(T) = 0.02$ K for ϵ_r^* and n_D^* measurements; $u(p) = 10$ kPa; $u(f) = 20$ Hz. Expanded uncertainties, U (coverage factor = 2, approx. 95% confidence level): $U(\rho^*) = 0.0006 \text{ g}\cdot\text{cm}^{-3}$, $U(c^*) = 1.3 \text{ m}\cdot\text{s}^{-1}$, $U(n_D^*) = 0.0003$. Relative expanded uncertainties, U_r (coverage factor = 2): $U_r(\alpha_p^*) = 0.028$; $U_r(\kappa_S^*) = 0.002$; $U_r(\kappa_T^*) = 0.015$; $U_r(\epsilon_r^*) = 0.01$.

^b Determined using experimental values measured in this work and C_{pm}^* values from the literature included in this table.

Table 3

Density (ρ), speed of sound (c) and excess molar volume (V_m^E) of alkan-1-ol (1) + benzylamine (2) liquid mixtures as functions of the mole fraction of the alkan-1-ol (x_1) at temperature T and pressure $p = 0.1$ MPa.^a

x_1	ρ /g·cm ⁻³	c /m·s ⁻¹	V_m^E /cm ³ ·mol ⁻¹	x_1	ρ /g·cm ⁻³	c /m·s ⁻¹	V_m^E /cm ³ ·mol ⁻¹
methanol (1) + benzylamine (2) ; $T/K = 293.15$							
0.0585	0.98040	1576.0	-0.2477	0.5507	0.94342	1484.2	-1.5547
0.1129	0.97838	1571.4	-0.4751	0.6059	0.93479	1461.7	-1.5588
0.1501	0.97677	1567.7	-0.6170	0.6501	0.92655	1440.1	-1.5227
0.2045	0.97418	1561.7	-0.8178	0.7037	0.91501	1409.8	-1.4511
0.2512	0.97161	1555.6	-0.9745	0.7481	0.90354	1380.1	-1.3423
0.3053	0.96823	1547.3	-1.1422	0.8011	0.88761	1339.1	-1.1785
0.3541	0.96459	1538.4	-1.2656	0.8499	0.86982	1294.8	-0.9616
0.4019	0.96060	1528.3	-1.3766	0.9010	0.84806	1242.5	-0.7035
0.4513	0.95573	1516.0	-1.4611	0.9498	0.82297	1185.4	-0.3898
0.4968	0.95063	1502.9	-1.5231				
methanol (1) + benzylamine (2) ; $T/K = 298.15$							
0.0521	0.97648	1556.2	-0.2399	0.5514	0.93872	1464.0	-1.5616
0.1123	0.97421	1551.3	-0.4901	0.6029	0.93078	1443.4	-1.5744
0.1523	0.97252	1547.4	-0.6494	0.6503	0.92202	1420.8	-1.5407
0.1970	0.97037	1542.5	-0.8138	0.7004	0.91113	1392.4	-1.4656
0.2527	0.96733	1535.4	-1.0047	0.7489	0.89868	1360.4	-1.3507
0.2970	0.96454	1528.8	-1.1408	0.7998	0.88328	1321.2	-1.1870
0.3542	0.96029	1518.5	-1.2873	0.8506	0.86497	1276.0	-0.9725
0.3994	0.95677	1509.1	-1.4151	0.9003	0.84366	1225.3	-0.7121
0.4505	0.95143	1496.4	-1.4784	0.9501	0.81807	1167.6	-0.3900
0.4999	0.94581	1482.1	-1.5424				
methanol (1) + benzylamine (2) ; $T/K = 303.15$							
0.0485	0.97209	1536.6	-0.2137	0.5526	0.93411	1445.3	-1.5834
0.1030	0.97010	1532.4	-0.4488	0.6051	0.92581	1424.0	-1.5847
0.1459	0.96831	1528.5	-0.6226	0.6530	0.91692	1401.2	-1.5523
0.1955	0.96589	1523.1	-0.8032	0.7005	0.90658	1374.5	-1.4825
0.2470	0.96308	1516.7	-0.9815	0.7524	0.89322	1340.4	-1.3605
0.2953	0.96007	1509.6	-1.1341	0.8006	0.87850	1303.4	-1.2008
0.3501	0.95614	1500.1	-1.2884	0.8504	0.86055	1259.5	-0.9897
0.3964	0.95215	1490.5	-1.3869	0.9000	0.83923	1209.1	-0.7237
0.4466	0.94730	1478.5	-1.4822	0.9498	0.81374	1152.0	-0.4027
0.4948	0.94186	1464.9	-1.5464				
propan-1-ol (1) + benzylamine (2) ; $T/K = 293.15$							
0.0515	0.97739	1570.8	-0.1570	0.5509	0.91138	1438.3	-1.0601
0.1016	0.97239	1561.0	-0.3129	0.6023	0.90196	1419.0	-1.0487
0.1509	0.96713	1550.9	-0.4489	0.6509	0.89245	1399.4	-1.0140
0.2006	0.96160	1539.9	-0.5821	0.7017	0.88184	1377.8	-0.9515
0.2448	0.95634	1529.5	-0.6828	0.7489	0.87133	1356.4	-0.8683
0.3011	0.94930	1515.3	-0.8018	0.7999	0.85925	1332.0	-0.7526
0.3532	0.94233	1501.3	-0.8928	0.8496	0.84671	1306.9	-0.6127
0.4003	0.93563	1487.7	-0.9589	0.9000	0.83311	1280.0	-0.4364
0.4505	0.92807	1472.5	-1.0143	0.9497	0.81885	1252.1	-0.2341
0.5007	0.92000	1455.9	-1.0485				
propan-1-ol (1) + benzylamine (2) ; $T/K = 298.15$							
0.0608	0.97211	1548.9	-0.1785	0.5536	0.90653	1418.5	-1.0521
0.1085	0.96730	1539.9	-0.3252	0.5994	0.89805	1401.0	-1.0322
0.1554	0.96228	1530.1	-0.4556	0.6517	0.88793	1380.5	-1.0035
0.2047	0.95670	1519.2	-0.5801	0.7000	0.87783	1359.8	-0.9407
0.2518	0.95112	1508.3	-0.6927	0.7505	0.86662	1337.2	-0.8527
0.3023	0.94471	1495.4	-0.7917	0.7985	0.85533	1314.4	-0.7487
0.3469	0.93881	1483.6	-0.8754	0.8425	0.84431	1292.4	-0.6263
0.3980	0.93153	1469.1	-0.9435	0.8987	0.82931	1262.7	-0.4364
0.4446	0.92457	1455.0	-0.9982	0.9499	0.81470	1234.4	-0.2305
0.4912	0.91706	1439.8	-1.0232				
propan-1-ol (1) + benzylamine (2) ; $T/K = 303.15$							
0.0511	0.96877	1530.9	-0.1592	0.5506	0.90270	1400.9	-1.0514
0.1010	0.96377	1521.5	-0.3131	0.6007	0.89358	1382.5	-1.0428
0.1530	0.95823	1510.9	-0.4585	0.6498	0.88398	1363.0	-1.0039
0.2000	0.95296	1500.7	-0.5810	0.6989	0.87379	1342.5	-0.9439
0.2525	0.94668	1488.6	-0.6991	0.7468	0.86325	1321.3	-0.8648
0.3042	0.94018	1475.7	-0.8073	0.8014	0.85032	1295.4	-0.7342
0.3476	0.93433	1464.3	-0.8776	0.8502	0.83805	1271.1	-0.5951
0.4168	0.92440	1444.5	-0.9690	0.9005	0.82464	1244.8	-0.4272
0.4517	0.91909	1433.9	-1.0047	0.9501	0.81055	1217.4	-0.2301
0.5020	0.91104	1417.8	-1.0417				
pentan-1-ol (1) + benzylamine (2) ; $T/K = 293.15$							
0.0513	0.97507	1567.1	-0.1382	0.5485	0.89705	1429.6	-0.7753
0.0974	0.96834	1555.2	-0.2467	0.5991	0.88834	1414.6	-0.7572
0.1492	0.96063	1541.5	-0.3544	0.6480	0.87980	1399.8	-0.7252
0.1986	0.95321	1528.2	-0.4517	0.6982	0.87090	1384.9	-0.6760
0.2499	0.94539	1514.5	-0.5419	0.7498	0.86156	1369.2	-0.6014
0.2978	0.93795	1501.3	-0.6123	0.7986	0.85262	1354.3	-0.5167

Table 3 (continued)

x_1	ρ /g·cm ⁻³	c /m·s ⁻¹	V_m^E /cm ³ ·mol ⁻¹	x_1	ρ /g·cm ⁻³	c /m·s ⁻¹	V_m^E /cm ³ ·mol ⁻¹
0.3478	0.93004	1487.3	-0.6709	0.8484	0.84335	1339.1	-0.4105
0.3999	0.92172	1472.6	-0.7247	0.8991	0.83380	1323.4	-0.2864
0.4490	0.91372	1458.6	-0.7582	0.9496	0.82415	1307.7	-0.1429
0.4961	0.90589	1444.9	-0.7731				
pentan-1-ol (1) + benzylamine (2) ; T/K = 298.15							
0.0506	0.97084	1547.4	-0.1297	0.5504	0.89259	1410.9	-0.7600
0.1001	0.96358	1534.7	-0.2400	0.6009	0.88402	1396.0	-0.7528
0.1477	0.95658	1522.5	-0.3462	0.6422	0.87679	1383.8	-0.7201
0.1983	0.94898	1509.1	-0.4432	0.6987	0.86684	1367.0	-0.6680
0.2486	0.94133	1495.7	-0.5309	0.7496	0.85759	1351.3	-0.5849
0.2968	0.93380	1482.5	-0.5937	0.8005	0.84841	1336.2	-0.5099
0.3476	0.92588	1468.7	-0.6636	0.8492	0.83929	1321.3	-0.3939
0.3987	0.91773	1454.4	-0.7142	0.9002	0.82983	1305.8	-0.2825
0.4501	0.90938	1439.9	-0.7487	0.9500	0.82034	1290.5	-0.1389
0.4985	0.90135	1425.9	-0.7624				
pentan-1-ol (1) + benzylamine (2) ; T/K = 303.15							
0.0482	0.96687	1528.2	-0.1291	0.5527	0.88816	1392.5	-0.7580
0.1009	0.95917	1514.9	-0.2464	0.6037	0.87938	1377.6	-0.7304
0.1497	0.95197	1502.4	-0.3494	0.6497	0.87145	1364.1	-0.7044
0.1995	0.94451	1489.6	-0.4437	0.7039	0.86188	1348.1	-0.6454
0.2494	0.93693	1476.6	-0.5281	0.7504	0.85356	1334.3	-0.5807
0.2989	0.92928	1463.3	-0.5985	0.8012	0.84424	1318.8	-0.4796
0.3521	0.92097	1448.9	-0.6658	0.8505	0.83512	1303.9	-0.3705
0.4003	0.91326	1435.6	-0.7070	0.9008	0.82580	1288.8	-0.2556
0.4529	0.90475	1420.9	-0.7419	0.9506	0.81650	1273.9	-0.1306
0.5006	0.89685	1407.4	-0.7527				

^a Standard uncertainties, u : $u(T) = 0.01$ K; $u(p) = 10$ kPa. Expanded uncertainties, U (coverage factor = 2, approx. 95% confidence level): $U(x_1) = 0.0010$; $U(\rho) = 0.0006$ g·cm⁻³; $U(c) = 1.3$ m·s⁻¹; $U(V_m^E) = 0.010 \cdot |V_m^E|_{\max} + 0.005$ cm³·mol⁻¹.

Table 4

Excess isentropic compressibility (κ_s^E) and excess speed of sound (c^E) of alkan-1-ol (1) + benzylamine (2) liquid mixtures as functions of the mole fraction of the alkan-1-ol (x_1) at temperature $T = 298.15$ K and pressure $p = 0.1$ MPa.^a

x_1	κ_s^E /TPa ⁻¹	c^E /m·s ⁻¹	x_1	κ_s^E /TPa ⁻¹	c^E /m·s ⁻¹
methanol (1) + benzylamine (2) ; T/K = 298.15					
0.0521	-10.0	16.3	0.5514	-121.0	136.5
0.1123	-22.1	35.0	0.6029	-132.0	140.3
0.1523	-30.4	47.1	0.6503	-140.4	140.4
0.1970	-39.9	60.2	0.7004	-146.8	136.5
0.2527	-52.3	76.1	0.7489	-149.6	128.6
0.2970	-62.4	88.2	0.7998	-147.2	115.2
0.3542	-75.6	102.4	0.8506	-136.5	96.1
0.3994	-86.5	112.7	0.9003	-113.9	71.2
0.4505	-98.3	122.7	0.9501	-72.3	39.4
0.4999	-109.8	130.8			
propan-1-ol (1) + benzylamine (2) ; T/K = 298.15					
0.0608	-10.2	16.8	0.5536	-75.0	80.3
0.1085	-18.3	29.1	0.5994	-76.6	78.3
0.1554	-25.9	39.5	0.6517	-77.5	74.9
0.2047	-33.7	49.3	0.7000	-75.9	69.6
0.2518	-41.0	57.7	0.7505	-72.2	62.6
0.3023	-48.2	65.0	0.7985	-66.2	54.2
0.3469	-54.3	70.5	0.8425	-57.9	44.9
0.3980	-60.7	75.3	0.8987	-42.9	30.9
0.4446	-65.9	78.4	0.9499	-24.3	16.4
0.4912	-70.2	79.9			
pentan-1-ol (1) + benzylamine (2) ; T/K = 298.15					
0.0506	-7.0	11.5	0.5504	-42.9	46.1
0.1001	-13.2	20.8	0.6009	-42.3	43.6
0.1477	-18.8	28.4	0.6422	-41.1	41.0
0.1983	-24.1	34.9	0.6987	-38.6	36.7
0.2486	-28.9	40.0	0.7496	-34.6	31.6
0.2968	-32.8	43.6	0.8005	-30.3	26.6
0.3476	-36.5	46.4	0.8492	-24.5	20.8
0.3987	-39.3	47.9	0.9002	-17.6	14.3
0.4501	-41.4	48.3	0.9500	-9.3	7.3
0.4985	-42.5	47.6			

^a Standard uncertainties, u : $u(T) = 0.01$ K; $u(p) = 10$ kPa. Expanded uncertainty, U (coverage factor = 2, approx. 95% confidence level): $U(x_1) = 0.0010$. Relative expanded uncertainties, U_r : $U_r(\kappa_s^E) = 0.015$, $U_r(c^E) = 0.02$.

Table 5

Volume fractions of alkan-1-ol (ϕ_1), relative permittivities at frequency $f = 1$ MHz (ε_r) and excess relative permittivities at $f = 1$ MHz (ε_r^E) of alkan-1-ol (1) + benzylamine (2) liquid mixtures as functions of the mole fraction of the alkan-1-ol (x_1), at temperature T and pressure $p = 0.1$ MPa.^a

x_1	ϕ_1	ε_r	ε_r^E	x_1	ϕ_1	ε_r	ε_r^E
methanol (1) + benzylamine (2) ; $T/K = 293.15$							
0.0491	0.0188	5.490	-0.066	0.5528	0.3144	14.111	0.114
0.1123	0.0448	6.119	-0.179	0.5952	0.3529	15.326	0.229
0.1520	0.0623	6.580	-0.218	0.6515	0.4095	17.197	0.484
0.2044	0.0870	7.216	-0.287	0.6944	0.4574	18.637	0.556
0.2499	0.1100	7.864	-0.296	0.7498	0.5264	20.808	0.757
0.2989	0.1366	8.595	-0.325	0.7983	0.5948	22.808	0.803
0.3617	0.1737	9.709	-0.270	0.8516	0.6804	25.256	0.807
0.3922	0.1931	10.264	-0.269	0.8991	0.7677	27.653	0.711
0.4506	0.2333	11.533	-0.148	0.9500	0.8757	30.509	0.483
0.4988	0.2696	12.652	-0.066				
methanol (1) + benzylamine (2) ; $T/K = 298.15$							
0.0491	0.0188	5.401	-0.064	0.5528	0.3148	13.740	0.082
0.1123	0.0449	6.011	-0.177	0.5952	0.3534	14.913	0.186
0.1520	0.0625	6.455	-0.220	0.6515	0.4100	16.725	0.432
0.2044	0.0872	7.069	-0.290	0.6944	0.4579	18.119	0.500
0.2499	0.1102	7.697	-0.298	0.7498	0.5270	20.241	0.709
0.2989	0.1368	8.405	-0.326	0.7983	0.5953	22.166	0.744
0.3617	0.1740	9.480	-0.281	0.8516	0.6808	24.545	0.756
0.3922	0.1935	10.019	-0.282	0.8991	0.7681	26.872	0.667
0.4506	0.2336	11.243	-0.168	0.9500	0.8760	29.659	0.467
0.4988	0.2700	12.329	-0.089				
methanol (1) + benzylamine (2) ; $T/K = 303.15$							
0.0491	0.0188	5.312	-0.063	0.5528	0.3150	13.375	0.058
0.1123	0.0450	5.903	-0.175	0.5952	0.3536	14.506	0.154
0.1520	0.0625	6.333	-0.214	0.6515	0.4102	16.262	0.392
0.2044	0.0872	6.924	-0.285	0.6944	0.4581	17.608	0.454
0.2499	0.1103	7.534	-0.294	0.7498	0.5272	19.658	0.651
0.2989	0.1369	8.216	-0.326	0.7983	0.5956	21.533	0.692
0.3617	0.1741	9.256	-0.283	0.8516	0.6810	23.844	0.713
0.3922	0.1936	9.775	-0.287	0.8991	0.7683	26.096	0.625
0.4506	0.2338	10.961	-0.179	0.9500	0.8761	28.824	0.462
0.4988	0.2702	12.006	-0.110				
propan-1-ol (1) + benzylamine (2) ; $T/K = 293.15$							
0.0515	0.0359	5.383	-0.218	0.5509	0.4569	10.880	-1.543
0.1016	0.0720	5.765	-0.421	0.6023	0.5094	11.736	-1.538
0.1494	0.1075	6.159	-0.602	0.6509	0.5611	12.629	-1.483
0.2006	0.1468	6.610	-0.788	0.7017	0.6173	13.631	-1.391
0.2448	0.1818	7.042	-0.923	0.7489	0.6716	14.646	-1.256
0.3011	0.2280	7.597	-1.117	0.7999	0.7327	15.833	-1.059
0.3532	0.2724	8.178	-1.255	0.8496	0.7948	17.054	-0.845
0.4003	0.3140	8.742	-1.365	0.9000	0.8606	18.390	-0.575
0.4505	0.3599	9.398	-1.453	0.9497	0.9283	19.774	-0.288
0.5007	0.4074	10.107	-1.514				
propan-1-ol (1) + benzylamine (2) ; $T/K = 298.15$							
0.0515	0.0359	5.295	-0.210	0.5509	0.4570	10.576	-1.499
0.1016	0.0720	5.663	-0.405	0.6023	0.5096	11.392	-1.504
0.1494	0.1076	6.045	-0.579	0.6509	0.5613	12.251	-1.451
0.2006	0.1469	6.477	-0.760	0.7017	0.6175	13.206	-1.373
0.2448	0.1820	6.897	-0.888	0.7489	0.6718	14.185	-1.241
0.3011	0.2282	7.427	-1.078	0.7999	0.7328	15.322	-1.056
0.3532	0.2726	7.986	-1.212	0.8496	0.7949	16.502	-0.845
0.4003	0.3141	8.525	-1.321	0.9000	0.8606	17.789	-0.583
0.4505	0.3600	9.156	-1.406	0.9497	0.9283	19.138	-0.290
0.5007	0.4076	9.832	-1.472				
propan-1-ol (1) + benzylamine (2) ; $T/K = 303.15$							
0.0515	0.0359	5.211	-0.199	0.5509	0.4571	10.277	-1.452
0.1016	0.0720	5.563	-0.388	0.6023	0.5097	11.056	-1.463
0.1494	0.1076	5.932	-0.553	0.6509	0.5614	11.880	-1.414
0.2006	0.1470	6.345	-0.732	0.7017	0.6176	12.790	-1.347
0.2448	0.1820	6.756	-0.846	0.7489	0.6719	13.733	-1.219
0.3011	0.2283	7.259	-1.037	0.7999	0.7329	14.821	-1.046
0.3532	0.2727	7.796	-1.167	0.8496	0.7950	15.957	-0.842
0.4003	0.3142	8.310	-1.275	0.9000	0.8607	17.198	-0.587
0.4505	0.3601	8.918	-1.356	0.9497	0.9284	18.508	-0.293
0.5007	0.4077	9.562	-1.426				
pentan-1-ol (1) + benzylamine (2) ; $T/K = 293.15$							
0.0513	0.0509	5.311	-0.254	0.5485	0.5466	8.830	-2.052
0.0974	0.0967	5.590	-0.466	0.5991	0.5972	9.335	-2.090
0.1492	0.1482	5.898	-0.711	0.6480	0.6462	9.860	-2.090
0.1986	0.1974	6.208	-0.928	0.6982	0.6965	10.475	-2.015
0.2499	0.2484	6.536	-1.147	0.7498	0.7483	11.164	-1.881
0.2978	0.2962	6.856	-1.340	0.7986	0.7973	11.908	-1.663

Table 5 (continued)

x_1	ϕ_1	ε_r	ε_r^E	x_1	ϕ_1	ε_r	ε_r^E
0.3478	0.3460	7.203	-1.527	0.8484	0.8474	12.727	-1.381
0.3999	0.3980	7.588	-1.700	0.8991	0.8984	13.672	-0.983
0.4490	0.4471	7.966	-1.849	0.9496	0.9492	14.675	-0.525
0.4961	0.4941	8.367	-1.952				
pentan-1-ol (1) + benzylamine (2); $T/K = 298.15$							
0.0513	0.0509	5.226	-0.239	0.5485	0.5466	8.596	-1.933
0.0974	0.0967	5.495	-0.438	0.5991	0.5973	9.072	-1.975
0.1492	0.1482	5.791	-0.668	0.6480	0.6463	9.572	-1.976
0.1986	0.1974	6.089	-0.873	0.6982	0.6966	10.150	-1.911
0.2499	0.2485	6.404	-1.080	0.7498	0.7484	10.805	-1.786
0.2978	0.2962	6.712	-1.259	0.7986	0.7974	11.501	-1.590
0.3478	0.3461	7.045	-1.436	0.8484	0.8474	12.283	-1.319
0.3999	0.3981	7.412	-1.600	0.8991	0.8984	13.173	-0.950
0.4490	0.4471	7.774	-1.739	0.9496	0.9492	14.135	-0.507
0.4961	0.4942	8.153	-1.841				
pentan-1-ol (1) + benzylamine (2); $T/K = 303.15$							
0.0513	0.0509	5.142	-0.223	0.5485	0.5466	8.366	-1.812
0.0974	0.0967	5.400	-0.410	0.5991	0.5972	8.813	-1.856
0.1492	0.1482	5.687	-0.623	0.6480	0.6462	9.289	-1.856
0.1986	0.1974	5.971	-0.817	0.6982	0.6966	9.832	-1.802
0.2499	0.2485	6.275	-1.009	0.7498	0.7483	10.451	-1.685
0.2978	0.2962	6.568	-1.179	0.7986	0.7974	11.104	-1.509
0.3478	0.3460	6.889	-1.341	0.8484	0.8474	11.845	-1.253
0.3999	0.3980	7.238	-1.497	0.8991	0.8984	12.682	-0.912
0.4490	0.4471	7.585	-1.627	0.9496	0.9492	13.603	-0.484
0.4961	0.4942	7.943	-1.726				

^a Standard uncertainties, u : $u(T) = 0.02 \text{ K}$; $u(p) = 10 \text{ kPa}$; $u(f) = 20 \text{ Hz}$. Expanded uncertainty, U (coverage factor = 2, approx. 95% confidence level): $U(x_1) = 0.0010$; $U(\phi_1) = 0.004$. Relative expanded uncertainty, U_r (coverage factor = 2): $U_r(\varepsilon_r) = 0.01$; $U_r(\varepsilon_r^E) = 0.03$.

tions using the reproducibility would give some unrealistically large uncertainties.

3. Equations

If dispersion and absorption of the acoustic wave are negligible, the Newton-Laplace equation allows to determine the isentropic compressibility (κ_s) from ρ and c experimental measurements:

$$\kappa_s = \frac{1}{\rho c^2} \quad (1)$$

The isothermal compressibility (κ_T) can be calculated through general thermodynamic relations involving κ_s , the molar isobaric heat capacity (C_{pm}), the molar volume (V_m) and the isobaric thermal expansion coefficient (α_p):

$$\kappa_T = \kappa_s + \frac{TV_m(\alpha_p)^2}{C_{pm}} \quad (2)$$

The values F^{id} of the thermodynamic properties of an ideal mixture at the same temperature and pressure as the real mixture are computed from the well-established formulae from Benson and Kiyohara [33–35]:

$$F^{id} = x_1 F_1^* + x_2 F_2^* \quad (F = V_m, C_{pm}) \quad (3)$$

$$F^{id} = \phi_1 F_1^* + \phi_2 F_2^* \quad (F = \alpha_p, \kappa_T) \quad (4)$$

where F_i^* is the value of the property F of pure component i , x_i represents the mole fraction of component i and $\phi_i = x_i V_m^*/V_m^{id}$ is the volume fraction of component i . Ideal values of κ_s and c are calculated using the equations [33]:

$$\kappa_s^{id} = \kappa_T^{id} - \frac{TV_m^{id}(\alpha_p^{id})^2}{C_{pm}^{id}} \quad (5)$$

$$c^{id} = \left(\frac{1}{\rho^{id} \kappa_s^{id}} \right)^{1/2} \quad (6)$$

being $\rho^{id} = (x_1 M_1 + x_2 M_2)/V_m^{id}$ (M_i , molar mass of the i component).

The ideal dielectric and refractive properties are calculated from [36,37]:

$$\varepsilon_r^{id} = \phi_1 \varepsilon_{r1}^* + \phi_2 \varepsilon_{r2}^* \quad (7)$$

$$n_D^{id} = [\phi_1 (n_{D1}^*)^2 + \phi_2 (n_{D2}^*)^2]^{1/2} \quad (8)$$

$$\left[\left(\frac{\partial \varepsilon_r}{\partial T} \right)_p \right]^{id} = \left(\frac{\partial \varepsilon_r^{id}}{\partial T} \right)_p \quad (9)$$

Lastly, the excess properties are defined by:

$$F^E = F - F^{id} \quad (10)$$

4. Results

The $\alpha_p = -(1/\rho)(\partial \rho / \partial T)_p$ values of the pure compounds at $T = 298.15 \text{ K}$ and $p = 0.1 \text{ MPa}$ were obtained from fittings of experimental ρ values to straight lines as functions of T in the range (293.15 to 303.15) K. An analogous procedure was used for the temperature derivative $(\partial \varepsilon_r^E / \partial T)_p = [(\partial \varepsilon_r / \partial T)_p]^E = (\partial \varepsilon_r / \partial T)_p - (\partial \varepsilon_r^{id} / \partial T)_p$.

The experimental values of the determined quantities at $p = 0.1 \text{ MPa}$ for alkan-1-ol (1) + benzylamine (2) liquid mixtures can be found in Tables 3–7 as functions of x_1 (mole fraction of the alkan-1-ol). Values of ρ , c , and V_m^E at $T = (293.15 \text{ to } 303.15) \text{ K}$ are collected in Table 3; κ_s^E and c^E values at $T = 298.15 \text{ K}$ are written in Table 4; ϕ_1 , ε_r and ε_r^E values at $T = (293.15 \text{ to } 303.15) \text{ K}$ are found in Table 5; whereas ϕ_1 , n_D and n_D^E values at the same conditions are written in Table 6. $(\partial \varepsilon_r^E / \partial T)_p$ results at $T = 298.15 \text{ K}$ are collected in Table 7.

Table 6

Volume fractions of alkan-1-ol (ϕ_1), refractive indices at the sodium D-line (n_D) and excess refractive indices at the sodium D-line (n_D^E) of alkan-1-ol (1) + benzylamine (2) liquid mixtures as functions of the mole fraction of the alkan-1-ol, x_1 , at temperature T and pressure $p = 0.1$ MPa.^a

x_1	ϕ_1	n_D	$10^5 n_D^E$	x_1	ϕ_1	n_D	$10^5 n_D^E$
methanol (1) + benzylamine (2) ; $T/K = 293.15$							
0.0491	0.0188	1.54122	135	0.5528	0.3144	1.48728	790
0.1520	0.0623	1.53423	311	0.6515	0.4095	1.46711	772
0.2499	0.1100	1.52608	461	0.7498	0.5264	1.44126	683
0.3617	0.1737	1.51461	613	0.8516	0.6804	1.40590	503
0.4506	0.2333	1.50312	690	0.9500	0.8757	1.35930	218
methanol (1) + benzylamine (2) ; $T/K = 298.15$							
0.0491	0.0188	1.53772	16	0.6515	0.4100	1.46470	766
0.1520	0.0625	1.53077	200	0.7498	0.5270	1.43893	685
0.2499	0.1102	1.52268	355	0.8516	0.6808	1.40271	512
0.3617	0.1740	1.51140	527	0.8991	0.7681	1.38261	339
0.4506	0.2336	1.50041	653	0.9500	0.8760	1.35672	183
0.5528	0.3148	1.48450	746				
methanol (1) + benzylamine (2) ; $T/K = 303.15$							
0.1123	0.0450	1.53158	166	0.4506	0.2338	1.49800	648
0.1520	0.0625	1.52900	260	0.5528	0.3150	1.48200	730
0.2044	0.0872	1.52487	345	0.6515	0.4102	1.46200	727
0.2989	0.1369	1.51643	508	0.7498	0.5272	1.43600	620
0.3922	0.1936	1.50582	604	0.8991	0.7683	1.38083	381
propan-1-ol (1) + benzylamine (2) ; $T/K = 293.15$							
0.0515	0.0359	1.53877	54	0.5509	0.4569	1.47666	329
0.1016	0.0720	1.53379	101	0.6023	0.5094	1.46835	327
0.1494	0.1075	1.52878	138	0.6509	0.5611	1.45996	309
0.2006	0.1468	1.52323	181	0.7017	0.6173	1.45086	297
0.2448	0.1818	1.51817	210	0.7489	0.6716	1.44175	259
0.3011	0.2280	1.51147	249	0.7999	0.7327	1.43199	271
0.3532	0.2724	1.50485	271	0.8496	0.7948	1.42108	192
0.4003	0.3140	1.49884	314	0.9000	0.8606	1.40987	151
0.4505	0.3599	1.49182	326	0.9497	0.9283	1.39810	93
0.5007	0.4074	1.48464	350				
propan-1-ol (1) + benzylamine (2) ; $T/K = 298.15$							
0.0515	0.0359	1.53644	52	0.5509	0.4570	1.47431	321
0.1016	0.0720	1.53154	107	0.6023	0.5096	1.46591	310
0.1494	0.1076	1.52648	140	0.6509	0.5613	1.45760	300
0.2006	0.1469	1.52096	186	0.7017	0.6175	1.44842	278
0.2448	0.1820	1.51594	219	0.7489	0.6718	1.43937	245
0.3011	0.2282	1.50936	269	0.7999	0.7328	1.42924	218
0.3532	0.2726	1.50256	273	0.8496	0.7949	1.41852	157
0.4003	0.3141	1.49647	305	0.9000	0.8606	1.40736	118
0.4505	0.3600	1.48942	313	0.9497	0.9283	1.39550	50
0.5007	0.4076	1.48216	330				
propan-1-ol (1) + benzylamine (2) ; $T/K = 303.15$							
0.0515	0.0359	1.53406	52	0.5509	0.4571	1.47207	321
0.1016	0.0720	1.52905	94	0.6023	0.5097	1.46371	313
0.1494	0.1076	1.52411	138	0.6509	0.5614	1.45539	299
0.2006	0.1470	1.51853	178	0.7017	0.6176	1.44631	286
0.2448	0.1820	1.51356	214	0.7489	0.6719	1.43718	243
0.3011	0.2283	1.50685	251	0.7999	0.7329	1.42723	231
0.3532	0.2727	1.50033	281	0.8496	0.7950	1.41628	144
0.4003	0.3142	1.49410	298	0.9000	0.8607	1.40520	111
0.4505	0.3601	1.48714	313	0.9497	0.9284	1.39349	56
0.5007	0.4077	1.47980	321				
pentan-1-ol (1) + benzylamine (2) ; $T/K = 293.15$							
0.0513	0.0509	1.53766	55	0.5485	0.5466	1.47359	155
0.0974	0.0967	1.53190	68	0.5991	0.5972	1.46663	140
0.1492	0.1482	1.52549	93	0.6480	0.6462	1.45989	128
0.1986	0.1974	1.51923	106	0.6982	0.6965	1.45288	109
0.2499	0.2484	1.51275	123	0.7498	0.7483	1.44573	101
0.2978	0.2962	1.50647	121	0.7986	0.7973	1.43869	68
0.3478	0.3460	1.50029	158	0.8484	0.8474	1.43171	60
0.3999	0.3980	1.49337	153	0.8991	0.8984	1.42443	38
0.4490	0.4471	1.48692	159	0.9496	0.9492	1.41706	7
0.4961	0.4941	1.48051	144				
pentan-1-ol (1) + benzylamine (2) ; $T/K = 298.15$							
0.0513	0.0509	1.53519	38	0.5485	0.5466	1.47134	143
0.0974	0.0967	1.52951	58	0.5991	0.5973	1.46439	127
0.1492	0.1482	1.52301	72	0.6480	0.6463	1.45779	128
0.1986	0.1974	1.51688	96	0.6982	0.6966	1.45077	107
0.2499	0.2485	1.51041	113	0.7498	0.7484	1.44361	95
0.2978	0.2962	1.50410	105	0.7986	0.7974	1.43664	68
0.3478	0.3461	1.49783	133	0.8484	0.8474	1.42971	61
0.3999	0.3981	1.49092	127	0.8991	0.8984	1.42245	39
0.4490	0.4471	1.48458	141	0.9496	0.9492	1.41515	13

Table 6 (continued)

x_1	ϕ_1	n_D	$10^5 n_D^E$	x_1	ϕ_1	n_D	$10^5 n_D^E$
0.4961	0.4942	1.47828	137				
pentan-1-ol (1) + benzylamine (2); $T/K = 303.15$							
0.0513	0.0509	1.53273	29	0.5485	0.5466	1.46909	134
0.0974	0.0967	1.52701	43	0.5991	0.5972	1.46225	127
0.1492	0.1482	1.52062	66	0.6480	0.6462	1.45555	115
0.1986	0.1974	1.51444	83	0.6982	0.6966	1.44867	107
0.2499	0.2485	1.50805	106	0.7498	0.7483	1.44142	83
0.2978	0.2962	1.50179	101	0.7986	0.7974	1.43459	69
0.3478	0.3460	1.49552	125	0.8484	0.8474	1.42770	64
0.3999	0.3980	1.48874	130	0.8991	0.8984	1.42048	43
0.4490	0.4471	1.48234	138	0.9496	0.9492	1.41324	21
0.4961	0.4942	1.47597	125				

^a Standard uncertainties, u : $u(T) = 0.02$ K; $u(p) = 10$ kPa. Expanded uncertainty, U (coverage factor = 2, approx. 95% confidence level): $U(x_1) = 0.0010$; $U(\phi_1) = 0.004$. $U(n_D) = 0.0003$. Relative expanded uncertainty, U_r (coverage factor = 2): $U_r(n_D^E) = 0.03$.

Table 7

Volume fraction of alkan-1-ol (ϕ_1) and temperature derivative of the excess relative permittivity at frequency $f = 1$ MHz ($(\partial \epsilon_r^E / \partial T)_p$) of alkan-1-ol (1) + benzylamine (2) liquid mixtures as functions of the mole fraction of the alkan-1-ol (x_1) at temperature T and pressure $p = 0.1$ MPa. ^a

x_1	ϕ_1	$(\partial \epsilon_r^E / \partial T)_p$	x_1	ϕ_1	$(\partial \epsilon_r^E / \partial T)_p$
methanol (1) + benzylamine (2); $T/K = 298.15$					
0.0491	0.0188	0.0003	0.5528	0.3148	-0.0056
0.1123	0.0449	0.0004	0.5952	0.3534	-0.0075
0.1520	0.0625	0.0004	0.6515	0.4100	-0.0092
0.2044	0.0872	0.0002	0.6944	0.4579	-0.0102
0.2499	0.1102	0.0002	0.7498	0.5270	-0.0111
0.2989	0.1368	-0.0001	0.7983	0.5953	-0.0106
0.3617	0.1740	-0.0013	0.8516	0.6808	-0.0094
0.3922	0.1935	-0.0018	0.8991	0.7681	-0.0086
0.4506	0.2336	-0.0031	0.9500	0.8760	-0.0021
0.4988	0.2700	-0.0044			
propan-1-ol (1) + benzylamine (2); $T/K = 298.15$					
0.0515	0.0359	0.0019	0.5509	0.4570	0.0091
0.1016	0.0720	0.0033	0.6023	0.5096	0.0075
0.1494	0.1076	0.0049	0.6509	0.5613	0.0069
0.2006	0.1469	0.0056	0.7017	0.6175	0.0044
0.2448	0.1820	0.0077	0.7489	0.6718	0.0037
0.3011	0.2282	0.0080	0.7999	0.7328	0.0013
0.3532	0.2726	0.0088	0.8496	0.7949	0.0003
0.4003	0.3141	0.0090	0.9000	0.8606	-0.0012
0.4505	0.3600	0.0097	0.9497	0.9283	-0.0005
0.5007	0.4076	0.0088			
pentan-1-ol (1) + benzylamine (2); $T/K = 298.15$					
0.0513	0.0509	0.0031	0.5485	0.5466	0.0240
0.0974	0.0967	0.0056	0.5991	0.5973	0.0234
0.1492	0.1482	0.0088	0.6480	0.6463	0.0234
0.1986	0.1974	0.0111	0.6982	0.6966	0.0213
0.2499	0.2485	0.0138	0.7498	0.7484	0.0196
0.2978	0.2962	0.0161	0.7986	0.7974	0.0154
0.3478	0.3461	0.0186	0.8484	0.8474	0.0128
0.3999	0.3981	0.0203	0.8991	0.8984	0.0071
0.4490	0.4471	0.0222	0.9496	0.9492	0.0041
0.4961	0.4942	0.0226			

^a Standard uncertainties, u : $u(T) = 0.02$ K; $u(p) = 10$ kPa; $u(f) = 20$ Hz. Expanded uncertainties, U (coverage factor = 2, approx. 95% confidence level): $U(x_1) = 0.0010$; $U(\phi_1) = 0.004$, $U[(\partial \epsilon_r^E / \partial T)_p] = 0.0008$ K⁻¹.

The F^E data were fitted by unweighted linear least-squares regressions to Redlich-Kister polynomials [38]:

$$F^E = x_1(1 - x_1) \sum_{i=0}^{k-1} A_i (2x_1 - 1)^i \quad (11)$$

The number, k , of appropriate coefficients for each system, property and temperature has been determined by the application of an F-test of additional term [39] at a 99.5% confidence level. Table 8 includes the parameters A_i obtained, and the standard deviations, $\sigma(F^E)$, defined by:

$$\sigma(F^E) = \left[\frac{1}{N-k} \sum_{j=1}^N (F_{\text{cal},j}^E - F_{\text{exp},j}^E)^2 \right]^{1/2} \quad (12)$$

where j indexes the N experimental data $F_{\text{exp},j}^E$, and $F_{\text{cal},j}^E$ is the corresponding value of the excess property F^E calculated from equation (11).

The values of the excess properties at $T = 298.15$ K and $p = 0.1$ MPa (V_m^E , κ_s^E and c^E versus x_1 ; and ϵ_r^E , $(\partial \epsilon_r^E / \partial T)_p$ and n_D^E versus ϕ_1) are plotted in Figures 1–6 with the corresponding Redlich-Kister fittings.

Table 8

Coefficients A_i and standard deviations, $\sigma(F^E)$ (equation (12)), for the representation of F^E at temperature T and pressure $p = 0.1$ MPa for alkan-1-ol (1) + benzylamine (2) liquid mixtures by equation (11).

Property F^E	alkan-1-ol	T/K	A_0	A_1	A_2	A_3	A_4	A_5	A_6	$\sigma(F^E)$
$V_m^E / \text{cm}^3 \cdot \text{mol}^{-1}$	methanol	293.15	-6.095	-1.97	-0.26					0.004
		298.15	-6.17	-1.87	-0.36					0.007
		303.15	-6.202	-2.00	-0.38					0.003
	propan-1-ol	293.15	-4.190	-0.915	0.09					0.0018
		298.15	-4.138	-0.91	0.11					0.004
		303.15	-4.156	-0.85	0.09					0.003
	pentan-1-ol	293.15	-3.096	-0.39	0.20	0.27				0.002
		298.15	-3.06	-0.31	0.26					0.006
		303.15	-3.026	-0.35	0.35	0.43				0.004
$\kappa_S^E / \text{TPa}^{-1}$	methanol	298.15	-439.1	-461	-362	-203	-80	-162	-148	0.15
	propan-1-ol	298.15	-284.0	-163.2	-71	-24				0.13
	pentan-1-ol	298.15	-170.2	-30.7						0.11
$c^E / \text{m} \cdot \text{s}^{-1}$	methanol	298.15	523.9	281.8	74					0.3
	propan-1-ol	298.15	320.3	28.2						0.17
	pentan-1-ol	298.15	190.4	-44.6	6.5					0.11
ε_r^E	methanol	293.15	-0.17	5.3	5.2	1.2				0.02
		298.15	-0.27	5.0	5.0	1.2				0.02
		303.15	-0.35	4.6	4.8	1.3				0.02
	propan-1-ol	293.15	-6.08	-1.89	0.87	1.19				0.006
		298.15	-5.90	-1.98	0.68	1.1				0.007
		303.15	-5.71	-2.04	0.51	1.0				0.008
	pentan-1-ol	293.15	-7.864	-4.15	-1.1	1.06	1.0			0.005
		298.15	-7.408	-3.97	-1.15	0.87	0.9			0.003
		303.15	-6.939	-3.78	-1.20	0.7	0.8			0.003
$10^5 n_D^E$	methanol	293.15	3038	1183						24
		298.15	2783	1746						22
		303.15	2812	1226						20
	propan-1-ol	293.15	1323	321						12
		298.15	1270	113						8
		303.15	1259	156						11
	pentan-1-ol	293.15	599	-180						9
		298.15	544	-88						7
		303.15	517							7
$(\partial \varepsilon_r^E / \partial T)_p / \text{K}^{-1}$	methanol	298.15	-0.019	-0.057	-0.036					0.0007
	propan-1-ol	298.15	0.0368	-0.022	-0.037					0.0005
	pentan-1-ol	298.15	0.0931	0.037	-0.024	-0.036				0.0004

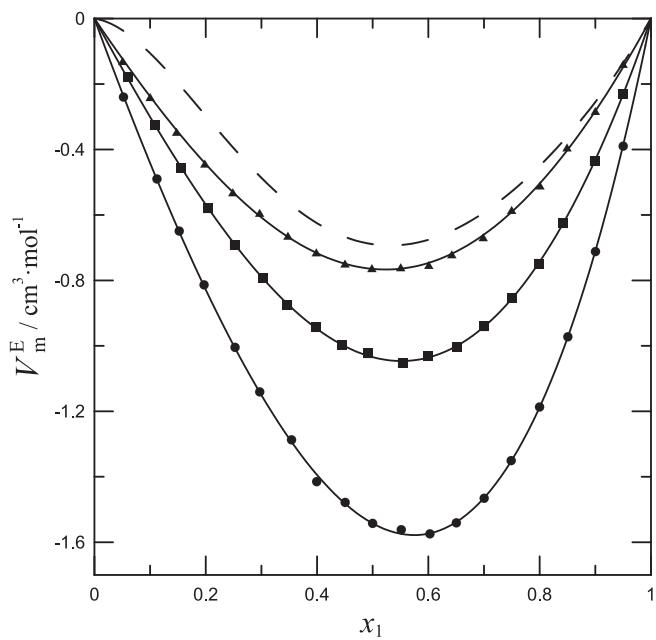


Figure 1. Excess molar volume (V_m^E) of alkan-1-ol (1) + benzylamine (2) liquid mixtures as a function of the alkan-1-ol mole fraction (x_1) at 298.15 K and 0.1 MPa. Symbols, experimental values (this work): (●), methanol; (■), propan-1-ol; (▲), pentan-1-ol. Solid lines, calculations with equation (11) using the coefficients from Table 8. Dashed line, pentan-1-ol (smoothed curve from reference [67]).

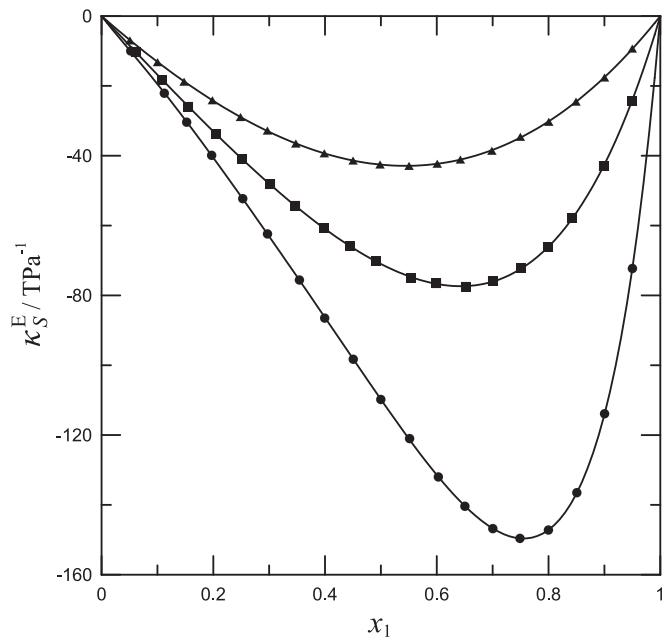


Figure 2. Excess isentropic compressibility (κ_S^E) of alkan-1-ol (1) + benzylamine (2) liquid mixtures as a function of the alkan-1-ol mole fraction (x_1) at 298.15 K and 0.1 MPa. Full symbols, experimental values (this work): (●), methanol; (■), propan-1-ol; (▲), pentan-1-ol. Solid lines, calculations with equation (11) using the coefficients from Table 8.

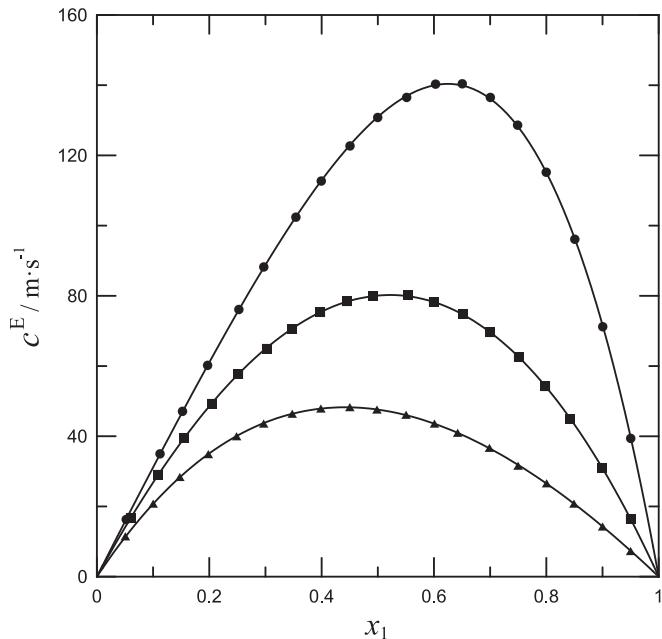


Figure 3. Excess speed of sound (c^E) of alkan-1-ol (1) + benzylamine (2) liquid mixtures as a function of the alkan-1-ol mole fraction (x_1) at 298.15 K and 0.1 MPa. Full symbols, experimental values (this work): (●), methanol; (■), propan-1-ol; (▲), pentan-1-ol. Solid lines, calculations with equation (11) using the coefficients from Table 8.

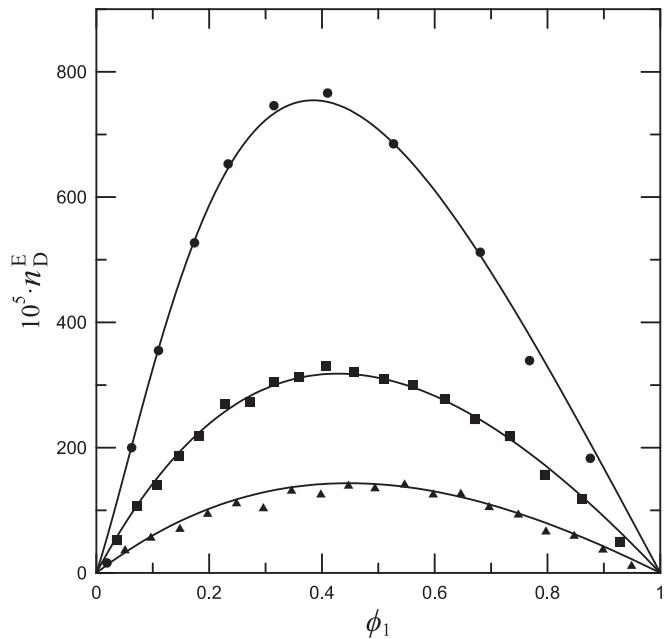


Figure 5. Excess refractive index at the sodium D-line (n_D^E) of alkan-1-ol (1) + benzylamine (2) liquid mixtures as a function of the alkan-1-ol volume fraction (ϕ_1) at 298.15 K and 0.1 MPa. Full symbols, experimental values (this work): (●), methanol; (■), propan-1-ol; (▲), pentan-1-ol. Solid lines, calculations with equation (11) using the coefficients from Table 8.

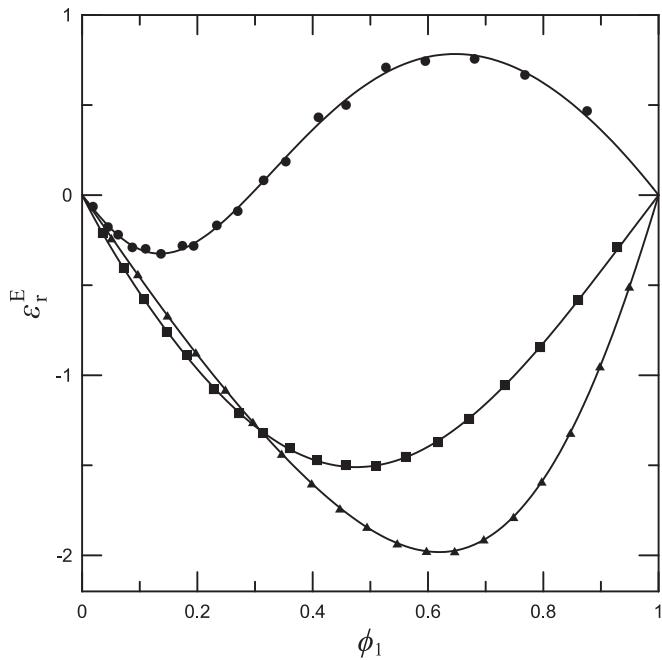


Figure 4. Excess relative permittivity (ϵ_r^E) of alkan-1-ol (1) + benzylamine (2) liquid mixtures as a function of the alkan-1-ol volume fraction (ϕ_1) at 298.15 K, 0.1 MPa, and 1 MHz. Symbols, experimental values (this work): (●), methanol; (■), propan-1-ol; (▲), pentan-1-ol. Solid lines, calculations with equation (11) using the coefficients from Table 8.

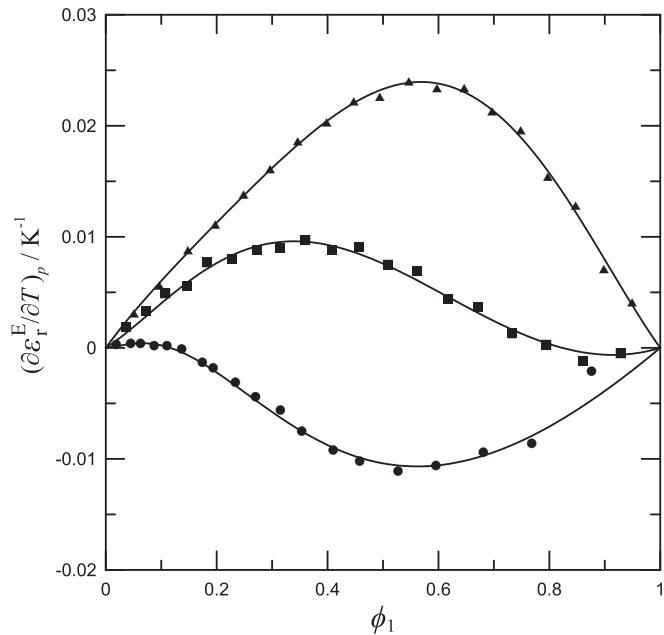


Figure 6. Temperature derivative of the excess relative permittivity $(\partial \epsilon_r^E / \partial T)_p$ of alkan-1-ol (1) + benzylamine (2) liquid mixtures as a function of the alkan-1-ol volume fraction (ϕ_1) at 298.15 K, 0.1 MPa, and 1 MHz. Full symbols, experimental values (this work): (●), methanol; (■), propan-1-ol; (▲), pentan-1-ol. Solid lines, calculations with equation (11) using the coefficients from Table 8.

5. Discussion

In this section, if nothing else is specified, the values of the thermophysical properties are given at $T = 298.15\text{ K}$ and $x_1 = 0.5$, except for dielectric properties, which will be given at $\phi_1 = 0.5$. As before, we will denote by n the number of carbon atoms of the alkan-1-ols and by $n\text{OH}$ the alkan-1-ol with n carbon atoms.

5.1. Excess molar volumes, isentropic compressibilities and speeds of sound

$V_m^E/\text{cm}^3\cdot\text{mol}^{-1}$ values of $n\text{OH} + \text{benzylamine}$ liquid mixtures (Figure 1) are negative, revealing a predominance of interactions between unlike molecules and structural effects: -1.543 (1OH), -1.035 (3OH), -0.765 (5OH). In the case of the mixture containing methanol, structural effects may be of free volume type, if one takes into account the large difference between the α_p values of the components of the system (Table 2). Values of V_m^E increase with n , a trend that may be explained by: i) a higher contribution from the rupture of benzylamine-benzylamine interactions in the mixing process by longer alkan-1-ols due to their larger aliphatic surface, and ii) a more important contribution from $n\text{OH}$ -benzylamine interactions created in mixtures with shorter alkan-1-ols, since the OH group is more sterically hindered in longer alkan-1-ols. A similar interpretation can be given to the negative values and parallel variation with n which is observed for $\kappa_S^E/\text{TPa}^{-1}$ (Figure 2): -109.8 (1OH), -71.0 (3OH), -42.6 (5OH). These $\kappa_S^E/\text{TPa}^{-1}$ values are similar to those of $n\text{OH} + \text{aniline}$ systems [15]: -109.7 (1OH), -72.5 (3OH), -37.3 (5OH). This is an interesting fact, as the $V_m^E/\text{cm}^3\cdot\text{mol}^{-1}$ values for the aniline mixtures are higher than those of benzylamine solutions: -0.902 (1OH, [15]), -0.593 (3OH, [15]), -0.398 (4OH [40]), -0.241 (5OH, [15]). The relation between V_m^E of benzylamine and aniline mixtures can be understood considering that in pure aniline there exist stronger dipolar interactions and that the breaking of such interactions contributes more positively to V_m^E . It is to be noted that large structural effects exist in alkan-1-ol + aniline systems, since from ethanol H_m^E values are positive [41,42] and V_m^E values are negative [15]. The positive $c^E/\text{m}\cdot\text{s}^{-1}$ obtained for alkan-1-ol + benzylamine (Figure 3) are consistent with the above results on κ_S^E : 131.0 (1OH), 80.1 (3OH), 47.6 (5OH).

The excess molar isobaric expansion, $A_p = (\partial V^E / \partial T)_p$, is negative for the 1OH + benzylamine mixture ($-2.7 \cdot 10^{-3} \text{ cm}^3 \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$) and very small and positive for the other two systems ($8.5 \cdot 10^{-4}$ and $1.8 \cdot 10^{-4} \text{ cm}^3 \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ for the 3OH and 5OH solutions, respectively). A similar trend is encountered for alkan-1-ol + aniline systems. Thus, $A_p/\text{cm}^3\cdot\text{mol}^{-1}\cdot\text{K}^{-1} = -2.8 \cdot 10^{-3}$ (1OH) [15,43]; $6.5 \cdot 10^{-3}$ (4OH) [40]; regarding these values, note that, as discussed in an earlier article [15], (i) they should be taken with caution because of the dispersion of the literature data, and (ii) reference [40] for the 4OH system seems to be the one that agrees best with our data. Negative A_p values have been linked to the existence of structural effects [44], but have also been interpreted in terms of a decrease in the molar volume of complex formation, which overcompensates for the decrease in the extent of complex formation, and have been encountered, e.g., in amine + trichloromethane mixtures [45,46].

5.2. Excess relative permittivities

Negative contributions to the value of ε_r^E appear when interactions between like molecules are broken, and either positive or negative ones when interactions between unlike molecules are created, depending on the effective response of the formed multimers to an electric field. Dominance of the rupture of interactions

between like molecules occurs in $n\text{OH} + \text{alkane}$ mixtures, e.g. heptane [9,47–49], or benzene [49,50]. For the latter, ε_r^E values (at 293.2 K) are: -1.28 (1OH), -2.15 (3OH), -2.78 (4OH), -2.83 (6OH), -2.23 (8OH). The values of this property for $n\text{OH} + \text{benzylamine}$ systems are: 0.524 (1OH), -1.507 (3OH), -1.856 (5OH). The benzylamine molecule can be obtained by adding a $-\text{CH}_2\text{NH}_2$ group to the phenyl ring, and the contribution from the rupture of interactions between molecules of the same species is expected to be more negative than in benzene solutions. The higher ε_r^E results for the benzylamine mixtures and the positive value for the 1OH + benzylamine mixture indicate that the contribution from the created interactions upon mixing to ε_r^E is positive. The variation of ε_r^E with n can be explained by the weaker and lower association of longer alkan-1-ols [10].

$n\text{OH} + \text{aniline}$ systems show lower negative ε_r^E values [15]: -0.778 (1OH), -1.850 (3OH), -2.082 (5OH). The ε_r^E curves of these systems, as well as those of excess molar enthalpies and excess molar internal energies at constant volume, are known to be characterized by the dominance of the rupture of the strong dipolar interactions between aniline molecules and of the alcohol network [15]. As said above, such strong dipolar interactions are not so relevant in liquid benzylamine, and this affects strongly the observed ε_r^E values. Therefore, one should expect a more important relative weight of the solvation effects between molecules of $n\text{OH}$ and benzylamine. This is also suggested by the Kirkwood-Fröhlich model applied to our data (see below), because the molecular dipoles in the benzylamine mixtures are, according to this theory, more parallelly aligned in average. Moreover, the breaking of aniline-aniline interactions by alkan-1-ol molecules should give a more negative contribution to ε_r^E than in the case of benzylamine-benzylamine interactions, which is also in accordance with the observed lower ε_r^E values of aniline systems.

The T derivative of the excess relative permittivity is negative for 1OH + benzylamine (excluding a short interval at low ϕ_1), and positive for the 3OH and 5OH mixtures. The curves are practically identical to the ones for $n\text{OH} + \text{aniline}$ systems, and an analogous interpretation can be given [15].

5.3. Molar refraction

The molar refraction, R_m , can be used to quantify the dispersive interactions [21,51], as it is proportional to the average electronic contribution to the polarizability from one molecule in a macroscopic sphere of liquid [20,21]:

$$R_m = \frac{n_D^2 - 1}{n_D^2 + 2} V_m = \frac{N_A \alpha_e}{3 \varepsilon_0} \quad (16)$$

(N_A is Avogadro's constant and ε_0 the vacuum permittivity). For $n\text{OH} + \text{benzylamine}$ mixtures at equimolar composition, $R_m/\text{cm}^3\cdot\text{mol}^{-1} = 21.1$ (1OH), 25.9 (3OH), 30.6 (5OH). They increase with n and are higher than the corresponding values in $n\text{OH} + \text{aniline}$ systems [15]: 19.5 (1OH), 24.2 (3OH), 28.9 (5OH). Dispersive interactions are therefore slightly stronger in benzylamine systems and also increase with n .

The substitution of ideal values in equation (16) allows to calculate the ideal molar refraction, R_m^{id} , and the excess value $R_m^E = R_m - R_m^{id}$. For $n\text{OH} + \text{benzylamine}$ mixtures at equimolar composition, $R_m^E/\text{cm}^3\cdot\text{mol}^{-1} = -0.44$ (1OH), -0.29 (3OH), -0.22 (5OH). Therefore, dispersive interactions are weaker than if the molecular dipoles of different species did not interact. The increase of R_m^E with n is similar to what is observed in $n\text{OH} + \text{aniline}$ mixtures [15]: -0.27 (1OH), -0.17 (3OH), -0.07 (5OH), and a similar interpretation can be given: it may be due to a more important solvation for small n values. Moreover, $R_m^E(n\text{OH} + \text{benzylamine}) < R_m^E(n\text{OH} + \text{aniline})$.

This fact may be interpreted as follows. The breaking of dipolar interactions of aniline on mixing might lead to an important positive contribution to R_m due to a corresponding increase of the freedom of the valence electrons to move, which is expected to be much less relevant in benzylamine systems because dipolar interactions do not play such a decisive role in this amine.

5.4. Kirkwood-Fröhlich model

The Kirkwood-Fröhlich model assumes that the molecules are in a spherical cavity of an infinitely large dielectric and treats macroscopically the induced contribution to the total polarizability by means of the high-frequency permittivity, ϵ_r^∞ . Long-range dipolar interactions are included in a local field in the cavity, considering its surroundings as a continuous medium of permittivity ϵ_r . The Kirkwood correlation factor, g_K , gathers information about short-range interactions by quantifying the deviations of the relative orientation of a dipole with respect to its neighbors from randomness. For a one-fluid model of a mixture of polar liquids [52], g_K can be determined using [20,21,52,53]:

$$g_K = \frac{9k_B T V_m \epsilon_0 (\epsilon_r - \epsilon_r^\infty)(2\epsilon_r + \epsilon_r^\infty)}{N_A \mu^2 \epsilon_r (\epsilon_r^\infty + 2)^2} \quad (17)$$

where k_B is Boltzmann's constant; N_A , Avogadro's constant; ϵ_0 , the vacuum permittivity; and V_m , the molar volume of the liquid at the working temperature, T . For polar compounds, ϵ_r^∞ has been estimated using $\epsilon_r^\infty = 1.1n_D^2$ [54]. μ represents the dipole moment of the solution, estimated from the equation [52]:

$$\mu^2 = x_1 \mu_1^2 + x_2 \mu_2^2 \quad (18)$$

where μ_i stands for the dipole moment of component i ($=1,2$). Calculations have been performed using smoothed values of V_m^E , n_D^E and ϵ_r^E at $\Delta x_1 = 0.01$. The source and values of μ_i used for alkan-1-ol + benzylamine mixtures are collected in Table 2.

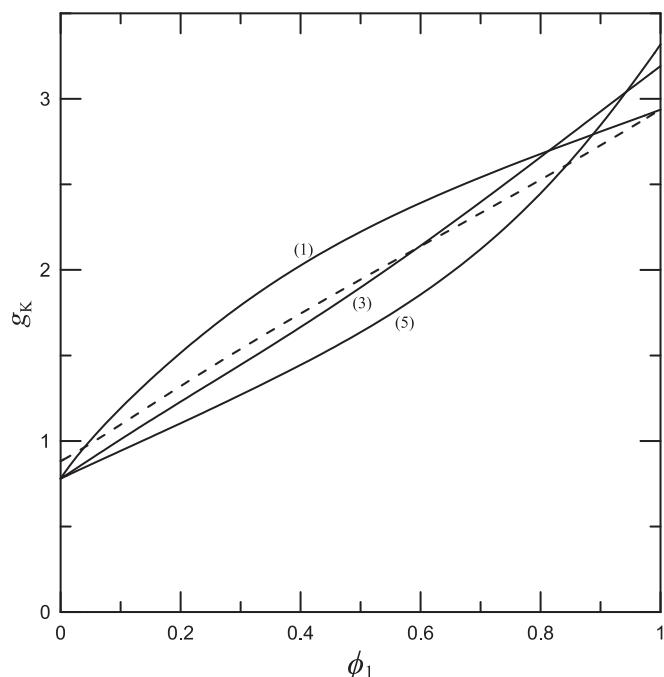


Figure 7. Kirkwood correlation factor (g_K) of alkan-1-ol (1) + amine (2) liquid mixtures as a function of the volume fraction of compound (1), ϕ_1 , at 0.1 MPa and 298.15 K: Solid lines, alkan-1-ol (1) + benzylamine (2) (this work); the numbers in parentheses indicate the number of carbon atoms of the alkan-1-ol. (— —), methanol (1) + aniline (2) ([15]).

For the mixtures $nOH + benzylamine$ under study, the g_K values decrease with n (Figure 7): 2.22 (1OH), 1.90 (3OH), 1.63 (5OH), due to a weaker and lower solvation and self-association of the alkan-1-ol when it is longer. These g_K values are somewhat higher than the ones found for the corresponding aniline solutions [15]: 1.94 (1OH), 1.72 (3OH), 1.50 (5OH). As a sample for comparison, the curve for 1OH + aniline is represented in Figure 7. Therefore, as advanced above, the Kirkwood-Fröhlich model suggests a more parallel alignment of molecular dipoles in benzylamine mixtures than in aniline systems. This is an interesting fact, pointing out to a more important relative weight of linear-like multimers against cyclic-like ones in the mixtures containing benzylamine.

6. Conclusions

Densities, speeds of sound, relative permittivities at 1 MHz and refractive indices at the sodium D-line have been measured for methanol, propan-1-ol or pentan-1-ol + benzylamine liquid mixtures at 0.1 MPa and $T = (293.15 \text{ to } 303.15) \text{ K}$. Negative values of V_m^E are found, revealing a predominance of interactions between unlike molecules and structural effects. The analysis of ϵ_r^E values reveals a positive contribution to this quantity from the creation of interactions between unlike molecules. The positive value for the methanol mixture underlines their importance in the systems under study. Alkan-1-ol + aniline systems show higher V_m^E and lower ϵ_r^E , a fact closely related to the stronger dipolar interactions between the molecules of this amine. The negative R_m^E values, which decrease with the length of the alkan-1-ol chain, indicate a loss of dispersive interactions and can be ascribed to the mentioned solvation effects. The Kirkwood-Fröhlich model supports the relevance of solvation in the dielectric properties of the mixtures, i.e., the presence linear-like multimers (especially for the methanol mixture) with dipole moments that respond effectively to an electric field and give a positive contribution to ϵ_r^E . Interestingly, the model suggests a less parallel alignment of molecular dipoles in the corresponding alkan-1-ol + aniline systems.

Funding

This work was supported by Consejería de Educación de Castilla y León, under Project VA100G19 (Apoyo a GIR, BDNS: 425389). Ana Cobos gratefully acknowledges the support from Ministerio de Educación, Cultura y Deporte through the grant FPU15/05456.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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