Density, Viscosity, and Speed of Sound of (Methyl Benzoate + Cyclohexane), (Methyl Benzoate + *n*-Hexane), (Methyl Benzoate + Heptane), and (Methyl Benzoate + Octane) at Temperatures of (303.15, 308.15, and 313.15) K

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Density, viscosity, and speed of sound data of (methyl benzoate + cyclohexane), (methyl benzoate + *n*-hexane), (methyl benzoate + heptane), and (methyl benzoate + octane) have been determined at $T =$ (303.15, 308.15, and 313.15) K. From this data, excess volume, V^E, and isentropic compressibility, K_s, have been estimated. The values of V^E for (methyl benzoate + cyclohexane) are very largely positive, while for (methyl benzoate + octane) they are both positive and negative and for the remaining mixtures negative. The V^E were fitted to the Redlich−Kister polynomial equation. The measured viscosities were correlated with Auslander and McAllister's four-body interaction models.

Introduction

Excess thermodynamic and transport properties of binary mixtures provide important information with which to speculate the molecular liquid structure. These properties are mostly dependent on the molecular interactions as well as the difference in the sizes of molecules. Several researchers[1−9] have measured the density, viscosity, and refractive index for a wide range of binary mixtures containing esters as one of the components, and these properties were interpreted in terms of specific or nonspecific interactions. In continuation of our research,[10−12] in this paper we report the results of excess volume, viscosity, and speed of sound for (methyl benzoate + cyclohexane), (methyl benzoate + *n*-hexane), (methyl benzoate + heptane), and (methyl benzoate + octane) at temperatures of (303.15, 318.15, and 313.15) K. Methyl benzoate is an important industrial chemical that is widely used in perfumery and pesticides. The excess volumes, V^E, were correlated using the Redlich−Kister[13] equation to estimate the binary interaction parameters and standard errors. Furthermore, the viscosity data were correlated with the Auslander[14] and McAllister's[15] four-body interaction models.

Experimental Procedure

Materials. Methyl benzoate, cyclohexane, *n*-hexane, heptane, and octane, all Fluka, with stated purities of better mass fraction purities of > 0.99 were used without further purification. The density and viscosity were determined at a temperature of 303.15 K to evaluate the samples for the presence of large quantities of impurities. Table 1 shows a comparison of densities and viscosities with the available literature data.[16−22] There is fairly good agreement between our data and the previously reported values.

Methods. All binary mixtures were prepared gravimetrically in air-tight bottles. The mass measurements were performed on a digital electronic balance (Mettler AE 240, Switzerland) with an uncertainty of ± 0.0001 g. The binary mixtures were prepared just before use. The uncertainty in mole fraction was estimated to be < ± 0.0001. Caution was taken to prevent the evaporation of the solutions after preparation. The required properties of the mixture were determined on the same day. The procedure for measuring viscosity has been described in our previous publication.[12] The densities of the pure and their mixtures were determined with a DMA 35 Anton Paar digital density meter. The instrument has a resolution of 0.0001 g·cm^{-3}. The average uncertainty in the measured density was ± 0.1 %. The speed of sound was measured with a single-crystal variable path interferometer (Mittal Enterprises, New Delhi, India) operating at a frequency of 2 MHz that had been calibrated with water and benzene. The uncertainty in the speed of sound was found to be ± 2 %. In all property measurements the temperature was controlled within ± 0.01 K using a constant temperature bath (INSREF model IRI O16C, India) by circulating water from the thermostat.

Table 1. Comparison of Experimental Densities, ρ, Viscosities, η, and Speed of Sound, u, of Pure Components with Available Literature Values at (303.15, 308.15, and 313.15) K

	T	ρ/(g·cm^{-3})		η/(mPa·s)		u/(m·s^{-1})	
liquid	K	exptl	lit.	exptl	lit.	exptl	lit.
methyl benzoate	303.15	1.0783	1.0788[16]	1.667	1.673[17]	1380	1392[16]
	308.15	1.0744	1.0739[16]	1.514		1364	1372[16]
	313.15	1.0703	1.0690[16]	1.376	1.365[16]	1348	1352[16]
cyclohexane	303.15	0.7683	0.7685[17]	0.823	0.820[17]	1228	1230[21]
	308.15	0.7644		0.772		1208	
	313.15	0.7605		0.685		1188	
n-hexane	303.15	0.6498	0.6502[18]	0.283	0.276[17]	1048	1054[19]
	308.15	0.6459	0.6453[18]	0.269		1028	1032[19]
	313.15	0.6422		0.252		1008	1010[19]
heptane	303.15	0.6745	0.6751[18]	0.375	0.375[17]	1116	1122[22]
	308.15	0.6703	0.6707[18]	0.350	0.349[18]	1092	
	313.15	0.6672		0.325		1072	
octane	303.15	0.6939	0.6945[17]	0.491	0.484[17]	1148	1152[20]
	308.15	0.6904	0.6906[18]	0.450	0.450[18]	1132	1133[20]
	313.15	0.6870		0.415		1112	

Results and Discussion

Table 2 lists the measured density, ρ, viscosity, η, and speed of sound, u, data at temperatures of (303.15, 308.15, and 313.15) K along with the corresponding excess volume,

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Table 2. Values of Density, ρ, Excess Volume, V^E, Viscosity, η, Speed of Sound, u, and Isentropic Compressibility, K_s, for the Binary Liquid Mixtures

x_1	ρ / g·cm^{-3}	V^E / cm^3·mol^{-1}	η / mPa·s	u / m·s^{-1}	K_s / TPa^{-1}	x_1	ρ / g·cm^{-3}	V^E / cm^3·mol^{-1}	η / mPa·s	u / m·s^{-1}	K_s / TPa^{-1}
Methyl Benzoate (1) + Cyclohexane (2)						Methyl Benzoate (1) + Heptane (2)					
					$T = 303.15$ K						
0.0000	0.7683		0.823	1228	863	0.0000	0.6745		0.375	1116	1190
0.0880	0.7969	0.343	0.802	1240	816	0.1154	0.7155	−0.141	0.413	1128	1098
0.1777	0.8259	0.573	0.820	1252	772	0.2273	0.7564	−0.215	0.467	1148	1003
0.2692	0.8558	0.655	0.854	1264	731	0.3350	0.7971	−0.276	0.533	1168	920
0.3668	0.8875	0.642	0.905	1276	692	0.4390	0.8377	−0.312	0.627	1192	840
0.4667	0.9188	0.583	0.977	1292	652	0.5404	0.8786	−0.345	0.723	1220	765
0.5639	0.9500	0.481	1.076	1308	615	0.6365	0.9183	−0.335	0.840	1248	699
0.6675	0.9817	0.376	1.181	1324	581	0.7324	0.9590	−0.294	0.990	1276	640
0.7727	1.0133	0.238	1.308	1340	550	0.8252	0.9994	−0.217	1.172	1308	585
0.8859	1.0461	0.125	1.478	1360	517	0.9133	1.0387	−0.126	1.404	1348	530
1.0000	1.0783		1.667	1380	487	1.0000	1.0783		1.667	1380	487
					$T = 308.15$ K						
0.0000	0.7644		0.772	1208	896	0.0000	0.6703		0.350	1092	1251
0.0880	0.7930	0.335	0.740	1220	847	0.1154	0.7113	−0.154	0.380	1108	1145
0.1777	0.8221	0.556	0.757	1232	801	0.2273	0.7524	−0.271	0.430	1128	1045
0.2692	0.8519	0.650	0.792	1244	759	0.3350	0.7931	−0.338	0.494	1148	957
0.3668	0.8836	0.632	0.840	1260	713	0.4390	0.8339	−0.406	0.575	1172	873
0.4667	0.9150	0.560	0.904	1276	671	0.5404	0.8748	−0.436	0.666	1196	799
0.5639	0.9461	0.470	0.999	1292	633	0.6365	0.9147	−0.437	0.781	1228	725
0.6675	0.9780	0.342	1.090	1308	598	0.7324	0.9553	−0.370	0.904	1260	659
0.7727	1.0095	0.220	1.197	1324	565	0.8252	0.9957	−0.277	1.068	1296	598
0.8859	1.0422	0.118	1.363	1344	531	0.9133	1.0348	−0.142	1.284	1336	541
1.0000	1.0744		1.514	1364	500	1.0000	1.0744		1.514	1364	500
					$T = 313.15$ K						
0.0000	0.7605		0.685	1188	932	0.0000	0.6672		0.325	1072	1304
0.0880	0.7891	0.333	0.675	1200	880	0.1154	0.7081	−0.163	0.350	1088	1193
0.1777	0.8182	0.549	0.704	1212	832	0.2273	0.7491	−0.289	0.401	1108	1087
0.2692	0.8481	0.613	0.736	1224	787	0.3350	0.7898	−0.371	0.458	1128	995
0.3668	0.8797	0.613	0.781	1240	739	0.4390	0.8305	−0.445	0.529	1152	907
0.4667	0.9111	0.539	0.835	1256	696	0.5404	0.8713	−0.468	0.615	1180	824
0.5639	0.9422	0.451	0.932	1276	652	0.6365	0.9111	−0.467	0.728	1212	747
0.6675	0.9741	0.317	1.004	1292	615	0.7324	0.9517	−0.405	0.849	1248	675
0.7727	1.0055	0.203	1.097	1312	578	0.8252	0.9921	−0.326	0.982	1284	611
0.8859	1.0383	0.092	1.234	1332	543	0.9133	1.0309	−0.156	1.150	1324	553
1.0000	1.0703		1.376	1348	514	1.0000	1.0703		1.376	1348	514
Methyl Benzoate (1) + *n*-Hexane (2)						Methyl Benzoate (1) + Octane (2)					
					$T = 303.15$ K						
0.0000	0.6498		0.283	1048	1401	0.0000	0.6939		0.491	1148	1094
0.1027	0.6927	−0.144	0.324	1072	1256	0.1290	0.7322	0.194	0.522	1160	1015
0.2076	0.7380	−0.464	0.384	1100	1120	0.2456	0.7700	0.149	0.576	1180	933
0.3084	0.7814	−0.641	0.441	1128	1006	0.3586	0.8092	0.003	0.644	1200	858
0.4087	0.8252	−0.828	0.527	1160	901	0.4655	0.8484	−0.098	0.727	1220	792
0.5114	0.8696	−0.873	0.624	1196	804	0.5672	0.8875	−0.144	0.832	1240	733
0.6115	0.9131	−0.881	0.751	1232	722	0.6620	0.9258	−0.168	0.936	1260	680
0.7092	0.9555	−0.820	0.915	1272	647	0.7528	0.9643	−0.174	1.084	1284	629
0.8069	0.9977	−0.692	1.104	1312	582	0.8382	1.0023	−0.165	1.245	1308	583
0.9024	1.0369	−0.275	1.370	1344	534	0.9213	1.0406	−0.100	1.434	1340	535
1.0000	1.0783		1.667	1380	487	1.0000	1.0783		1.667	1380	487
					$T = 308.15$ K						
0.0000	0.6459		0.269	1028	1465	0.0000	0.6904		0.450	1132	1130
0.1027	0.6890	−0.201	0.301	1052	1311	0.1290	0.7288	0.151	0.484	1140	1056
0.2076	0.7342	−0.512	0.358	1080	1168	0.2456	0.7664	0.125	0.529	1160	970
0.3084	0.7776	−0.701	0.409	1108	1048	0.3586	0.8055	0.002	0.596	1180	892
0.4087	0.8213	−0.870	0.487	1140	937	0.4655	0.8445	−0.080	0.668	1200	822
0.5114	0.8656	−0.905	0.579	1180	830	0.5672	0.8836	−0.135	0.762	1224	755
0.6115	0.9092	−0.911	0.688	1220	739	0.6620	0.9219	−0.165	0.857	1248	697
0.7092	0.9515	−0.843	0.844	1256	666	0.7528	0.9603	−0.160	0.990	1272	644
0.8069	0.9937	−0.711	1.016	1292	603	0.8382	0.9981	−0.134	1.125	1300	593
0.9024	1.0330	−0.294	1.238	1328	549	0.9213	1.0364	−0.059	1.303	1328	547
1.0000	1.0744		1.514	1364	500	1.0000	1.0744		1.514	1364	500
					$T = 313.15$ K						
0.0000	0.6422		0.252	1008	1533	0.0000	0.6870		0.415	1112	1177
0.1027	0.6851	−0.184	0.283	1036	1360	0.1290	0.7253	0.160	0.447	1124	1091
0.2076	0.7303	−0.533	0.335	1064	1209	0.2456	0.7629	0.118	0.492	1144	1002
0.3084	0.7737	−0.723	0.383	1096	1076	0.3586	0.8018	0.005	0.556	1164	920
0.4087	0.8173	−0.899	0.454	1128	962	0.4655	0.8407	−0.078	0.621	1184	849
0.5114	0.8617	−0.944	0.542	1164	857	0.5672	0.8797	−0.129	0.705	1204	784
0.6115	0.9053	−0.954	0.635	1200	767	0.6620	0.9180	−0.170	0.788	1228	722
0.7092	0.9473	−0.844	0.780	1240	687	0.7528	0.9565	−0.187	0.913	1252	667
0.8069	0.9897	−0.730	0.935	1280	617	0.8382	0.9943	−0.165	1.033	1280	614
0.9024	1.0291	−0.320	1.135	1316	561	0.9213	1.0324	−0.072	1.193	1308	566
1.0000	1.0703		1.376	1348	514	1.0000	1.0703		1.376	1348	514

Table 3. Fitting Parameters and Standard Deviations (σ) of Equation 3 for the Least-Squares Representation of V^E

	T	$cm^3 \cdot mol^{-1}$			
system	K	A_0	A_1	A_2	σ
methyl benzoate (1) + cyclohexane (2)	303.15	2.221	−1.938	0.805	0.011
	308.15	2.147	−1.951	0.774	0.012
	313.15	2.052	−2.031	0.708	0.008
methyl benzoate (1) + *n*-hexane (2)	303.15	−3.729	1.145	1.735	0.061
	308.15	−3.848	0.929	1.304	0.055
	313.15	−3.984	1.077	1.435	0.055
methyl benzoate (1) + heptane (2)	303.15	−1.329	−0.182	−0.199	0.011
	308.15	−1.734	−0.283	0.123	0.015
	313.15	−1.885	−0.370	0.101	0.015
methyl benzoate (1) + octane (2)	303.15	−0.409	−1.903	1.083	0.031
	308.15	−0.422	−1.495	1.179	0.012
	313.15	−0.427	−1.644	1.045	0.019

V^E, and isentropic compressibility, K_s, for (methyl benzoate + cyclohexane), (methyl benzoate + hexane), (methyl benzoate + heptane), and (methyl benzoate + octane) as a function of mole fraction of methyl benzoate. The density values have been used to calculate excess volumes, V^E, using the following equation.

$$V^E = (x_1M_1 + x_2M_2)/\rho - (x_1M_1/\rho_1 + x_2M_2/\rho_2) \quad (1)$$

where ρ is the density of the mixture and x_1, M_1, and ρ_1 and x_2, M_2, and ρ_2 are the mole fraction, molar mass, and density of pure components 1 and 2, respectively.

The speed of sound, u, was used to calculate the isentropic compressibility, K_s using

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

The excess volumes V^E were fitted by the method of nonlinear least-squares to a Redlich−Kister type polynomial.[13]

$$V^E = x_1x_2\sum A_i(x_1 - x_2)^i \quad (3)$$

The values of coefficient A_i were determined by a regression analysis based on the least-squares method and are reported along with the corresponding standard deviations between the experimental and the calculated values of the respective functions in Table 3.

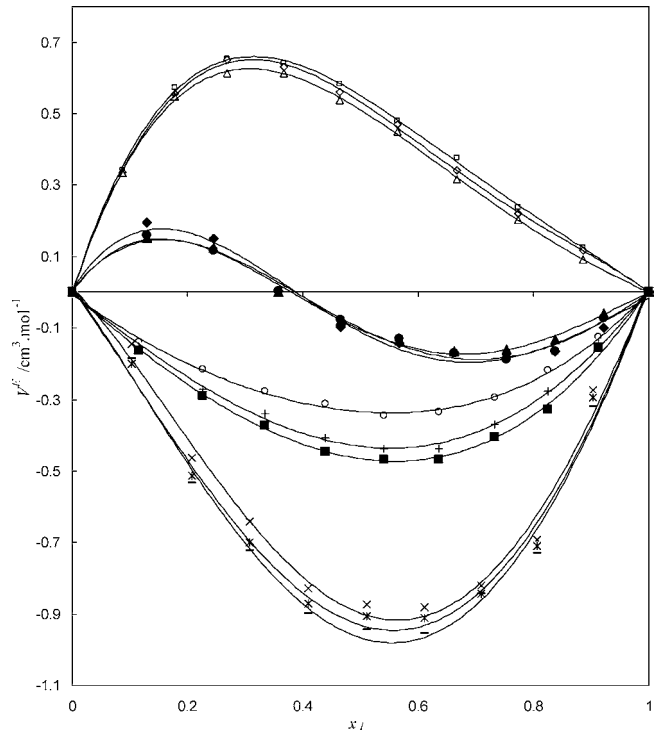

Figure 1. Excess volume, V^E, from eq 3 as a function of methyl benzoate mole fraction. (Methyl benzoate + cyclohexane): □, T = 303.15 K; ◇, T = 308.15 K; △, T = 313.15 K. (Methyl benzoate + *n*-hexane): ×, T = 303.15 K; ∗, T = 308.15 K; −, T = 313.15 K. (Methyl benzoate + heptane): ○, T = 303.15 K; +, T = 308.15 K; ■, T = 313.15 K. (Methyl benzoate + octane): ◆, T = 303.15 K; ▲, T = 308.15 K; ●, T = 313.15 K.

The standard deviation (σ) was calculated using the relation

$$\sigma(V^E) = [\sum (V^E_{obsd} - V^E_{calcd})^2/n - m]^{1/2} \quad (4)$$

where n represents the number of experimental points and m is the number of adjustable parameters.

Figure 1 displays the dependence of V^E on the mole fraction of ester (x_1) for all of the studied systems at temperatures of (303.15, 308.15, and 313.15) K. The V^E for (methyl benzoate + cyclohexane) are positive, while for (methyl benzoate + *n*-hexane) and (methyl benzoate + heptane) they are negative over the whole composition range. However, for (methyl

Table 4. Adjustable Parameters and Percentage Standard Deviation σ (%) of Equations 5 and 6 for the Viscosities of Binary Liquid Mixtures

T	Auslander				McAllister four-body model			
K	B_{12}	A_{21}	B_{21}	σ (%)	ν_{1112}	ν_{1122}	ν_{2221}	σ (%)
			Methyl Benzoate (1) + Cyclohexane (2)					
303.15	−0.2461	0.4832	4.4488	0.17	1.2487	1.1052	0.8728	0.18
308.15	−0.3076	0.2909	7.2194	0.30	1.1434	1.0746	0.7793	0.36
313.15	−0.1446	0.3368	5.9487	0.40	1.0544	0.9793	0.7707	0.41
			Methyl Benzoate (1) + *n*-Hexane (2)					
303.15	0.2319	1.1546	2.3359	0.57	1.0124	0.6556	0.5315	0.34
308.15	0.1728	0.9792	2.7348	0.59	0.9220	0.6332	0.4776	0.42
313.15	0.2025	1.03936	2.4919	0.59	0.8611	0.5885	0.4535	0.43
			Methyl Benzoate (1) + Heptane (2)					
303.15	0.0724	0.6128	4.6227	0.32	1.0047	0.7818	0.5833	0.29
308.15	0.1581	0.9827	2.7894	0.31	1.0636	0.7944	0.6854	0.25
313.15	0.0010	0.3581	7.5721	0.38	0.9339	0.7780	0.6351	0.40
			Methyl Benzoate (1) + Octane (2)					
303.15	0.1298	1.0164	2.7663	0.20	1.0505	0.8750	0.6904	0.19
308.15	0.1274	0.8533	3.3532	0.20	0.9548	0.8092	0.6470	0.19
313.15	0.1742	0.9257	2.9851	0.27	0.8852	0.7567	0.6080	0.26

benzoate + octane), the V^E values are both positive and negative at all of the studied temperatures displaying an s-shaped curve. The magnitude of V^E increases with an increase in chain length of the alkanes (*n*-hexane to octane). The more negative value of V^E for *n*-hexane may possibly be explained as a result of an increase in the number of carbon atoms in methyl benzoate, leading to low polarity and weakening of the association between benzoate molecules and hence forming induced dipole−dipole complexes between methyl benzoate and *n*-hexane. In case of cyclohexane, the excess volumes are positive over the whole composition range.

The experimental viscosity data were correlated with the three-parameter equation of Auslander as

$$\eta = \frac{\eta_1 x_1(x_1 + B_{12}x_2) + \eta_2(A_{21}x_2(B_{21}x_1 + x_2))}{x_1(x_1 + B_{12}x_2) + (A_{21}x_2)(B_{21}x_1 + x_2)} \quad (5)$$

where η, η_1, and η_2 are the dynamic viscosities of the mixture and pure components, respectively. B_{12}, A_{21}, and B_{21} are the parameters representing binary 1,2 interactions.

The kinematic viscosities were correlated to McAllister four-body interaction model given by

$$\begin{aligned}\ln \nu = {x_1}^4 \ln \nu_1 + 4{x_1}^3 x_2 \ln \nu_{1112} + 6{x_1}^2{x_2}^2 \ln \nu_{1122} + \\ 4x_1{x_2}^3 \ln \nu_{2221} + {x_2}^4 \ln \nu_2 - \ln[x_1 + (x_2M_2/M_1)] + \\ 4{x_1}^3 x_2 \ln[\{3 + (M_2/M_1)\}/4] + 6{x_1}^2{x_2}^2 \times \\ \ln[\{1 + (M_2/M_1)\}/2] + 4x_1{x_2}^3 \ln[\{1 + (3M_2/M_1)\}/4] + \\ {x_2}^4 \ln(M_2/M_1) \quad (6)\end{aligned}$$

where ν, ν_1, and ν_2 are the kinematic viscosities of the mixture and pure components 1 and 2, respectively. ν_{1112}, ν_{1122}, and ν_{2221} are the model parameters. The correlating ability of eqs 5 and 6 was tested by calculating the standard percentage deviation σ between the experimental and the calculated viscosity as

$$\sigma = [1/(n - m)\sum\{(100(\nu_{exptl} - \nu_{calcd})/\nu_{exptl})^2\}^{1/2}] \quad (7)$$

where n represents the number of data points and m the number of numerical coefficients in the respective equations. Table 4 records the model parameter along with standard percentage deviations σ. It is observed that the values of the parameter B_{12} of the Auslander model for the cyclohexane mixture are negative, whereas A_{21} and B_{21} are positive. However, for the remaining system, they are (B_{12}, A_{21}, and B_{21}) are positive, while for the McAllister model the parameters (ν_{1112}, ν_{1122}, and ν_{2221}) are positive for all of the studied systems. The proposed models provide a reasonably accurate estimation of viscosity of the binary liquid mixtures under consideration at the studied temperatures, which is evident from the small standard percentage deviations between the experimental and the calculated viscosity values. The values of σ for the Auslander (three-parameter) equation are in the range from 0.17 to 0.59, while for the McAllister four-body interaction model they are in the range from 0.18 to 0.43. As could be expected, the McAllister's four-body interaction model is adequate for correlating the kinematic viscosities of the binary mixtures under study.

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Supporting Information Available:

Experimental data summary. This material is available free of charge via the Internet at http://pubs.acs.org.

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