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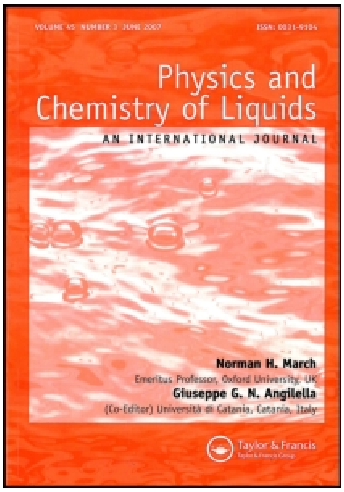

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Volumetric and Ultrasonic Behaviour of 1,2-Dibromoethane With a Ketone

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Volumetric and Ultrasonic Behaviour of 1,2-Dibromoethane With a Ketone

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New experimental data for excess volume and sound velocity in binary liquid mixtures of 1,2-dibromoethane with methyl ethyl ketone, diethyl ketone, methyl propyl ketone, methyl isobutyl ketone, cyclopentanone and cyclohexanone at 303.15 K have been reported. The results have been analysed in terms of free length theory (FLT) and collision factor theory (CFT). Molar sound velocity (R), molar compressibility (W) and van der Waals constant (b) have also been computed. The sound velocity data were also used along with densities to compute isentropic compressibilities (k_s). The excess volumes (V^E) and the deviation in isentropic compressibilities (Δk_s) are used to understand the interactions between unlike molecules.

1 INTRODUCTION

In continuation to our studies on excess thermodynamic properties and ultrasonic behaviour of binary mixtures of haloethane with a series of ketones,[1,2] we report here excess volumes, ultrasonic sound velocities and isentropic compressibilities for mixtures of 1,2-dibromoethane (DBE) with methyl ethyl ketone (MEK), diethyl ketone (DEK), methyl propyl ketone (MPK), methyl isobutyl ketone (MIBK), cyclopentanone and cyclohexanone at 303.15 K. Sound velocity data have been analysed in terms of free length theory (FLT) due to Jacobson[3] and collision factor theory (CFT) developed by Schaaffs.[4] The molar sound velocity[5] (R), molar compressibilities[6] (W) and van der Waals constant[7] (b) which give information on the complex formation association or deassociation of the components, have also been computed.

2 THEORETICAL ASPECTS

The theoretical aspects of FLT and CFT have been described in detail by Choudary and Naidu.[8] Values of R and W of the pure liquids as well as of mixtures can be calculated by using the following expressions:

$$R = \frac{M}{\rho} u^{1/3} \tag{1}$$

and

$$W = \frac{M}{\rho} k_s^{-1/7} \tag{2}$$

where M, ρ, u and k_s stand for molecular weight, density, sound velocity and isentropic compressibility respectively. For pure liquids, b can be calculated using the relation

$$b = \tfrac{16}{3}\pi N r^3 \tag{3}$$

where r is the effective molecular radius. The value of b for the binary mixtures can be calculated from pure liquid values using the mole fractions of the components.

3 EXPERIMENTAL SECTION

1,2-Dibromoethane and ketones were purified by the methods described by Reddick and Bunger.[9] The purity of the samples are checked by comparing the measured densities of the components with those reported in the literature.[9,10]

Excess volumes were measured by using the dilatometer described by Rao and Naidu.[11] The V^E values were accurate to $\pm 0.003\ \mathrm{cm^3\ mol^{-1}}$. Isentropic compressibilities were computed from the density and sound velocity data, density data being computed from the measured excess volume data. The following relation was employed for computing density:

$$\rho = \frac{X_1 M_1 + X_2 M_2}{V_m + V^E} \tag{4}$$

where X, M and V_m denote the mole fraction, molecular weight and molar volume of the mixtures respectively. The ultrasonic sound velocity was measured with a single-crystal interferometer at a frequency of 2 MHz and the data were accurate to $\pm 0.15\%$. All the measurements were made at constant temperature employing a thermostat that could be maintained to within ± 0.01 K.

4 RESULTS AND DISCUSSION

(a) Excess volume

The experimental V^E values of the six binary mixtures are given in Table I and are also graphically represented in Figure 1. The dependence of V^E on composition may be expressed by an empirical equation of the form

$$V^E/\text{cm}^3\,\text{mol}^{-1} = x_1(1 - x_1)[(a_0 + a_1(2x_1 - 1) + a_2(2x_1 - 1)^2] \quad (5)$$

where a_0, a_1 and a_2 are adjustable parameters and x_1 is the mole fraction of DBE. The values of the parameters obtained by the least squares method are included in Table II along with the standard deviation $\sigma(V^E)$. The values of $\sigma(V^E)$ were obtained by the equation

$$\sigma(V^E) = \frac{\sum (V^E_{\text{calcd.}} - V^E_{\text{exptl.}})^{1/2}}{n - P} \quad (6)$$

where n is the number of experimental data and P is the number of parameters.

TABLE I

Excess volumes (V^E) for the binary mixtures of 1,2-dibromoethane with ketones at 303.15 K

x_1	V^E cm^3 mol^{-1}	x_1	V^E cm^3 mol^{-1}	x_1	V^E cm^3 mol^{-1}
1,2-dibromoethane + methyl ethyl ketone		1,2-dibromoethane + methyl propyl ketone		1,2-dibromoethane' + cyclopentanone	
0.1596	−0.030	0.1532	0.018	0.0979	0.042
0.2852	−0.005	0.2687	0.029	0.2116	0.064
0.4164	−0.064	0.3394	0.036	0.2438	0.065
0.4925	−0.070	0.4445	0.045	0.4445	0.083
0.5665	−0.078	0.5053	0.047	0.5421	0.078
0.7403	−0.077	0.6350	0.040	0.6065	0.075
0.7990	−0.074	0.8073	0.022	0.7630	0.060
0.9142	−0.047	0.9179	0.010	0.9137	0.028
1,2-dibromoethane + diethyl ketone		1,2-dibromoethane + methyl isobutyl ketone		1,2-dibromoethane + cyclohexanone	
0.1534	−0.018	0.1405	0.012	0.1310	0.062
0.2249	−0.030	0.2969	0.023	0.2804	0.103
0.3352	−0.034	0.3235	0.028	0.3434	0.113
0.4521	−0.039	0.5088	0.031	0.4915	0.120
0.5021	−0.037	0.5725	0.030	0.6046	0.112
0.6553	−0.026	0.6813	0.026	0.6481	0.106
0.8072	−0.012	0.8395	0.016	0.7943	0.075
0.9269	−0.004	0.9306	0.009	0.9107	0.037

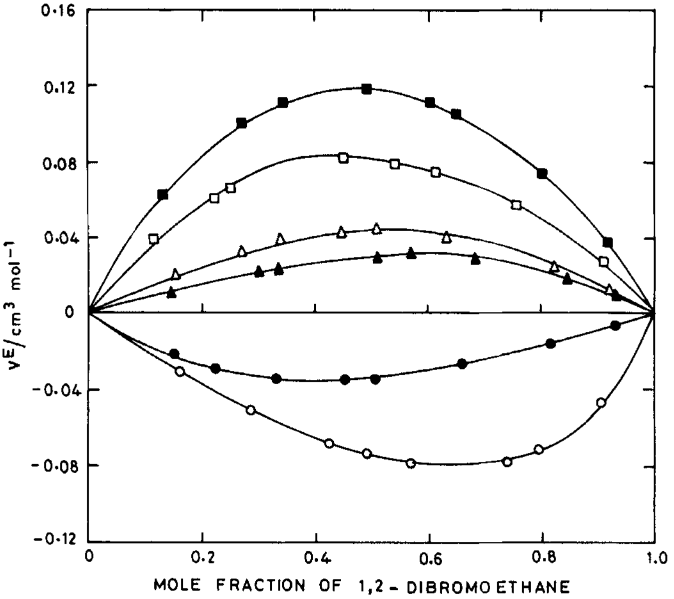

FIGURE 1 The values of V^E against mole fraction (x_1) for the system 1,2-dibromoethane with: ○, methyl ethyl ketone; ●, diethyl ketone; △, methyl propyl ketone; ▲, methyl isobutyl ketone; □, cyclopentanone; ■, cyclohexanone.

TABLE II

Values of parameters for Eq. (5) calculated by the method of least squares along with the standard deviation $\sigma(V^E)$ at 303.15 K

System	a_0	a_1	a_2	$\sigma(V^E)$
	(cm^3 mol^{-1})			
1,2-dibromoethane + methyl ethyl ketone	−0.282	−0.199	−0.195	0.003
1,2-dibromoethane + diethyl ketone	−0.146	0.055	0.066	0.002
1,2-dibromoethane + methyl propyl ketone	0.178	0.008	−0.082	0.002
1,2-dibromoethane + methyl isobutyl ketone	0.128	0.024	−0.021	0.002
1,2-dibromoethane + cyclopentanone	0.311	−0.065	0.138	0.003
1,2-dibromoethane + cyclohexanone	0.473	−0.059	0.038	0.002

The results included in Figure 1 show that V^E is negative in the systems of DBE with MEK and DEK at all concentrations. The mixtures of DBE and MPK, MIBK, cyclopentanone and cyclohexanone results in positive excess volumes throughout the mole fraction range studied. The values of V^E may be explained in terms of (i) differences in size of the unlike molecules (ii) difference in shape of component molecules (iii) deassociation of self associated bromoalkane[12] and ketones,[13] (iv) dipole induced dipole interactions[2] between ketoxy group and free electrons of bromine atoms of 1,2-dibromoethane. The first three factors contribute to expansion in volume while the last factor contributes to contraction in volume.

Values of V^E change from negative to positive as the ketone is changed from MEK to cyclic ketones indicating the varying contributions from the above factors. The excess volume fall in order: MEK < DEK < MIBK < MPK < cyclopentanone < cyclohexanone.

(b) Sound velocities

The surface area (Y) and collision factor (s) of the pure components used in FLT and CFT were calculated using the experimental sound velocities and densities. Critical temperature surface tension and the ratio of specific heats, which were used in the calculation of molar volume at absolute zero (V_0) and the average molecular radius (r_m), were taken from the literature.[10,14–16] The values of molar volume (V_m), available volume (V_a) free length (L_f), surface area (Y), collision factor (s) and the average radius of the molecules (r_m) of the pure components are given in Table III. The free length of the binary mixtures and sound velocity data predicted in terms of FLT and CFT are given in Table IV along with experimental results in columns 2 to 5. The percentage deviation between the experimental sound

TABLE III

Values of v_m, v_0, v_a, L_f, Y, S and r_m of the pure liquid components at 303.15 K

Component	v_m	v_0	v_a	L_f Å	Y	S	r_m Å
	Volume, cm^3 mol^{-1}						
1,2-dibromoethane	86.990	69.797	17.193	0.4364	78.804	1.2262	2.586
Methyl ethyl ketone	90.767	70.663	20.104	0.6051	66.452	1.6577	2.513
Diethyl ketone	107.054	84.785	22.269	0.5877	75.783	1.6128	2.700
Methyl propyl ketone	108.122	85.790	22.332	0.5994	74.513	1.9046	2.565
Methyl isobutyl ketone	126.585	100.896	25.689	0.6068	84.667	3.1590	2.264
Cyclopentanone	89.587	71.884	17.703	0.4739	74.707	1.7736	2.581
Cyclohexanone	104.686	85.946	18.740	0.4695	79.827	1.7653	2.732

TABLE IV

Values of L_f^M, u_{exptl}, u_{theo}(FLT) and u_{theo}(CFT), u%(FLT) and u%(CFT), R, W and b for 1,2-dibromoethane with Ketones at 303.15 K

		Values of u, m sec^{-1}							
			Theoretical						
x_1	L_f^M Å	Expt.	FLT	CFT	u% (FLT)	u% (CFT)	R	W	b (Cm^3 mol^{-1})
			1,2-dibromoethane + methyl ethyl ketone						
0.0000	0.6051	1170	1170	1170	0.00	0.00	956	1773	160.15
0.1596	0.5732	1100	1098	1145	0.17	4.13	930	1790	162.39
0.2852	0.5495	1060	1061	1125	0.04	6.12	914	1800	164.17
0.4164	0.5260	1031	1033	1102	0.16	6.89	900	1812	166.04
0.4925	0.5129	1018	1021	1088	0.25	6.90	894	1818	167.13
0.5665	0.5004	1008	1011	1074	0.32	6.59	888	1825	168.19
0.7403	0.4728	996	996	1040	0.01	4.44	878	1843	170.71
0.7990	0.4639	992	993	1028	0.06	3.64	874	1848	171.57
0.9142	0.4475	990	987	1003	0.26	1.35	870	1860	173.26
1.0000	0.4364	984	984	984	0.00	0.00	865	1866	174.52
			1,2-dibromoethane + diethyl ketone						
0.0000	0.5877	1197	1197	1197	0.00	0.00	1337	2110	198.63
0.1534	0.5632	1132	1133	1165	0.04	2.93	1084	2073	194.80
0.2249	0.5518	1112	1109	1150	0.28	3.45	1062	2058	193.03
0.3352	0.5348	1076	1078	1127	0.16	4.76	1028	2029	190.32
0.4521	0.5168	1047	1051	1103	0.41	5.31	995	2000	187.47
0.5021	0.5093	1037	1041	1092	0.43	5.29	981	1987	186.27
0.6553	0.4865	1012	1017	1059	0.45	4.65	943	1950	182.60
0.8072	0.4643	994	999	1026	0.46	3.23	907	1912	179.01
0.9269	0.4469	987	989	1000	0.16	1.32	881	1884	176.21
1.0000	0.4364	984	984	984	0.00	0.00	865	1866	174.52

1,2-dibromoethane + methyl propyl ketone									
0.0000	0.5994	1200	1200	1200	0.00	0.00	1149	2129	170.03
0.1532	0.5753	1130	1134	1174	0.29	3.86	1093	2089	170.09
0.2687	0.5572	1091	1095	1153	0.36	5.63	1055	2058	171.43
0.3394	0.5482	1070	1075	1139	0.48	6.41	1033	2039	171.73
0.4445	0.5463	1045	1050	1118	0.51	6.93	1002	2012	172.17
0.5053	0.5206	1034	1038	1105	0.41	6.84	986	1996	172.43
0.6350	0.5004	1012	1017	1076	0.51	6.33	951	1961	172.97
0.8073	0.4736	995	997	1035	0.24	4.02	909	1918	173.70
0.9179	0.4565	986	999	1007	0.27	2.07	883	1887	174.17
1.0000	0.4364	984	984	984	0.00	0.00	865	1866	174.52
1,2-dibromoethane + methyl isobutyl ketone									
0.0000	0.6068	1169	1169	1169	0.00	0.00	1334	2471	1171.08
0.1405	0.5846	1123	1120	1142	0.30	1.69	1258	2390	1242.73
0.2969	0.5593	1078	1076	1112	0.16	3.16	1178	2296	1325.36
0.3235	0.5593	1071	1070	1107	0.11	3.36	1164	2280	1340.28
0.5088	0.5239	1030	1033	1072	0.27	4.09	1075	2166	1444.16
0.5725	0.5130	1018	1023	1061	0.46	4.15	1046	2126	1481.07
0.6813	0.4940	1002	1008	1041	0.62	3.82	997	2059	1545.57
0.8395	0.4658	989	993	1012	0.39	2.33	930	1963	1642.64
0.9306	0.4492	983	987	996	0.40	1.32	892	1907	1700.34
1.0000	0.4364	984	984	984	0.00	0.00	865	1866	174.52

(continued over page)

TABLE IV (*continued*)

		Values of u, m sec^{-1}							
			Theoretical						
x_1	L_f^M Å	Expt.	FLT	CFT	$u\%$ (FLT)	$u\%$ (CFT)	R	W	b (Cm^3 mol^{-1})
			1,2-dibromoethane + cyclopentanone						
0.0000	0.4739	1374	1374	1374	0.00	0.00	996	1877	173.51
0.0979	0.4712	1305	1304	1336	0.09	2.41	977	1876	173.61
0.2116	0.4673	1239	1238	1293	0.12	4.35	957	1875	173.72
0.2438	0.4661	1222	1221	1281	0.08	4.79	952	1874	173.76
0.4445	0.4589	1135	1134	1203	0.13	6.00	923	1872	173.96
0.5421	0.4551	1100	1100	1165	0.05	5.91	911	1871	174.06
0.6065	0.4526	1077	1079	1140	0.19	5.82	903	1869	174.12
0.7630	0.4464	1034	1036	1078	0.16	4.27	886	1868	174.28
0.9137	0.4402	999	1001	1018	0.21	1.94	872	1866	174.43
1.0000	0.4364	984	984	984	0.00	0.00	865	1866	174.52
			1,2-dibromoethane + cyclohexanone						
0.0000	0.4695	1388	1388	1388	0.00	0.00	1168	2199	205.79
0.1310	0.4668	1309	1305	1335	0.30	1.97	1121	2156	201.49
0.2804	0.4629	1229	1227	1274	0.20	3.68	1065	2105	196.67
0.3434	0.4610	1198	1198	1248	0.43	4.24	1049	2083	194.66
0.4915	0.4563	1137	1137	1189	0.03	4.57	1003	2033	189.99
0.6046	0.4524	1096	1096	1143	0.01	4.33	970	1996	186.47
0.6481	0.4508	1081	1082	1126	0.06	4.15	958	1981	185.68
0.7943	0.4451	1034	1037	1067	0.32	3.19	917	1931	180.67
0.9107	0.4403	1004	1006	1020	0.19	1.60	887	1894	177.17
1.0000	0.4364	984	984	984	0.00	0.00	865	1866	174.52

velocity data and those of the FLT and CFT values are given in columns 6 and 7. Values of R, W and b are also given in columns 8–10 of Table IV.

The results included in columns 3–7 of the Table IV show that the FLT sound velocities are in excellent agreement with experimental results in all the systems. However, the sound velocities estimated by CFT are not in good agreement with experimental results.

The data included in columns 8 to 10 of Table IV show that the values of R, W and b exhibit linear relationship with mole fraction in all the systems studied.

(c) Isentropic compressibility

Isentropic compressibilities (k_s) were calculated using the relation

$$k_s = u^{-2}\rho^{-1} \tag{7}$$

where u and ρ denote the sound velocity and density. The values of k_s are accurate to $\pm 2\,\mathrm{TPa}^{-1}$. The deviation in k_s from ideal value, assumed to be additive in terms of volume fraction, is estimated using the equation

$$\Delta k_s = k_s - \varphi_1 k_{s1} - (1 - \varphi_1)k_{s2} \tag{8}$$

where k_s, k_{s1} and k_{s2} are the isentropic compressibilities of the mixture and the pure components respectively and φ_1 is the volume fraction of DBE.

Experimental data for ρ, u, k_s and Δk_s at 303.15 K are included in Table V and also graphically represented in Figure 2. The dependence of Δk_s on volume fraction is expressed by an empirical equation of the form

$$\Delta k_s = \varphi_1(1 - \varphi_1)[b_0 + b_1(2\varphi_1 - 1) + b_2(2\varphi_1 - 1)^2] \tag{9}$$

The values of b_0, b_1 and b_2 calculated by the method of least squares are given in Table VI along with standard deviation $\sigma(\Delta k_s)$.

The results included in Figure 2 show that Δk_s is negative over the volume fraction range studied in the systems of DBE with MEK, DEK, MPK and MIBK. The mixtures of DBE with cyclopentanone and cyclohexanone exhibit small positive values. These are attributed to weak dipolar interactions between unlike molecules which lead to decrease in free length, increase in sound speed and negative deviation in isentropic compressibility. These interactions are expected to decrease with increase in chain length. Hence the increase in Δk_s values from MEK to MPK. In the case of MIBK, values of Δk_s are more negative and this may be probably due to the presence of branched chain. For the systems containing cyclic ketones the small positive Δk_s values can be attributed to the fact that the free length is not affected much.

TABLE V

Volume fractions (φ_1), densities (ρ), sound velocities (u), and the deviation in isentropic compressibilities (Δk_s) of binary liquid mixtures of 1,2-dibromoethane with ketones at 303.15 K.

φ_1	ρ (gcm^{-3})	u (ms^{-1})	K_s (TPa^{-1})	ΔK_s (TPa^{-1})
1,2-dibromoethane + methyl ethyl ketone				
0.0000	0.79445	1170	920	0
0.1540	1.00500	1100	822	−29
0.2766	1.17274	1060	759	−39
0.4061	1.34985	1031	697	−44
0.4819	1.45347	1018	664	−43
0.5560	1.55493	1008	633	−41
0.7320	1.79542	996	562	−34
0.7921	1.87741	992	541	−30
0.9108	2.03900	990	500	−17
1.0000	2.15967	984	479	0
1,2-dibromoethane + diethyl ketone				
0.0000	0.80455	1197	867	0
0.1283	0.97864	1132	797	−20
0.1908	1.06341	1112	762	−30
0.2888	1.19880	1076	721	−33
0.4014	1.34900	1047	676	−35
0.4504	1.41541	1037	657	−35
0.6070	1.62761	1012	600	−32
0.7728	1.85208	994	546	−21
0.9115	2.03988	987	503	−10
1.0000	2.15967	984	479	0
1,2-dibromoethane + methyl propyl ketone				
0.0000	0.79660	1200	872	0
0.1271	0.96961	1130	808	−14
0.2282	1.10727	1091	759	−23
0.2924	1.19480	1070	731	−26
0.3916	1.32980	1045	688	−30
0.4511	1.41075	1034	664	−31
0.5833	1.59095	1012	614	−29
0.7712	1.84733	995	547	−20
0.8999	2.02305	986	508	−10
1.0000	2.15967	984	479	0
1,2-dibromoethane + methyl isobutyl ketone				
0.0000	0.79125	1169	925	0
0.1010	0.92936	1123	853	−27
0.2249	1.09882	1078	784	−41
0.2473	1.12944	1071	772	−43
0.4158	1.35989	1030	694	−46
0.4793	1.44665	1018	667	−44
0.5950	1.60503	1002	621	−39
0.7823	1.86151	989	550	−26
0.9021	2.02551	983	511	−12
1.0000	2.15967	984	479	0

TABLE V (*continued*)

φ_1	(gcm^{-3})	u (ms^{-1})	K_s (TPa^{-1})	ΔK_s (TPa^{-1})
	1,2-dibromoethane + cyclopentanone			
0.0000	0.93898	1374	564	0
0.0953	1.05486	1305	557	1
0.2067	1.19048	1239	547	1
0.2384	1.22912	1222	545	1
0.4373	1.47135	1135	528	1
0.5348	1.59039	1100	520	1
0.5995	1.66932	1077	516	3
0.7576	1.86255	1034	502	2
0.9114	2.05080	999	489	2
1.0000	2.15967	984	479	0
	1,2-dibromoethane + cyclohexanone			
0.0000	0.93757	1388	554	0
0.1113	1.07297	1309	544	−2
0.2446	1.23522	1229	536	0
0.3029	1.30629	1198	533	2
0.4454	1.48008	1137	523	2
0.5596	1.61952	1096	514	2
0.6048	1.67480	1081	511	2
0.7624	1.86775	1034	501	4
0.8945	2.02983	1004	489	2
1.0000	2.15967	984	479	0

TABLE VI

Values of parameters in Eq. (9) and the standard deviation $\sigma(\Delta k_s)$ at 303.15 K.

	b_0	b_1	b_2	$\sigma(\Delta k_s)$
System	TPa^{-1}			
1,2-dibromoethane + methyl ethyl ketone	−169.710	23.478	−81.583	1
1,2-dibromoethane + diethyl ketone	−130.000	39.771	−51.331	2
1,2-dibromoethane + methyl propyl ketone	−123.006	11.640	6.247	0
1,2-dibromo + methyl isobutyl ketone	−172.839	90.900	−67.804	1

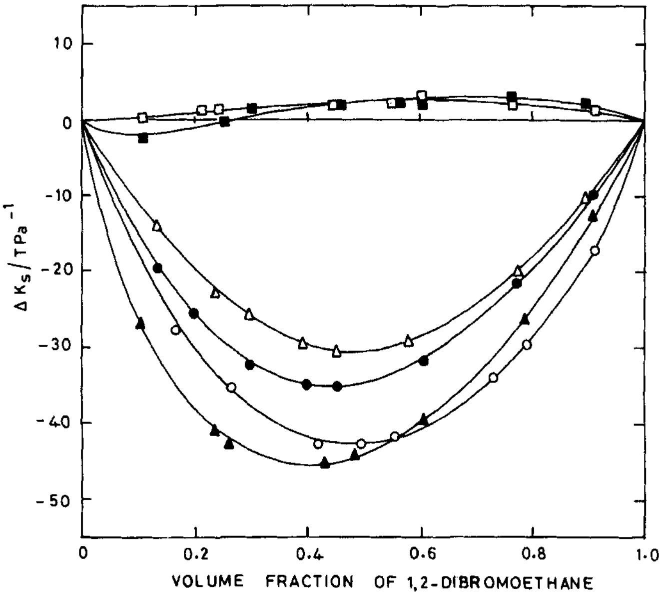

FIGURE 2 The values of Δk_s against volume fraction (φ_1) for the system of 1,2-dibromoethane with: ○, methyl ethyl ketone; ●, diethyl ketone; △, methyl propyl ketone; ▲, methyl isobutyl ketone; □, cyclopentanone; ■, cyclohexanone.

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