Densities, Speeds of Sound, Excess Molar Volumes, and Excess Isentropic Compressibilities at T = (298.15 and 308.15) K for Methyl Methacrylate + 1-Alkanols (1-Butanol, 1-Pentanol, and 1-Heptanol) + Cyclohexane, + Benzene, + Toluene, + *p*-Xylene, and + Ethylbenzene

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The densities and speeds of sound for ternary mixtures of methyl methacrylate (MMA) + 1-alkanols (1-butanol, 1-pentanol, 1-heptanol) + aromatic organic solvents (benzene, toluene, *p*-xylene, and ethylbenzene) or + cyclohexane have been measured at T/K = (298.15 and 308.15). Excess molar volumes and excess isentropic compressibilities were calculated across the mole fraction range, and the same were compared with the values calculated using similar data for the binary pairs through Redlich−Kister (RK), Tsao and Smith (TS), and Kohler (K) equations. As compared to others, the RK equation predicted the excess molar volumes and excess isentropic compressibilities adequately.

Introduction

Studies on thermodynamic and thermophysical behavior of binary liquid mixtures of acrylic esters + aliphatic and aromatic organic solvents and acrylic esters + alkanols are of great utility from the practical as well as theoretical point of view. The production of higher homologues of acrylic esters is done by the trans-esterification reaction in which a methyl ester is reacted with an alkanol of the desired chain characteristics in an inert medium consisting of an aliphatic or aromatic organic solvent. The knowledge of various excess thermodynamic and thermophysical functions for such mixtures thus is of great help in optimizing the process parameters needed for an efficient design of the trans-esterification process at the industrial scale. Acrylic esters differ from aliphatic esters by the fact that, in the former, there is an unsaturation alongside the esteric functional group in the same molecule. Therefore, the acrylic esters are best candidates for studying the proximity effects due to unsaturation on ester linkage or vice versa. The binary systems of methyl methacrylate (MMA) with methanol and ethanol exhibited positive excess molar and isobaric heat capacities.[1] Our own research group has reported systematic measurements of volumetric, transport, acoustic, and dielectric properties of MMA + 1-alkanols (methanol to 1-hexanol)[2,3] and alkyl (methyl-, ethyl-, and butyl) acrylates + 1-alkanols (1-heptanol to 1-dodecanol)[4−6] and ternary mixtures[7] of methyl acrylate + 1-propanol (or 1-butanol) + organic solvents (*n*-hexane, *n*-heptane, cyclohexane, benzene, and toluene). The analysis of the excess and deviation functions revealed that even though the thermophysical behavior of acrylic ester + 1-alkanol mixtures is similar to that of their counterpart alkyl alkanoate + 1-alkanol systems, the presence of unsaturation in the acrylic esters seems to produce additional specific interactions between the ester and −OH groups. Packing effects in terms of $n-\pi$ interactions between the lone pair electrons of oxygen of alcoholic −OH group and π-electron clouds of ester molecules are also possible. More precise studies on acrylic ester + 1-alkanols and alkyl alkanoate + same 1-alkanols under identical experimental conditions are needed so that one can make direct quantitative comparison to ascertain the role of unsaturation in the interactions between acrylic esters and 1-alkanols.

As far as we are aware, the studies dealing with the thermophysical behavior of acrylic esters + 1-alkanols + organic solvents (aliphatic as well as aromatic) are scarce in the literature. Therefore, as a continuation of our previous work, this paper reports densities, ρ, and speeds of sound, u, excess molar volumes, V_m^E, and excess isentropic compressibilities, κ_s^E, for the ternary systems of MMA + 1-alkanols (1-butanol, 1-pentanol, 1-heptanol) + aromatic organic solvents (benzene, toluene, *p*-xylene, and ethylbenzene) or + cyclohexane at T/K = (298.15 and 308.15). The applicability of semi-empirical relations based on Redlich−Kister (RK), Tsao and Smith (TS), and Kohler (K) equations was tested by calculating the ternary excess quantities from the similar data of respective binary mixtures.[8−11]

Experimental Section

Materials. MMA of puriss grade (> 99.5 % pure on a mole basis) was supplied by Fluka, and it was used without any further purification. 1-Butanol (1-BuOH) was a technical grade (> 99 % pure on a mole basis) chemical from SD Fine Chemicals, India.

l-Pentanol (1-PeOH) and l-heptanol (1-HtOH) of analytical reagent quality (> 99 % pure on mole basis) were purchased from local suppliers. Cyclohexane of a reagent grade (with 99.5 % pure on mole basis), benzene, toluene, *p*-xylene, and ethylbenzene of analytical reagent grade (with 99.5 % pure on mole basis) were purchased locally. The above chemicals were purified by standard procedures.[6,7,12]

Methods. The ternary mixtures were prepared by mass in hermetically sealed glass vials of about 15 cm^3. The solutions of each composition were prepared fresh, and the primary properties were measured on the same day to avoid any error in the measurement due to evaporation loss. The uncertainty in the mole fraction was estimated to be less than ± 0.0001.

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Table 1. Densities ρ and Speeds of Sound u for Pure Components at T = (298.15 and 308.15) K

	T/K = 298.15		T/K = 308.15	
	exp.	lit.	exp.	lit.
		MMA		
ρ/(g·cm^{-3})	0.93760	0.937615[17]	0.92570	0.92574[18]
u/(m·s^{-1})	1182	1181[18]	1152	1153[18]
		1-Butanol		
ρ/(g·cm^{-3})	0.80591	0.80586[19]	0.79821	0.79820[20]
u/(m·s^{-1})	1244	1244[21]	1212	1205.5[19]
		1-Pentanol		
ρ/(g·cm^{-3})	0.81076	0.81075[22]	0.80341	0.803442[23]
u/(m·s^{-1})	1274	1274.32[22]	1240	1241.84[24]
		1-Heptanol		
ρ/(g·cm^{-3})	0.81891	0.818813[25]	0.81194	0.811730[25]
u/(m·s^{-1})	1330	1330[21]	1292	1295[26]
		Cyclohexane		
ρ/(g·cm^{-3})	0.77386	0.77393[27]	0.76446	0.7644[28]
u/(m·s^{-1})	1254	1255[29]	1212	1208[28]
		Benzene		
ρ/(g·cm^{-3})	0.87358	0.87357[30]	0.86296	0.86290[31]
u/(m·s^{-1})	1300	1299.73[32]	1260	
		Toluene		
ρ/(g·cm^{-3})	0.86221	0.86220[32]	0.85286	0.85287[33]
u/(m·s^{-1})	1304	1305.38[32]	1262	1262.6[33]
		p-Xylene		
ρ/(g·cm^{-3})	0.85670	0.85668[34]	0.84788	0.84783[35]
u/(m·s^{-1})	1314	1316[36]	1272	1266.4[35]
		Ethylbenzene		
ρ/(g·cm^{-3})	0.86263	0.86252[37]	0.85389	0.85415[37]
u/(m·s^{-1})	1319	1318[37]	1276	1276[37]

Densities, ρ, and speeds of sound, u, of the pure liquids and their mixtures were measured with a high-precision vibrating tube digital densimeter (Anton Paar, DMA 5000) and an ultrasonic interferometer (Mittal Enterprises, New Delhi, India) operating at a fixed frequency of 2 MHz. The densimeter was calibrated with air and freshly degassed, four times distilled water at measuring temperatures. The instrument has a built-in temperature adjustment unit, and the temperature around the tube was maintained to a precision of ± 0.001 K, but the accuracy in the temperature was ± 0.01 K as Pt100 sensors were employed. The temperature within the measuring cell of interferometer was maintained by circulating water from thermostatic baths maintained within ± 0.01 K. The repeatability in the measured densities and speed of sound were $3 \cdot 10^{-6}$ g·cm^{-3} and 1.1 m·s^{-1}. The experimental ρ and u for the pure components along with the comparison of the literature values are given in Table 1. The absolute mean deviations between our experimental and literature reported densities and speeds of sound were $6.2 \cdot 10^{-5}$ g·cm^{-3} and 1.6 m·s^{-1}.

Equations

The excess molar volumes of the ternary mixture V^{E}_{m123} were calculated from the relation;

$$V^{E}_{m123} = V - \sum_{i=1}^{i=3} x_i V_i \quad (1)$$

where V represents the molar volume of ternary mixtures, V_i and x_i refer to the molar volume and mole fraction of the i^{th} component. The V is calculated from the measured densities of ternary mixtures, ρ_{123}, and molar masses of respective components, M_i, through the relation;

$$V = (\sum_{i=1}^{i=3} x_i M_i)/\rho_{123} \quad (2)$$

The experimental ρ_{123} and V^{E}_{m123} data for 15 ternary mixtures are listed in Table 2 and Table S1 (Supporting Information), respectively. The V^{E}_{m123} values are also calculated from the binary contributions, that is, $V_{m12}{}^{E}$, $V_{m23}{}^{E}$, and $V_{m13}{}^{E}$ using the RK,[13] TS,[14] and K[15] equations:

$$V^{E}_{m123}(\mathrm{RK}) = V^{E}_{m12} + V^{E}_{m23} + V^{E}_{m13} \quad (3)$$

$$V^{E}_{m123}(\mathrm{TS}) = [x_2 V^{E}_{m12}/(1 - x_1)] + V^{E}_{m13}[x_3/(1 - x_1)] + (1 - x_1)V^{E}_{m23} \quad (4)$$

where $V_{mij}{}^{E}$ refer to the excess molar volumes for the binary mixtures at compositions $x_i{}^{o}$ and $x_j{}^{o}$ such that $x_i{}^{o} = x_1$ for 1 + 2 and 1 + 3 pairs and $x_j{}^{o} = x_2/(x_2 + x_3)$ for the 2 + 3 pair.

$$V^{E}_{m123}(\mathrm{K}) = (x_1 + x_2)^2 V^{E}_{m12} + (x_1 + x_3)^2 V^{E}_{m13} + (x_2 + x_3)^2 V^{E}_{m23} \quad (5)$$

where $V_{mij}{}^{E}$ is the binary contribution of the ternary property at $x_i{}^{o}$ and $x_j{}^{o}$; $x_i{}^{o} = 1 - x_i{}^{o} = x_i/(x_i + x_j)$. The subscripts i and j represents for example 1 and 2, 1 and 3, and 2 and 3 for 1 + 2, 1 + 3, and 2 + 3 pair of mixtures. The binary contributions of respective 1 + 2, 1 + 3, and 2 + 3 pairs to the excess molar volumes of ternary mixtures were then calculated using the equation,

$$V^{E}_{ij} = x_i x_j \sum_{n=0}^{n=2} a_n (2x_1 - 1)^n \quad (6)$$

where i and j are either 1 + 2, 1 + 3, or 2 + 3 for respective binary systems and a_n are the coefficients obtained by a multiple regression analysis based on least-squares method.[8−11] The summary of the coefficients along with the values of standard deviations, σ, is given in Table S3.

The experimental speeds of sound for ternary mixtures were used in conjunction with density values to calculate the isentropic compressibilities, κ^{E}_{s123}, using the Laplace equation, that is, $\kappa^{E}_{s123}\mathrm{E} = 1/(u2_{123}\rho_{123})$. The κ^{E}_{s123} were calculated using the relation,

$$\kappa^{E}_{s123}/(\mathrm{TPa}^{-1}) = \kappa_s - \kappa_s{}^{id} \quad (7)$$

where κ_{s123} is the isentropic compressibility for a given composition of the ternary mixture and $\kappa_s{}^{id}$ is the ideal contribution calculated from the relations;[16]

$$\kappa_s{}^{id} = \sum_{i=1}^{3} \phi_i[\kappa_{si} + TV_i(\alpha_i{}^2)/C_{pi}] - \{T(\sum_{i=1}^{3} x_i V_i)(\sum_{i=1}^{3} \phi_i \alpha_i)^2 / \sum_{i=1}^{3} x_i C_{pi}\} \quad (8)$$

Table 2. Densities, ρ_{123}, and Speed of Sound, u_{123}, for MMA (1) + 1-Alkanols (2) + Organic Solvents (3) at T = (298.15 and 308.15) K

		ρ_{123} / g·cm^{-3}		u_{123} / m·s^{-1}				ρ_{123} / g·cm^{-3}		u_{123} / m·s^{-1}	
x_1	x_2					x_1	x_2				
	T/K	298.15	308.15	298.15	308.15		T/K	298.15	308.15	298.15	308.15
MMA (1) + 1-Butanol (2) + Cyclohexane (3)						MMA (1) + 1-Butanol (2) + Benzene (3)					
0.0512	0.1911	0.78450	0.77446	1226	1182	0.0490	0.1901	0.86128	0.85093	1273	1233
0.1005	0.1845	0.79114	0.78085	1213	1170	0.1004	0.1801	0.86572	0.85528	1272	1230
0.2006	0.1574	0.80508	0.79433	1176	1140	0.2005	0.1588	0.87441	0.86382	1266	1221
0.2805	0.1425	0.81703	0.80603	1148	1117	0.2812	0.1439	0.88116	0.87047	1259	1212
0.3721	0.1245	0.83118	0.82005	1126	1100	0.3723	0.1257	0.88877	0.87794	1250	1202
0.4870	0.1098	0.84978	0.83871	1123	1097	0.4879	0.1027	0.89826	0.88720	1238	1192
0.5910	0.0865	0.86652	0.85564	1135	1107	0.5907	0.0817	0.90656	0.89526	1226	1184
0.6921	0.0695	0.88340	0.87273	1156	1124	0.6917	0.0617	0.91450	0.90295	1215	1177
0.7642	0.0451	0.89499	0.88441	1168	1133	0.7642	0.0475	0.92007	0.90836	1207	1172
0.0545	0.3785	0.78994	0.78033	1222	1181	0.0546	0.3770	0.84852	0.83859	1258	1222
0.1154	0.3561	0.79850	0.78859	1215	1173	0.1152	0.3530	0.85466	0.84463	1256	1220
0.2064	0.3146	0.81151	0.80117	1199	1159	0.2080	0.3169	0.86391	0.85373	1253	1215
0.3065	0.2665	0.82615	0.81546	1181	1144	0.3055	0.2766	0.87363	0.86328	1247	1207
0.4036	0.2345	0.84125	0.83043	1174	1139	0.4039	0.2384	0.88310	0.87256	1239	1198
0.5001	0.1925	0.85598	0.84513	1168	1134	0.5005	0.1996	0.89232	0.88156	1230	1190
0.5840	0.1664	0.86940	0.85865	1174	1138	0.5981	0.1609	0.90143	0.89043	1220	1183
0.7045	0.1198	0.88845	0.87783	1179	1142	0.7049	0.1178	0.91128	0.90001	1209	1174
0.8156	0.0710	0.90605	0.89538	1181	1144	0.8199	0.0720	0.92165	0.91010	1198	1164
0.0545	0.5625	0.79563	0.78658	1225	1185	0.0528	0.5646	0.83637	0.82698	1247	1214
0.1132	0.5340	0.80431	0.79503	1224	1183	0.1134	0.5302	0.84317	0.83368	1244	1213
0.2018	0.4785	0.81719	0.80750	1222	1179	0.2021	0.4772	0.85317	0.84349	1238	1211
0.3925	0.3684	0.84567	0.83539	1213	1169	0.3924	0.3648	0.87403	0.86389	1224	1200
0.4869	0.3078	0.85949	0.84905	1206	1164	0.4873	0.3084	0.88420	0.87382	1217	1192
0.5789	0.2484	0.87290	0.86239	1200	1159	0.5873	0.2471	0.89488	0.88423	1210	1183
0.6874	0.1895	0.88937	0.87884	1193	1155	0.6872	0.1887	0.90522	0.89431	1202	1174
0.9354	0.0375	0.92706	0.91578	1183	1147	0.9353	0.0374	0.93102	0.91934	1186	1152
0.3046	0.4485	0.83390	0.82397	1218	1174	0.3042	0.4190	0.86432	0.85440	1231	1206
0.8111	0.1241	0.90852	0.89782	1187	1150	0.8101	0.1142	0.91798	0.90672	1194	1163
0.0445	0.7245	0.80005	0.79145	1228	1191	0.0445	0.7646	0.82353	0.81488	1240	1208
0.0908	0.7965	0.81198	0.80356	1230	1192	0.0919	0.7264	0.82956	0.82080	1237	1206
0.1992	0.6801	0.82631	0.81741	1217	1177	0.1994	0.6402	0.84302	0.83393	1226	1203
0.3135	0.5590	0.84120	0.83176	1199	1160	0.3137	0.5492	0.85698	0.84752	1214	1198
0.4152	0.4977	0.85681	0.84714	1182	1146	0.4153	0.4675	0.86921	0.85942	1206	1191
0.2084	0.3949	0.81490	0.80487	1214	1171	0.5087	0.3931	0.88025	0.87017	1201	1184
0.6016	0.3175	0.88106	0.87082	1184	1148	0.6012	0.3190	0.89108	0.88073	1197	1176
0.7065	0.2312	0.89546	0.88502	1187	1150	0.7062	0.2302	0.90359	0.89291	1194	1167
0.7935	0.1756	0.90849	0.89783	1186	1150	0.7931	0.1655	0.91333	0.90240	1190	1160
0.7086	0.2453	0.89674	0.88632	1184	1149	0.7081	0.2332	0.90350	0.89284	1193	1167
0.2586	0.6050	0.83343	0.82420	1208	1169	0.2588	0.6037	0.84966	0.84043	1219	1199
0.1922	0.4440	0.81413	0.80432	1220	1176	0.1921	0.4469	0.85432	0.84454	1242	1212
0.1345	0.3126	0.80005	0.78992	1209	1167	0.1349	0.3155	0.85864	0.84850	1258	1220
0.0695	0.1633	0.78626	0.77611	1223	1179	0.0692	0.1604	0.86498	0.85453	1275	1234
0.3762	0.3786	0.84327	0.83303	1214	1170	0.3761	0.3762	0.87216	0.86207	1225	1201
0.2751	0.2964	0.82212	0.81158	1192	1153	0.2751	0.2762	0.87148	0.86115	1250	1209
0.1780	0.1765	0.80228	0.79164	1186	1148	0.1783	0.1781	0.87142	0.86090	1266	1222
0.5879	0.2601	0.87519	0.86473	1199	1159	0.5881	0.2529	0.89455	0.88392	1209	1183
0.4480	0.1931	0.84698	0.83606	1158	1126	0.4489	0.1928	0.88929	0.87857	1237	1194
0.3092	0.1379	0.82146	0.81040	1140	1111	0.3091	0.1330	0.88389	0.87314	1257	1209
MMA (1) + 1-Butanol (2) + Toluene (3)						MMA (1) + 1-Butanol (2) + *p*-Xylene (3)					
0.0501	0.1915	0.85637	0.82384	1281	1235	0.0540	0.1916	0.85236	0.84338	1294	1246
0.1002	0.1849	0.86065	0.82931	1278	1230	0.0980	0.1845	0.84959	0.84076	1299	1252
0.2003	0.1578	0.86991	0.84047	1271	1221	0.2009	0.1564	0.86445	0.85498	1279	1227
0.2810	0.1429	0.87695	0.84922	1263	1212	0.2795	0.1423	0.87096	0.86124	1271	1217
0.3722	0.1247	0.88485	0.85902	1253	1203	0.3715	0.1212	0.87898	0.86900	1261	1207
0.4874	0.1097	0.89418	0.87102	1240	1191	0.4865	0.1095	0.88874	0.87849	1247	1195
0.5906	0.0867	0.90287	0.88184	1229	1183	0.5945	0.0856	0.89872	0.88822	1234	1187
0.6912	0.0697	0.91094	0.89222	1218	1175	0.6945	0.0654	0.90810	0.89737	1222	1179
0.7641	0.0455	0.91745	0.90001	1210	1171	0.7655	0.0421	0.91523	0.90430	1214	1174
0.0546	0.3787	0.84679	0.82003	1265	1222	0.0556	0.3654	0.84487	0.83602	1280	1234
0.1155	0.3563	0.85274	0.82702	1262	1216	0.1123	0.3564	0.84937	0.84036	1273	1226
0.2060	0.3149	0.86198	0.83751	1257	1207	0.2056	0.3123	0.85825	0.84892	1265	1214
0.3050	0.2666	0.87215	0.84892	1250	1198	0.3046	0.2564	0.86822	0.85856	1257	1204
0.4035	0.2344	0.88126	0.85975	1241	1189	0.4021	0.2465	0.87635	0.86645	1244	1194
0.5001	0.1926	0.89058	0.87045	1232	1183	0.5478	0.1925	0.89050	0.88023	1228	1182
0.5950	0.1669	0.89878	0.88054	1222	1176	0.5845	0.1654	0.89468	0.88430	1226	1180
0.7042	0.1198	0.90909	0.89249	1211	1169	0.7045	0.1178	0.90681	0.89615	1213	1172
0.8195	0.0712	0.92000	0.90517	1200	1162	0.8156	0.0745	0.91822	0.90722	1201	1164
0.0522	0.5626	0.83653	0.81559	1253	1213	0.0465	0.5426	0.83594	0.82729	1266	1224
0.1131	0.5342	0.84279	0.82272	1249	1207	0.3326	0.5321	0.85814	0.84888	1220	1189
0.2020	0.4789	0.85267	0.83337	1242	1199	0.2045	0.4589	0.85163	0.84251	1250	1207
0.3921	0.3688	0.87298	0.85555	1226	1184	0.3945	0.3645	0.87073	0.86103	1230	1191

Table 2. Continued

		ρ_{123}		u_{123}				ρ_{123}		u_{123}	
x_1	x_2	g·cm^{-3}		m·s^{-1}		x_1	x_2	g·cm^{-3}		m·s^{-1}	
	T/K	298.15	308.15	298.15	308.15		T/K	298.15	308.15	298.15	308.15
MMA (1) + 1-Butanol (2) + Toluene (3)						MMA (1) + 1-Butanol (2) + *p*-Xylene (3)					
0.4872	0.3090	0.88319	0.86656	1219	1177	0.4856	0.3046	0.88069	0.87071	1223	1184
0.5870	0.2491	0.89362	0.87794	1212	1171	0.5879	0.2561	0.89119	0.88096	1212	1176
0.6871	0.1898	0.90397	0.88930	1204	1166	0.6895	0.1798	0.90294	0.89241	1207	1170
0.9352	0.0379	0.93042	0.91797	1187	1151	0.9345	0.0345	0.93030	0.91881	1188	1152
0.3045	0.4487	0.86205	0.84473	1230	1189	0.3012	0.4326	0.86030	0.85092	1237	1198
0.8105	0.1242	0.91639	0.90323	1194	1158	0.7956	0.1168	0.91466	0.90380	1198	1162
0.0449	0.7246	0.82669	0.81099	1245	1208	0.0445	0.7156	0.82690	0.81846	1251	1213
0.0910	0.7964	0.82582	0.81401	1237	1200	0.0890	0.7895	0.82595	0.81763	1239	1203
0.1998	0.6802	0.84092	0.82839	1226	1190	0.1897	0.6856	0.83929	0.83060	1230	1196
0.3136	0.5592	0.85639	0.84310	1216	1183	0.3156	0.5487	0.85600	0.84680	1221	1190
0.4151	0.4975	0.86728	0.85493	1206	1175	0.4189	0.4985	0.86662	0.85713	1209	1181
0.5089	0.3951	0.87982	0.86680	1202	1171	0.5145	0.3565	0.88081	0.87088	1210	1179
0.6019	0.3178	0.89075	0.87793	1199	1166	0.6012	0.3048	0.89020	0.88005	1203	1172
0.7067	0.2315	0.90298	0.89041	1195	1161	0.7065	0.2312	0.90216	0.89171	1196	1164
0.7939	0.1795	0.91208	0.90044	1189	1155	0.7926	0.1658	0.91226	0.90152	1192	1158
0.7085	0.2452	0.90233	0.89031	1193	1159	0.7056	0.2398	0.90171	0.89128	1195	1163
0.2585	0.6045	0.84972	0.83633	1222	1187	0.2565	0.6021	0.84875	0.83977	1226	1194
0.1924	0.4444	0.85385	0.83315	1247	1202	0.1948	0.4498	0.85131	0.84220	1252	1209
0.1345	0.3125	0.85657	0.83003	1264	1216	0.1323	0.3098	0.85289	0.84377	1275	1226
0.0699	0.1634	0.85940	0.82659	1282	1235	0.0678	0.1548	0.85489	0.84585	1295	1247
0.3766	0.3782	0.87132	0.85375	1228	1185	0.3564	0.3598	0.86789	0.85826	1237	1195
0.2754	0.2962	0.86830	0.84518	1251	1200	0.2745	0.2345	0.86680	0.85719	1263	1210
0.1782	0.1765	0.86721	0.83773	1271	1222	0.1795	0.1548	0.86293	0.85352	1282	1230
0.5888	0.2589	0.89319	0.87791	1210	1171	0.5842	0.2346	0.89179	0.88153	1216	1177
0.4482	0.1935	0.88677	0.86521	1239	1188	0.4479	0.1958	0.88214	0.87207	1243	1192
0.3098	0.1389	0.87937	0.85229	1259	1209	0.3012	0.1354	0.87291	0.86313	1269	1215
MMA (1) + 1-Butanol (2) + Ethylbenzene (3)						MMA (1) + 1-Pentanol (2) + Cyclohexane (3)					
0.0481	0.1900	0.86115	0.84756	1299	1252	0.0611	0.1915	0.78726	0.77763	1231	1187
0.1005	0.1806	0.86477	0.85127	1295	1245	0.0987	0.1849	0.79205	0.78220	1225	1182
0.2006	0.1584	0.87211	0.85874	1285	1231	0.1999	0.1578	0.80547	0.79510	1212	1171
0.2815	0.1438	0.87802	0.86478	1276	1219	0.2798	0.1429	0.81702	0.80636	1203	1163
0.3722	0.1254	0.88490	0.87180	1265	1209	0.3698	0.1247	0.83052	0.81968	1195	1157
0.4877	0.1028	0.89381	0.88093	1250	1197	0.4854	0.1097	0.84884	0.83801	1188	1151
0.5906	0.0815	0.90200	0.88934	1237	1188	0.5912	0.0867	0.86572	0.85503	1184	1147
0.6915	0.0615	0.91020	0.89779	1224	1180	0.6901	0.0697	0.88212	0.87160	1183	1145
0.7642	0.0480	0.91624	0.90401	1215	1174	0.7654	0.0455	0.89448	0.88400	1182	1144
0.0545	0.3774	0.85135	0.83893	1284	1237	0.0541	0.3787	0.79270	0.78356	1229	1189
0.1151	0.3528	0.85638	0.84402	1279	1229	0.1123	0.3563	0.79999	0.79056	1221	1183
0.2079	0.3165	0.86417	0.85187	1271	1217	0.2015	0.3149	0.81158	0.80170	1211	1174
0.3052	0.2765	0.87259	0.86034	1262	1205	0.3035	0.2666	0.82554	0.81525	1201	1166
0.4037	0.2385	0.88113	0.86894	1251	1195	0.4032	0.2344	0.84034	0.82987	1195	1160
0.5008	0.1997	0.88974	0.87763	1240	1187	0.4926	0.1926	0.85343	0.84288	1190	1155
0.5984	0.1612	0.89852	0.88651	1228	1180	0.5941	0.1669	0.86955	0.85910	1187	1151
0.7045	0.1175	0.90836	0.89646	1216	1173	0.7056	0.1198	0.88695	0.87655	1184	1148
0.8201	0.0740	0.91923	0.90745	1202	1164	0.8154	0.0712	0.90466	0.89415	1183	1145
0.0525	0.5645	0.84031	0.82912	1268	1225	0.0518	0.5626	0.79910	0.79043	1233	1195
0.1135	0.5301	0.84611	0.83495	1262	1218	0.1126	0.5342	0.80681	0.79791	1225	1190
0.2025	0.4775	0.85479	0.84361	1253	1208	0.2018	0.4789	0.81815	0.80886	1216	1181
0.3921	0.3648	0.87358	0.86229	1233	1191	0.3918	0.3688	0.84411	0.83420	1201	1167
0.4872	0.3083	0.88311	0.87177	1224	1183	0.4865	0.309	0.85735	0.84726	1195	1161
0.5874	0.2470	0.89332	0.88195	1215	1176	0.5789	0.2491	0.87046	0.86027	1191	1156
0.6875	0.1890	0.90350	0.89213	1207	1169	0.6865	0.1898	0.88658	0.87634	1187	1151
0.9352	0.0376	0.93025	0.91857	1187	1152	0.9425	0.0379	0.92753	0.91628	1182	1146
0.3042	0.4191	0.86472	0.85350	1242	1198	0.3012	0.4487	0.83271	0.82316	1209	1175
0.7989	0.1140	0.91561	0.90411	1199	1162	0.8095	0.1242	0.90585	0.89541	1185	1148
0.0445	0.7648	0.82668	0.81697	1252	1214	0.0435	0.7246	0.80440	0.79615	1242	1206
0.0915	0.7269	0.83213	0.82238	1247	1209	0.0909	0.7964	0.81482	0.80674	1245	1209
0.1996	0.6409	0.84457	0.83466	1233	1199	0.1985	0.6802	0.82648	0.81793	1230	1195
0.3135	0.5490	0.85771	0.84754	1220	1190	0.3088	0.5592	0.83890	0.82981	1217	1182
0.4150	0.4678	0.86930	0.85891	1210	1183	0.4098	0.4975	0.85291	0.84358	1211	1175
0.5052	0.3936	0.87968	0.86911	1204	1176	0.2102	0.3951	0.81605	0.80643	1212	1177
0.6011	0.3195	0.89044	0.87974	1199	1170	0.6048	0.3178	0.87798	0.86809	1196	1160
0.7056	0.2305	0.90270	0.89178	1196	1164	0.7071	0.2315	0.89204	0.88193	1190	1153
0.7929	0.1654	0.91258	0.90153	1191	1158	0.7945	0.1795	0.90552	0.89519	1187	1150
0.7085	0.2334	0.90276	0.89188	1195	1163	0.7023	0.2452	0.89182	0.88175	1191	1154
0.2584	0.6039	0.85070	0.84075	1225	1193	0.2578	0.6045	0.83260	0.82372	1222	1187
0.1926	0.4465	0.85596	0.84451	1259	1211	0.1922	0.4444	0.81526	0.80586	1215	1180
0.1345	0.3154	0.85963	0.84708	1280	1228	0.1365	0.3125	0.80186	0.79220	1218	1180
0.0695	0.1609	0.86397	0.85027	1299	1251	0.0659	0.1634	0.78696	0.77725	1231	1187
0.3759	0.3761	0.87188	0.86060	1234	1192	0.3765	0.3782	0.84199	0.83212	1202	1168
0.2754	0.2756	0.87070	0.85834	1266	1209	0.2732	0.2962	0.82193	0.81180	1204	1169
0.1782	0.1782	0.86966	0.85636	1287	1232	0.1795	0.1765	0.80316	0.79291	1214	1173
0.5882	0.2526	0.89309	0.88176	1214	1176	0.5846	0.2589	0.87187	0.86173	1192	1156
0.4482	0.1935	0.88647	0.87412	1249	1193	0.4501	0.1935	0.84639	0.83579	1191	1156
0.3078	0.1336	0.88025	0.86702	1273	1217	0.3084	0.1389	0.82132	0.81059	1200	1161

Table 2. Continued

		ρ_{123}		u_{123}				ρ_{123}		u_{123}	
x_1	x_2	g·cm^{-3}		m·s^{-1}		x_1	x_2	g·cm^{-3}		m·s^{-1}	
	T/K	298.15	308.15	298.15	308.15		T/K	298.15	308.15	298.15	308.15
MMA (1) + 1-Pentanol (2) + Benzene (3)						MMA (1) + 1-Pentanol (2) + Toluene (3)					
0.0430	0.1898	0.86020	0.85011	1273	1229	0.0431	0.1912	0.85460	0.82247	1282	1237
0.1001	0.1784	0.86414	0.85469	1272	1221	0.1012	0.1835	0.85840	0.82859	1280	1230
0.2012	0.1580	0.87247	0.86292	1267	1208	0.2011	0.1565	0.86703	0.83948	1273	1219
0.2811	0.1432	0.87725	0.86932	1260	1199	0.2795	0.1432	0.87376	0.84782	1266	1209
0.3720	0.1256	0.88636	0.87663	1251	1190	0.3719	0.124	0.88114	0.85770	1256	1199
0.4875	0.1025	0.89509	0.88588	1238	1182	0.4874	0.1087	0.89093	0.86968	1243	1187
0.5905	0.0815	0.90362	0.89405	1227	1176	0.5895	0.0865	0.89925	0.88046	1231	1180
0.6915	0.0621	0.91188	0.90186	1216	1171	0.6915	0.0684	0.90803	0.89112	1219	1173
0.7642	0.0478	0.91745	0.90744	1208	1167	0.7645	0.0445	0.91541	0.89924	1211	1169
0.0542	0.3775	0.84824	0.83838	1262	1216	0.0541	0.3784	0.84546	0.81993	1272	1226
0.1152	0.3529	0.85280	0.84387	1261	1208	0.1154	0.3559	0.85027	0.82637	1269	1219
0.2079	0.3156	0.86104	0.85234	1258	1197	0.2059	0.3138	0.85824	0.83618	1264	1208
0.3052	0.2765	0.86959	0.86132	1252	1187	0.3045	0.2654	0.86747	0.84708	1257	1198
0.4035	0.2384	0.87818	0.87031	1243	1179	0.4032	0.2323	0.87615	0.85766	1247	1188
0.5002	0.1998	0.88714	0.87922	1234	1173	0.4995	0.1920	0.88529	0.86823	1237	1181
0.6985	0.1611	0.89605	0.89424	1207	1163	0.5948	0.1658	0.89378	0.87830	1226	1174
0.7056	0.1175	0.90641	0.89812	1211	1165	0.7046	0.1189	0.90433	0.89059	1213	1167
0.8195	0.0716	0.91763	0.90868	1198	1159	0.8154	0.0722	0.91604	0.90331	1201	1160
0.0521	0.5648	0.83818	0.82755	1257	1214	0.0531	0.5636	0.83724	0.81648	1265	1222
0.1132	0.5315	0.84334	0.83325	1254	1206	0.1132	0.5341	0.84167	0.82266	1260	1215
0.2019	0.4769	0.85049	0.84204	1247	1198	0.2019	0.4784	0.84933	0.83216	1253	1206
0.3920	0.3649	0.86880	0.86103	1230	1183	0.3915	0.3678	0.86679	0.85282	1235	1190
0.4871	0.3082	0.87750	0.87071	1221	1177	0.4869	0.3078	0.87626	0.86355	1226	1182
0.5871	0.2469	0.88823	0.88112	1213	1170	0.5481	0.2484	0.88601	0.87116	1224	1177
0.6870	0.1891	0.89838	0.89139	1204	1164	0.6871	0.1878	0.89656	0.88649	1207	1166
0.9352	0.0378	0.92878	0.91842	1186	1150	0.9351	0.0375	0.92836	0.91708	1187	1150
0.3042	0.4189	0.86025	0.85206	1237	1190	0.3045	0.4478	0.85694	0.84255	1240	1197
0.8102	0.1146	0.91237	0.90453	1195	1157	0.8104	0.1252	0.90990	0.90084	1195	1156
0.0450	0.7642	0.82910	0.81691	1258	1219	0.0442	0.7245	0.83044	0.81267	1262	1223
0.0912	0.7269	0.83322	0.82159	1253	1213	0.0894	0.7954	0.82972	0.81518	1256	1218
0.1991	0.6401	0.84187	0.83276	1238	1203	0.1995	0.6806	0.83928	0.82740	1241	1206
0.3135	0.5489	0.85236	0.84492	1222	1196	0.3134	0.5595	0.85118	0.84060	1227	1196
0.4150	0.4656	0.86234	0.85611	1211	1188	0.4098	0.4986	0.86038	0.85100	1215	1187
0.5090	0.3928	0.87238	0.86641	1204	1180	0.5079	0.3945	0.87210	0.86299	1209	1179
0.6011	0.3194	0.88241	0.87682	1199	1172	0.6015	0.3168	0.88208	0.87411	1203	1171
0.7065	0.2301	0.89523	0.88944	1195	1162	0.7065	0.2323	0.89405	0.88687	1198	1162
0.7934	0.1652	0.90595	0.89946	1191	1155	0.7937	0.1789	0.90374	0.89725	1191	1154
0.7086	0.2337	0.89525	0.88928	1195	1162	0.7084	0.2457	0.89292	0.88660	1196	1160
0.2598	0.6035	0.84686	0.83853	1229	1200	0.2584	0.6035	0.84601	0.83452	1234	1201
0.1924	0.4462	0.85176	0.84323	1250	1198	0.1925	0.4245	0.85030	0.83244	1258	1208
0.1348	0.3154	0.85663	0.84755	1262	1207	0.1341	0.3154	0.85346	0.82904	1270	1218
0.0690	0.1602	0.86392	0.85401	1274	1228	0.0687	0.1652	0.85640	0.82568	1283	1236
0.3748	0.3759	0.86645	0.85924	1231	1185	0.3759	0.3784	0.86499	0.85106	1236	1191
0.2760	0.2758	0.86775	0.85946	1254	1190	0.2752	0.2954	0.86350	0.84345	1258	1200
0.1780	0.1784	0.86923	0.85986	1267	1209	0.1778	0.1754	0.86388	0.83662	1274	1220
0.5886	0.2525	0.88718	0.88081	1212	1170	0.5879	0.2568	0.88562	0.87471	1215	1173
0.4485	0.1926	0.88456	0.87649	1241	1177	0.4451	0.1932	0.88231	0.86289	1245	1186
0.3096	0.1335	0.88091	0.87194	1257	1196	0.3078	0.1365	0.87692	0.85088	1263	1206
MMA (1) + 1-Pentanol (2) + *p*-Xylene (3)						MMA (1) + 1-Pentanol (2) + Ethylbenzene (3)					
0.0432	0.1946	0.85029	0.84231	1292	1248	0.0430	0.1901	0.85946	0.84742	1298	1251
0.1050	0.1851	0.84880	0.84067	1296	1252	0.1010	0.1800	0.86270	0.85126	1293	1242
0.2004	0.1523	0.86240	0.85441	1279	1231	0.2012	0.1588	0.86973	0.85835	1284	1226
0.2787	0.1421	0.86792	0.86033	1271	1222	0.2798	0.1439	0.87541	0.86404	1275	1215
0.3564	0.1221	0.87622	0.86688	1263	1214	0.3659	0.1257	0.88224	0.87057	1265	1205
0.4854	0.1094	0.88581	0.87729	1247	1200	0.4879	0.1027	0.89115	0.88003	1250	1193
0.5898	0.0845	0.89552	0.88680	1235	1190	0.5915	0.0817	0.89936	0.88846	1237	1185
0.6978	0.0645	0.90497	0.89668	1222	1180	0.6948	0.0617	0.90782	0.89715	1224	1177
0.7598	0.0418	0.91239	0.90310	1215	1175	0.7559	0.0475	0.91420	0.90260	1216	1173
0.0549	0.3665	0.84400	0.83585	1281	1237	0.0554	0.3771	0.84925	0.83967	1284	1237
0.1145	0.3545	0.84747	0.84007	1275	1231	0.1146	0.3530	0.85349	0.84408	1279	1228
0.2046	0.3102	0.85469	0.84784	1268	1221	0.2074	0.3169	0.86039	0.85121	1271	1214
0.3048	0.2560	0.86417	0.85711	1259	1211	0.3049	0.2766	0.86822	0.85916	1262	1201
0.4035	0.2456	0.87126	0.86469	1247	1201	0.4039	0.2384	0.87655	0.86742	1252	1191
0.5578	0.1931	0.88437	0.87885	1230	1186	0.5011	0.1996	0.88477	0.87589	1241	1183
0.5848	0.1655	0.88939	0.88232	1228	1185	0.5978	0.1609	0.89129	0.88465	1229	1176
0.7051	0.1174	0.90238	0.89440	1215	1174	0.7045	0.1178	0.90384	0.89474	1216	1170
0.8168	0.0751	0.91379	0.90588	1202	1163	0.8198	0.0720	0.91568	0.90618	1202	1162
0.0487	0.5421	0.83674	0.82798	1273	1232	0.0519	0.5646	0.83975	0.83049	1273	1231
0.3333	0.5332	0.85330	0.84623	1230	1199	0.1132	0.5302	0.84406	0.83539	1267	1222
0.2048	0.4584	0.84924	0.84130	1257	1216	0.2014	0.4772	0.85106	0.84291	1259	1212
0.3949	0.3641	0.86452	0.85852	1236	1198	0.3948	0.3648	0.86750	0.86024	1240	1194
0.4879	0.3035	0.89341	0.86814	1228	1190	0.4856	0.3084	0.87668	0.86905	1231	1186
0.5978	0.2550	0.88361	0.87877	1215	1178	0.5946	0.2471	0.88624	0.87969	1219	1176
0.6889	0.1784	0.89573	0.88981	1210	1171	0.6848	0.1887	0.89682	0.88930	1211	1169
0.9365	0.0343	0.92752	0.91819	1187	1151	0.9605	0.0374	0.92797	0.91989	1183	1146

Table 2. Continued

		ρ_{123}		u_{123}				ρ_{123}		u_{123}	
x_1	x_2	g·cm^{-3}		m·s^{-1}		x_1	x_2	g·cm^{-3}		m·s^{-1}	
	T/K	298.15	308.15	298.15	308.15		T/K	298.15	308.15	298.15	308.15
MMA (1) + 1-Pentanol (2) + *p*-Xylene (3)						MMA (1) + 1-Pentanol (2) + Ethylbenzene (3)					
0.3102	0.4316	0.85650	0.84950	1243	1206	0.3048	0.419	0.85988	0.85190	1249	1202
0.7948	0.1162	0.90887	0.90167	1200	1162	0.7984	0.1142	0.91119	0.90204	1200	1160
0.0441	0.7154	0.83042	0.81989	1267	1229	0.0441	0.7646	0.83021	0.81940	1266	1229
0.0885	0.7890	0.82977	0.81881	1259	1222	0.0912	0.7264	0.83378	0.82375	1260	1222
0.1886	0.6845	0.83868	0.82972	1246	1211	0.1201	0.6402	0.84235	0.82995	1260	1220
0.3154	0.5481	0.85112	0.84433	1232	1201	0.3129	0.5492	0.85258	0.84528	1235	1203
0.4178	0.4952	0.85982	0.85378	1220	1189	0.4195	0.4675	0.86233	0.85610	1224	1194
0.5201	0.3562	0.87309	0.86788	1216	1183	0.5084	0.3931	0.87188	0.86581	1217	1185
0.6045	0.2989	0.88214	0.87699	1208	1175	0.6032	0.3175	0.88236	0.87630	1209	1175
0.7048	0.2215	0.89398	0.88868	1201	1165	0.7054	0.2316	0.89445	0.88832	1201	1165
0.7921	0.1645	0.90449	0.89862	1193	1157	0.7898	0.1665	0.90529	0.89832	1194	1157
0.7048	0.2356	0.89359	0.88790	1199	1164	0.7065	0.2348	0.89445	0.88821	1200	1164
0.2555	0.6012	0.84573	0.83790	1239	1206	0.2564	0.6048	0.84708	0.83902	1241	1208
0.1932	0.4489	0.84865	0.84097	1259	1218	0.1918	0.4475	0.85224	0.84388	1263	1213
0.1321	0.3045	0.85088	0.84332	1276	1231	0.1345	0.3154	0.85691	0.84698	1280	1226
0.0690	0.1578	0.85366	0.84548	1292	1247	0.0659	0.1601	0.86228	0.85011	1298	1249
0.3551	0.3578	0.86286	0.85602	1243	1203	0.3742	0.3754	0.86596	0.85840	1242	1196
0.2715	0.2365	0.86336	0.85564	1265	1216	0.2748	0.2765	0.86655	0.85729	1266	1205
0.1785	0.1546	0.86090	0.85282	1281	1234	0.1776	0.1778	0.86733	0.85599	1285	1228
0.5812	0.2315	0.88511	0.87871	1221	1182	0.5875	0.2521	0.88600	0.87893	1220	1177
0.4432	0.1954	0.87722	0.86998	1246	1198	0.4456	0.1924	0.88219	0.87253	1250	1188
0.3001	0.1364	0.87045	0.86212	1268	1219	0.3098	0.1321	0.87783	0.86656	1272	1212
MMA (1) + 1-Heptanol (2) + Cyclohexane (3)						MMA (1) + 1-Heptanol (2) + Benzene (3)					
0.0501	0.1900	0.78896	0.77961	1237	1192	0.0485	0.1906	0.86020	0.85045	1283	1234
0.1002	0.1835	0.79489	0.78527	1230	1184	0.1012	0.1812	0.86414	0.85427	1280	1225
0.2003	0.1565	0.80714	0.79698	1217	1170	0.2015	0.1554	0.87247	0.86234	1272	1208
0.2810	0.1421	0.81818	0.80771	1208	1161	0.2561	0.1398	0.87725	0.86697	1267	1199
0.3722	0.1240	0.83126	0.82059	1200	1154	0.3721	0.1198	0.88636	0.87584	1254	1183
0.4874	0.1101	0.84888	0.83823	1193	1148	0.4856	0.1025	0.89509	0.88435	1241	1175
0.5906	0.0865	0.86487	0.85437	1188	1146	0.5898	0.0812	0.90362	0.89264	1229	1171
0.6912	0.0687	0.88122	0.87092	1186	1145	0.6915	0.0612	0.91188	0.90067	1217	1169
0.7640	0.0451	0.89327	0.88299	1184	1144	0.7598	0.0478	0.91745	0.90608	1209	1168
0.0546	0.3784	0.79809	0.78932	1242	1197	0.0546	0.3765	0.84824	0.83935	1280	1230
0.1155	0.3556	0.80461	0.79555	1235	1189	0.1099	0.3521	0.85280	0.84375	1277	1221
0.2060	0.3145	0.81488	0.80535	1225	1178	0.2075	0.3126	0.86104	0.85167	1270	1206
0.3052	0.2656	0.82693	0.81697	1214	1168	0.3065	0.2748	0.86959	0.85991	1261	1193
0.4035	0.2342	0.84042	0.83026	1206	1162	0.4035	0.2378	0.87818	0.86824	1250	1184
0.5002	0.1923	0.85382	0.84359	1199	1156	0.5012	0.1989	0.88714	0.87695	1238	1177
0.5950	0.1656	0.86806	0.85796	1195	1154	0.5946	0.1598	0.89605	0.88564	1227	1173
0.7042	0.1187	0.88468	0.87468	1189	1150	0.7045	0.1178	0.90641	0.89576	1213	1168
0.8195	0.0710	0.90359	0.89347	1186	1148	0.8146	0.0719	0.91763	0.90665	1201	1163
0.0522	0.5623	0.80578	0.79754	1255	1212	0.0525	0.5645	0.83818	0.83002	1284	1238
0.1131	0.5321	0.81193	0.80347	1249	1205	0.1126	0.5219	0.84334	0.83497	1279	1229
0.2021	0.4778	0.82127	0.81241	1239	1195	0.2015	0.4773	0.85049	0.84183	1271	1219
0.3921	0.3598	0.84359	0.83402	1219	1177	0.3921	0.3546	0.86880	0.85939	1250	1199
0.4872	0.3084	0.85599	0.84631	1211	1171	0.4875	0.3075	0.87750	0.86787	1238	1191
0.5871	0.2475	0.86939	0.85967	1203	1164	0.5874	0.2415	0.88823	0.87835	1225	1181
0.6871	0.1884	0.88367	0.87399	1196	1158	0.6868	0.1874	0.89838	0.88833	1213	1173
0.9352	0.0365	0.92466	0.91383	1184	1147	0.9365	0.0365	0.92878	0.91755	1187	1152
0.3045	0.4465	0.83428	0.82512	1232	1189	0.3048	0.4098	0.86025	0.85117	1260	1208
0.8105	0.1243	0.90293	0.89314	1189	1153	0.8106	0.1146	0.91237	0.90199	1199	1163
0.0449	0.7232	0.81178	0.80396	1273	1233	0.0436	0.7632	0.82910	0.82157	1296	1255
0.0910	0.7954	0.82044	0.81284	1284	1245	0.0918	0.7154	0.83322	0.82553	1289	1247
0.1998	0.6812	0.82954	0.82144	1265	1225	0.1987	0.6393	0.84187	0.83379	1276	1235
0.3136	0.5584	0.84012	0.83144	1245	1205	0.3129	0.5502	0.85236	0.84378	1260	1222
0.4151	0.4965	0.85227	0.84334	1235	1196	0.4141	0.4702	0.86234	0.85339	1245	1210
0.5089	0.3899	0.86222	0.85297	1220	1182	0.5056	0.3898	0.87238	0.86315	1233	1198
0.6019	0.3165	0.87392	0.86460	1210	1173	0.6015	0.3201	0.88241	0.87303	1220	1186
0.7067	0.2308	0.88797	0.87858	1200	1163	0.7056	0.2301	0.89523	0.88562	1208	1173
0.7939	0.1787	0.90113	0.89165	1194	1159	0.7926	0.1645	0.90595	0.89612	1198	1164
0.7085	0.2456	0.88864	0.87933	1201	1165	0.7054	0.2298	0.89525	0.88564	1208	1173
0.2585	0.6041	0.83429	0.82583	1253	1212	0.2578	0.5998	0.84686	0.83853	1268	1229
0.1924	0.4432	0.81851	0.80954	1236	1191	0.1915	0.4456	0.85176	0.84299	1272	1217
0.1345	0.3098	0.80505	0.79574	1231	1184	0.1345	0.3145	0.85663	0.84737	1275	1217
0.0699	0.1602	0.78998	0.78044	1235	1188	0.0694	0.1589	0.86392	0.85398	1283	1232
0.3766	0.3784	0.84202	0.83254	1222	1180	0.3756	0.3754	0.86645	0.85715	1252	1202
0.2754	0.2954	0.82387	0.81409	1218	1173	0.2748	0.2741	0.86775	0.85811	1264	1196
0.1782	0.1754	0.80491	0.79491	1221	1173	0.1777	0.1769	0.86923	0.85923	1274	1211
0.5888	0.2588	0.87004	0.86039	1204	1165	0.5847	0.2521	0.88718	0.87735	1225	1183
0.4482	0.1945	0.84579	0.83549	1201	1157	0.4456	0.1910	0.88456	0.87435	1245	1178
0.3098	0.1365	0.82225	0.81170	1206	1159	0.3045	0.1329	0.88091	0.87054	1262	1191

Table 2. Continued

		ρ_{123}		u_{123}				ρ_{123}		u_{123}	
x_1	x_2	g·cm^{-3}		m·s^{-1}		x_1	x_2	g·cm^{-3}		m·s^{-1}	
	T/K	298.15	308.15	298.15	308.15		T/K	298.15	308.15	298.15	308.15
MMA (1) + 1-Heptanol (2) + Toluene (3)						MMA (1) + 1-Heptanol (2) + *p*-Xylene (3)					
0.0512	0.1911	0.85460	0.82423	1289	1241	0.0398	0.1904	0.85029	0.84199	1361	1254
0.0998	0.1840	0.85840	0.82901	1286	1236	0.0950	0.1821	0.84880	0.84057	1367	1258
0.1987	0.1580	0.86703	0.83915	1277	1224	0.2005	0.1554	0.86240	0.85344	1337	1235
0.2789	0.1432	0.87376	0.84735	1269	1215	0.2698	0.1420	0.86792	0.85869	1325	1227
0.3645	0.1252	0.88114	0.85621	1260	1205	0.3698	0.1232	0.87622	0.86665	1307	1216
0.4875	0.1089	0.89093	0.86861	1245	1193	0.4851	0.1089	0.88581	0.87592	1284	1204
0.5815	0.0854	0.89925	0.87859	1234	1185	0.5886	0.0845	0.89552	0.88538	1264	1194
0.6901	0.0684	0.90803	0.88980	1220	1177	0.6874	0.0645	0.90497	0.89459	1245	1184
0.7632	0.0432	0.91541	0.89827	1212	1172	0.7548	0.0418	0.91239	0.90177	1233	1178
0.0548	0.3778	0.84546	0.82181	1288	1240	0.0548	0.3652	0.84400	0.83613	1338	1252
0.1150	0.3554	0.85027	0.82735	1283	1233	0.1089	0.3562	0.84747	0.83942	1331	1244
0.2054	0.3141	0.85824	0.83612	1274	1221	0.1956	0.3120	0.85469	0.84622	1320	1232
0.3045	0.2674	0.86747	0.84617	1264	1209	0.2989	0.2550	0.86417	0.85522	1306	1220
0.4032	0.2332	0.87615	0.85615	1252	1199	0.3945	0.2455	0.87126	0.86202	1288	1209
0.4999	0.1924	0.88529	0.86640	1241	1190	0.5348	0.1921	0.88437	0.87473	1263	1194
0.5984	0.1635	0.89378	0.87659	1228	1182	0.5798	0.1653	0.88939	0.87962	1256	1190
0.7051	0.1187	0.90433	0.88860	1215	1173	0.7065	0.1175	0.90238	0.89232	1232	1178
0.8145	0.0706	0.91604	0.90170	1203	1164	0.8065	0.0742	0.91379	0.90339	1215	1168
0.0512	0.5616	0.83724	0.81947	1291	1246	0.0451	0.5432	0.83674	0.82925	1325	1256
0.1098	0.5321	0.84167	0.82439	1286	1239	0.3326	0.5317	0.85330	0.84488	1271	1223
0.2001	0.4787	0.84933	0.83250	1276	1229	0.2106	0.4578	0.84924	0.84108	1304	1233
0.3912	0.3678	0.86679	0.85104	1253	1207	0.3845	0.3632	0.86452	0.85562	1277	1212
0.4871	0.3089	0.87626	0.86108	1241	1196	0.4865	0.3026	0.89341	0.88472	1188	1148
0.5789	0.2478	0.88601	0.87133	1229	1186	0.5789	0.2554	0.88361	0.87419	1244	1189
0.6778	0.1884	0.89656	0.88270	1216	1176	0.6784	0.1789	0.89573	0.88602	1230	1179
0.9401	0.0375	0.92836	0.91653	1187	1151	0.9265	0.0335	0.92752	0.91647	1193	1154
0.3012	0.4465	0.85694	0.84149	1264	1219	0.3045	0.4236	0.85650	0.84796	1288	1222
0.8015	0.1235	0.90990	0.89744	1201	1164	0.7895	0.1156	0.90887	0.89882	1212	1167
0.0439	0.7244	0.83044	0.81735	1298	1256	0.0441	0.7164	0.83042	0.82317	1317	1262
0.0890	0.7963	0.82972	0.81965	1299	1258	0.0887	0.7894	0.82977	0.82250	1308	1260
0.1974	0.6912	0.83928	0.82872	1282	1241	0.1900	0.6865	0.83868	0.83097	1292	1244
0.3035	0.5589	0.85118	0.83922	1265	1225	0.3098	0.5501	0.85112	0.84281	1275	1226
0.4098	0.4955	0.86038	0.84911	1250	1212	0.4145	0.4978	0.85982	0.85114	1255	1213
0.5075	0.3898	0.87210	0.86015	1236	1198	0.5098	0.3565	0.87309	0.86400	1247	1199
0.5984	0.3165	0.88208	0.87036	1223	1186	0.6015	0.3042	0.88214	0.87291	1231	1187
0.6995	0.2335	0.89405	0.88258	1210	1174	0.7055	0.2301	0.89398	0.88458	1215	1174
0.7865	0.1789	0.90374	0.89308	1199	1164	0.7854	0.1645	0.90449	0.89483	1205	1166
0.6954	0.2445	0.89292	0.88172	1210	1174	0.7045	0.2345	0.89359	0.88421	1214	1174
0.2514	0.6065	0.84601	0.83424	1273	1232	0.2554	0.6018	0.84573	0.83768	1283	1234
0.1859	0.4421	0.85030	0.83197	1277	1228	0.1942	0.4489	0.84865	0.84052	1308	1235
0.1285	0.3098	0.85346	0.82932	1282	1231	0.1326	0.3092	0.85088	0.84262	1331	1241
0.0548	0.1632	0.85640	0.82507	1290	1241	0.0682	0.1545	0.85366	0.84515	1362	1251
0.3698	0.3765	0.86499	0.84897	1256	1209	0.3559	0.3589	0.86286	0.85402	1284	1215
0.2698	0.2956	0.86350	0.84231	1268	1214	0.2712	0.2335	0.86336	0.85442	1314	1223
0.1698	0.1752	0.86388	0.83593	1280	1227	0.1765	0.1526	0.86090	0.85201	1342	1238
0.5850	0.2578	0.88562	0.87147	1227	1186	0.5798	0.2323	0.88511	0.87560	1247	1189
0.4472	0.1865	0.88231	0.86179	1248	1194	0.4395	0.1956	0.87722	0.86773	1284	1204
0.3102	0.1298	0.87692	0.85078	1266	1211	0.3002	0.1352	0.87045	0.86110	1320	1224
MMA (1) + 1-Heptanol (2) + Ethylbenzene (3)											
0.0491	0.1889	0.85946	0.84596	1309	1253	0.6872	0.1879	0.89682	0.88620	1219	1174
0.1004	0.1805	0.86270	0.84929	1306	1240	0.9353	0.0365	0.92797	0.91671	1188	1152
0.2005	0.1578	0.86973	0.85642	1296	1222	0.3042	0.4125	0.85988	0.84939	1276	1212
0.2812	0.1435	0.87541	0.86222	1286	1212	0.8101	0.1123	0.91119	0.90051	1203	1162
0.3723	0.1246	0.88224	0.86919	1273	1205	0.0445	0.7656	0.83021	0.82183	1310	1264
0.4879	0.1016	0.89115	0.87836	1256	1196	0.0919	0.7254	0.83378	0.82531	1302	1256
0.5907	0.0815	0.89936	0.88685	1242	1186	0.1994	0.6412	0.84235	0.83356	1282	1254
0.6917	0.0613	0.90782	0.89561	1228	1177	0.3137	0.5489	0.85258	0.84337	1259	1257
0.7642	0.0465	0.91420	0.90220	1217	1171	0.4153	0.4678	0.86233	0.85282	1241	1250
0.0546	0.3765	0.84925	0.83762	1308	1249	0.5087	0.3926	0.87188	0.86220	1228	1234
0.1152	0.3526	0.85349	0.84183	1304	1230	0.6012	0.3121	0.88236	0.87252	1219	1209
0.2081	0.3165	0.86039	0.84865	1297	1207	0.7062	0.2307	0.89445	0.88457	1208	1184
0.3055	0.2764	0.86822	0.85638	1285	1190	0.7931	0.1645	0.90529	0.89531	1199	1167
0.4039	0.2354	0.87655	0.86467	1272	1179	0.7081	0.2321	0.89445	0.88461	1207	1184
0.5005	0.1992	0.88477	0.87300	1257	1172	0.2588	0.6023	0.84708	0.83814	1269	1258
0.5980	0.1902	0.89129	0.88001	1239	1170	0.1920	0.4456	0.85224	0.84173	1294	1220
0.7049	0.1174	0.90384	0.89243	1224	1166	0.1349	0.3098	0.85691	0.84487	1303	1225
0.8199	0.0724	0.91568	0.90441	1206	1162	0.0692	0.1598	0.86228	0.84859	1308	1250
0.0528	0.5643	0.83975	0.82987	1308	1253	0.3761	0.3751	0.86596	0.85539	1265	1206
0.1134	0.5302	0.84406	0.83410	1302	1239	0.2750	0.2756	0.86655	0.85460	1289	1195
0.2021	0.4765	0.85106	0.84087	1291	1224	0.1783	0.1756	0.86733	0.85411	1299	1223
0.3924	0.3645	0.86750	0.85689	1262	1205	0.5881	0.2525	0.88600	0.87537	1232	1186
0.4873	0.3038	0.87668	0.86594	1249	1193	0.4489	0.1921	0.88219	0.87009	1265	1177
0.5873	0.2487	0.88624	0.87556	1233	1184	0.3091	0.1325	0.87783	0.86464	1282	1211

Table 3. Isobaric Thermal Expansion Coefficient, α_i, and Molar Heat Capacities, C_{pi}, for Pure Components at T = (298.15 and 308.15) K

	α_i[a]		C_{pi}	
	kK^{-1}		$J \cdot K^{-1} \cdot mol^{-1}$	
T/K	298.15	308.15	298.15	308.15
MMA	1.236	1.263	191.1[38]	198.2[38]
1-butanol	0.836	0.856	177.2[39]	184.6[39]
1-pentanol	0.838	0.857	208.1[39]	216.5[39]
1-heptanol	0.876	0.894	272.1[39]	283.1[39]
benzene	1.232	1.254	135.74[42]	137.82[42]
toluene	1.087	1.110	157.06[42]	159.66[42]
p-xylene	1.022	1.036	181.9[41]	185.2[41]
ethylbenzene	1.026	1.040	185.6[12]	190.5[12]
cyclohexane	1.229	1.258	155.9[40]	159.9[40]

[a] α_i values were calculated from measured densities at different temperatures ((288.15 to 323.15) K for MMA, (288.15 to 338.15) K for organic solvents, and (288.15 to 353.15) K for 1-alcohols) using the relation, $\alpha_i = [(\rho_1/\rho_2) - 1]/\Delta T$.

and the ϕ_i is the ideal state volume fraction and is defined by the relation,

$$\phi_i = x_i V_i / (\sum_{i=1}^{3} x_i V_i) \quad (9)$$

α_i are the isobaric thermal expansion coefficient, and C_{pi} are the molar heat capacities. The values of α_i and $C_{p,i}$ for pure components at T/K = (298.15 and 308.15) are listed in Table 3. The κ_{s123}^{E} values are also calculated from the binary contributions, that is, κ_{s12}^{E}, κ_{s23}^{E}, and κ_{s13}^{E} using eqs 3 to 5. The excess isentropic compressibilities of respective binary 1 + 2, 1 + 3, and 2 + 3 binary pairs were obtained from the literature data[8−11] in a similar way as in the case of V_{ij}^{E}. The respective coefficients are listed in Table S3.

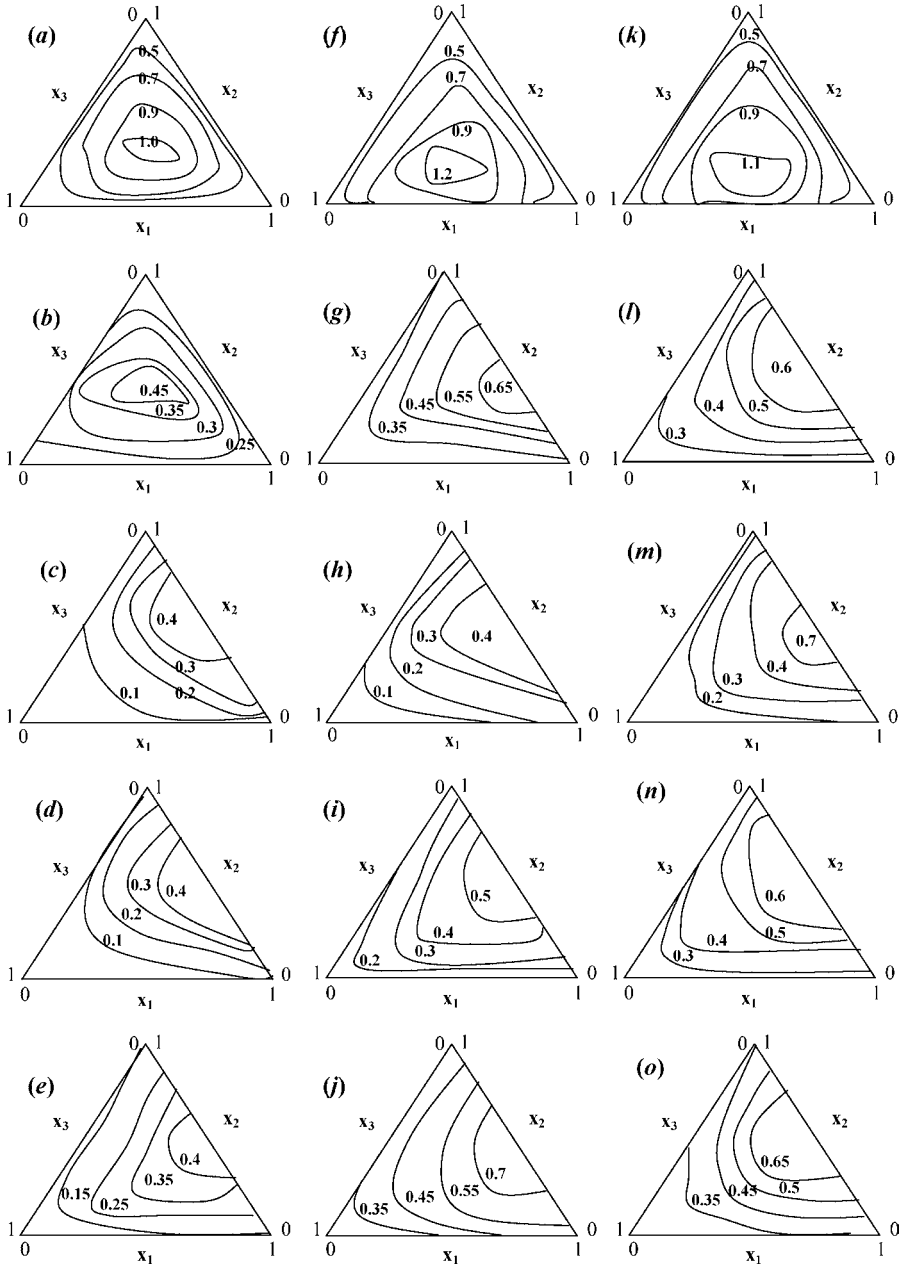

Figure 1. Curves of constant excess molar volumes, V_{m123}^{E}, for MMA (x_1) + 1-alkanols (x_2) (a to e, 1-butanol; f to j, 1-pentanol; k to o, 1-heptanol) and + organic solvents (x_3) (a, f, k, cyclohexane; b, g, l, benzene; c, h, m, toluene; d, i, n, *p*-xylene; e, j, o, ethylbenzene) at 298.15 K, calculated with eq 3.

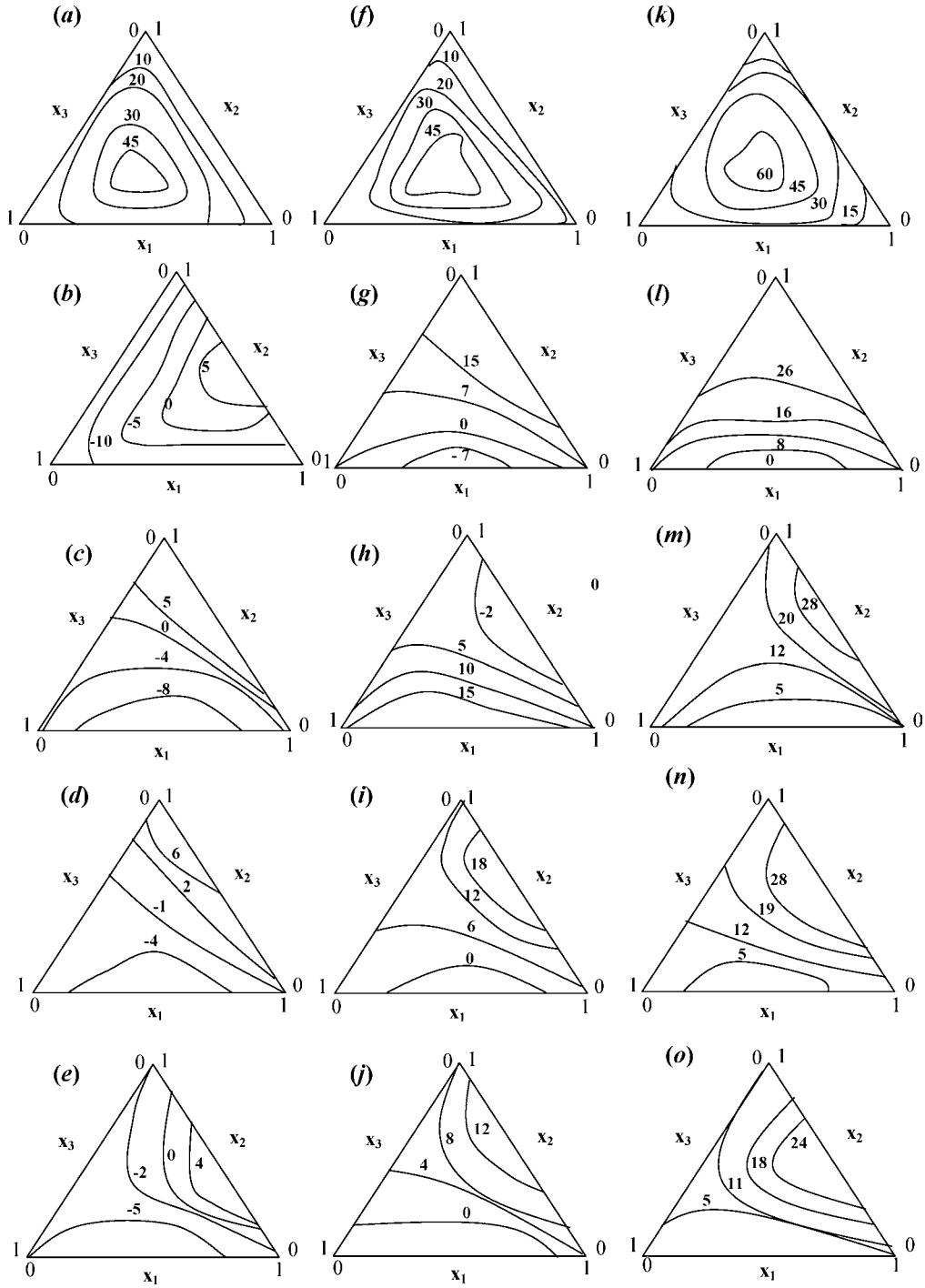

Figure 2. Curves of constant excess isentropic compressibilities, κ_s^E for MMA (x_1) + 1-alkanols (x_2) (a to e, 1-butanol; f to j, 1-pentanol; k to o, 1-heptanol) and + organic solvents (x_3) (a, f, k, cyclohexane; b, g, l, benzene; c, h, m, toluene; d, i, n, *p*-xylene; e, j, o, ethylbenzene) at 298.15 K, calculated with eq 3.

Results and Discussion

Excess Molar Volumes ($V_m^{E}{}_{123}$). Table S1 of the Supporting Information also lists the calculated excess molar volumes for the 15 ternary mixtures. The deviations in excess molar volumes of ternary mixtures, $\Delta V^E_{123} = V_m{}^E{}_{123(exp.)} - V_m{}^E{}_{123(cal)}$ (as calculated from RK, TS, and K equations) were found to vary from 0 to 0.008, 0.006 to 0.15, and 0.001 to 0.093 $cm^3 \cdot mol^{-1}$, respectively. Therefore, it is concluded that the experimental ternary excess molar volumes are the best reproduced by the RK equation. Figure 1 shows the curves of constant ternary excess molar volumes (calculated using the RK equation) for the 15 ternary mixtures of MMA (1) + 1-alkanols (2) + organic solvents (3). The vertical columns in the figures, namely, a to e, f to j, and k to o represent mixtures containing 1-butanol, 1-pentanol, and 1-heptanol and while each of the horizontal columns give ternary diagrams of mixtures with MMA + 1-alkanols (1-butanol, 1-pentanol and 1-heptanol) + cyclohexane, + benzene, + toluene, + *p*-xylene, and + ethylbenzene, respectively. It can be observed that the mixtures of MMA + 1-alkanol (1-butanol, 1-pentanol, and 1-heptanol) + organic solvents are mostly characterized by positive excess molar volumes. However, there exists a large difference in positive values for mixtures containing cyclohexane as compared to other organic solvents. No such large differences were visible in

ternary mixtures containing benzene and its substituted homologues. Other notable observations are: (i) the maximum value of V_{m123}^{E} for systems with cyclohexane follow the order 1-butanol < 1-heptanol < 1-pentanol, (ii) a discontinuity in $V_{m123}^{E} = 0.7$ for MMA + 1-heptanol + cyclohexane is clearly visible, (iii) the shape of curves of constant excess molar volumes for MMA + 1-butanol + benzene look different than MMA + 1-pentanol or +1-heptanol + benzene mixtures, (iv) the effect of chain length of 1-alkanols on constant excess molar volumes is clearly evident in MMA + 1-alkanols + *p*-xylene systems, where in the increase of chain length from butyl to heptyl systematically resulted in more positive values, and (v) the ternary mixtures of MMA + 1-heptanol + toluene and MMA + 1-heptanol + *p*-xylene are characterized by more positive values as compared to other two alkanols. These observed trends clearly indicate that the ternary mixtures of type presented in the work are characterized by complex interactions in the bulk state. Practically, the ternary mixtures consisting of MMA + 1-alkanols + cyclohexane are characterized by more and large positive values than mixtures with other organic solvents. These results indicate that cyclohexane is an effective structure breaker, while the aromatic organic solvents would probably interact specifically with either MMA or 1-alkanols even though structure-breaking effects between unlike molecules are quite possible.

Speeds of Sound, u_{123}, and Excess Isentropic Compressibilities κ_{s123}^{E}. The experimental u_{123} are listed in Table 2. The experimental κ_{s123}^{E} values along with the calculated values, κ_{s123}^{E} from eqs 3 to 5 are listed in Table S2 (Supporting Information). The $\Delta\kappa_{s123}^{E}$, that is, the difference between $\kappa_{s123(exp)}^{E}$ and $\kappa_{s123(cal.)}^{E}$, was at a minimum with the RK equation calculated values as compared to other two equations. Figure 2 displays the curves for constant excess isentropic compressibilities (calculated values using the RK equation) for the ternary mixtures at T/K = 298.15. A perusal of parts a to e reveals that κ_{s123}^{E} values are positive across the composition for MMA + 1-alkanols + cyclohexane, and the same were decreased or even became negative at certain compositions upon the replacement of cyclohexane with aromatic organic solvents. Other certain striking features in the curves are: (i) among the three alkanols, 1-heptanol containing ternary mixtures are characterized by more and positive values irrespective of the nature of the organic solvents, (ii) for 15 investigated ternary mixtures, the constant values of κ_{s123}^{E} increase with the increase in alkyl chain length from 1-butanol to 1-heptanol. These results support our earlier conclusion that structure-breaking interactions are predominant between unlike components, namely, MMA, 1-alkanols, and cyclohexane in the mixed state. The introduction of aromatic organic solvents as a third component into the ternary system shifted the overall balance of the net bulk interactions in favor of weak structure-making interactions even though the constituent binary mixtures of MMA + 1-alkanols or MMA + aromatic organic solvents were characterized by the possible predominance of structure-breaking effects.

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Supporting Information Available:

Experimental and calculated excess molar volumes and excess isentropic compressibilities and values of binary constants. This material is available free of charge via the Internet at http://pubs.acs.org.

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