

Molecular Interactions Studies Between Some Esters and Hydrocarbon Solvents by Ultrasound Velocities Measurements

¹S.S. Yadava, ²Y. Singh and ¹Neetu Kushwaha

¹Department of Chemistry, D.D.U.Gorakhpur, University, Gorakhpur-273009 (U.P.), India

²Department of Chemistry, U.P. Autonomous College, Varanasi, India

Abstract: Ultrasound velocities (u_{12}) and densities (ρ_{12}) of the twelve binary mixtures of some esters viz. methylethanoate, ethylethanoate and n-propylethanoate and symmetrical hydrocarbon solvents viz. cyclohexane, benzene, 1, 4-dimethylbenzene and 1, 3, 5-trimethylbenzene have been measured at 308.15 K over complete concentration range. Adiabatic compressibilities (β_{ad}), free lengths (L_f), acoustical impedances (Z), available volumes (V_a) and Wada's constants (W) have been determined from the measured values of ultrasound velocities and densities. Ultrasound velocities increase with mole fractions of hydrocarbon solvents for all the binary mixtures. Deviations from additive values in velocities (Δu), in free lengths (ΔL_f), in acoustical impedances (ΔZ) and in available volumes (ΔV_a) and excess adiabatic compressibilities (β_{ad}^E) have also been evaluated. Experimental results and the variations of several evaluated parameters with mole fractions are discussed on the basis of molecular interactions.

Key words: Ultrasound velocity • Liquid mixtures and molecular interaction

INTRODUCTION

Increasing interests have been shown by several workers for the study of molecular interactions in binary [1-3] and ternary [4, 5] liquid mixtures in recent past employing ultrasonic velocities measurements. Binary mixtures of several alkylethanoate with 1, 1, 1-trichloroethane [6] at 303.15 K and with several glycols [7] at 298.15 and 313.15 K have been studied by sound velocity measurements and isentropic compressibilities have been evaluated. In our laboratory binary mixtures of different type of components with aromatic hydrocarbons employing dipole moment [8, 9] viscosity [10-12] excess molar volume [13] and ultrasound velocity [14, 15] have been studied. However, studies on binary mixtures of esters with hydrocarbons [16-19] are limited.

In view of the importance of ultrasound velocity for the study of molecular interactions in binary mixtures and the limited study on the mixtures of esters with hydrocarbons, present work have been undertaken. In the present work, twelve binary mixtures containing methylethanoate, ethylethanoate and n-propylethanoate as esters and nonpolar hydrocarbon solvents viz. cyclohexane, benzene, 1, 4-dimethylbenzene, 1, 3, 5-trimethylbenzene have been studied at 308.15K employing

ultrasound velocity measurements. Esters with increasing alkyl groups and aromatic hydrocarbons with successive methylation have been deliberately chosen to show the effect of steric factors on the molecular interactions. Methylated benzenes have gradually changing polarisabilities. Thus the present work is expected to throw light on the effects of polarisabilities over such interactions. Cyclohexane has been selected as an inert solvent because it has ring structure as benzene without any π -electron and serve as reference point for comparison of the molecular interactions.

Experimental: Chemicals used in the present investigation were cyclohexane, Merck-HPLC grade (99.7%), benzene, Merck-GR (99.7%), 1, 4-dimethylbenzene, Merck-synthesis (99%), 3, 5-trimethylbenzene, Merck-synthesis (> 98%), methylethanoate, Merck-synthesis (> 99%), ethylethanoate, Merck-GR (99.5%) and n-propylethanoate, Merck-synthesis (> 98%). All the chemicals except cyclohexane, which was of HPLC grade, were fractionally distilled over one meter long column and only middle fractions were used. Purities of the chemicals were checked by densities measurements which are compared with the literature values as follows:

Densities (ρ) and ultrasound velocity (u) of pure components at 308.15 K.

Component	$\rho \times 10^{-3} \text{ (kg m}^{-3}\text{)}$		$u \text{ (ms}^{-1}\text{)}$	
	Expt.	Lit.	Expt.	Lit.
Methylethanoate	0.91561	0.91522 ^a	1106	1112.1 ^g
Ethylethanoate	0.88252	0.88250 ^b	1102	1097.8 ^g
n-propylethanoate	0.87105	0.8718 ^c	1124	1139 ^g
Cyclohexane	0.76477	0.76447 ^d	1202	1204 ^h
Benzene	0.86302	0.86295 ^e	1252	1252.6 ^f
1, 4-dimethylbenzene	0.84878	0.84787 ^e	1269	1276 ⁱ
1, 3, 5-trimethylbenzene	0.85447	0.8525 ^f	1309	1303.5 ^f

a= S.J.Tangeda, S.N.Nallani, J.Chem.Thermodyn. 38 (2006) 272-277.

b= Reference [7].

c= M.I.Aralaguppi, C.V.Jadar, T.M.Aminabhavi, J.Chem.Eng.Data 44 (1999) 441-445.

d= (estimated value) J.Timmermans, Physico-Chemical Constants of Pure Organic Compounds, Elsevier publishing company, Inc., New York, 1950.

e= J.George, N.V.Sastry, J.Chem.Eng.Data 48 (2003) 977-989.

f= B.R.Kumar, B.Satyanarayana, S.A.Banu, K.A.Jyothi, T.S.Jyostana, N. Satyanarayana, Indian J. of Pure and Applied Phys.47(2009)511-516.

g= V.Jaana, S.N.Nallani, Rasayan J.Chem. 3(2008) 612-617.

h= A.Ali, A.K.Nain, N.Kumar, M.I.Brahim, Chin. J. Chem. 21(2003) 253.

i= A.M.Zeliznyi, F.V.Dyakiv, E.F.Shevchenko, Zh.Obshch.Khim 46 (1976)1913-1919.

Binary mixture of each esters with different solvents were made gravimetrically by weighing on a single pan analytical balance (Model K-15 Deluxe, K. Roy

Instruments Pvt. Ltd., Varanasi) with uncertainty ± 0.01 kg after injecting the components with syringe in a rubber sealed glass vessel. Ultrasound velocities were measured for different samples covering the complete mole fraction range using M-82 multifrequency ultrasonic interferometer (M/S Mittal Enterprises, New Delhi) at a constant frequency of 1 MHz. The ultrasound velocities were accurate to $\pm 0.03\%$. The densities accurate to $\pm 0.00001 \times 10^3 \text{ kg m}^{-3}$ of pure liquids and their binary mixtures were determined taking into account of buoyancy correction [20] using a single stem pycnometer. The temperature of the samples during measurements was kept constant at 308.15 ± 0.03 K employing a water thermostat.

RESULTS AND DISCUSSION

Experimental values of ultrasound velocities (u_{12}) and densities (ρ_{12}) for all the binary mixtures studied at 308.15 K with composition of binary mixtures are given in Table 1. Ultrasound velocities in a medium depend [15] inversely on the density and compressibility of the medium as given in equation 1.

Table 1: Mole fraction (x_1), Volume fraction (\bar{O}_1) of hydrocarbons, ultrasound velocities (u_{12}), densities (ρ_{12}), adiabatic compressibilities (β_{ad}), free lengths (L_f), available volumes (V_a), acoustical impedances (Z) and Wada's constant (W) at 308.15K for binary mixtures of different esters + hydrocarbon solvents

x_1	\bar{O}_1	$u_{12} \text{ ms}^{-1}$	$\rho_{12} \times 10^{-3} \text{ kgm}^{-3}$	$\beta_{ad} \text{ (TPa)}^{-1}$	$L_f \times 10^{10} \text{ m}$	$V_a \times 10^6 \text{ m}^3 \text{ mol}^{-1}$	$Z \times 10^{-3} \text{ kgm}^{-2} \text{ s}^{-1}$	$W \times 10^6 \{ \text{m}^3 \text{ mol} \text{ TP a} \}^{-1/2}$
Cyclohexane (1) + Methylethanoate (2)								
0.00000	0.00000	1106	0.9156	893	0.6015	24.9803	1012.6647	30.6507
0.10176	0.13350	1106	0.88989	919	0.6102	26.0582	984.2183	31.8424
0.19971	0.25341	1107	0.86793	940	0.6171	27.0139	960.7985	32.9707
0.30251	0.37104	1109	0.84792	959	0.6233	27.9143	940.3433	34.1107
0.40017	0.47574	1111	0.83067	975	0.6285	28.7401	922.8744	35.1802
0.49683	0.57319	1118	0.81571	981	0.6304	29.2080	911.9638	36.2405
0.60098	0.67198	1132	0.80211	973	0.6279	29.2233	907.9885	37.3879
0.70000	0.76040	1144	0.79075	966	0.6256	29.2428	904.6180	38.4370
0.79112	0.83745	1165	0.78142	943	0.6181	28.5488	910.3543	39.4719
0.89061	0.91717	1183	0.77293	924	0.6118	28.0062	914.3762	40.5108
1.00000	1.00000	1202	0.76477	905	0.6055	27.3739	919.2547	41.6098
Benzene (1) + Methylethanoate (2)								
0.00000	0.00000	1106	0.91561	893	0.6015	24.9803	1012.6647	30.6507
0.10333	0.11418	1125	0.90885	869	0.5934	24.3342	1022.4563	31.1733
0.20217	0.22087	1136	0.90220	859	0.5899	24.0739	1024.8992	31.6233
0.30174	0.32586	1150	0.89597	844	0.5848	23.6358	1030.3655	32.0944
0.40155	0.42878	1173	0.89124	815	0.5746	22.6673	1045.4245	32.5995
0.49845	0.52646	1183	0.88577	807	0.5718	22.3880	1047.8659	33.0165
0.59729	0.62395	1195	0.88135	795	0.5675	21.9672	1053.2133	33.4273
0.69852	0.72160	1217	0.87656	770	0.5585	20.9989	1066.7735	33.9438
0.79661	0.81397	1224	0.87279	765	0.5567	20.8105	1068.2950	34.2976
0.89755	0.90742	1234	0.87093	754	0.5527	20.4072	1074.7276	34.6233
1.00000	1.00000	1252	0.86302	739	0.5472	19.6854	1080.5010	35.2273

Table I: Continued

1, 4-dimethylbenzene (1) + Methyleneoate (2)								
0.00000	0.00000	1106	0.91561	893	0.6015	24.9803	1012.6647	30.6507
0.10315	0.15095	1125	0.90577	872	0.5945	25.3650	1018.9913	32.4762
0.20182	0.28102	1145	0.89724	850	0.5869	25.5312	1027.3398	34.2518
0.30094	0.39958	1164	0.88904	830	0.5799	25.6654	1034.8426	36.0543
0.39561	0.50296	1181	0.88195	813	0.5739	25.7647	1041.5830	37.7754
0.49725	0.60459	1197	0.87508	798	0.5684	25.9139	1047.4708	39.6105
0.60002	0.69871	1210	0.86957	785	0.5641	26.1610	1052.1797	41.4115
0.69291	0.77719	1225	0.86425	771	0.5589	26.1178	1058.7063	43.1106
0.79307	0.85560	1241	0.85988	755	0.5531	25.9690	1067.1111	44.9091
0.89376	0.92860	1255	0.85537	742	0.5484	25.9021	1073.4894	46.7257
1.00000	1.00000	1269	0.84878	732	0.5446	25.8746	1077.1018	48.7472
1, 3, 5-trimethylbenzene (1) + Methyleneoate (2)								
0.00000	0.00000	1106	0.91561	893	0.6015	24.9803	1012.6647	30.6507
0.10375	0.16756	1131	0.90463	864	0.5916	25.5544	1023.1365	33.1827
0.20201	0.30561	1150	0.89499	845	0.5851	26.2074	1029.2385	35.5802
0.30079	0.42790	1172	0.88732	820	0.5764	26.5150	1039.9390	38.0109
0.39898	0.53579	1192	0.88039	799	0.5690	26.7866	1049.4249	40.4322
0.49755	0.63258	1209	0.87450	782	0.5629	27.1138	1057.2705	42.8369
0.59872	0.72177	1231	0.86997	759	0.5545	26.9583	1070.9331	45.3235
0.69588	0.79913	1246	0.86539	744	0.5490	27.1450	1078.2759	47.7069
0.79113	0.86817	1265	0.86241	725	0.5418	26.8433	1090.9487	50.0371
0.88838	0.93260	1282	0.85960	708	0.5355	26.6015	1102.0072	52.4144
1.00000	1.00000	1309	0.85447	683	0.5260	25.5847	1118.5012	55.3720
Cyclohexane (1) + Ethyleneoate (2)								
0.00000	0.00000	1102	0.88252	933	0.6148	31.0749	972.5370	37.5865
0.10463	0.11412	1103	0.86615	949	0.6201	31.4504	955.3635	38.0248
0.20008	0.21610	1107	0.85151	958	0.6230	31.5973	942.6216	38.4603
0.30369	0.32466	1109	0.83660	972	0.6275	31.8798	927.7894	38.8816
0.40048	0.42405	1113	0.82399	980	0.6301	31.9628	917.1009	39.2570
0.49957	0.52388	1120	0.81191	982	0.6308	31.8274	909.3392	39.6493
0.59675	0.61993	1128	0.80098	981	0.6304	31.5827	903.5054	40.0171
0.69868	0.71876	1147	0.78942	963	0.6246	30.6108	905.4647	40.5195
0.79949	0.81464	1165	0.78061	944	0.6184	29.5875	909.42107	40.9019
0.89495	0.90375	1181	0.77284	928	0.6132	28.6580	912.7240	41.2303
1.00000	1.00000	1202	0.76477	905	0.6055	27.3739	919.2535	41.6098
Benzene (1) + Ethyleneoate (2)								
0.00000	0.00000	1102	0.88252	933	0.6148	31.0749	972.5370	37.5865
0.10020	0.09170	1108	0.88116	924	0.6120	30.3983	976.3253	37.2679
0.19794	0.18282	1122	0.87827	904	0.6053	29.2981	985.4189	37.0868
0.29588	0.27585	1131	0.87606	892	0.6013	28.4911	990.8239	36.8278
0.39660	0.37341	1153	0.87417	860	0.5904	26.8915	1007.9180	36.6617
0.50010	0.47554	1165	0.87183	845	0.5852	25.9170	1015.6820	36.3994
0.59110	0.56719	1186	0.87029	817	0.5753	24.4390	1032.1639	36.2385
0.70227	0.68136	1198	0.86811	803	0.5702	23.4683	1039.9958	35.9268
0.79038	0.77365	1216	0.86624	781	0.5624	22.2218	1053.3478	35.7547
0.89202	0.88219	1232	0.86459	762	0.5556	21.0660	1065.1749	35.4937
1.00000	1.00000	1252	0.86302	739	0.5473	19.6854	1080.5010	35.2273

Table I: Continued

1, 4-dimethylbenzene (1) + Ethylethanoate (2)								
0.00000	0.00000	1102	0.88252	933	0.6148	31.0749	972.5370	37.5865
0.10408	0.12703	1125	0.87904	899	0.6035	30.3915	988.9200	38.7448
0.20239	0.24119	1139	0.87493	881	0.5974	30.2187	996.5453	39.8093
0.30413	0.35382	1154	0.87128	862	0.5910	29.9455	1005.4570	40.9033
0.40065	0.45577	1170	0.86821	841	0.5837	29.5126	1015.8060	41.9595
0.49394	0.55010	1188	0.86393	820	0.5764	28.9191	1026.3488	43.0674
0.60558	0.65794	1210	0.86080	793	0.5668	28.0450	1041.5680	44.3333
0.69676	0.74216	1228	0.85818	773	0.5596	27.2782	1053.8450	45.3728
0.79593	0.83011	1244	0.85477	756	0.5534	26.6751	1063.3340	46.5112
0.88762	0.90819	1257	0.85228	743	0.5487	26.1923	1071.3160	47.5181
1.00000	1.00000	1269	0.84878	732	0.5446	25.8746	1077.1020	48.7472
1, 3, 5-trimethylbenzene (1) + Ethylethanoate (2)								
0.00000	0.00000	1102	0.88252	933	0.61481	31.0749	972.5370	37.5865
0.10355	0.1399	1117	0.87881	912	0.6079	31.4076	981.6308	39.2963
0.19815	0.2583	1134	0.87434	889	0.6003	31.4681	991.5016	40.9574
0.30053	0.3771	1161	0.87069	852	0.5875	30.8046	1010.8711	42.8190
0.39593	0.4801	1188	0.86842	816	0.5749	29.8932	1031.6829	44.5492
0.49361	0.5787	1204	0.86525	797	0.5683	29.7340	1041.7610	46.2577
0.60022	0.6790	1223	0.86256	775	0.5604	29.3305	1054.9109	48.1216
0.69472	0.7623	1247	0.86064	747	0.5502	28.3021	1073.2181	49.8527
0.79823	0.8479	1265	0.85872	728	0.5430	27.7287	1086.2808	51.6569
0.88565	0.9160	1283	0.85681	709	0.5360	26.9459	1099.2872	53.2498
1.00000	1.00000	1309	0.85447	683	0.5260	25.5847	1118.5012	55.3720
Cyclohexane (1) + n-propylethanoate (2)								
0.00000	0.00000	1124	0.87105	909	0.6068	34.8817	979.0602	44.3056
0.10248	0.09680	1127	0.86001	915	0.6090	34.4738	969.2313	44.0238
0.19888	0.18896	1130	0.84916	922	0.6113	34.0935	959.5508	43.7684
0.29761	0.28453	1132	0.83625	933	0.6149	33.8520	946.6350	43.5702
0.39914	0.38403	1136	0.82460	940	0.6170	33.3952	936.7456	43.3065
0.49975	0.48390	1142	0.81585	940	0.6171	32.6825	931.7007	42.9386
0.59907	0.58374	1153	0.80517	934	0.6152	31.7014	928.3610	42.7111
0.70004	0.68657	1160	0.79233	938	0.6164	31.0809	919.1028	42.5168
0.79722	0.78677	1174	0.78512	924	0.6118	29.7761	921.7309	42.1610
0.89387	0.88769	1191	0.77614	908	0.6065	28.3466	924.4383	41.9096
1.00000	1.00000	1202	0.76477	905	0.6056	27.3739	919.2535	41.6098
Benzene (1) + n-propylethanoate (2)								
0.00000	0.00000	1124	0.87105	909	0.6068	34.8817	979.0602	44.3056
0.10878	0.08610	1160	0.87809	846	0.5856	31.1662	1018.5960	43.2669
0.20393	0.16510	1172	0.87682	830	0.5791	29.6643	1027.6096	42.4521
0.30294	0.25121	1182	0.87626	817	0.5753	28.2788	1035.7866	41.5310
0.40286	0.34243	1192	0.87552	804	0.5707	26.9279	1043.5960	40.6093
0.50159	0.43722	1205	0.87475	787	0.5648	25.4217	1054.1340	39.7207
0.60094	0.53287	1220	0.87396	769	0.5581	23.8305	1066.2800	38.8325
0.70145	0.64460	1235	0.87307	751	0.5516	22.2821	1078.2785	37.9295
0.80146	0.75706	1242	0.86530	749	0.5509	21.4309	1074.7026	37.2081
0.89719	0.87074	1248	0.86448	743	0.5485	20.5065	1078.8710	36.2516
1.00000	1.00000	1252	0.86302	739	0.5473	19.6854	1080.4760	35.2273

Table 1: Continued

1, 4-dimethylbenzene (1) + n-propylethanoate (2)								
0.00000	0.00000	1124	0.87105	909	0.6068	34.8817	979.0602	44.3056
0.10242	0.10852	1143	0.87012	880	0.5971	33.6606	994.5472	44.7389
0.20099	0.21156	1156	0.86810	862	0.5910	32.9062	1003.5236	45.1498
0.30236	0.31616	1171	0.86523	843	0.5844	32.0265	1013.1843	45.6244
0.40210	0.41773	1187	0.86305	822	0.5771	31.0302	1024.4404	46.0833
0.49886	0.51501	1202	0.86100	804	0.5707	30.0870	1034.9220	46.5136
0.60032	0.61573	1213	0.85882	791	0.5661	29.4398	1041.7487	46.9158
0.69962	0.71301	1225	0.85607	778	0.5614	28.7331	1048.6858	47.3668
0.79703	0.80728	1243	0.85394	758	0.5542	27.5247	1061.4474	47.8402
0.89420	0.90017	1259	0.85169	741	0.5479	26.4585	1072.2777	48.3012
1.00000	1.00000	1269	0.84878	732	0.5446	25.87460	1077.1018	48.7472
1, 3, 5-trimethylbenzene (1) + n-propylethanoate (2)								
0.00000	0.00000	1124	0.87105	909	0.6068	34.8817	979.0602	44.3056
0.10218	0.12014	1144	0.86930	879	0.5968	34.0887	994.4792	45.4146
0.19832	0.22885	1158	0.86660	861	0.5906	33.6988	1003.5228	46.4543
0.30194	0.34164	1175	0.86437	838	0.5827	33.0617	1015.6348	47.7390
0.39689	0.44120	1190	0.86259	819	0.5760	32.4703	1026.4821	48.6004
0.49766	0.54310	1211	0.86090	792	0.5665	31.3819	1042.5499	49.7447
0.59783	0.64075	1233	0.85951	765	0.5567	30.1381	1059.7758	50.8883
0.69925	0.73612	1244	0.85810	753	0.5523	29.7580	1067.4764	51.9162
0.79367	0.82191	1256	0.85631	740	0.5475	29.2434	1075.5254	52.9296
0.89187	0.90822	1278	0.85505	716	0.5386	27.8312	1092.7539	54.0693
1.00000	1.00000	1309	0.85447	683	0.5260	25.5847	1118.5012	55.3720

$$u^2 = 1/\beta_{ad}\rho \quad (1)$$

Cyclohexane < 1, 4-dimethylbenzene ~ 1, 3, 5-trimethylbenzene < Benzene

β_{ad} evaluated on the basis of equation (1) for all the systems with the concentrations are also reported in Table 1. Variations of u_{12} , ρ_{12} and β_{ad} with concentrations of hydrocarbons for binary systems with methylethanoate only are shown in Figures 1-3 respectively for the sake of brevity. A perusal of Table 1 shows that the values of u_{12} increase with mole fractions of hydrocarbon solvents (x_1) in all the twelve binary mixtures studied. Ultrasound waves are high frequency mechanical waves and propagates as either longitudinal or shear waves. The increase in ultrasound velocities with composition is quite expected as the densities decrease with composition. Comparison of u_{12} values in binary mixtures of a particular ester with different hydrocarbons having fixed composition shows the following order except a few stray values:

Cyclohexane < Benzene < 1, 4-dimethylbenzene < 1, 3, 5-trimethylbenzene

Table 1 shows the following order about densities for the above mixtures.

Perusal of Table 1 shows that β_{ad} values lies on the following order:

Cyclohexane > Benzene > 1, 4-dimethylbenzene > 1, 3, 5-trimethylbenzene with a common ester in the binary mixture except the binary mixture of n-propylethanoate with benzene which has lowest β_{ad} values. However, a few of the β_{ad} values for this system at higher concentration of hydrocarbon are higher than the mixture of ester with 1, 4-dimethylbenzene and 1, 3, 5-trimethylbenzene. The order of β_{ad} and u_{12} values for the studied binary systems above mentioned except for the mixture of n-propylethanoate and benzene binary system shows that the compressibilities play a dominant role to affect the velocities rather than the densities. At a fixed mole fraction of hydrocarbon in binary mixture for a common ester with different hydrocarbons systems, the largest values of β_{ad} with cyclohexane show that when cyclohexane is mixed into ester, self association of the ester decreases causing an increase in interspace between ester molecules in the mixture compare to that in pure

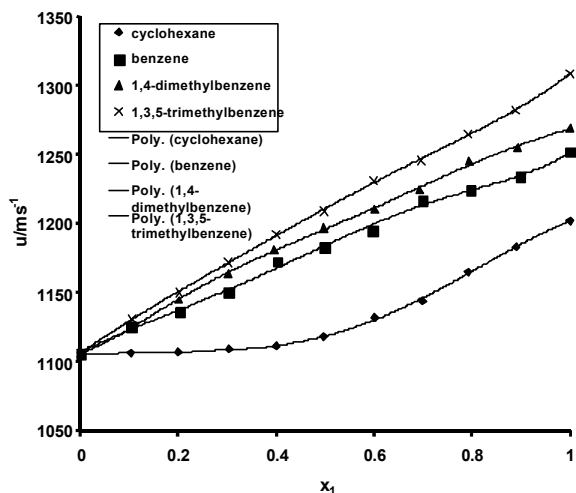


Fig. 1: Ultrasound velocities (u_{12}) of binary mixtures of methylethanoate+ hydrocarbons versus mole fractions of hydrocarbon (x_1) at 308.15 K.

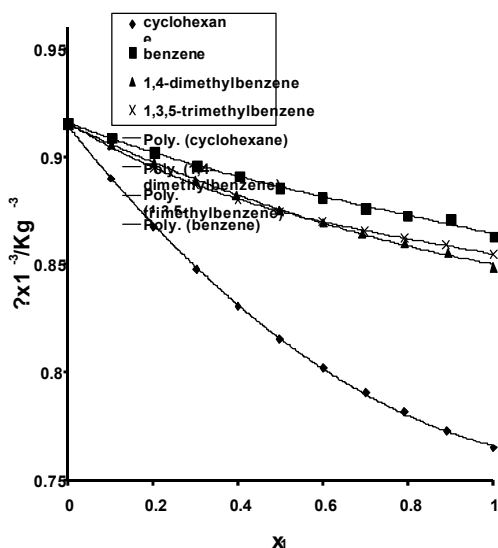


Fig. 2: Densities (ρ_{12}) of binary mixtures of methylethanoate + hydrocarbons versus mole fractions of hydrocarbon (x_1) at 308.15 K.

ester. Similar suggestion [21] has been made on the basis of the study of binary systems of alcohol with cyclohexane. When cyclohexane is replaced with benzene, 1, 4-dimethylbenzene and 1, 3, 5-trimethylbenzene rupturing of intermolecular association of ester decreases causing a decrease in the adiabatic compressibilities.

Free lengths (L_f), which is an intermolecular property defined as the distance between the surfaces of the molecules, are evaluated from the values of adiabatic compressibilities employing equation

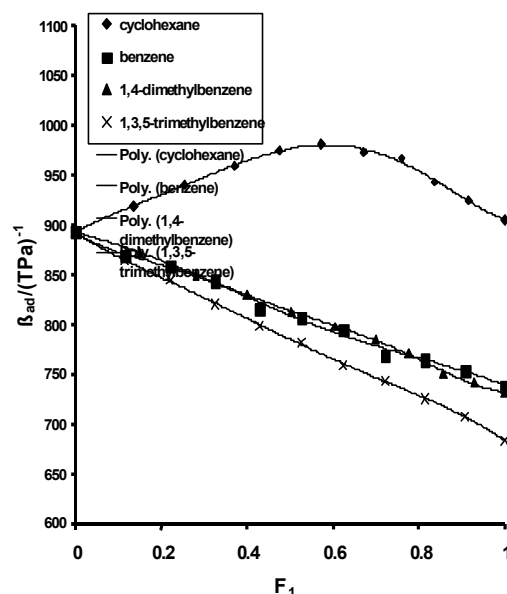


Fig. 3: Adiabatic compressibilities (β_{ad}) of binary mixtures of methylethanoate + hydrocarbons versus volume fractions of hydrocarbon (\bar{O}_1) at 308.15 K.

$$L_f = K (\beta_{ad})^{1/2} \quad (2)$$

Where K is Jacobson's constant which is dependent on temperature. Evaluated L_f values are given in Table 1. For the sake of brevity variation of L_f values with composition for binary mixtures of methylethanoate only with several hydrocarbons is shown in Figure 4. A perusal of the Table 1 shows that for a common ester in the binary mixture L_f values follow the order Cyclohexane > Benzene > 1, 4-dimethylbenzene > 1, 3, 5-trimethylbenzene except the binary mixtures of n-propylethanoate and benzene were L_f values are lowest. However, a few of the L_f values for the system at higher concentrations of hydrocarbons are higher than the mixture of ester with 1, 4-dimethylbenzene and 1, 3, 5-trimethylbenzene. The above order of free lengths is quite similar to that observed for β_{ad} as expected.

Available volume (V_a) is the difference of molar volume at temperature T and that at absolute zero temperature. which is evaluated from the measured values of ultrasound velocities at experimental temperature by equation

$$V_a = V_T [1 - (u/u_0)] \quad (3)$$

where V_T is the molar volume at experimental temperature T and $u_0 = 1600 \text{ ms}^{-1}$. The values of V_a for all

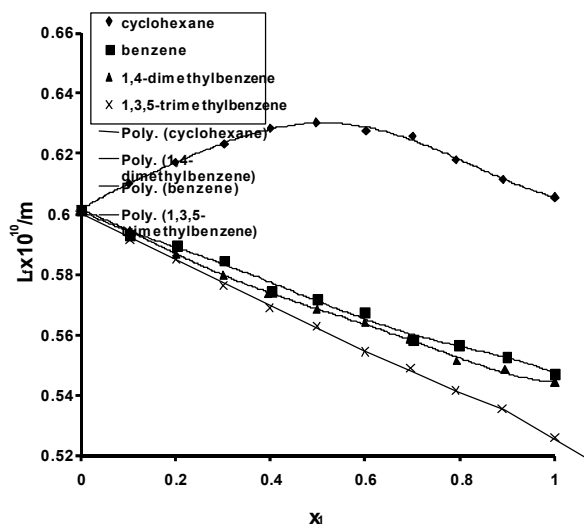


Fig. 4: Free lengths (L_f) of binary mixtures of methylethanoate+hydrocarbons versus mole fractions of hydrocarbon (x_1) at 308.15 K.

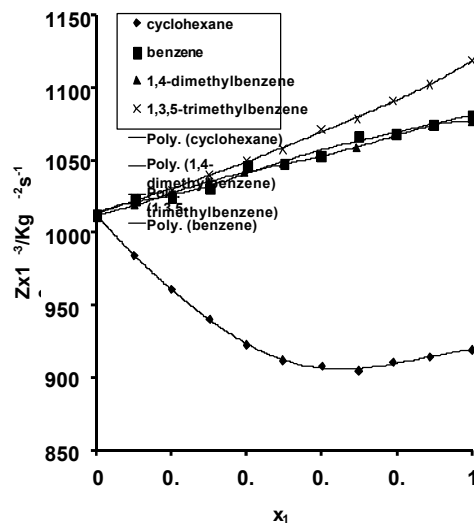


Fig. 6: Acoustical impedances (Z) of binary mixtures of methylethanoate + hydrocarbons versus mole fractions of hydrocarbon (x_1) at 308.15 K.

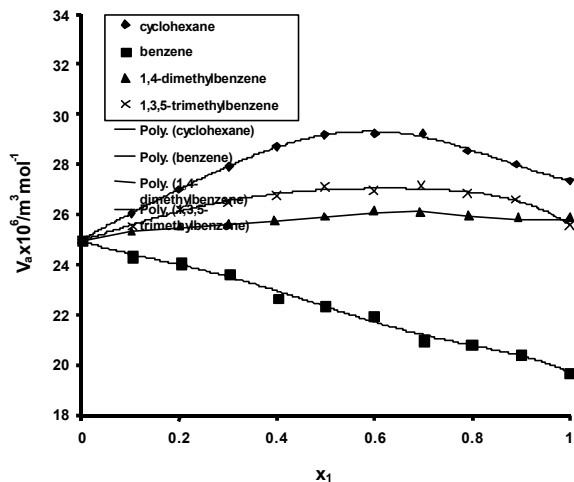


Fig. 5: Available volumes (V_a) of binary mixtures of methylethanoate+hydrocarbons versus mole fractions of hydrocarbon (x_1) at 308.15 K.

the systems are also recorded in table 1 and the variation of V_a with concentrations of hydrocarbons for binary systems with methylethanoate ester only is shown in Figure 5. Perusal of V_a values shows that they have following order for the binary mixtures of a common ester with several hydrocarbons:

Cyclohexane > 1, 3, 5-trimethylbenzene > 1, 4-dimethylbenzene > Benzene

The observed trend of V_a for the mixtures of an ester with aromatic solvents is just the reverse of that for L_f and β_{ad} .

Kinsler *et al.* [22] have suggested that acoustical impedance is more significant parameter to describe the medium and the intermolecular interactions than the ultrasound velocity and density individually. Thus acoustical impedances, Z for all the binary mixtures are evaluated from the experimental values of ultrasound velocities and densities employing following equation

$$Z = u \cdot \rho \quad (4)$$

Values of Z for the mixtures studied are given in Table 1. For the sake of clarity and to avoid repetitions of figures of similar type only the variations of Z values of binary mixtures of different hydrocarbons with methylethanoate are represented in Figure 6. It is evident that the values of Z follow the similar trend as that of u for all the mixtures studied i.e. Cyclohexane < Benzene < 1, 4-dimethylbenzene < 1, 3, 5-trimethylbenzene except n-propylethanoate and benzene binary mixture for which Z values are highest. However, at higher mole fractions of hydrocarbon a few Z values are lower for this system than n-propylethanoate and 1, 3, 5-trimethylbenzene system.

Wada's constant, W are evaluated from experimental ρ and evaluated β_{ad} values by equation

$$W = (x_1 M_1 + x_2 M_2 / \rho_{12}) \cdot (\beta_{ad})^{-1/7} \dots \quad (5)$$

Where M_1 , M_2 are mass of component 1 and 2 respectively. The values of W for all the systems studied are given in Table 1. Variation of W with mole fractions of

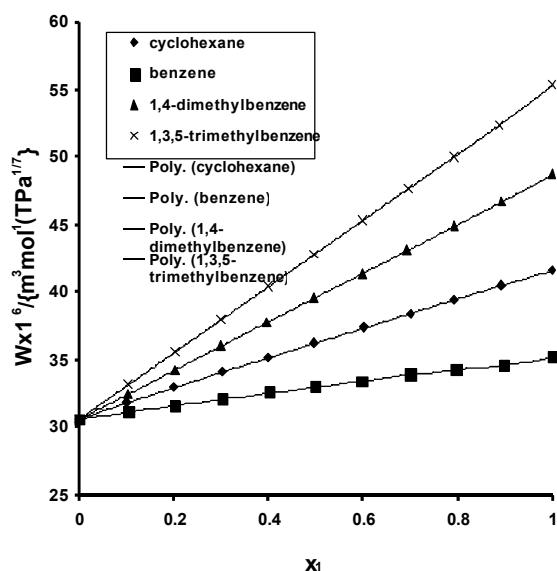


Fig. 7: Wada's constants (W) of binary mixtures of methylethanoate + hydrocarbons versus mole fractions of hydrocarbon (x_1) at 308.15 K.

hydrocarbon solvents are linear for all the mixtures studied. Such variations of binary mixtures of methylethanoate only with different hydrocarbons are shown in figure 7. The values of W for binary mixtures of

common ester, with several hydrocarbons follow the trend exactly similar to that for molar volumes of the hydrocarbons. It is interesting to note that when methylethanoate is replaced by ethylethanoate for a common hydrocarbon solvent in a binary mixture, the slopes of straight lines decrease with almost the same amount. Similarly when ethylethanoate is replaced by *n*-propylethanoate in the binary mixture, the slopes of the straight lines further decrease with almost the same amount. It seems that for a common hydrocarbon decreasing in slopes of straight lines are dependent on the chain length of the esters i.e. on the number of $>CH_2$ groups present in the esters.

Deviations from additive values in any parameter 'A' except adiabatic compressibility are evaluated by equation (6)

$$\Delta A \text{ or } A^E = A_{\text{exp}} - x_1 A_1 - x_2 A_2 \quad (6)$$

where A_1 , A_2 and x_1 , x_2 are the values of parameter A and mole fraction of component 1 and 2 respectively. The deviations in ultrasound velocities Δu from additive values with concentrations of hydrocarbons for all the systems are recorded in Table 2. Table 2 shows that the values of Δu follow the similar trend as that of

Table 2: Mole fraction (x_1), Volume fraction (\bar{O}_1) of hydrocarbons, Δu , β_{ad}^E , ΔL_f , ΔZ and ΔV_a at 308.15 K for binary mixtures of different esters + hydrocarbon solvents

x_1	\bar{O}_1	$\Delta u \text{ ms}^{-1}$	$\beta_{\text{ad}}^E (\text{T Pa})^{-1}$	$\Delta L_f \times 10^{10} \text{ m}$	$\Delta Z \times 10^{-3} \text{ kgm}^{-2} \text{ s}^{-1}$	$\Delta V_a \times 10^6 \text{ m}^3 \text{ mol}^{-1}$
Cyclohexane (1) + Methylethanoate (2)						
0.00000	0.00000	---	---	----	----	----
0.10176	0.13350	-10	24	0.0083	-18.9410	0.8343
0.19971	0.25341	-18	44	0.0148	-33.2112	1.5556
0.30251	0.37104	-26	62	0.0206	-44.0639	2.2099
0.40017	0.47574	-33	76	0.0254	-52.4104	2.8020
0.49683	0.57319	-36	81	0.0269	-54.2920	3.0385
0.60098	0.67198	-32	72	0.0240	-48.5386	2.8045
0.70000	0.76040	-29	64	0.0213	-42.6597	2.5870
0.79112	0.83745	-17	40	0.0134	-28.4119	1.6749
0.89061	0.91717	-9	20	0.0068	-15.0966	0.8941
1.00000	1.00000	---	---	----	----	----
Benzene (1) + Methylethanoate (2)						
0.00000	0.00000	---	---	----	----	----
0.10333	0.11418	4	-8	-0.0025	2.7821	-0.0990
0.20217	0.22087	1	-3	-0.0006	-1.4800	0.1641
0.30174	0.32586	0	-3	-0.0003	-2.7681	0.2532
0.40155	0.42878	8	-16	-0.0051	5.5203	-0.1868
0.49845	0.52646	4	-9	-0.0026	1.3882	0.0469
0.59729	0.62395	2	-6	-0.0015	0.0306	0.1495
0.69852	0.72160	9	-15	-0.0050	6.7239	-0.2828
0.79661	0.81397	2	-5	-0.0015	1.5913	0.0482
0.89755	0.90742	-3	-1	-0.0001	1.1764	0.1793
1.00000	1.00000	---	---	----	----	----

Table 2: Continued

1, 4-dimethylbenzene (1) + Methylthanoate (2)						
0.00000	0.00000	---	---	----	----	----
0.10315	0.15095	2	3	-0.0011	-0.3201	0.2925
0.20182	0.28102	6	2	-0.0031	1.6704	0.3704
0.30094	0.39958	9	1	-0.0043	2.7863	0.4160
0.39561	0.50296	11	1	-0.0050	3.4264	0.4306
0.49725	0.60459	10	2	-0.0047	2.7648	0.4889
0.60002	0.69871	6	5	-0.0031	0.8514	0.6441
0.69291	0.77719	6	3	-0.0030	1.3925	0.5178
0.79307	0.85560	6	-0	-0.0033	3.3433	0.2795
0.89376	0.92860	3	-2	-0.0021	3.2334	0.1225
1.00000	1.00000	---	---	----	----	----
1, 3, 5-trimethylbenzene (1) + Methylthanoate (2)						
0.00000	0.00000	---	---	----	----	----
0.10375	0.16756	4	6	-0.0020	-0.5087	0.5114
0.20201	0.30561	3	16	-0.0011	-4.8062	1.1050
0.30079	0.42790	5	17	-0.0024	-4.5602	1.3529
0.39898	0.53579	5	19	-0.0024	-5.4664	1.5652
0.49755	0.63258	2	22	-0.0011	-8.0531	1.8327
0.59872	0.72177	4	18	-0.0018	-5.0980	1.6162
0.69588	0.79913	-1	19	0.0001	-8.0382	1.7441
0.79113	0.86817	-2	14	0.0000	-5.4465	1.3849
0.88838	0.93260	-4	11	0.0011	-4.6805	1.0842
1.00000	1.00000	---	---	----	----	----
Cyclohexane (1) + Ethylethanoate (2)						
0.00000	0.00000	---	---	----	----	----
0.10463	0.11412	-10	19	0.0062	-11.5985	0.7628
0.20008	0.21610	-15	31	0.0100	-19.2545	1.2628
0.30369	0.32466	-23	48	0.0155	-28.5660	1.9288
0.40048	0.42405	-29	59	0.0190	-34.0972	2.3700
0.49957	0.52388	-32	64	0.0206	-36.5790	2.6014
0.59675	0.61993	-34	65	0.0212	-37.2347	2.7163
0.69868	0.71876	-25	50	0.0163	-29.8442	2.1217
0.79949	0.81464	-17	34	0.0111	-20.5268	1.4716
0.89495	0.90375	-10	20	0.0067	-12.1269	0.8953
1.00000	1.00000	---	---	----	----	----
Benzene (1) + Ethylethanoate (2)						
0.00000	0.00000	---	---	----	----	----
0.10020	0.09170	-9	9	0.0040	-7.0297	0.4646
0.19794	0.18282	-10	7	0.0039	-8.4885	0.4772
0.29588	0.27585	-15	13	0.0065	-13.6575	0.7864
0.39660	0.37341	-8	-1	0.0024	-7.4375	0.3337
0.50010	0.47554	-12	4	0.0042	-10.8478	0.5379
0.59110	0.56719	-5	-6	0.0004	-4.1906	0.0964
0.70227	0.68136	-9	2	0.0028	-8.3609	0.3923
0.79038	0.77365	-5	-2	0.0010	-4.5217	0.1492
0.89202	0.88219	-4	0	0.0010	-3.6681	0.1505
1.00000	1.00000	---	---	----	----	----

Table 2: Continued

1, 4-dimethylbenzene (1) + Ethylethanoate (2)						
0.00000	0.00000	---	---	----	----	----
0.10408	0.12703	6	-8	-0.0040	5.4999	-0.1421
0.20239	0.24119	3	-4	-0.0032	2.8454	0.1963
0.30413	0.35382	1	0	-0.0025	1.1186	0.4521
0.40065	0.45577	1	0	-0.0030	1.3751	0.5211
0.49394	0.55010	4	-2	-0.0037	2.1628	0.4129
0.60558	0.65794	7	-8	-0.0055	5.7085	0.1193
0.69676	0.74216	10	-11	-0.0063	8.4512	-0.1734
0.79593	0.83011	9	-10	-0.0055	7.5706	-0.2608
0.88762	0.90819	7	-7	-0.0038	5.9650	-0.2667
1.00000	1.00000	---	---	----	----	----
1, 3, 5-trimethylbenzene (1) + Ethylethanoate (2)						
0.00000	0.00000	---	---	----	----	----
0.10355	0.1399	-6	14	0.0023	-6.0208	0.9015
0.19815	0.2583	-9	21	0.0031	-9.9583	1.4814
0.30053	0.3771	-3	13	-0.0006	-5.5326	1.3795
0.39593	0.4801	4	3	-0.0047	1.3544	0.9918
0.49361	0.5787	0	9	-0.0027	-2.8254	1.3691
0.60022	0.6790	-3	12	-0.0011	-5.2368	1.5508
0.69472	0.7623	1	5	-0.0029	-0.7232	1.0413
0.79823	0.8479	-2	7	-0.0009	-2.7693	1.0361
0.88565	0.9160	-2	5	-0.0002	-2.5229	0.7331
1.00000	1.00000	---	---	----	----	----
Cyclohexane (1) + n-propylethanoate (2)						
0.00000	0.00000	---	---	----	----	----
0.10248	0.09680	-5	6	0.0024	-3.7000	0.3615
0.19888	0.18896	-10	14	0.0047	-7.6153	0.7050
0.29761	0.28453	-15	25	0.0085	-14.6265	1.2047
0.39914	0.38403	-19	33	0.0107	-18.4438	1.5102
0.49975	0.48390	-21	33	0.0109	-17.4717	1.5528
0.59907	0.58374	-18	27	0.0091	-14.8715	1.3174
0.70004	0.68657	-19	32	0.0105	-18.0912	1.4550
0.79722	0.78677	-12	18	0.0060	-9.6502	0.8798
0.89387	0.88769	-3	3	0.0008	-1.1625	0.1759
1.00000	1.00000	---	---	----	----	----
Benzene (1) + n-propylethanoate (2)						
0.00000	0.00000	---	---	----	----	----
0.10878	0.08610	22	-48	-0.0147	28.5039	-2.0625
0.20393	0.16510	22	-51	-0.0155	27.8677	-2.1184
0.30294	0.25121	19	-49	-0.0134	26.0035	-1.9993
0.40286	0.34243	16	-47	-0.0121	23.6794	-1.8318
0.50159	0.43722	17	-48	-0.0121	24.2047	-1.8377
0.60094	0.53287	19	-49	-0.0129	26.2750	-1.9191
0.70145	0.64460	21	-48	-0.0135	28.0802	-1.9402
0.80146	0.75706	15	-31	-0.0082	14.3618	-1.2716
0.89719	0.87074	9	-18	-0.0049	8.8215	-0.7412
1.00000	1.00000	---	---	----	----	----

Table 2: Continued

1, 4-dimethylbenzene (1) + n-propylethanoate (2)						
0.00000	0.00000	---	---	---	---	---
0.10242	0.10852	4	-10	-0.0033	5.4456	-0.2986
0.20099	0.21156	3	-10	-0.0033	4.7581	-0.1652
0.30236	0.31616	3	-10	-0.0036	4.4803	-0.1318
0.40210	0.41773	5	-13	-0.0045	5.9577	-0.2298
0.49886	0.51501	6	-14	-0.0050	6.9528	-0.3014
0.60032	0.61573	2	-9	-0.0033	3.8322	-0.0347
0.69962	0.71301	0	-5	-0.0018	1.0337	-0.1529
0.79703	0.80728	3	-8	-0.0030	4.2451	-0.1781
0.89420	0.90017	5	-9	-0.0032	5.5487	-0.3690
1.00000	1.00000	---	---	---	---	---
1, 3, 5-trimethylbenzene (1) + n-propylethanoate (2)						
0.00000	0.00000	---	---	---	---	---
0.10218	0.12014	1	-3	-0.0018	1.1709	0.1570
0.19832	0.22885	-3	4	-0.0001	-3.1913	0.6609
0.30194	0.34164	-5	6	0.0003	-5.5283	0.9871
0.39689	0.44120	-7	10	0.0013	-7.9209	1.2785
0.49766	0.54310	-5	6	-0.0001	-5.9045	1.1270
0.59783	0.64075	-2	1	-0.0018	-2.6464	0.8144
0.69925	0.73612	-9	10	0.0020	-9.0879	1.3772
0.79367	0.82191	-15	17	0.0049	-14.2050	1.7405
0.89187	0.90822	-11	12	0.0038	-10.6696	1.2412
1.00000	1.00000	---	---	---	---	---

the ultrasound velocities u for a common ester with different hydrocarbon solvents. A perusal of Table 2 also shows that the values of Δu for all the esters are negative for binary mixtures with cyclohexane and positive for the mixture with 1, 4-dimethylbenzene. It has been suggested [23] that the negative deviation in sound velocity and positive deviation in compressibility may be due to presence of dispersion forces and a positive deviation in sound velocity and negative deviation in compressibility may be due to charge-transfer, dipole-dipole and dipole-induced dipole interactions. In the present study negative Δu values of binary mixtures for all the esters with cyclohexane indicate the presence of dispersion forces between the unlike molecules. This can be understood because cyclohexane has no polarisable π -electrons. The positive Δu values for the binary mixtures of all the esters with 1, 4-dimethylbenzene indicate the presence of dipole-induced dipole interactions between unlike molecules. The esters are polar molecules having significant values of dipole moments [24]. Aromatic hydrocarbon 1, 4-dimethylbenzene has π -electrons which are easily polarisable due to presence of two methyl groups. Thus in the binary systems of esters with 1, 4-dimethylbenzene, the dipole-induced dipole interactions are quite expected.

However, Δu values for binary mixtures of esters with benzene are positive with exception of binary mixtures with ethylethanoate and negative with 1, 3, 5-trimethylbenzene with some stray values. Although 1, 3, 5-trimethylbenzene has polarisable π -electrons, the negative Δu values for the binary mixtures of esters with 1, 3, 5-trimethylbenzene may be due to blocking of the mutual approach of unlike molecules reducing the dipole-induced dipole interactions between the component molecules.

Excess adiabatic compressibility β_{ad}^E , for the systems studied are evaluated using volume fractions in place of mole fractions employing equation (6). The evaluated values of β_{ad}^E are given in Table 2. Table 2 shows that β_{ad}^E values of the binary mixtures for all the esters with cyclohexane are positive which indicates that presence of cyclohexane reduces the self association of ester molecules and consequently enlarging the volume and increasing the compressibility. Values of β_{ad}^E for binary mixtures of esters with benzene and 1, 4-dimethylbenzene are negative except the mixture of ethylethanoate with benzene and methylethanoate with 1, 4-dimethylbenzene. This shows that the mixtures are more compact due to dipole-induced dipole interaction between

unlike molecules. Fort and Moore [25] and Kaulgud [26] have suggested that β_{ad}^E becomes increasingly negative with increasing strength of interaction between the component liquids. This is similar to our observation for the binary mixtures of esters with benzene. β_{ad}^E values of binary mixtures with 1, 3, 5-trimethylbenzene are positive and smaller than that for cyclohexane. This also confirms the same contention as observed on the basis of deviations in velocities. Our observation that positive deviation in compressibility is due to dispersion forces while negative deviation is the result of stronger specific interaction between the component molecules is similar to that by Kalra *et al.* [27].

The deviations in free lengths from additive values, ΔL_f are also evaluated employing equation (6) and recorded in Table 2. Table 2 shows that ΔL_f values for all the binary mixtures of esters with cyclohexane are positive and for the other binary mixtures these are negative. However ΔL_f values for ethylethanoate + benzene mixture are positive but smaller than that for mixture of ester with cyclohexane. This also shows that interactions between unlike molecules in such mixtures are stronger than that in the mixture with cyclohexane. This once again confirm the contention that there are graded interactions in the binary mixtures studied.

Values of deviations in acoustical impedance, ΔZ with concentration of hydrocarbons are recorded in Table 2. Perusal of Table 2 shows that ΔZ values follow the similar trend as that for Δu values for the binary mixture of ethylethanoate and n-propylethanoate with all the hydrocarbon solvents. However, the binary mixtures of hydrocarbons with methylethanoate show some exceptional behavior. The lowest values of ΔZ for the binary mixtures of all the esters with cyclohexane show the presence of weak dispersion interactions between unlike molecules. Higher ΔZ values for the binary mixtures of esters with aromatic hydrocarbons than that with cyclohexane once again show the existence of stronger specific interactions between esters and aromatic hydrocarbon molecules.

Deviations in available volume from additive values, ΔV_a for all the mixtures are evaluated and recorded in Table 2. A close observation of table 2 reveals that ΔV_a values are highest for the binary mixtures of esters with cyclohexane showing larger void in the mixture. These voids in the binary mixtures of esters with aromatic hydrocarbons reduce showing compact packing of unlike molecules in the binary mixtures.

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