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Effect of temperature on density, sound velocity, refractive index and their derived properties for the binary systems (heptanoic acid + propanoic or butanoic acids)

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ABSTRACT

In this work, the effect of temperature on density (ρ), sound velocity (u), refractive index (n) and their derived properties for carboxylic acid mixtures was studied. The thermophysical properties: density, sound velocity and refractive index were measured over the entire composition range at T = (293.15, 298.15, 303.15, 308.15 and 313.15) K and at p = 0.1 MPa for the binary systems (heptanoic acid + propanoic or butanoic acids). The mass fraction of water was found to be unusually large and could not be reduced further. The Lorentz–Lorenz approximation was used to predict the density from refractive index or the refractive index from density of the binary mixtures. Sound velocity mixing rules were applied to the experimental sound velocity data. Excess molar volumes, V_m^E, isentropic compressibilities, κ_s, excess isentropic compressibilities, κ_s^E, and deviation in refractive indices, Δn, were also calculated from the experimental data. The Redlich–Kister polynomial equation was fitted to the excess properties and the deviation in refractive index data. Thermophysical properties are useful in understanding the intermolecular interactions between the components of mixtures.

1. Introduction

Carboxylic acids are important chemicals used: in a variety of industrial applications for example in: food and beverages as an acidulant, in the manufacture of polyester resins, pharmaceuticals and chemicals [1–3]. Carboxylic acids are used for the synthesis of esters and as catalysts in the synthesis of biodiesel and biodegradable polymers [1,2,4,5]. Carboxylic acids are produced via different methods either from petroleum-based feed stocks or from carbohydrates through fermentation [6].

The experimental values of thermophysical properties: density, sound velocity, refractive index, viscosity; and the thermodynamic properties: heat capacity, Gibbs energy, excess molar volume, excess molar enthalpy [7,8] allow for new correlations and/or predictive models to be developed. These properties also provide information about the intermolecular interactions that can occur in binary or ternary systems especially where hydrogen bonding occurs [9–12].

In this work, the densities, sound velocities and refractive indices for the binary systems (heptanoic acid + propanoic or butanoic acids) over the entire composition range and at T = (293.15, 298.15, 303.15, 308.15, and 313.15) K and at p = 0.1 MPa are reported. These results were used to calculate the excess molar volumes, isentropic compressibilities, excess isentropic compressibilities and deviation in refractive indices, respectively, over the entire composition range and at each temperature. The Redlich–Kister polynomial equation was fitted to the excess molar volume, excess isentropic compressibility and deviation in refractive index data. The Lorentz–Lorenz approximation was used to predict the density from refractive index or the refractive index from the density of the binary mixtures. Four sound velocity mixing rules were applied to the experimental data. This work is a continuation of our research group's studies on carboxylic acid mixtures [2,13–19].

2. Experimental

2.1. Chemicals

The propanoic acid (CAS No. 79-09-4), butanoic acid (CAS No. 107-92-6), heptanoic acid (CAS No. 111-14-8), diethyl carbonate (CAS No. 105-58-8), and ethanol (CAS No. 64-17-5), were purchased from Fluka or Aldrich. The mass fraction purity of all chemicals used in this study was ⩾0.99 and values are given in

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TABLE 1
Pure component specifications: suppliers, CAS number, specified purity and GC purity.

Chemical name	Supplier	CAS No.	Mass fraction purity	
			Initial	GC analysis
Propanoic acid	Fluka	79-09-4	⩾0.99	>0.99
Butanoic acid	Aldrich	107-92-6	⩾99.5	>0.99
Heptanoic acid	Aldrich	111-14-8	⩾99.0	>0.99
Diethyl carbonate	Aldrich	105-58-8	⩾99.0	>0.99
Ethanol	Aldrich	64-17-5	⩾99.5	>0.99

table 1. All the chemicals were stored over 0.4 nm molecular sieves to prevent the absorption of moisture. The water content mass fraction was determined by a Metrohm 702 SM Titrino titrator and found to be 0.36% in propanoic acid 0.39% in butyric acid, 0.26% in heptanoic acid, 0.50% in diethyl carbonate and 0.36% in ethanol. The purities of the chemicals were checked by comparing the experimental density, sound velocity and refractive index values of the pure chemicals with the literature values at various temperatures and show fairly well with those reported in literature [20–31] and are given in table 2. No further purification of these chemicals was carried out.

2.2. Apparatus and procedure

Binary mixtures were prepared by mass, using an OHAUS analytical balance with a precision of ±0.0001 g. The estimated error in the mole fraction was 0.0005. The standard uncertainty in pressure was ±0.04 MPa. The details of the experimental procedure can be found elsewhere [15]. A binary test system (diethyl carbonate + ethanol) [32] was done at T = 298.15 K to validate the experimental technique by the calculation of the excess molar volumes, isentropic compressibilities, excess isentropic compressibilities and deviation in refractive index and compared with

TABLE 2
Comparison of experimental density, ρ, sound velocity, u, and refractive index, n, of the pure component with the corresponding literature values at T = (293.15, 298.15, 303.15, 308.15 and 313.15) K and at pressure p = 0.1 MPa.

Component	T/K	ρ/(g · cm^{-3})		u/(m · s^{-1})		n	
		Expt.	Lit.	Expt.	Lit.	Expt.	Lit.
Propanoic acid	293.15	0.9939	0.9934[a]	1166	1162.0[c] 1176.0[d] 1142.0[e]	1.3865	1.3865[b]
	298.15	0.9885	0.9882[a]	1147	1152.4[f]	1.3848	1.3850[b]
	303.15	0.9831	0.9829[a]	1128	1121[g] 1145.0[c] 1199.3[h]	1.3830	1.3830[b]
	308.15	0.9777	0.9774[a]		1109	1.3811	1.3813[b]
	313.15	0.9723	0.9721[a]	1090	1092.0[i]	1.3791	1.3790[b]
Butanoic acid	293.15	0.9578	0.9576[a]	1196	1197.0[j] 1203.0[d] 1206.2[k]	1.3983	1.3980[b]
	298.15	0.9528	0.9528[a]	1177	1188.0[k] 1209.6[f]	1.3963	1.3963[b]
	303.15	0.9479	0.9479[a]	1158	1170.1[k] 1174[g]	1.3942	1.3950[b]
	308.15	0.9429	0.9429[a]	1139		1.3928	1.3938[b]
	313.15	0.9381	0.9379[a]	1121		1.3901	1.3921[b]
Heptanoic acid	293.15	0.9176	0.9176[b]	1295	1294.0[j] 1304.0[l] 1312.0[d] 1326.0[f]	1.4237	1.4230[b]
	298.15	0.9135	0.9143[b]	1278		1.4214	1.4212[b]
	303.15	0.9093	0.9082[a]	1260	1268.0[h] 1289.0[f]	1.4193	1.4192[b]
	308.15	0.9051	0.9040[a]	1242		1.4173	1.4174[b]
	313.15	0.9009	0.8998[a]	1225	1253.0[f]	1.4153	1.4152[b]

Standard uncertainties u are $u(T)$ = ±0.03 K, $u(p)$ = ±0.04 MPa and the combined expanded uncertainty U_c in density, sound velocity and refractive index measurements were less than $U_c(\rho)$ = ±2 · 10^{-4} g · cm^{-3}, $U_c(u)$ = ±1.0 m · s^{-1} and $U_c(n)$ = ±3 · 10^{-4}, respectively (0.95 level of confidence).

[a] Reference [20].
[b] Reference [21].
[c] Reference [22].
[d] Reference [23].
[e] Reference [24].
[f] Reference [25].
[g] Reference [26].
[h] Reference [27].
[i] Reference [28].
[j] Reference [29].
[k] Reference [30].
[l] Reference [31].

TABLE 3
Densities, ρ, sound velocity, u, refractive index, n, excess molar volume, V^{E}_{m}, deviation in isentropic compressibility, $\Delta\kappa_s$, and deviation in refractive index Δn, for the binary test system {diethyl carbonate (x_1) + ethanol (x_2)} at $T = 298.15$ K and at pressure $p = 0.1$ MPa.

x_1	$\rho/(g \cdot cm^{-3})$	$u/(m \cdot s^{-1})$	n	$V^{E}_{m}/(cm^3 \cdot mol^{-1})$	$\Delta\kappa_s/(TPa^{-1})$	Δn
0.0000	0.7852	1142	1.3594	0.000	0.0	0.0000
0.0415	0.8003	1145	1.3614	0.011	−12.8	0.0010
0.0943	0.8177	1147	1.3635	0.022	−24.4	0.0019
0.1932	0.8459	1150	1.3670	0.046	−38.1	0.0031
0.2929	0.8696	1154	1.3698	0.071	−44.2	0.0036
0.3938	0.8900	1156	1.3724	0.096	−45.0	0.0039
0.4965	0.9078	1159	1.3746	0.111	−42.0	0.0037
0.5970	0.9230	1162	1.3765	0.125	−36.5	0.0033
0.6924	0.9357	1166	1.3781	0.127	−29.9	0.0027
0.7911	0.9476	1169	1.3796	0.113	−21.9	0.0019
0.8990	0.9593	1173	1.3811	0.074	−11.7	0.0009
0.9413	0.9636	1175	1.3817	0.046	−7.0	0.0005
1.0000	0.9693	1177	1.3825	0.000	0.0	0.0000

Standard uncertainties u are $u(x_1) = 0.0005$, $u(T) = \pm 0.03$ K, $u(p) = \pm 0.04$ MPa and the combined expanded uncertainty U_c in density, sound velocity and refractive index measurements were less than $U_c(\rho) = \pm 2 \cdot 10^{-4}$ g · cm^{-3}, $U_c(u) = \pm 1.0$ m · s^{-1} and $U_c(n) = \pm 3 \cdot 10^{-4}$, respectively (0.95 level of confidence).

literature values. The results for the test system are given in table 3. The difference between experimental and literature excess molar volumes, isentropic compressibilities, excess isentropic compressibilities and deviation in refractive index for the test system was within the experimental error. The estimated error in density, sound velocity and refractive index was less than $\pm 2 \cdot 10^{-4}$ g · cm^{-3}, ± 1.0 m · s^{-1}, and $\pm 3 \cdot 10^{-4}$, respectively.

Density and sound velocity of pure components and binary mixtures were measured using a digital vibrating-tube densimeter and sound velocity analyzer (Anton Paar DSA 5000M) with an accuracy of ±0.02 K in temperature. The sound velocity was measured at a frequency of approximately 3 MHz [33]. The details regarding sound velocity measurement using the Anton Paar DSA 5000M are described in a previous study in literature [33]. Measurement of the refractive index of pure components and binary mixtures were obtained by a digital automatic refractometer (Anton Paar RXA 156) with an accuracy of ±0.03 K in temperature. The calibration for the DSA 5000M together with RXA 156 was done by measuring the density, sound velocity and refractive index of Anton Paar ultrapure water and dry air at $T = 298.15$ K and checked every week. The estimated error in excess molar volume, isentropic compressibility, excess isentropic compressibility and deviation in refractive index is ± 0.003 cm^3 · mol^{-1}, ± 1 TPa^{-1}, ± 0.6 TPa^{-1}, and ±0.00008, respectively.

3. Results and discussion

3.1. Thermophysical properties

3.1.1. Density

The thermophysical properties ρ, u and n were measured over the entire composition range at $T = (293.15, 298.15, 303.15, 308.15$ and $313.15)$ K and at $p = 0.1$ MPa for the binary systems (heptanoic acid + propanoic or butanoic acids) and are given in tables 4 and 5. It was found that the ρ values for all the binary systems decreases with an increase in both temperature and composition.

The framework of the Lorentz–Lorenz approximation was used to predict the density from the refractive index data. The predictive expression for ρ can be obtained within the framework of the Lorentz–Lorenz approximation [34] was given in equation (1):

$$\rho = \frac{\left(\frac{n^2-1}{n^2+2}\right)(x_1M_1 + x_2M_2)}{\left(\frac{n_1^2-1}{n_1^2+2}\right)\frac{x_1M_1}{\rho_1} + \left(\frac{n_2^2-1}{n_2^2+2}\right)\frac{x_2M_2}{\rho_2}}. \quad (1)$$

The root mean square deviation (*rmsd*) calculated from equation (1) is given in table 6. The maximum root mean square deviation for the density of the mixtures is 0.00069 g · cm^{-3} at $T = 313.15$ K and occurred for (heptanoic acid + propanoic acid). The root mean square deviation shows that density predicted from the refractive index gave good results for both binary systems.

3.1.2. Sound velocity

Sound velocity data provides information about (solvent + solvent), (ion + solvent) and (ion + ion) interactions in mixture [35]. From tables 4 and 5, it can be seen that the u values decreases as the temperature increases for both binary systems. The values of u increase with an increase in the composition of heptanoic acid at all temperatures but decreases with temperature at a fixed composition.

Sound velocity mixing rules were applied to calculate the sound velocity of the binary mixture from pure component data. The mixing rules used were the equation of Rao [36] equation (2), Wada [37] equation (3), Nomoto [38] equation (4), and Berryman [39] equation (5). Values of the sound velocity calculated from the above four sound velocity mixing rules are given below:

$$u^{1/3}V_m = \sum_{i=1}^{2} u_i^{1/3}x_iV_i \therefore u = \left(\sum_{i=1}^{2} u_i^{1/3}x_iV_i \Big/ V_m\right)^3, \quad (2)$$

$$\kappa_s^{-1/7}V_m = \sum_{i=1}^{2} \kappa_{s,i}^{-1/7}x_iV_i \therefore u = \left(\sum_{i=1}^{2} u_i^{2/7}\rho_i^{1/7}x_iV_i \Big/ (\rho^{1/7}V_m)\right)^{7/2}, \quad (3)$$

$$u = \left(\sum_{i=1}^{2} \phi_i u_i^{1/3}\right)^3, \quad (4)$$

$$\kappa_s = \sum_{i=1}^{2} \phi_i\kappa_{s,i} \therefore u = \left(\rho\sum_{i=1}^{2} \phi_i\kappa_{s,i}\right)^{-1/2}, \quad (5)$$

where V_i, u_i, x_i, and ρ_i are the molar volume, sound velocity, mole fraction and density of the pure component i, respectively. The root mean square deviation (*rmsd*) for the calculated speed of sound data is given in table 7. These results showed that the Rao and Wada mixing rules gave better correlation while the Nomoto and Berryman mixing rules gave larger *rmsd* values. The largest root mean square deviation was obtained using the Nomoto mixing rule for the (heptanoic acid + propanoic acid) system at $T = 293.15$ K.

TABLE 4
Densities, ρ, excess molar volume, V^{E}_{m}, sound velocity, u, isentropic compressibility, $\kappa_{s,}$ excess isentropic compressibility, κ^{E}_{s}, refractive index, n, and deviation in refractive index, Δn, for the binary system {heptanoic acid (x_1) + propanoic acid (x_2)} at T = (293.15, 298.15, 303.15, 308.15 and 313.15) K and at pressure p = 0.1 MPa.

x_1	ρ/(g · cm^{-3})	V^{E}_{m}/(cm^3 · mol^{-1})	u/(m · s^{-1})	κ_s/(TPa^{-1})	κ^{E}_{s}/(TPa^{-1})	n	Δn
			T = 293.15 K				
0.0000	0.9939	0.000	1166	741	0.0	1.3865	0.0000
0.0539	0.9858	0.049	1176	733	1.1	1.3900	0.0015
0.1023	0.9793	0.080	1185	727	2.2	1.3929	0.0026
0.2037	0.9675	0.134	1202	715	3.6	1.3982	0.0042
0.3021	0.9578	0.162	1217	705	4.0	1.4026	0.0050
0.4007	0.9496	0.171	1231	695	3.9	1.4066	0.0053
0.5015	0.9423	0.169	1244	685	3.5	1.4102	0.0051
0.6051	0.9359	0.150	1257	677	2.9	1.4135	0.0046
0.7009	0.9307	0.122	1267	669	2.1	1.4162	0.0039
0.8016	0.9258	0.093	1277	662	1.6	1.4188	0.0028
0.9002	0.9215	0.053	1287	656	0.8	1.4212	0.0015
0.9500	0.9195	0.026	1291	652	0.4	1.4223	0.0007
1.0000	0.9177	0.000	1295	649	0.0	1.4234	0.0000
			T = 298.15 K				
0.0000	0.9885	0.000	1147	769	0.0	1.3848	0.0000
0.0539	0.9805	0.050	1158	761	1.1	1.3880	0.0012
0.1023	0.9742	0.081	1166	755	2.2	1.3909	0.0023
0.2037	0.9625	0.136	1184	742	3.6	1.3961	0.0038
0.3021	0.9530	0.165	1199	730	4.1	1.4006	0.0047
0.4007	0.9449	0.173	1213	719	4.0	1.4045	0.0050
0.5015	0.9377	0.171	1226	709	3.6	1.4081	0.0049
0.6051	0.9311	0.152	1239	700	3.0	1.4114	0.0044
0.7009	0.9263	0.123	1249	692	2.2	1.4142	0.0037
0.8016	0.9214	0.094	1260	684	1.6	1.4169	0.0027
0.9002	0.9172	0.054	1269	677	0.9	1.4192	0.0014
0.9500	0.9153	0.026	1273	674	0.4	1.4203	0.0007
1.0000	0.9135	0.000	1278	671	0.0	1.4214	0.0000
			T = 303.15 K				
0.0000	0.9831	0.000	1128	800	0.0	1.3830	0.0000
0.0539	0.9752	0.051	1139	791	1.1	1.3859	0.0009
0.1023	0.9690	0.083	1148	784	2.3	1.3888	0.0020
0.2037	0.9575	0.138	1165	770	3.8	1.3940	0.0035
0.3021	0.9481	0.167	1180	757	4.2	1.3985	0.0044
0.4007	0.9402	0.175	1195	745	4.2	1.4024	0.0048
0.5015	0.9332	0.174	1208	735	3.7	1.4060	0.0047
0.6051	0.9269	0.154	1220	724	3.1	1.4094	0.0043
0.7009	0.9219	0.125	1231	716	2.3	1.4121	0.0036
0.8016	0.9171	0.095	1242	707	1.7	1.4148	0.0026
0.9002	0.9130	0.054	1251	700	0.9	1.4171	0.0014
0.9500	0.9111	0.026	1256	696	0.4	1.4183	0.0007
1.0000	0.9093	0.000	1260	693	0.0	1.4194	0.0000
			T = 308.15 K				
0.0000	0.9777	0.000	1109	832	0.0	1.3811	0.0000
0.0539	0.9700	0.053	1120	822	1.1	1.3838	0.0007
0.1023	0.9638	0.085	1129	814	2.4	1.3866	0.0017
0.2037	0.9525	0.141	1146	799	4.0	1.3919	0.0033
0.3021	0.9433	0.170	1162	785	4.4	1.3963	0.0042
0.4007	0.9355	0.178	1176	773	4.4	1.4003	0.0046
0.5015	0.9286	0.176	1190	761	3.9	1.4039	0.0046
0.6051	0.9224	0.156	1202	750	3.2	1.4073	0.0042
0.7009	0.9175	0.126	1213	740	2.4	1.4101	0.0035
0.8016	0.9128	0.097	1224	732	1.8	1.4127	0.0026
0.9002	0.9088	0.055	1233	724	1.0	1.4151	0.0014
0.9500	0.9069	0.027	1238	720	0.4	1.4162	0.0007
1.0000	0.9051	0.000	1242	716	0.0	1.4173	0.0000
			T = 313.15				
0.0000	0.9723	0.000	1090	865	0.0	1.3792	0.0000
0.0539	0.9647	0.054	1101	855	1.1	1.3816	0.0005
0.1023	0.9586	0.087	1110	846	2.4	1.3845	0.0016
0.2037	0.9475	0.143	1128	829	4.1	1.3897	0.0032
0.3021	0.9384	0.173	1144	815	4.7	1.3942	0.0041
0.4007	0.9308	0.181	1158	801	4.5	1.3982	0.0045
0.5015	0.9240	0.179	1172	788	4.1	1.4018	0.0045
0.6051	0.9180	0.158	1185	776	3.4	1.4052	0.0041
0.7009	0.9131	0.128	1196	766	2.5	1.4080	0.0035
0.8016	0.9085	0.098	1206	757	1.8	1.4107	0.0025
0.9002	0.9045	0.055	1216	748	1.0	1.4131	0.0013
0.9500	0.9027	0.027	1220	744	0.4	1.4142	0.0007
1.0000	0.9009	0.000	1225	740	0.0	1.4153	0.0000

Standard uncertainties u are $u(x_1)$ = 0.0005, $u(T)$ = ±0.03 K, $u(p)$ = ±0.04 MPa and the combined expanded uncertainty U_c in density, sound velocity and refractive index measurements were less than $U_c(\rho)$ = ±2 · 10^{-4} g · cm^{-3}, $U_c(u)$ = ±1.0 m · s^{-1} and $U_c(n)$ = ±3 · 10^{-4}, respectively (0.95 level of confidence).

TABLE 5
Densities, ρ, excess molar volume, V^{E}_{m}, sound velocity, u, isentropic compressibility, $\kappa_{s,}$ excess isentropic compressibility, κ^{E}_{s}, refractive index, n, and deviation in refractive index, Δn, for the binary system {heptanoic acid (x_1) + butanoic acid (x_2)} at T = (293.15, 298.15, 303.15, 308.15 and 313.15) K and at pressure p = 0.1 MPa.

x_1	ρ/(g · cm^{-3})	V^{E}_{m}/(cm^3 · mol^{-1})	u/(m · s^{-1})	κ_s/(TPa^{-1})	κ^{E}_{s}/(TPa^{-1})	n	Δn
			T = 293.15 K				
0.0000	0.9578	0.000	1196	730	0.0	1.3983	0.0000
0.0559	0.9542	0.019	1203	724	−0.3	1.4005	0.0008
0.1059	0.9513	0.033	1209	719	−0.5	1.4024	0.0012
0.2061	0.9458	0.058	1222	709	−1.2	1.4054	0.0019
0.3087	0.9408	0.070	1233	699	−1.9	1.4084	0.0023
0.4037	0.9367	0.074	1244	690	−2.2	1.4109	0.0025
0.5037	0.9327	0.074	1254	682	−2.2	1.4134	0.0025
0.6006	0.9292	0.066	1263	675	−2.2	1.4157	0.0023
0.7049	0.9258	0.055	1272	668	−1.9	1.4179	0.0019
0.8050	0.9228	0.039	1280	661	−1.4	1.4199	0.0014
0.9052	0.9201	0.016	1289	655	−1.0	1.4217	0.0007
0.9575	0.9187	0.008	1292	652	−0.4	1.4226	0.0003
1.0000	0.9177	0.000	1295	649	0.0	1.4234	0.0000
			T = 298.15 K				
0.0000	0.9528	0.000	1177	757	0.0	1.3963	0.0000
0.0559	0.9493	0.023	1185	751	−0.5	1.3984	0.0006
0.1059	0.9464	0.037	1191	745	−0.8	1.4001	0.0011
0.2061	0.9410	0.062	1203	734	−1.5	1.4033	0.0018
0.3087	0.9362	0.073	1215	723	−2.1	1.4063	0.0022
0.4037	0.9321	0.076	1225	714	−2.4	1.4089	0.0024
0.5037	0.9282	0.076	1236	706	−2.4	1.4114	0.0024
0.6006	0.9248	0.070	1245	698	−2.3	1.4136	0.0022
0.7049	0.9215	0.058	1254	690	−1.9	1.4159	0.0018
0.8050	0.9185	0.041	1263	683	−1.5	1.4179	0.0013
0.9052	0.9159	0.019	1271	676	−1.0	1.4197	0.0007
0.9575	0.9145	0.010	1275	673	−0.5	1.4207	0.0003
1.0000	0.9135	0.000	1278	671	0.0	1.4214	0.0000
			T = 303.15 K				
0.0000	0.9479	0.000	1158	787	0.0	1.3942	0.0000
0.0559	0.9444	0.028	1166	779	−0.6	1.3963	0.0006
0.1059	0.9416	0.042	1172	773	−1.1	1.3980	0.0011
0.2061	0.9363	0.065	1185	761	−1.7	1.4012	0.0017
0.3087	0.9315	0.076	1197	749	−2.3	1.4042	0.0022
0.4037	0.9276	0.080	1207	740	−2.5	1.4068	0.0024
0.5037	0.9238	0.079	1217	731	−2.5	1.4093	0.0024
0.6006	0.9204	0.073	1227	722	−2.4	1.4116	0.0022
0.7049	0.9171	0.062	1236	714	−2.0	1.4138	0.0018
0.8050	0.9142	0.044	1245	706	−1.5	1.4158	0.0013
0.9052	0.9116	0.022	1253	699	−1.0	1.4177	0.0007
0.9575	0.9103	0.012	1257	695	−0.5	1.4186	0.0003
1.0000	0.9093	0.000	1260	693	0.0	1.4194	0.0000
			T = 308.15 K				
0.0000	0.9429	0.000	1139	817	0.0	1.3922	0.0000
0.0559	0.9395	0.030	1147	809	−0.8	1.3942	0.0005
0.1059	0.9367	0.045	1154	802	−1.4	1.3958	0.0010
0.2061	0.9315	0.069	1167	789	−2.0	1.3991	0.0017
0.3087	0.9269	0.078	1179	777	−2.6	1.4021	0.0021
0.4037	0.9230	0.082	1189	766	−2.7	1.4047	0.0023
0.5037	0.9193	0.082	1199	756	−2.6	1.4072	0.0023
0.6006	0.9160	0.075	1209	747	−2.5	1.4095	0.0022
0.7049	0.9128	0.064	1218	738	−2.1	1.4117	0.0018
0.8050	0.9100	0.047	1227	730	−1.5	1.4138	0.0013
0.9052	0.9074	0.024	1235	722	−1.0	1.4157	0.0007
0.9575	0.9061	0.014	1239	719	−0.5	1.4166	0.0003
1.0000	0.9051	0.000	1242	716	0.0	1.4173	0.0000
			T = 313.15 K				
0.0000	0.9381	0.000	1121	849	0.0	1.3901	0.0000
0.0559	0.9346	0.033	1129	840	−0.9	1.3920	0.0005
0.1059	0.9319	0.046	1136	832	−1.7	1.3937	0.0009
0.2061	0.9268	0.070	1148	818	−2.3	1.3970	0.0016
0.3087	0.9223	0.080	1161	805	−2.8	1.4000	0.0021
0.4037	0.9185	0.083	1171	794	−3.0	1.4026	0.0023
0.5037	0.9148	0.082	1182	783	−2.8	1.4051	0.0023
0.6006	0.9116	0.077	1191	773	−2.6	1.4074	0.0021
0.7049	0.9084	0.066	1201	764	−2.1	1.4097	0.0018
0.8050	0.9057	0.048	1209	755	−1.6	1.4117	0.0013
0.9052	0.9031	0.025	1218	747	−1.1	1.4136	0.0007
0.9575	0.9019	0.015	1222	743	−0.5	1.4146	0.0003
1.0000	0.9009	0.000	1225	740	0.0	1.4153	0.0000

Standard uncertainties u are $u(x_1)$ = 0.0005, $u(T)$ = ±0.03 K, $u(p)$ = ±0.04 MPa and the combined expanded uncertainty U_c in density, sound velocity and refractive index measurements were less than $U_c(\rho) = \pm 2 \cdot 10^{-4}$ g · cm^{-3}, $U_c(u) = \pm 1.0$ m · s^{-1} and $U_c(n) = \pm 3 \cdot 10^{-4}$, respectively (0.95 level of confidence).

TABLE 6
Root mean square deviation, *rmsd*, between the experimental and the predicted density, ρ, or refractive index, n, of the binary systems at different temperatures and at pressure $p = 0.1$ MPa.

Properties	T/K = 293.15	*rmsd* (ρ and n)			
		298.15	303.15	308.15	313.15
{Heptanoic acid (x_1) + propanoic acid (x_2)}					
ρ/(g · cm^{-3})	0.0006	0.0002	0.0002	0.0005	0.0007
N	0.0003	0.0001	0.0001	0.0002	0.0003
{Heptanoic acid (x_1) + butanoic acid (x_2)}					
ρ/(g · cm^{-3})	0.0003	0.0002	0.0002	0.0001	0.0001
N	0.0002	0.0001	0.0001	0.0001	0.0001

Standard uncertainties u are $u(T) = \pm 0.03$ K, $u(p) = \pm 0.04$ MPa and the combined expanded uncertainty U_c in density and refractive index measurements were less than $U_c(\rho) = \pm 2 \cdot 10^{-4}$ g · cm^{-3}, and $U_c(n) = \pm 3 \cdot 10^{-4}$, respectively (0.95 level of confidence).

TABLE 7
Root mean square deviation, *rmsd*, in sound velocity, u, at different temperatures and at pressure $p = 0.1$ MPa for the studied mixing rules.

Equation	T/K = 293.15	*rmsd* (u)			
		298.15	303.15	308.15	313.15
{Heptanoic acid (x_1) + propanoic acid (x_2)}					
Rao	0.2	0.3	0.3	0.3	0.3
Wada	0.8	0.8	0.8	0.8	0.8
Nomoto	4.2	4.1	4.1	4.1	4.0
Berryman	3.2	3.1	3.0	2.9	2.8
{Heptanoic acid (x_1) + butanoic acid (x_2)}					
Rao	0.2	0.1	0.3	0.4	0.7
Wada	0.1	0.2	0.5	0.6	0.9
Nomoto	1.9	1.7	1.5	1.4	1.3
Berryman	1.1	0.9	0.7	0.5	0.3

Standard uncertainties u are $u(T) = \pm 0.03$ K, $u(p) = \pm 0.04$ MPa and the combined expanded uncertainty U_c in sound velocity was less than $U_c(u) = \pm 1.0$ m · s^{-1} (0.95 level of confidence).

3.1.3. Refractive index

Refractive index data are given in tables 4 and 5 and show an increase with an increase in the composition of heptanoic acid and decreases as the temperature increases for both binary systems.

The inverse predictive expression for n obtained from equation (1) within the framework of the Lorentz–Lorenz approximation [34] was used to predict the refractive index from the density and is given in equation (6):

$$n = \left(\frac{2\left[\left(\frac{n_1^2-1}{n_1^2+2}\right)x_1\rho\frac{M_1}{\rho_1} + x_2\left(\frac{n_2^2-1}{n_2^2+2}\right)\rho\frac{M_2}{\rho_2}\right] + [x_1M_1 + x_2M_2]}{[x_1M_1 + x_2M_2] - \left[\left(\frac{n_1^2-1}{n_1^2+2}\right)x_1\rho\frac{M_1}{\rho_1} + x_2\left(\frac{n_2^2-1}{n_2^2+2}\right)\rho\frac{M_2}{\rho_2}\right]} \right)^{1/2}. \quad (6)$$

The root mean square deviation (*rmsd*) is also presented in table 6. The maximum root mean square deviation value is 0.0003 for the (heptanoic acid + propanoic acid) system at $T = 313.15$ K. The root mean square deviations values show that refractive index predicted from density gave good results for both systems. Therefore the Lorentz–Lorenz approximation is the most suitable equation for the prediction of density from the refractive index or refractive index from density.

3.2. Derived properties

3.2.1. Excess molar volume

The extent of deviation of liquid mixtures from ideal behavior is expressed by excess functions. The excess molar volumes can be interpreted in three areas, namely physical, chemical, and structural effects. The excess molar volumes, V_m^E, for the binary systems were calculated from the experimental density of the mixture and the pure components using equation (7):

$$V_m^E = \frac{x_1M_1 + x_2M_2}{\rho} - \frac{x_1M_1}{\rho_1} - \frac{x_2M_2}{\rho_2}, \quad (7)$$

where x_1 and x_2 are mole fractions; M_1 and M_2 denote molar masses; ρ_1 and ρ_2 are the densities; where 1 refers to heptanoic acid and 2 refers to propanoic acid or butanoic acid, and ρ is the density of the binary mixtures.

The results of excess molar volume, V_m^E, for the binary systems (heptanoic acid + propanoic acid or butanoic acid) are given in tables 4 and 5 and also plotted in figure 1(a) and (b), respectively. The V_m^E values for both binary systems are positive and depend on composition and temperature as shown in figure 1(a) and (b). The positive contribution is due to the breakdown of intramolecular hydrogen bonds between the heptanoic or propanoic or butanoic acids monomers [40]. The V_m^E value for (heptanoic acid + propanoic acid) > (heptanoic acid + butanoic acid) and is possibly due to the additional CH_2 group on butanoic acid resulting in weaker intramolecular hydrogen bonding in the latter binary system. The $V_{m,max}^E$ values occurred at $x_1 = 0.4007$ and 0.4037, respectively, at each temperature and increased slightly with an increase in temperature for both the systems.

3.2.2. Isentropic compressibilities, deviation in isentropic compressibilities and excess isentropic compressibilities

The isentropic compressibilities, κ_s, were calculated using the Newton–Laplace equation (8):

$$\kappa_s = \frac{1}{\rho u^2}, \quad (8)$$

where ρ is the density and u is the sound velocity of the binary mixtures.

The deviation in isentropic compressibility, $\Delta\kappa_s$ (ϕ), was calculated using the equation (9):

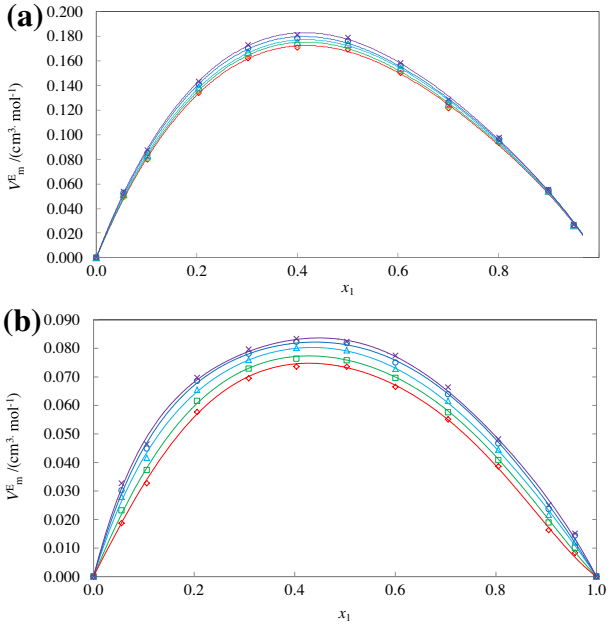

FIGURE 1. Plot of excess molar volumes, V_m^E, of binary mixtures of (a) {heptanoic acid (x_1) + propanoic acid (x_2)} and (b) {heptanoic acid (x_1) + butanoic acid (x_2)} against mole fraction of heptanoic acid at $T = 293.15$ K (◊), 298.15 K (□), 303.15 K (△), 308.15 K (○) and 313.15 K (×). The solid lines were generated using Redlich–Kister curve-fitting.

TABLE 8
Heat capacity, C_p, values of pure component at several temperatures and at pressure $p = 0.1$ MPa used in this work.

T/K	293.15	298.15	303.15	308.15	313.15
		Propanoic acid			
C_p/J · K^{-1} · kmol^{-1}	1.521 · 10^{5}[a]	1.538 · 10^{5}[a]	1.554 · 10^{5}[a]	1.571 · 10^{5}[a]	1.587 · 10^{5}[a]
		Butanoic acid			
C_p/J · K^{-1} · kmol^{-1}	1.761 · 10^{5}[a]	1.781 · 10^{5}[a]	1.800 · 10^{5}[a]	1.820 · 10^{5}[a]	1.839 · 10^{5}[a]
		Heptanoic acid			
C_p/J · K^{-1} · kmol^{-1}	1.594 · 10^{5}[a]	1.620 · 10^{5}[a]	1.644 · 10^{5}[a]	1.670 · 10^{5}[a]	1.695 · 10^{5}[a]

[a] Reference [42].

$$\Delta\kappa_s(\phi) = \kappa_s - \sum_i^2 \phi_i \kappa_{s,i}, \tag{9}$$

where $\kappa_{s,i}$ and ϕ_i are the isentropic compressibility and volume fractions of the pure component i, respectively. The volume fractions were calculated assuming an ideal mixture.

The excess isentropic compressibility [41] for the binary system was calculated using equation (10):

$$\kappa_s^E = \Delta\kappa_s(\phi) - T\left[\sum_i \phi_i \frac{(\alpha^*_{p,i})^2}{\sigma^*_{p,i}} - \frac{\left(\sum_i \phi_i \alpha^*_{p,i}\right)^2}{\sum_i \phi_i \sigma^*_{p,i}}\right], \tag{10}$$

where $\sigma^*_{p,i} = C_{p,m}/V_m$ is the heat capacitance or heat capacity per unit volume of the mixture, since heat capacities were not measured in this work, heat capacity values from literature was used or calculated from literature values at the experimental temperature [42] and are given in table 8. The thermal expansion coefficients defined as $\alpha^*_{p,i} = 1/V_m(\delta V_m/\delta T)_P = -1/\rho(\delta\rho/\delta T)_P$ has been calculated at each temperature from the experimental density [41]. The results for isentropic compressibility, κ_s, and excess isentropic compressibility, κ_s^E, for the binary systems (heptanoic acid + propanoic acid or butanoic acid) at $T = (293.15, 298.15, 303.15, 308.15$ and $313.15)$ K are given in tables 4 and 5.

The isentropic compressibility values, κ_s increases with an increase in temperature at a fixed composition due to an increase in thermal agitation making the solution more compressible [43]. The κ_s values decrease with an increase in composition of heptanoic acid at a fixed temperature for all the binary systems. At a particular temperature, the (heptanoic acid + propanoic acid) solution is more compressible than (heptanoic acid + butanoic acid) solution due to the more compact propanoic acid.

The excess isentropic compressibility κ_s^E for both the binary systems is plotted in figure 2(a) and (b). From the figure 2(a) and (b), it can be seen that the values of κ_s^E are positive for the system (heptanoic acid + propanoic acid) and negative for the system (heptanoic acid + butanoic acid). In general the κ_s^E values increase with an increase in temperature for the system (heptanoic acid + propanoic acid) and decrease for the system (heptanoic acid + butanoic acid) at a fixed composition of heptanoic acid are shown in tables 4 and 5. The κ_s^E values show that the

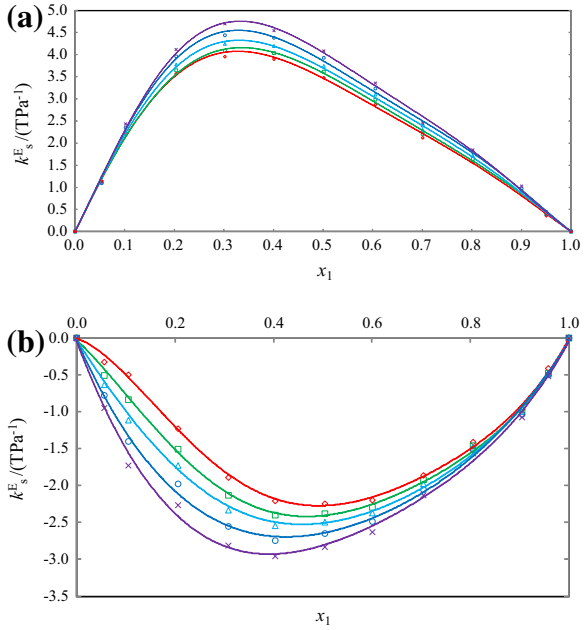

FIGURE 2. Plot of excess isentropic compressibility, κ_s^E, of binary mixtures of (a) {heptanoic acid (x_1) + propanoic acid (x_2)} and (b) {heptanoic acid (x_1) + butanoic acid (x_2)} against mole fraction of heptanoic acid at $T = 293.15$ K (◊), 298.15 K (□), 303.15 K (△), 308.15 K (○) and 313.15 K (×). The solid lines were generated using Redlich–Kister curve-fitting.

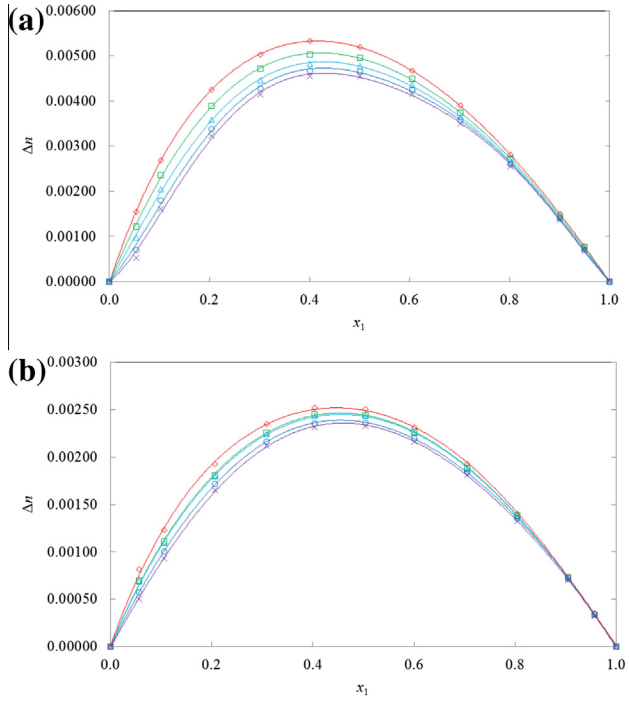

FIGURE 3. Plot of the deviation in refractive index, Δn, of binary mixtures of (a) {heptanoic acid (x_1) + propanoic acid (x_2)} and (b) {heptanoic acid (x_1) + butanoic acid (x_2)} against mole fraction of heptanoic acid at $T = 293.15$ K (◊), 298.15 K (□), 303.15 K (△), 308.15 K (○) and 313.15 K (×). The solid lines were generated using Redlich–Kister curve-fitting.

TABLE 9
Coefficients A_i, and standard deviations, σ, obtained for the {heptanoic acid (x_1) + propanoic or butanoic acid (x_2)} at different temperatures and at pressure $p = 0.1$ MPa for the Redlich–Kister equation.

	T/K	A_0	A_1	A_2	A_3	A_4	σ
				{Heptanoic acid (x_1) + propanoic acid (x_2)}			
V_m^E/(cm^3 · mol^{-1})	293.15	0.671	0.226	0.053	−0.047	0.073	0.002
	298.15	0.679	0.234	0.060	−0.053	0.069	0.002
	303.15	0.689	0.231	0.054	−0.031	0.079	0.002
	308.15	0.698	0.241	0.062	−0.036	0.093	0.002
	313.15	0.709	0.246	0.058	−0.027	0.097	0.002
κ_s^E/(TPa^{-1})	293.15	13.9	11.5	7.7	−3.9	−6.3	0.1
	298.15	14.3	11.6	6.6	−4.6	−5.2	0.1
	303.15	14.8	12.0	8.4	−4.5	−7.9	0.1
	308.15	15.4	12.9	9.6	−5.9	−9.3	0.1
	313.15	16.2	13.6	9.5	−6.6	−10.9	0.1
Δn	293.15	0.02100	0.00636	0.00334	0.00216	0.00023	0.00001
	298.15	0.02000	0.00581	0.00297	−0.00053	−0.00257	0.00004
	303.15	0.01900	0.00529	0.00279	−0.00261	−0.00541	0.00004
	308.15	0.01900	0.00514	0.00333	−0.00508	−0.00899	0.00006
	313.15	0.01800	0.00495	0.00343	−0.00659	−0.01100	0.00008
				{Heptanoic acid (x_1) + butanoic acid (x_2)}			
V_m^E/(cm^3 · mol^{-1})	293.15	0.292	0.079	0.059	0.025	−0.120	0.001
	298.15	0.303	0.073	0.062	0.055	−0.053	0.001
	303.15	0.318	0.064	0.045	0.090	0.049	0.001
	308.15	0.326	0.066	0.060	0.094	0.080	0.001
	313.15	0.331	0.059	0.065	0.113	0.104	0.002
κ_s^E/(TPa^{-1})	293.15	−9.1	−0.4	2.4	5.4	−1.1	0.1
	298.15	−9.8	−1.4	2.9	4.5	−5.1	0.1
	303.15	−10.2	−1.6	1.7	2.4	−5.1	0.1
	308.15	−10.7	−2.6	−0.1	0.8	−4.8	0.1
	313.15	−11.5	−4.0	−0.1	1.4	−7.1	0.1
Δn	293.15	0.01000	0.00184	0.00009	0.00182	0.00244	0.00003
	298.15	0.00981	0.00171	0.00007	0.00076	0.00096	0.00002
	303.15	0.00977	0.00167	−0.00013	0.00070	0.00120	0.00001
	308.15	0.00947	0.00172	0.00031	−0.00051	−0.00026	0.00001
	313.15	0.00932	0.00165	0.00038	−0.00095	−0.00143	0.00001

Standard uncertainties u are $u(x_1) = 0.0005$, $u(T) = \pm 0.03$ K, $u(p) = \pm 0.04$ MPa and the combined expanded uncertainty U_c in density, sound velocity and refractive index measurements were less than $U_c(\rho) = \pm 2 \cdot 10^{-4}$ g · cm^{-3}, $U_c(u) = \pm 1.0$ m · s^{-1} and $U_c(n) = \pm 3 \cdot 10^{-4}$, respectively (0.95 level of confidence).

(heptanoic + propanoic acid) is more compressible than (heptanoic acid + butanoic acid) solution. The κ_s^E, maximum and κ_s^E, minimum values occur at $x_1 = 0.3021$ and 0.4037, respectively, for both systems at all temperatures.

3.2.3. Deviation in refractive index

The deviation in refractive index, Δn, was calculated using equation (11):

$$\Delta n = n - x_1 n_1 - x_2 n_2, \tag{11}$$

where n_1 and n_2 are the refractive index of pure components and n is the refractive index of the mixtures. The results for deviation in refractive index, Δn, for the binary systems (heptanoic acid + propanoic acid or butanoic acid) at $T = (293.15, 298.15, 303.15, 308.15$ and $313.15)$ K are given in tables 4 and 5, and plotted in figure 3(a) and (b), respectively. The Δn values are positive for both the systems at each temperature. In general, Δn values decrease with an increase in temperature at fixed composition for both systems. The Δn_{max} values occur at $x_1 = 0.4007$ and 0.4037, respectively at each temperature and decrease with an increase in temperature for both systems.

3.3. Correlation

The Redlich–Kister polynomial equation given below [44] was fitted to V_m^E, κ_s^E, and Δn data

$$X = x_1 x_2 \sum_{i=1}^{k} A_i (1 - 2x_1)^{i-1}, \tag{12}$$

where X is excess molar volumes, V_m^E, or excess isentropic compressibility, κ_s^E, or deviation in refractive index, Δn. The coefficients of A_i are the parameters that were obtained by fitting the equation to the experimental data with least-squares method using commercial software (MathCAD).

The standard deviation σ is defined as:

$$\sigma(X) = \sum_{i=1}^{N} \left[\frac{(X_{expt} - X_{calc})^2}{(N - k)} \right]^{1/2}, \tag{13}$$

where N is the number of experimental points and k is the number of coefficients used in the Redlich–Kister equation. Redlich–Kister parameters, A_i, determined by fitting the experimental data to equation (12) together with the standard deviation, σ, for the binary systems (heptanoic acid + propanoic acid or butanoic acid) at $T = (293.15, 298.15, 303.15, 308.15$ and $313.15)$ K are presented in table 9.

4. Conclusions

In this work, new experimental data for density, sound velocity, and refractive index over the entire composition range at $T = (293.15, 298.15, 303.15, 308.15$ and $313.15)$ K and at atmospheric pressure for the binary systems (heptanoic acid + propanoic acid or butanoic acid) are reported. The Lorentz–Lorenz approximation predicted the refractive index from density more accurately. Four sound velocity mixing rules were applied and the Rao and Wada mixing rules gave better correlation than the Nomoto and Berryman mixing rules better correlation results. Further, the derived properties such as excess molar volume, isentropic compressibility, excess isentropic compressibility and deviation in refractive index were also calculated. The Redlich–Kister polynomial equation was fitted to the excess molar volume,

excess isentropic compressibility and the deviation in refractive index and provides an excellent description for all systems studied. The value of excess molar volume, excess isentropic compressibility and deviation in refractive index are positive for all the systems except for the excess isentropic compressibility for the (heptanoic acid + butanoic acid) system.

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