Densities and Speeds of Sound for Binary Liquid Mixtures of Thiolane-1,1-dioxide with Butanone, Pentan-2-one, Pentan-3-one, and 4-Methyl-pentan-2-one at T = (303.15 or 308.15 or 313.15) K

Patwari Murali Krishna,[†] Bachu Ranjith Kumar,[†] Boodida Sathyanarayana,[†] Kanthala Sampath Kumar,[‡] and Nallani Satyanarayana*,[†]

Department of Chemistry, Kakatiya University, Warangal, 506 009, Andhra Pradesh, India, and Department of Applied Statistics, Telangana University, Nizamabad, 503 001, India

Experimental data on density (ρ) and speed of sound (u) at (303.15, 308.15, and 308.15) K are presented for the binary mixtures of (thiolane-1,1-dioxide + butanone), (thiolane-1,1-dioxide + pentan-2-one), (thiolane-1,1-dioxide + pentan-3-one), and (thiolane-1,1-dioxide + 4-methyl-pentan-2-one). From this data, excess molar volume and deviation in isentropic compressibility have been calculated. The Redlich−Kister polynomial equation was fitted to the experimental data.

Introduction

Studies on thermodynamic properties of mixtures are useful for designing transport and process equipment in the chemical industry and the theoretical interest in studying the structure of such solutions.[1,2] The interactions between the carbonyl group of ketone and solvents with polar groups, such as thiolane-1,1-dioxide, play a crucial role in the structural effects,[3,4] in the molecular level, and for practical applications. Our current project is devoted to the systematic study of liquid systems containing aliphatic ketones with respect to their environmental importance. Ketones are highly efficient solvents used to make high solid coatings[5] with lower solvent content and lower solvent emissions. To the best of our knowledge, no extensive studies have been made on the mixtures of thiolane-1,1-dioxide with aliphatic ketones. This prompted us to study the thermodynamic excess properties of binary liquid mixtures of thiolane-1,1-dioxide with butanone, pentan-2-one, pentan-3-one, and 4-methyl-pentan-2-one.

In continuation of our ongoing program of research, this paper reports the systematic measurements of the properties of organic solvents.[6−12] The present study is undertaken to determine experimentally density and speed of sound of four pure liquid ketones, butanone, pentan-2-one, pentan-3-one, and 4-methyl-pentan-2-one, and their binary mixtures with thiolane-1,1-dioxide at temperatures of (303.15, 308.15, and 313.15) K at a pressure of 0.1 MPa. With this data, the excess molar volume and deviation in isentropic compressibility of the mixtures were computed. The Redlich−Kister polynomial equation[13] was fitted to the results to derive the binary coefficients and estimate the standard deviation between experimental and calculated data.

Experimental

Materials. Thiolane-1,1-dioxide [CAS# 126-33-0] of mole fraction purity 0.99 was furnished by Sigma-Aldrich Chemicals, USA, and used without further purification. To minimize the contact of this deliquescent reagent with moist air, the product was kept in sealed bottles in a desiccator. Butanone [CAS # 78-93-3] of mole fraction purity 0.99 was purchased from Merck, India. Pentan-2-one [CAS # 107-87-9] of mole fraction purity 0.99, pentan-3-one [CAS # 96-22-0] of mole fraction purity 0.98, and 4-methyl-pentan-2-one [CAS # 108-10-1] of mole fraction purity 0.99 were obtained from Sigma-Aldrich Chemicals Pvt Ltd., Germany. All the ketones were used without further purification. The purity of the substances was verified by GLC. Densities and speeds of sound of the pure substances and their comparison with literature values are listed in Table 1.[14−25]

Table 1. Experimental Densities (ρ) and Speeds of Sound (u) of Pure Components at Different Temperatures

		$\rho \cdot 10^{-3}$/kg·m^{-3}		u/m·s^{-1}	
component	T/K	exptl	lit.	exptl	lit.
thiolane-1,1-dioxide	298.15	1.2639	1.2640[14]	1601	
	303.15	1.2618	1.2618[15]	1588	1588[25]
	313.15	1.2516	1.2519[16]	1558	
butanone	298.15	0.8012	0.8009[17]	1192	1192[24]
	303.15	0.7946	0.7945[18]	1178	
pentan-2-one	298.15	0.8019	0.8016[20]	1213	1213[24]
	303.15	0.7966	0.7965[18]	1192	1192[24]
pentan-3-one	298.15	0.8096	0.8097[21]	1218	
	303.15	0.8045	0.8045[18]	1197	1197[23]
4-methyl-pentan-2-one	298.15	0.7962	0.7963[22]	1184	
	303.15	0.7917	0.7916[19]	1175	

Apparatus and Procedure. Binary mixtures were prepared by mass in airtight bottles. The mass measurements were performed on a Dhona 100 DS, India, single-pan analytical balance with a resolution of $0.01 \cdot 10^{-6}$ kg. The required properties of the mixture were measured on the same day. The uncertainty in mole fraction was estimated to be less than $\pm\ 1 \cdot 10^{-4}$.

Densities of pure liquids and their mixtures were determined by using a $1 \cdot 10^{-5}$ m^3 double arm pycnometer.[9] The density values from triplicate replication at the measured temperatures were reproducible within $\pm\ 2 \cdot 10^{-2}$ kg·m^{-3}. The uncertainty in density and excess molar volume values was found to be $\pm$ 0.04 kg·m^{-3} and $0.001 \cdot 10^{-6}$ m^3·mol^{-1}

Speed of sound was determined by using an ultrasonic interferometer [model M-82, Mittal Enterprises, India], working at 2 MHz frequency. The detailed procedure for measuring speed of

* Corresponding author. Tel.: +91-870-2461437. Fax: +91-870-2438800. E-mail: ns_narayana@yahoo.com.
[†] Kakatiya University.
[‡] Telangana University.

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Table 2. Values of Density (ρ), Speed of Sound (u), Excess Molar Volume (V^E), and Deviation in Isentropic Compressibility ($\Delta\kappa_s$) for the Binary Liquid Mixtures at Various Temperatures

x_1	$\rho \cdot 10^{-3}$ / kg·m^{-3}	u / m·s^{-1}	$V^E \cdot 10^6$ / m^3·mol^{-1}	$\Delta\kappa_s \cdot 10^{11}$ / m^2·N^{-1}	x_1	$\rho \cdot 10^{-3}$ / kg·m^{-3}	u / m·s^{-1}	$V^E \cdot 10^6$ / m^3·mol^{-1}	$\Delta\kappa_s \cdot 10^{11}$ / m^2·N^{-1}
thiolane-1,1-dioxide (1) + butanone (2)					thiolane-1,1-dioxide (1) + pentan-3-one (2)				
		T/K = 303.15					T/K = 303.15		
0.0000	0.7946	1178	0.000	0.0000	0.0000	0.8045	1197	0.000	0.0000
0.0293	0.8106	1191	−0.189	−1.0178	0.0252	0.8159	1208	−0.140	−1.1402
0.1391	0.8697	1234	−0.778	−4.7296	0.1627	0.8781	1257	−0.737	−4.9989
0.2575	0.9316	1288	−1.219	−7.6132	0.2922	0.9374	1300	−1.108	−6.9261
0.3803	0.9935	1341	−1.480	−8.7651	0.4201	0.9962	1345	−1.276	−7.6701
0.4834	1.0431	1386	−1.518	−8.8811	0.5234	1.0441	1388	−1.314	−7.8577
0.5854	1.0897	1431	−1.385	−8.1139	0.6292	1.0935	1435	−1.266	−7.4860
0.7119	1.1452	1481	−1.102	−6.4330	0.7487	1.1494	1490	−1.099	−6.2095
0.8428	1.2005	1533	−0.724	−3.9221	0.8644	1.2028	1536	−0.768	−3.7636
0.9756	1.2527	1581	−0.131	−0.7057	0.9804	1.2535	1579	−0.133	−0.5164
1.0000	1.2618	1588	0.000	0.0000	1.0000	1.2618	1588	0.000	0.0000
		T/K = 308.15					T/K = 308.15		
0.0000	0.7904	1158	0.000	0.0000	0.0000	0.7996	1179	0.000	0.0000
0.0293	0.8062	1170	−0.174	−0.9440	0.0252	0.8111	1191	−0.171	−1.2830
0.1391	0.8656	1215	−0.819	−5.1188	0.1627	0.8731	1240	−0.759	−5.2334
0.2575	0.9275	1266	−1.271	−7.7244	0.2922	0.9319	1283	−1.083	−7.2630
0.3803	0.9896	1319	−1.560	−8.9378	0.4201	0.9911	1331	−1.298	−8.2159
0.4834	1.0399	1366	−1.662	−9.1848	0.5234	1.0391	1376	−1.343	−8.5373
0.5854	1.0866	1413	−1.536	−8.4709	0.6292	1.0884	1421	−1.283	−7.9043
0.7119	1.1420	1465	−1.240	−6.7787	0.7487	1.1442	1473	−1.098	−6.3484
0.8428	1.1968	1517	−0.821	−4.0271	0.8644	1.1981	1520	−0.797	−3.8161
0.9756	1.2490	1568	−0.219	−0.7321	0.9804	1.2500	1566	−0.232	−0.5208
1.0000	1.2570	1576	0.000	0.0000	1.0000	1.2570	1576	0.000	0.0000
		T/K = 313.15					T/K = 313.15		
0.0000	0.7831	1136	0.000	0.0000	0.0000	0.7943	1160	0.000	0.0000
0.0293	0.7992	1149	−0.209	−1.1691	0.0252	0.8058	1170	−0.171	−1.1209
0.1391	0.8587	1195	−0.871	−5.6340	0.1627	0.8676	1218	−0.758	−5.2177
0.2575	0.9209	1247	−1.353	−8.4438	0.2922	0.9266	1263	−1.119	−7.5296
0.3803	0.9835	1300	−1.678	−9.6598	0.4201	0.9856	1315	−1.320	−8.8733
0.4834	1.0344	1346	−1.812	−9.7906	0.5234	1.0337	1361	−1.371	−9.2597
0.5854	1.0818	1395	−1.720	−9.1509	0.6292	1.0831	1406	−1.323	−8.5246
0.7119	1.1372	1450	−1.385	−7.4330	0.7487	1.1393	1460	−1.166	−6.9436
0.8428	1.1918	1502	−0.906	−4.4485	0.8644	1.1932	1506	−0.854	−4.1969
0.9756	1.2437	1552	−0.234	−0.8803	0.9804	1.2446	1550	0.242	−0.6139
1.0000	1.2516	1558	0.000	0.0000	1.0000	1.2516	1558	0.000	0.0000
thiolane-1,1-dioxide (1) + pentan-2-one (2)					thiolane-1,1-dioxide (1) + 4-methyl-pentan-2-one (2)				
		T/K = 303.15					T/K = 303.15		
0.0000	0.7966	1192	0.000	0.0000	0.0000	0.7917	1175	0.000	0.0000
0.0261	0.8082	1203	−0.118	−1.1257	0.0287	0.8034	1185	−0.235	−1.0067
0.1620	0.8692	1246	−0.597	−4.6907	0.1852	0.8666	1233	−0.867	−5.1396
0.2945	0.9292	1290	−0.845	−6.5876	0.3272	0.9273	1281	−1.207	−7.4162
0.4450	0.9985	1352	−0.957	−7.5805	0.4634	0.9892	1335	−1.393	−8.5679
0.5296	1.0382	1384	−0.972	−7.7936	0.5671	1.0389	1381	−1.444	−8.7810
0.6319	1.0866	1429	−0.918	−7.3198	0.6671	1.0888	1426	−1.393	−8.0576
0.7498	1.1432	1484	−0.793	−5.9923	0.7777	1.1451	1479	−1.132	−6.3634
0.8677	1.2002	1536	−0.569	−3.6779	0.8809	1.1985	1530	−0.682	−3.9694
0.9799	1.2532	1580	−0.147	−0.6286	0.9829	1.2523	1578	−0.082	−0.5860
1.0000	1.2618	1588	0.000	0.0000	1.0000	1.2618	1588	0.000	0.0000
		T/K = 308.15					T/K = 308.15		
0.0000	0.7927	1176	0.000	0.0000	0.0000	0.7877	1156	0.000	0.0000
0.0261	0.8042	1183	−0.103	−0.6667	0.0287	0.7993	1166	−0.218	−1.1511
0.1620	0.8646	1225	−0.538	−4.2263	0.1852	0.8625	1215	−0.888	−5.5063
0.2945	0.9248	1271	−0.832	−6.4559	0.3272	0.9233	1263	−1.260	−7.8340
0.4450	0.9946	1340	−1.014	−8.0965	0.4634	0.9851	1316	−1.446	−8.8571
0.5296	1.0345	1373	−1.055	−8.3436	0.5671	1.0346	1361	−1.480	−9.0202
0.6319	1.0828	1418	−0.995	−7.7977	0.6671	1.0839	1407	−1.383	−8.2929
0.7498	1.1392	1469	−0.855	−6.1604	0.7777	1.1407	1463	−1.167	−6.6652
0.8677	1.1958	1520	−0.600	−3.6464	0.8809	1.1951	1513	−0.804	−4.0292
0.9799	1.2485	1562	−0.159	−0.4275	0.9829	1.2492	1564	−0.213	−0.5850
1.0000	1.2570	1576	0.000	0.0000	1.0000	1.2570	1576	0.000	0.0000
		T/K = 313.15					T/K = 313.15		
0.0000	0.7855	1151	0.000	0.0000	0.0000	0.7826	1138	0.000	0.0000
0.0261	0.7978	1161	−0.215	−1.0987	0.0287	0.7939	1147	−0.174	−1.0577
0.1620	0.8588	1203	−0.724	−4.8542	0.1852	0.8568	1198	−0.836	−5.8793
0.2945	0.9191	1251	−1.011	−7.3488	0.3272	0.9176	1247	−1.229	−8.4018
0.4450	0.9891	1318	−1.184	−8.7934	0.4634	0.9797	1300	−1.465	−9.4837
0.5296	1.0291	1353	−1.213	−9.1249	0.5671	1.0293	1345	−1.519	−9.5766
0.6319	1.0780	1401	−1.179	−8.6414	0.6671	1.0785	1391	−1.407	−8.8144
0.7498	1.1347	1450	−1.025	−6.6908	0.7777	1.1341	1447	−1.080	−7.0651
0.8677	1.1916	1503	−0.748	−4.0370	0.8809	1.1878	1496	−0.655	−4.2500
0.9799	1.2436	1549	−0.206	−0.6956	0.9829	1.2423	1550	−0.102	−0.7584
1.0000	1.2516	1558	0.000	0.0000	1.0000	1.2516	1558	0.000	0.0000

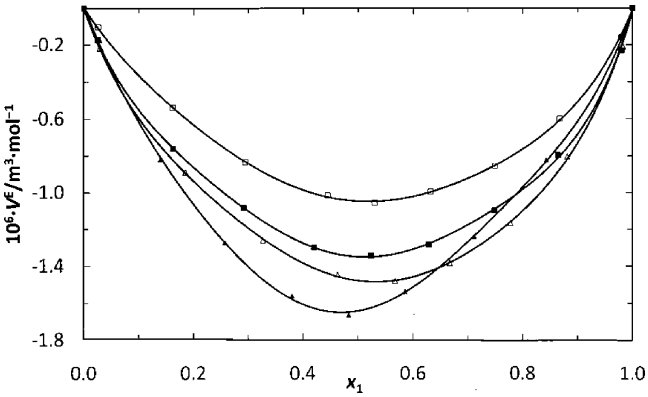

Figure 1. Plots of excess molar volume, V^E, as a function of mole fraction x at $T = 308.15$ K: ▲, {thiolane-1,1-dioxide (1) + butanone (2)}; □, {thiolane-1,1-dioxide (1) + pentan-2-one (2)}; ■, {thiolane-1,1-dioxide (1) + pentan-3-one}; Δ, {thiolane-1,1-dioxide (1) + 4-methyl-pentan-2-one (2)}; the symbols represent experimental values and lines represent the smoothed data of this work.

Table 3. Binary Coefficients (A_i) and Standard Errors (σ) of Thiolane-1,1-dioxide (1) + Ketones (2)

function	T/K	A_0	A_1	A_2	A_3	A_4	σ
thiolane-1,1-dioxide (1) + butanone (2)							
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	303.15	−5.996	1.4	0.82	−1.33	−1.42	0.010
	308.15	−6.56	1.07	1.83	−1.73	−3.5	0.022
	313.15	−7.213	0.56	2.46	−0.91	−4.37	0.018
$\Delta\kappa_s \cdot 10^{11}/m^2 \cdot N^{-1}$	303.15	−35.28	8.63	−1.2	−6.6	5.7	0.048
	308.15	−36.34	6.8	−3.2	−1.3	7.9	0.12
	313.15	−39.01	7.2	−6.2	−0.6	8.3	0.11
thiolane-1,1-dioxide (1) + pentan-2-one (2)							
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	303.15	−3.884	0.15	−0.86	−0.98	−1.33	0.011
	308.15	−4.187	−0.30	0.29	−1.13	−2.10	0.012
	313.15	−4.876	−0.50	−0.01	−0.17	−4.51	0.016
$\Delta\kappa_s \cdot 10^{11}/m^2 \cdot N^{-1}$	303.15	−31.0	1.70	−1.6	9.0	−7.1	0.038
	308.15	−33.16	1.53	4.6	4.0	0.1	0.087
	313.15	−36.5	2.17	7.6	7	−12	0.072
thiolane-1,1-dioxide (1) + pentan-3-one (2)							
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	303.15	−5.248	−0.254	−1.56	−0.88	0.32	0.0061
	308.15	−5.401	−0.3	0.67	−1.09	−4.6	0.020
	313.15	−5.505	−0.38	−0.09	−1.54	−3.7	0.020
$\Delta\kappa_s \cdot 10^{11}/m^2 \cdot N^{-1}$	303.15	−31.39	1.66	−8.6	11.9	3.3	0.043
	308.15	−33.9	2.84	0.0	12.2	−8	0.070
	313.15	−36.69	2.72	2.3	7.4	−6.7	0.067
thiolane-1,1-dioxide (1) + 4-methyl-pentan-2-one (2)							
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	303.15	−5.704	−1.59	−1.20	2.67	−0.13	0.017
	308.15	−5.917	−0.7	0.16	−0.36	−3.9	0.020
	313.15	−6.02	−1.253	0.72	1.64	−1.09	0.0071
$\Delta\kappa_s \cdot 10^{11}/m^2 \cdot N^{-1}$	303.15	−34.87	5.36	1.1	4.5	−5.6	0.044
	308.15	−35.85	5.45	−2.7	8.0	−1.9	0.040
	313.15	−38.13	7.28	−3.9	5.0	0.1	0.056

sound was discussed in our previous paper.[6] The speed of sound was measured with relative uncertainty of ± 0.3 %.

In all the property measurements, the temperature was controlled within ± 0.01 K using a constant temperature bath [INSREF model IRI-016 C, India], and the temperature was monitored with a platinum resistance thermometer [KI-TP303R, UK] with an accuracy of ± 0.001 K and an uncertainty of ± 0.004 K.

Results and Discussion

Experimental values of densities ρ, speeds of sound u, excess molar volumes V^E, and deviation in isentropic compressibilities $\Delta\kappa_s$ for (thiolane-1,1-dioxide + butanone), (thiolane-1,1-dioxide + pentan-2-one), (thiolane-1,1-dioxide + pentan-3-one), and (thiolane-1,1-dioxide + 4-methyl-pentan-2-one) at temperatures of (303.15, 308.15, and 313.15) K are listed in Table 2.

The density values have been used to calculate excess molar volumes V^E using the following equation

$$V^E/(m^3 \cdot mol^{-1}) = (x_1M_1 + x_2M_2)/\rho_m - (x_1M_1/\rho_1 + x_2M_2/\rho_2) \quad (1)$$

where ρ_m is the density of the mixture; x_1, M_1, ρ_1 and x_2, M_2, and ρ_2 are the mole fraction, molecular weight, and density of pure components 1 and 2, respectively.

The speed of sound u was used to calculate the isentropic compressibility κ_s using the equation

$$\kappa_s = 1/u^2\rho \quad (2)$$

The deviation in isentropic compressibility $\Delta\kappa_s$ has been evaluated using the equation

$$\Delta\kappa_s/(m^2 \cdot N^{-1}) = \kappa_s - (\Phi_1\kappa_{s1} + \Phi_2\kappa_{s2}) \quad (3)$$

where κ_{s1}, κ_{s2}, and κ_s are the isentropic compressibility of the pure components and observed isentropic compressibility of the liquid mixture, respectively.

Φ_i is the volume fraction and is calculated from the individual pure molar volumes, V_i, with the relation

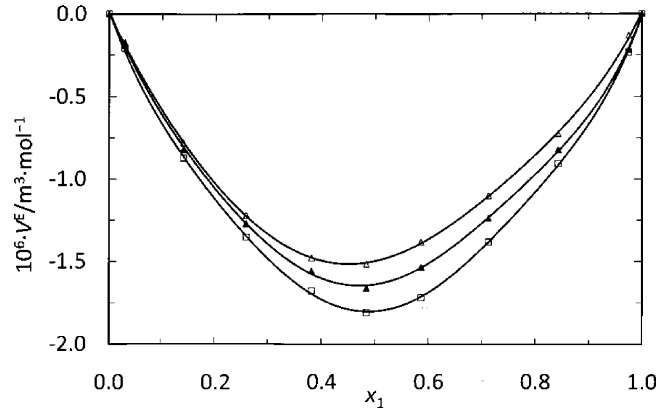

Figure 2. Plots of excess molar volumes, V^E, of thiolane-1,1-dioxide (1) + butanone (2) at Δ, 303.15; ▲, 308.15; □, 313.15 K; the symbols represent experimental values and lines represent the smoothed data of this work.

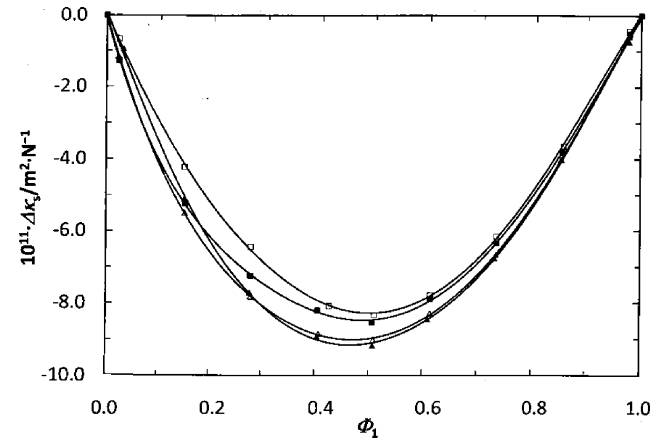

Figure 3. Plots of deviation in isentropic compressibility as function of volume fraction at $T = 308.15$ K: ▲, {thiolane-1,1-dioxide (1) + butanone (2)}; □, {thiolane-1,1-dioxide (1) + pentan-2-one (2)}; ■, {thiolane-1,1-dioxide (1) + pentan-3-one (2)}; Δ, {thiolane-1,1-dioxide (1) + 4-methyl-pentan-2-one (2)}; the symbols represent experimental values and lines represent the smoothed data of this work.

$$\Phi_i = x_i V_i / (\sum x_i V_i) \quad (4)$$

The Redlich−Kister equation[13] was fitted to the excess properties (Y^E) by the nonlinear least-squares method

$$Y^E = x_1 x_2 \sum A_i (x_1 x_2)^i \quad (5)$$

In each case, the optimum number of coefficients A_i was determined from an examination of the variation of standard deviation (σ) as calculated by

$$\sigma(Y^E) = \left[\sum (Y^E_{obs} - Y^E_{cal})^2 / (n - m) \right]^{1/2} \quad (6)$$

where n represents the number of experimental points and m is the number of coefficients used in fitting the data. It is found that for the solution of the fifth degree polynomial, the agreement between the experimental values and the calculated ones is satisfactory. The derived parameters (A_i) and the estimated standard deviation (σ) for V^E and $\Delta\kappa_s$ are summarized in Table 3.

Figure 1 shows that the excess molar volumes for (thiolane-1,1-dioxide + butanone), (thiolane-1,1-dioxide + pentan-2-one), (thiolane-1,1-dioxide + pentan-3-one), and (thiolane-1,1-dioxide + 4-methyl-pentan-2-one) are negative over the entire range of composition at $T = 308.15$ K. The variation of V^E with the temperature for (thiolane-1,1-dioxide + butanone) is shown in Figure 2. The absolute values of V^E vary in the following order: butanone > 4-methyl-pentan-2-one > pentan-3-one > pentan-2-one.

The variation of $\Delta\kappa_s$ with volume fraction, Φ_1, of thiolane-1,1-dioxide with aliphatic ketones is, as shown in Figure 3, negative over the whole composition range for all mixtures at $T = 308.15$ K. The negative $\Delta\kappa_s$ values become more negative at higher temperatures and exhibit minima at the volume fraction range (0.48 to 0.56) observed from Table 2. Inspection of Figure 3 at equimolar compositions reveals that the $\Delta\kappa_s$ values decrease in the order: butanone < 4-methyl-pentan-2-one < pentan-3-one < pentan-2-one.

Conclusion

The experimental values of density and speed of sound for (thiolane-1,1-dioxide + butanone), (thiolane-1,1-dioxide + pentan-2-one), (thiolane-1,1-dioxide + pentan-3-one), and (thiolane-1,1-dioxide + 4-methyl-pentan-2-one) at $T =$ (303.15, 308.15, and 313.15) K were determined as a function of compositions. From these data, V^E and $\Delta\kappa_s$ have been calculated, and the Redlich−Kister polynomial equation was fitted to the data. In the present study, V^E and $\Delta\kappa_s$ are negative over the entire range of composition and at all temperatures.

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