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Excess/deviation properties of binary mixtures of 2,5-dimethylfuran with furfuryl alcohol, methyl isobutyl ketone, 1-butanol and 2-butanol at temperature range of (293.15–323.15) K

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Abstract. Experimental values of density and speed of sound for binary liquid mixtures of 2,5-dimethylfuran (2,5-DMF) with furfuryl alcohol (FA), methyl isobutyl ketone (MIBK), 1-butanol and 2-butanol and over the entire composition range of 2,5-DMF and at the temperature range of 293.15–323.15 K at 10 K intervals and at pressure $p = 0.1$ MPa were reported. Experimental data were used to assess the thermodynamics properties of studied mixtures. These properties were used to interpret the molecular interactions among component of liquids. The values of excess/deviation functions have been fitted to Redlich−Kister type polynomial equation. From the obtained results, a discussion was carried out in terms of nature of intermolecular interactions and structure factors in the binary mixtures.

1 Introduction

Nowadays, fossil energies such as petroleum, natural gas and coal dominate approximately more than 80% of primary energy consumption estimated by sources [1]. This excessive reliance on fossil fuels in the world has serious reservations about their depletion and green house emission. Now, with increasing the demand for environmental concerns about global warming, the development of eco-friendly and renewable energy sources have been a most important topic in the last few years [2,3]. The energy-efficient processes for the sustainable production of fuels derived from biomass such as biodiesel fuel (BDF), bio-ethanol and liquid alkanes have also attracted attention over the year [4].

5-hydroxymethylfurfural (HMF), obtained by dehydration of monosaccharides, is considered to be most promising important intermediate for the synthesis of a wide variety of chemicals and alternative fuels based on bio-refinery [5]. To upgrade furanic compounds toward bio-fuels, hydrogenation is the most versatile reaction. Among several furan-based biofuel candidates, which include 2,5-dimethylfuran (2,5-DMF), 2-methylfuran (2-MF), 5-ethoxymethylfurfural (EMF) and ethyl levulinate (EL), 2,5-dimethylfuran is known as one of the potential transportation fuels because of its high energy density ($30\,\mathrm{kJ\cdot cm^{-3}}$) togther octane number (RON = 119), these values are similar to the gasoline which also have high energy density ($34\mathrm{ckJ\cdot cm^{-3}}$) together octane number (RON = 96). Moreover, DMF is nearly immiscible with water and thus easier to blend with gasoline than ethanol.

To explore the application of DMF as a fuel or as a gasoline additive, it is necessary to characterize its thermophysical properties including density, sound velocity, refractive indices, viscosity, surface tension and vapor–liquid equilibrium as a pure fluid as well as mixed with hydrocarbons or other gasoline additives. In the face of their importance, experimental and theoretical investigations concerning key properties are scarce and limited to narrow experimental conditions.

For the case of binary systems {2,5-dimethylfuran (2,5-DMF) + FA or MIBK or 1-butanol, or 2-butanol}, neither excess molar volumes (V_m^E), nor isentropic compressibility (ks) data have been previously reported. Consequently, and as continuation of our systematic studies on thermodynamic and thermophysical properties of binary mixtures containing solvents derived from biomass, the

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Table 1. Molar mass, CAS number, suppliers and purities of chemicals used in this study.

	Compound				
	2,5-DMF	Furfuryl alcohol	MIBK	1-Butanol	2-Butanol
Supplier	Sigma-Aldrich	Sigma-Aldrich	Fluka	Biochem	Sigma-Aldrich
*Purity mass fraction	0.990	0.980	0.990	0.995	0.990
M ($g\,mol^{-1}$)	96.13	98.10	100.16	74.12	74.12
CAS number	625-86-5	98-00-0	108-10-1	71-36-3	78-92-2
Lot#	WXBB6542V	MKBP8421V	SZBF1820V	–	STBG2236V

* No further purification was done.

present work is undertaken to measure new experimental data of densities, sound velocity, refractive indices of pure DMF and of the binary systems {2,5-dimethylfuran (2,5-DMF) + FA or MIBK or 1-butanol, or 2-butanol} over the entire composition range at (293.15, 303.15, 313.15 and 323.15) K and at pressure $p = 0.1$ MPa. Reliable density and speed of sound data for the measured systems are needed for optimized design of several industrial processes such as separation, storage, mixing processes, and transport. These data also used to develop accurate empirical equations, models and simulation programs. The results from these studies can provide valuable information about fluid at different temperature conditions including room temperatures to higher temperatures at 50 °C.

2 Experimental details

2.1 Chemicals

2,5-dimethylfuran (2,5-DMF), FA and 2-butanol were purchased from *Sigma Aldrich*, while 1-butanol was from *Biochem* and MIBK was from *Fluka*. The purity of these chemicals were declared to be more than 0.99 on mass fraction basis. The source and the purity of the utilized chemicals are shown in Table 1. As well the purity was checked by comparing the measured densities, speed of sound and refractive index, which are in good accord with literature values [6–65] and are presented in Table 2. All chemicals were kept in bottles to avoid contamination and evaporation during mixing.

2.2 Apparatus and procedure

The binary mixtures were prepared by mass measurement using an OHAUS analytical mass balance with a precision of ±0.0001 g. The uncertainty in the mole fraction was ±0.0005. Density and speed of sound for pure components and their binary mixtures were measured using a digital vibrating-tube densimeter and sound velocity analyzer (Anton Paar DSA 5000M) with uncertainty of ±0.02 K in temperature. The speed of sound was measured using a propagation time technique with frequency of 3 MHz. The estimated uncertainty in density and speed of sound were ±0.003 $g{\cdot}cm^{-3}$ and ±1.2 $m{\cdot}s^{-1}$, respectively. The refractive indices of the pure liquids used in the present work were measured using an Abbe digital refractometer (Model Abbemat 300, Anton Paar), with uncertainty of ±0.02 K in temperature. The measured values of the refractive indices using the method and apparatus were estimated to be ±0.005 of their true values.

3 Results and discussion

3.1 Density

The values of density ρ were measured at (293.15, 303.15, 313.15 and 323.15) K, and at pressure $p = 0.1$ MPa for the binary systems {2,5-dimethylfuran (2,5-DMF) + FA or MIBK or 1-butanol, or 2-butanol} and are given in Table 3. The plots of density *versus* concentration at investigated temperatures are given in Figure S1 (a)–(d). From Figure S1 (a)–(d), it can be seen that the ρ value decreases with an increase in temperature, and increases with an increase in concentration for all investigated binary systems, except for the system containing FA whereas a decreasing value of ρ with an increase in concentration was observed.

3.2 Speed of sound

The measurement of speed of sound, u, has been successfully employed in understanding the nature of molecular interactions in pure liquids and their liquid mixtures [66–68]. Speed of sound measurements are highly sensitive to molecular interactions and can be used to provide qualitative information about the physical nature and strength of molecular interaction in liquid mixtures [66–68]. In this regards, the speed of sound data, were also measured in the same conditions for all binary systems and are also given in Table 3. The plots of speed of sound *versus* concentration, at investigated temperatures, are given in Figure S2 (a)–(d). From Figure S2 (a)–(d), it can be seen that the u value also decreases with an increase in temperature, and in general, decreases with an increase in concentration for all binary systems excluding the {2,5-dimethylfuran + MIBK) system, whereas an increasing value of u with an increase in concentration was observed. Figure S2 (a)–(d), representing the variation of u as a mole fraction of 2,5-dimethylfuran for the systems containing FA or MIBK or 1-butanol, or 2-butanol, shows that at a given temperature, the curve of u as a function of x_1 has a maximum of $x_1 = 0.6773$ for MIBK whereas a minimum of $x_1 = 0.9004$ for FA, $x_1 = 0.7944$ for 1-butanol and $x_1 = 0.6860$ for 2-butanol. The minimum of x_1 observed for the systems whereas u value decreases with an increase in concentration while maximum of x_1 observed for the systems whereas an increasing value of u with increasing concentration was observed.

Table 2. Comparison of experimental density, ρ, sound velocity, u, and refractive indices, n_D, of the pure component with the corresponding literature values at 293.15, 298.15, 303.15, 313.15 and 323.15 K and at pressure $p = 0.1$ MPa.

Component	T (K)	ρ ($g \cdot cm^{-3}$)		u($m \cdot s^{-1}$)		n_D	
		Exp	Lit.	Exp.	Lit.	Exp.	Lit.
2,5- DMF	293.15	0.901	0.90118 [7] 0.90300 [8] 0.901016 [11]	1214.3	–	1.442	1.4415 [11]
	298.15	0.896	0.89563 [6] 0.89579 [7] 0.89588 [9] 0.89654 [10] 0.895633 [11]	1193.1	–	1.439	1.44012 [6] 1.44040 [7] 1.44016 [9] 1.44012 [10] 1.4388 [11]
	303.15	0.890	0.89037 [7] 0.890220 [11]	1171.2	–	1.436	1.4361 [11]
	313.15	0.879	0.87943 [7] 0.879290 [11]	1128.4	–	1.431	1.4305 [11]
	323.15	0.868	0.86836 [7] 0.868221 [11]	1086.0	–	1.425	1.4250 [11]
Furfuryl alcohol	293.15	1.133	1.13226 [25] 1.13200 [26] 1.134904 [27] 1.12850 [28]	1465.9	1464.4 [25] 1466.11 [27]	1.487	–
	298.15	1.128	1.12499 [12] 1.1260 [13]	1449.8	–	1.485	–
	303.15	1.124	1.12247 [12] 1.1238 [13] 1.12299 [25] 1.12247 [29] 1.125616 [27]	1433.6	1431.9 [25] 1.433.65 [27]	1.483	–
	313.15	1.114	1.11363 [25] 1.116244 [27]	1401.7	1400.0 [25] 1401.71 [27]	1.479	–
	323.15	1.105	–	1370.0	–	1.475	–
1- butanol	293.15	0.810	0.8098 [19] 0.80954 [22] 0.8098 [23] 0.80965 [32] 0.8095 [33] 0.809530 [34]	1258.6	1257.5 [32] 1257 [33] 1256.8 [34] 1257.66 [35]	1.399	–

Table 2. (continued).

Component	T (K)	ρ ($g \cdot cm^{-3}$)		u($m \cdot s^{-1}$)		n_D	
		Exp	Lit.	Exp.	Lit.	Exp.	Lit.
	298.15	0.806	0.805877 [11] 0.80584 [14] 0.8070 [15] 0.80575 [16] 0.8055 [17] 0.8060 [18,19] 0.87071 [20] 0.80589 [30] 0.80577 [31] 0.8057 [34] 0.80607 [36] 0.80548 [37] 0.80581 [38] 0.80601 [39] 0.8055 [40]	1241.8	1239.8 [34] 1240.37 [37] 1240.25 [38]	1.397	1.3973 [11] 1.3969 [14] 1.39732 [30] 1.3971 [31] 1.3969 [36] 1.3973 [39] 1.39716 [20] 1.3973 [40]
	303.15	0.802	0.8037 [15] 0.8021 [19] 0.80206 [20] 0.8022 [21] 0.80200 [32] 0.8019 [34] 0.802407 [41]	1224.9	1223.6 [32] 1224 [42] 1223.55 [43] 1222.9 [34]	1.395	1.395280 [41]
	313.15	0.795	0.7967 [15] 0.7943 [19] 0.79421 [32] 0.7946 [45,46]	1191.6	1190.2 [32] 1190 [42] 1190.28 [43]	1.391	–
	323.15	0.787	0.7864 [19] 0.7861 [34]	1158.4	1156.5 [34]	1.387	–
2-butanol	293.15	0.806	0.8073 [32] 0.8067 [42] 0.8063 [34]	1229.8	1232.4 [32] 1230 [42] 1230.1 [34]	1.397	–
	298.15	0.802	0.80235 [32] 0.80260 [31] 0.80250 [36] 0.80239 [47] 0.80241 [18] 0.80235 [48] 0.8022 [34]	1212.0	1230.18 [18] 1212.1 [34]	1.395	1.39519 [30] 1.3949 [31] 1.3951 [42] 1.3952 [49] 1.3947 [47] 1.39503 [18] 1.39530 [48]
	303.15	0.798	0.79899 [32] 0.7984 [42] 0.7980 [34]	1193.9	1196.1 [32] 1195 [42] 1194 [34]	1.393	–

Table 2. (continued).

Component	T (K)	ρ (g·cm^{-3})		u(m·s^{-1})		n_D	
		Exp	Lit.	Exp.	Lit.	Exp.	Lit.
	313.15	0.789	0.79028 [32] 0.7895 [42] 0.7893 [34] 0.78965 [49]	1157.5	1159.5 [32] 1157 [42] 1157.6 [34]	1.389	–
	323.15	0.780	0.7802 [34] 0.7805549 [49]	1120.5	1120.6 [34]	1.384	–
MIBK	293.15	0.801	0.8007 [56] 0.8007 [57] 0.8010 [58] 0.8008 [59] 0.80083 [65]	1211.3	1212.3 [65]	1.396	1.3959 [56] 1.3958 [58] 1.3962 [60] 1.39593 [65]
	298.15	0.796	0.79594 [30] 0.79640 [31]	1191.2	–	1.393	1.39355 [30] 1.3933 [31]
	303.15	0.791	0.79169 [50] 0.7917 [51] 0.7913 [43] 0.7913 [42,13] 0.79163 [54] 0.7916 [56] 0.7920 [58] 0.79100 [61] 0.78986 [62] 0.798 [63] 0.7916 [65] 0.79191 [55] 0.79163 [65]	1171.0	1175.0 [50] 1175 [51] 1180.0 [53] 1172 [13] 1129.35 [63] 1170 [64] 1171.4 [65]	1.391	1.3912 [56] 1.3915 [58] 1.3919 [61] 1.3912 [62] 1.39124 [65]
	313.15	0.782	0.7826 [51] 0.7870 [54] 0.7822 [56] 0.7823 [57] 0.78237 [65]	1131.1	1138.0 [51] 1131.4 [65]	1.386	1.3865 [56] 1.38645 [65]
	323.15	0.773	–	1091.6	–	1.382	–

Standard uncertainties u are $u(T) = \pm 0.02$ K, $u(p) = \pm 0.04$ MPa and the combined expanded uncertainty Uc in mole fraction, density, sound velocity and refractive index were $Uc(x) = \pm 0.0005$, $Uc(\rho) = \pm 0.003$ g·cm^{-3}, $Uc(u) = \pm 1.2$ m·s^{-1} and $Uc(n) = \pm 0.005$ respectively, (0.95 level of confidence).

3.3 Excess molar volumes

The excess molar volumes,V_m^E, were calculated from the density data of the mixture and the pure components using Equation (1):

$$V_m^E = \frac{x_1 M_1 + x_2 M_2}{\rho} - \frac{x_1 M_1}{\rho_1} - \frac{x_2 M_2}{\rho_2}. \quad (1)$$

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 2,5-dimethylfuran and 2 refers to FA or 1-butanol or 2-butanol or MIBK, and ρ is the density of the mixtures. Table S1 represents the results of excess molar volume, V_m^E, for the studied system and is also plotted in Figure S1 (a)–(d). The V_m^E values are positive for the systems (2,5-dimethylfuran + 1-butanol, or 2-butanol) and negative for the

Table 3. Densities, ρ, sound velocity, u, and isentropic compressibility, κs, for the binary systems {2,5-DMF (1)+FA (2), or MIBK (2), or 1-butanol (2), or 2-butanol (2)} at (293.15, 303.15, 313.15 and 323.15) K and at pressure $p = 0.1$ MPa.

x_1	$\rho/(\mathrm{g.cm^{-3}})$	$u/(\mathrm{ms^{-1}})$	κ_s $/(10^{12} \times \mathrm{Pa^{-1}})$
{2,5-DMF (1)+FA (2)}			
$T = 293.15$ K			
0.0000	1.133	1465.9	410.7
0.0996	1.107	1430.0	441.7
0.1996	1.081	1395.2	475.2
0.2964	1.057	1363.4	509.1
0.4031	1.031	1331.2	547.6
0.4954	1.009	1305.9	581.3
0.5973	0.985	1280.5	618.8
0.7022	0.962	1258.4	656.2
0.7916	0.943	1242.3	687.0
0.9004	0.921	1226.6	721.9
1.0000	0.901	1214.3	752.7
$T = 303.15$ K			
0.0000	1.124	1433.6	432.9
0.0996	1.098	1396.9	466.9
0.1996	1.071	1361.1	503.8
0.2964	1.047	1328.3	541.4
0.4031	1.021	1295.1	584.1
0.4954	0.999	1269.0	621.8
0.5973	0.975	1242.7	664.0
0.7022	0.952	1219.6	706.2
0.7916	0.933	1202.7	741.4
0.9004	0.910	1185.7	781.6
1.0000	0.890	1171.2	818.9
$T = 313.15$ K			
0.0000	1.114	1401.7	456.7
0.0996	1.088	1363.9	494.1
0.1996	1.062	1327.1	534.8
0.2964	1.037	1293.5	576.4
0.4031	1.011	1259.3	623.7
0.4954	0.988	1232.3	666.2
0.5973	0.965	1205.3	713.5
0.7022	0.941	1181.2	761.3
0.7916	0.922	1163.3	801.6
0.9004	0.899	1145.0	848.3
1.0000	0.879	1128.4	893.2
$T = 323.15$ K			
0.0000	1.105	1370.0	482.1
0.0996	1.078	1331.3	523.2
0.1996	1.052	1293.6	568.2
0.2964	1.027	1259.1	614.3
0.4031	1.000	1223.9	667.4
0.4954	0.978	1196.1	714.7
0.5973	0.954	1168.2	767.9
0.7022	0.931	1143.2	822.2
0.7916	0.911	1124.4	868.2
0.9004	0.888	1104.6	922.9

Table 3. (continued).

x_1	$\rho/(\mathrm{g.cm^{-3}})$	$u/(\mathrm{ms^{-1}})$	$\kappa_s\ /(10^{12}\times\mathrm{Pa^{-1}})$
1.0000	0.868	1086.0	976.6
{2,5-DMF (1)+MIBK (2)}			
$T=293.15\,\mathrm{K}$			
0.0000	0.801	1211.3	851.3
0.1015	0.810	1213.2	838.7
0.2002	0.819	1214.8	827.0
0.3044	0.829	1216.3	815.0
0.3976	0.839	1217.2	805.0
0.5000	0.849	1217.5	795.0
0.6004	0.859	1217.5	785.7
0.6779	0.867	1217.5	778.3
0.7885	0.878	1216.6	769.3
0.8536	0.885	1216.0	764.2
0.9276	0.893	1214.8	758.8
1.0000	0.901	1214.3	752.7
$T=303.15\,\mathrm{K}$			
0.0000	0.791	1171.0	921.4
0.1015	0.801	1172.6	908.3
0.2002	0.810	1173.8	896.2
0.3044	0.820	1174.9	883.8
0.3976	0.829	1175.5	873.4
0.5000	0.839	1175.5	863.1
0.6004	0.848	1175.2	853.4
0.6779	0.857	1175.0	845.7
0.7885	0.868	1173.8	836.4
0.8536	0.875	1173.1	831.0
0.9276	0.882	1171.8	825.4
1.0000	0.890	1171.2	818.9
$T=313.15\,\mathrm{K}$			
0.0000	0.782	1131.1	999.3
0.1015	0.791	1132.2	985.9
0.2002	0.800	1133.1	973.3
0.3044	0.810	1133.8	960.5
0.3976	0.819	1134.0	949.8
0.5000	0.828	1133.7	939.1
0.6004	0.838	1133.1	929.2
0.6779	0.846	1132.7	921.1
0.7885	0.857	1131.3	911.6
0.8536	0.864	1130.4	906.0
0.9276	0.871	1129.1	900.1
1.0000	0.879	1128.4	893.2
$T=323.15\,\mathrm{K}$			
0.0000	0.773	1091.6	1085.9
0.1015	0.782	1092.3	1072.1
0.2002	0.790	1092.8	1059.1
0.3044	0.800	1093.1	1046.1
0.3976	0.809	1093.0	1035.0
0.5000	0.818	1092.4	1024.1
0.6004	0.828	1091.5	1014.0

Table 3. (continued).

x_1	$\rho/(\mathrm{g.cm^{-3}})$	$u/(\mathrm{ms^{-1}})$	$\kappa_s\ /(10^{12}\times \mathrm{Pa^{-1}})$
0.6779	0.836	1090.9	1005.6
0.7885	0.846	1089.2	995.8
0.8536	0.853	1088.2	990.0
0.9276	0.861	1086.8	983.9
1.0000	0.868	1086.0	976.6
{2,5-DMF (1)+1-butanol (2)}			
$T=293.15\,\mathrm{K}$			
0.0000	0.810	1258.6	779.3
0.1053	0.821	1248.4	781.5
0.2013	0.831	1239.6	783.6
0.3027	0.840	1231.1	785.3
0.3969	0.849	1223.9	786.4
0.5099	0.859	1216.6	786.5
0.6051	0.867	1211.4	785.8
0.6947	0.875	1208.0	783.4
0.7944	0.883	1206.0	778.5
0.8938	0.891	1207.4	769.5
1.0000	0.901	1214.3	752.7
$T=303.15\,\mathrm{K}$			
0.0000	0.802	1224.9	830.6
0.1053	0.813	1213.2	835.8
0.2013	0.822	1203.3	840.2
0.3027	0.831	1193.7	844.2
0.3969	0.840	1185.6	847.3
0.5099	0.849	1177.4	849.4
0.6051	0.857	1171.6	849.9
0.6947	0.865	1167.6	848.4
0.7944	0.873	1165.2	844.0
0.8938	0.881	1166.1	835.0
1.0000	0.890	1171.2	818.9
$T=313.15\,\mathrm{K}$			
0.0000	0.795	1191.6	886.4
0.1053	0.804	1178.4	895.0
0.2013	0.813	1167.3	902.3
0.3027	0.822	1156.6	909.2
0.3969	0.830	1147.6	914.6
0.5099	0.840	1138.5	919.0
0.6051	0.847	1132.1	921.1
0.6947	0.854	1127.6	920.6
0.7944	0.862	1124.8	916.9
0.8938	0.870	1125.1	908.0
1.0000	0.879	1128.4	893.2
$T=323.15\,\mathrm{K}$			
0.0000	0.787	1158.4	947.3
0.1053	0.796	1143.8	960.1
0.2013	0.805	1131.6	970.8
0.3027	0.813	1119.9	980.9
0.3969	0.821	1110.1	989.0
0.5099	0.830	1100.1	996.2

Table 3. (continued).

x_1	$\rho/(\mathrm{g.cm^{-3}})$	$u/(\mathrm{ms^{-1}})$	κ_s $/(10^{12}\times \mathrm{Pa^{-1}})$
0.6051	0.837	1093.1	1000.1
0.6947	0.844	1088.2	1001.0
0.7944	0.851	1084.8	998.1
0.8938	0.859	1084.6	989.7
1.0000	0.868	1086.0	976.6
{2,5-DMF (1)+2-butanol (2)}			
$T=293.15\,\mathrm{K}$			
0.0000	0.806	1229.8	819.9
0.1033	0.817	1220.7	821.7
0.1972	0.826	1213.3	822.6
0.2955	0.835	1206.7	822.4
0.3920	0.844	1201.7	820.4
0.5124	0.855	1197.4	815.8
0.5938	0.862	1196.0	810.6
0.6860	0.871	1196.3	802.3
0.7963	0.881	1199.0	789.6
0.8957	0.890	1204.4	774.4
1.0000	0.901	1214.3	752.7
$T=303.15\,\mathrm{K}$			
0.0000	0.798	1193.9	879.0
0.1033	0.808	1183.3	884.0
0.1972	0.817	1174.9	887.1
0.2955	0.825	1167.5	888.8
0.3920	0.834	1161.9	888.0
0.5124	0.845	1157.2	884.0
0.5938	0.852	1155.6	878.9
0.6860	0.860	1155.6	870.3
0.7963	0.870	1157.9	857.0
0.8957	0.879	1163.0	840.8
1.0000	0.890	1171.2	818.9
$T=313.15\,\mathrm{K}$			
0.0000	0.789	1157.5	945.4
0.1033	0.799	1145.6	954.1
0.1972	0.807	1136.3	959.7
0.2955	0.816	1128.2	963.3
0.3920	0.824	1122.3	963.6
0.5124	0.834	1117.2	960.3
0.5938	0.842	1115.4	955.1
0.6860	0.850	1115.2	946.4
0.7963	0.859	1117.1	932.4
0.8957	0.868	1121.6	915.3
1.0000	0.879	1128.4	893.2
$T=323.15\,\mathrm{K}$			
0.0000	0.780	1120.5	1020.6
0.1033	0.789	1107.5	1033.1
0.1972	0.797	1097.6	1041.3
0.2955	0.805	1089.1	1046.9
0.3920	0.814	1082.8	1048.4

Table 3. (continued).

x_1	$\rho/(\mathrm{g.cm^{-3}})$	$u/(\mathrm{ms^{-1}})$	$\kappa_s\ /(10^{12}\times \mathrm{Pa^{-1}})$
0.5124	0.824	1077.5	1045.6
0.5938	0.831	1075.6	1040.5
0.6860	0.839	1075.1	1031.5
0.7963	0.848	1076.6	1016.9
0.8957	0.857	1080.5	999.1
1.0000	0.868	1086.0	976.6

Standard uncertainties u are $u(T) = \pm 0.02\,\mathrm{K}$, $u(p) = \pm 0.04\,\mathrm{MPa}$ and the combined expanded uncertainty Uc in mole fraction, density and sound velocity were $Uc(x) = \pm 0.0005$, $Uc(\rho) = \pm 0.003\,\mathrm{g\cdot cm^{-3}}$ and $Uc(u) = \pm 1.2\,\mathrm{m\cdot s^{-1}}$, respectively, (0.95 level of confidence).

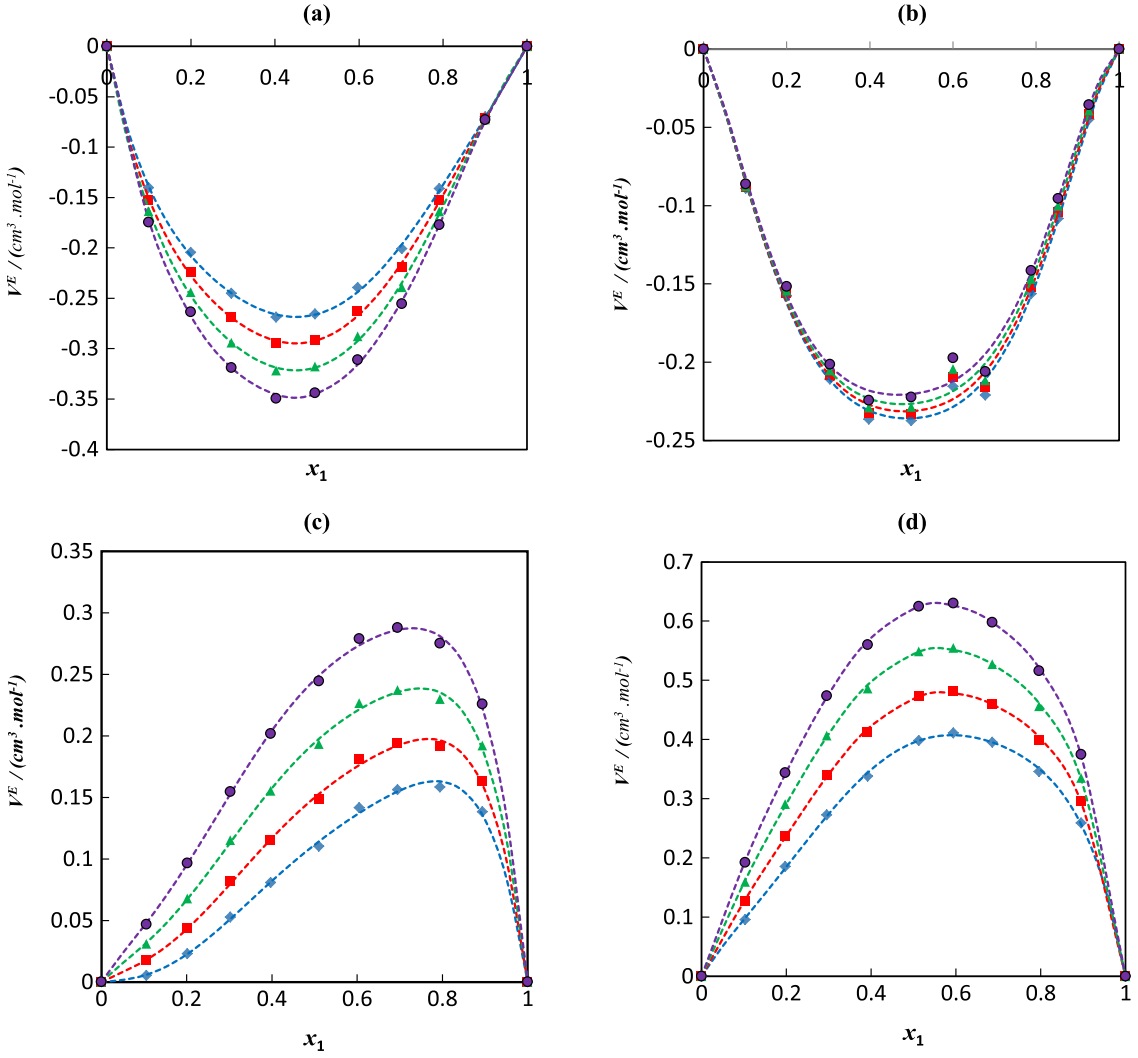

Fig. 1. Plot of excess molar volumes, V_m^E, for the binary mixtures: (a) {2,5-DMF (1)+FA (2)}; (b) {2,5-DMF (1)+MIBK (2)}; (c) {2,5-DMF (1)+1-butanol (2)} and (d) {2,5-DMF (1)+2-butanol (2)} as function of the composition expressed in the mole fraction at 293.15 K (♦); 303.15 K (■); 313.15 K (▲) and 323.15 K (●). The dotted lines were generated using Redlich-Kister polynomial curve-fitting.

systems (2,5-dimethylfuran + MIBK, or FA). The positive V_m^E values can be explained by (i) mutual loss of dipolar association due to addition of the 1-butanol or 2-butanol and contributions due to difference in size and shape of the components in the mixtures, and (ii) dipole–dipole and dipole-induced dipole interaction between unlike molecules. The first factor contributes to expansion in volume and second factor contributes to decrease in volume, which will cause contraction in volume. The experimental results in this work suggested that the factors responsible for expansion

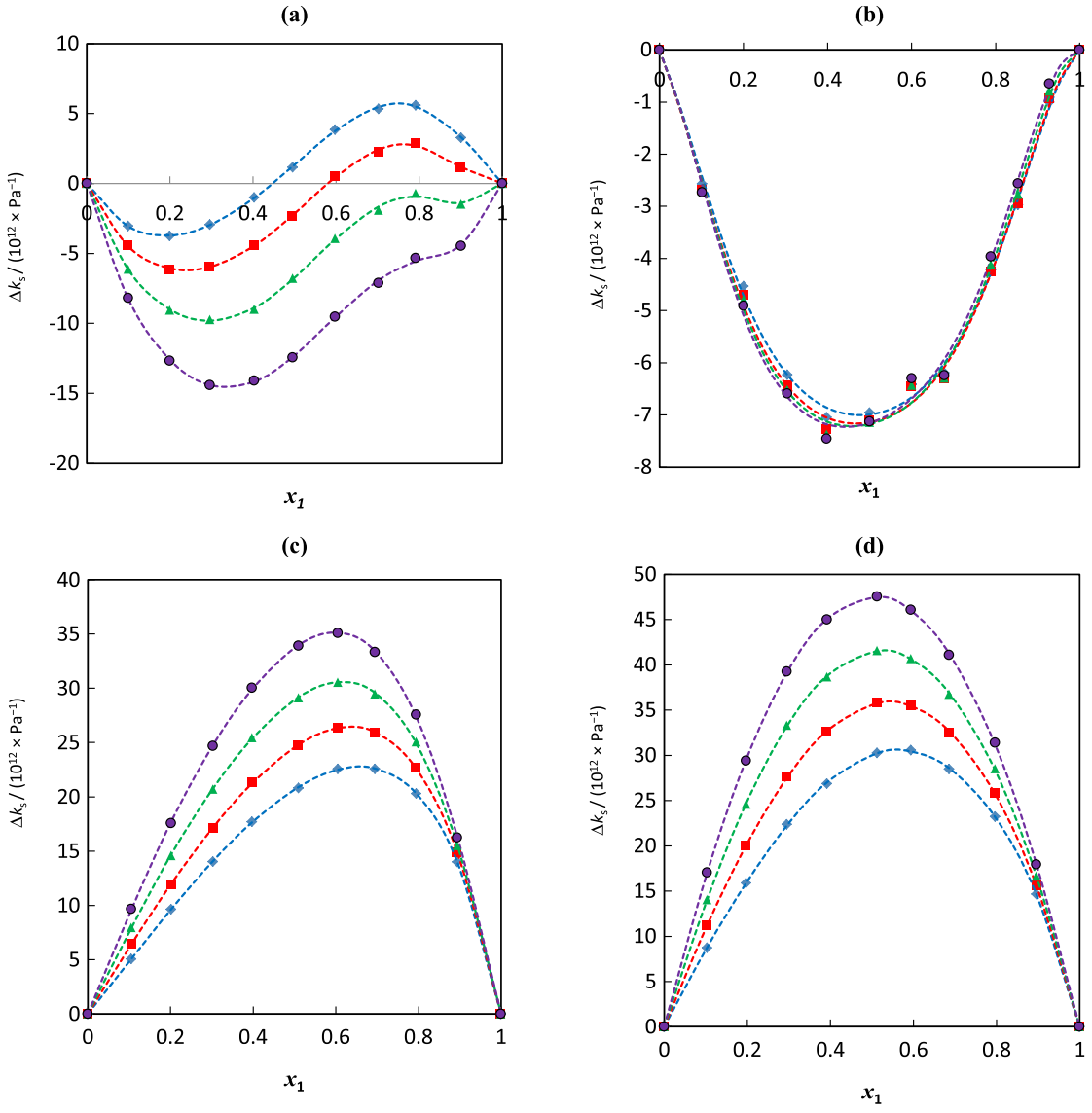

Fig. 2. Plot of deviation in isentropic compressibility,$\Delta\kappa_s$, for the binary mixtures: (a) {2,5-DMF (1)+FA (2)}; (b) {2,5-DMF (1) +MIBK (2)}; (c) {2,5-DMF (1)+1-butanol (2)} and (d) {2,5-DMF (1)+2-butanol (2)} as function of the composition expressed in the mole fraction at 293.15 K (♦); 303.15 K (■); 313.15 K (▲) and 323.15 K (●). The dotted lines were generated using Redlich-Kister polynomial curve-fitting.

in volume are dominant over the entire composition range in the mixtures (2,5-DMF + the 1-butanol or 2-butanol) systems whereas an inversion in sign for (2,5-DMF + MIBK or FA) systems suggested that factors responsible for decrease in volume are dominant over the composition range. As can been seen the results in Table S1, the V_m^E values at $x_1 = 0.5938$ for 2-butanol>1-butanol>MIBK>FA indicating that the interaction between 2,5-DMF with FA or MIBK or 1-butanol or 2-butanol as in order FA>MIBK>1-butanol>2-butanol. This observation based on fact that lower the V_m^E values have stronger interaction and vice versa. From Figure 1 (a)–(d), shows that the V_m^E values increase with the temperature for all systems except MIBK whereas decrease with temperature. The V_m^E minimum and maximum values increase with an increase in temperature for all the systems except the system containing FA. The excess molar volume of an equimolar mixtures for studied systems at 293.15, 303.15, 313.15 and 323.15 K are −0.266, −0.292, −0.318, −0.344 for FA; −0.231, −0.229, −0.223, −0.222 for MIBK; 0.112, 0.150, 0.195, 0.245 for 1-butanol and 0.395, 0.472, 0.549, 0.625 for 2-butanol were observed.

3.4 Isentropic compressibility, and deviation in isentropic compressibility

The Newton–Laplace equation was used to calculate the isentropic compressibility, κ_s,

$$\kappa_s = \frac{1}{\rho u^2} \tag{2}$$

Table 4. Coefficients A_i, and standard deviations, obtained for the binary systems studied in this work at different temperatures and at pressure $p = 0.1$ MPa for the Redlich–Kister equation.

	T /(K)	A_1	A_2	A_3	A_4	A_5	**σ**
{2,5-DMF (1)+FA (2)}							
V_m^E/(cm^3.mol^{-1})	293.15	−1.063	0.225	0.110	0.392	−0.421	0.004
	303.15	−1.167	0.249	0.114	0.464	−0.359	0.004
	313.15	−1.273	0.263	0.090	0.554	−0.223	0.004
	323.15	−1.377	0.297	0.075	0.603	−0.107	0.004
$\Delta\kappa_s$/(10^{-12}×Pa^{-1})	293.15	5.4	51.5	7.5	−11.2	−21.4	0.1
	303.15	−8.6	52.9	6.4	−21.0	−34.0	0.1
	313.15	−26.9	53.3	4.0	−31.4	−42.8	0.2
	323.15	−49.1	50.4	−4.1	−36.7	−45.0	0.2
{2,5-DMF (1)+MIBK (2)}							
V_m^E/(cm^3.mol^{-1})	293.15	−0.944	0.021	−0.385	0.130	0.828	0.008
	303.15	−0.925	0.033	−0.391	0.140	0.846	0.009
	313.15	−0.906	0.045	−0.393	0.150	0.863	0.009
	323.15	−0.881	0.058	−0.416	0.156	0.914	0.009
$\Delta\kappa_s$/(10^{-12}×Pa^{-1})	293.15	−28.0	2.0	−6.2	4.1	21.7	0.2
	303.15	−28.6	2.8	−6.9	4.4	23.4	0.2
	313.15	−28.7	3.7	−7.2	5.1	26.4	0.3
	323.15	−28.6	5.0	−8.2	5.3	29.8	0.3
{2,5-DMF (1)+1-butanol (2)}							
V_m^E/(cm^3.mol^{-1})	293.15	0.446	0.542	0.208	0.525	0.446	0.004
	303.15	0.599	0.583	0.207	0.594	0.553	0.004
	313.15	0.778	0.629	0.215	0.679	0.658	0.005
	323.15	0.982	0.679	0.258	0.782	0.737	0.005
$\Delta\kappa_s$/(10^{-12}×Pa^{-1})	293.15	82.7	48.3	26.2	18.0	4.6	0.1
	303.15	98.2	51.3	25.4	8.0	−3.9	0.1
	313.15	115.7	53.1	24.5	−3.5	−17.8	0.1
	323.15	134.9	53.8	22.0	−14.7	−29.4	0.2
{2,5-DMF (1)+2-butanol (2)}							
V_m^E/(cm^3.mol^{-1})	293.15	1.579	0.574	-0.125	0.770	1.021	0.005
	303.15	1.878	0.529	-0.072	0.904	1.112	0.005
	313.15	2.181	0.505	0.027	1.007	1.158	0.005
	323.15	2.487	0.508	0.158	1.076	1.160	0.005
$\Delta\kappa_s$/(10^{-12}×Pa^{-1})	293.15	120.2	31.3	−0.6	12.8	14.9	0.1
	303.15	142.5	23.6	−1.3	9.1	6.0	0.1
	313.15	165.8	14.2	−0.2	5.0	−1.6	0.1
	323.15	190.0	3.8	0.5	3.4	−5.6	0.2

Standard uncertainties u are $u(T) = \pm 0.02$ K, $u(p) = \pm 0.04$ MPa and the combined expanded uncertainty Uc in mole fraction, density and sound velocity were $Uc(x) = \pm 0.0005$, $Uc(\rho) = \pm 0.003$ g. cm^{-3} and $Uc(u) = \pm 1.2$ m · s^{-1}, respectively, (0.95 level of confidence).

The deviations in isentropic compressibility, $\Delta\kappa_s$, were calculated using the equation given below:

$$\Delta\kappa_{(s)} = \kappa_s - \sum_i^2 x_i \kappa_{s,i} \tag{3}$$

where $\kappa_{s,i}$ and x_i are the isentropic compressibility and mole fractions of the pure component i, respectively. The results of isentropic compressibility, κ_s, for studied systems at 293.15, 303.15, 313.15 and 323.15 K are given in Table 3 and are also plotted in Figure S3 (a)–(d). The isentropic compressibility, κ_s, value increases with an increase in

temperature at a fixed composition for all binary systems due to an increase in thermal agitation, making the solution more compressible [69]. The κ_s value increases with an increase in temperature and increases with an increase in the concentration of 2,5-DMF at a fixed temperature for the system of 2,5-DMF with FA, 1-butanol and 2-butanol except for the 1-butanol, 2-butanol systems whereas start decreasing from $x_1 = 0.5099$, $x_1 = 0.1972$ upwards respectively, while for the MIBK solution of 2,5-DMF, decreases with concentration.

It is well known that the addition of 2,5-DMF molecules to self-associated hydrogen bonded FA,1-butanol and 2-butanol will induce breaking of clusters of these molecules thereby releasing so many dipoles, which interact with dipoles of 2,5-DMF. This causes an increase in free space, decrease in speed of sound and positive deviation in isentropic compressibility [70]. The calculated $\Delta\kappa_s$ values for studied system at (293.15, 303.15, 313.15 and 323.15) K are also given in Table S1 and are graphically presented in (Figure 2 (a)–(d)). It is observed from Figure 2 (a)–(d), the values of $\Delta\kappa_s$ are negative for (2,5-DMF + MIBK) binary system, and both positive and negative for the systems (2,5-DMF + FA). The positive values of $\Delta\kappa_s$ are also observed for (2,5-DMF + 1-butanol or 2-butanol) systems. The negative values of deviations in isentropic compressibility, $\Delta\kappa_s$, indicate that there is strong unlike dipole–dipole interaction in the mixtures which compensates greater to the positive contribution to $\Delta\kappa_s$ arising from the mutual rupturing of the dipolar aggregates in components 1 and 2 by each other [71]. The positive values of $\Delta\kappa_s$ may be due to rupture of hydrogen bonded associates of 1-butanol or 2-butanol dominated over hydrogen bonding between unlike molecules.

3.5 Correlation of derived properties

Experimental excess/deviation properties of the {2,5-dimethylfuran (DMF)+FA or 1-butanol or 2-butanol, or MIBK} were correlated by Redlich–Kister Equation (4):

$$X = x_1 x_2 \sum_{i=1}^{k} A_i (1 - 2x_1)^{i-1} \quad (4)$$

where X is excess molar volumes, V_m^E and deviation in isentropic compressibility, $\Delta\kappa_s$. The values of the fitting parameters A_i have been evaluated using a least-square method. These results are summarized in Table 4, together with the corresponding standard deviations, σ, which was determined using Equation (5):

$$\sigma(X) = \left[\frac{\sum_{i=1}^{N} \left(X_{expt} - X_{calc}\right)^2}{(N-k)}\right]^{1/2} \quad (5)$$

where N is the number of experimental points and k is the number of coefficients used in the Redlich-Kister equation. The values of V_m^E and $\Delta\kappa_s$ as well as the plots of the Redlich-Kister model are displayed in Figures 1 (a)–(d) and 2 (a)–(d), respectively. The standard deviations, between the experimental data and those calculated using Redlich–Kister equation are also given in Table 4, show very small values at the investigated temperatures for all the systems.

4 Conclusion

In this work, density and speed of sound of {2,5-dimethylfuran (DMF) + FA or 1-butanol or 2-butanol, or MIBK} systems were measured over the temperature range of 293.15–323.15 K and at atmospheric pressure. The experimental values were used to calculate the excess functions, which were then correlated using a Redlich-Kister-type polynomial equation. The excess molar volumes were negative for {2,5-dimethylfuran (DMF) +F) or MIBK} systems and positive for {2,5-dimethylfuran (DMF)+1-butanol or 2-butanol} systems; deviations in isentropic were negative for (2,5-DMF+MIBK) binary system, and both positive and negative for the system (2,5-DMF+FA). The positive values of $\Delta\kappa_s$ are also observed for (2,5-DMF+1-butanol or 2-butanol) systems.

Supplementary Material

Supplementary figures and table.

The Supplementary Material is available at https://ogst.ifpenergiesnouvelles.fr/10.2516/ogst/2018012/olm.

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