

Excess Molar Volumes and Kinematic Viscosities of Binary Liquid Mixtures of Cyclohexanone + Dichloromethane, + Trichloromethane, + 1,2-Dichloroethane, + Trichloroethene, + 1,1,1-Trichloroethane, and + Cyclohexane

Jagan Nath* and Sanjay Kumar Mishra

Chemistry Department, Gorakhpur University, Gorakhpur 273009, India

Excess molar volumes, V_m^E , at $T = 298.15$ K, and kinematic viscosities, ν , at $T = 303.15$ K, have been measured for binary liquid mixtures of cyclohexanone ($C_6H_{10}O$) + dichloromethane (CH_2Cl_2), + trichloromethane ($CHCl_3$), + 1,2-dichloroethane (CH_2ClCH_2Cl), + trichloroethene ($CHClCCl_2$), + 1,1,1-trichloroethane (CCl_3CH_3), and + cyclohexane ($c-C_6H_{12}$). The values of ν and V_m^E have been fitted in appropriate equations using a least-squares method. V_m^E has been found to be negative throughout the entire range of composition for mixtures of $C_6H_{10}O + CH_2Cl_2$, + $CHCl_3$, + $CHClCCl_2$, and + CCl_3CH_3 , and positive for $C_6H_{10}O + c-C_6H_{12}$. For $C_6H_{10}O + CH_2ClCH_2Cl$, V_m^E has been found to be positive at lower mole fractions of $C_6H_{10}O$, and negative at higher mole fractions. The analysis has shown that McAllister's approach correlates ν of the present mixtures throughout the entire composition range to a significantly high degree of accuracy.

Introduction

Mixtures of cyclohexanone ($C_6H_{10}O$) with dichloromethane (CH_2Cl_2), trichloromethane ($CHCl_3$), 1,2-dichloroethane (CH_2ClCH_2Cl), trichloroethene ($CHClCCl_2$), 1,1,1-trichloroethane (CCl_3CH_3), and cyclohexane ($c-C_6H_{12}$) are of particular interest from the viewpoint of electron donor–acceptor interaction leading to the formation of intermolecular adducts between the components in the liquid state. The specific interaction of $C_6H_{10}O$ with the above chloro-compounds can be visualized as being due to the presence of lone-pair electrons on the oxygen atom of $C_6H_{10}O$, which can make this species act as an n-donor toward these chloro-compounds. On the other hand, CH_2Cl_2 , $CHCl_3$, CH_2ClCH_2Cl , $CHClCCl_2$, and CCl_3CH_3 can be involved in the formation of H-bond with, and act as σ -acceptors toward, $C_6H_{10}O$. The system $C_6H_{10}O + c-C_6H_{12}$ can be treated as a reference system since the nonspecific interactions are believed to be present between the components in this case. Although Nath and Dixit (1984a, 1985) have measured excess molar volumes, V_m^E , speeds of sound, u , relative permittivities, ϵ_r , and refractive indexes, n_D , for dimethyl ketone (CH_3COCH_3) + CH_2Cl_2 , + CH_2ClCH_2Cl , + $CHClCCl_2$, and + CCl_2CCl_2 , Nath and Saini (1989, 1990) have measured V_m^E , u , ϵ_r , and n_D for mixtures of methyl-ethyl ketone ($CH_3COC_2H_5$) + CH_2Cl_2 , + CH_2ClCH_2Cl , + $CHClCCl_2$, and + CCl_2CCl_2 , and Chadha and Tripathi (1995) have measured excess molar enthalpies, H_m^E , for mixtures of $CHCl_2CHCl_2$ with cyclopentanone, extensive studies concerning interactions between the components of mixtures of chloroalkanes and chloroalkenes with ketones of more complexity have not been made. Hence, in this work, we have measured V_m^E and kinematic viscosities, ν , of $C_6H_{10}O + CH_2Cl_2$, + $CHCl_3$, + CH_2ClCH_2Cl , + $CHClCCl_2$, + CCl_3CH_3 , and + $c-C_6H_{12}$, and the results of these measurements are reported and interpreted here.

Table 1. Kinematic Viscosities, ν , of Pure Liquids at $T = 303.15$ K

liquid	$\nu/mm^2 \cdot s^{-1}$	
	this work	lit. ^a
CH_2Cl_2	0.301	0.301
CH_2ClCH_2Cl	0.588	0.590
$CHClCCl_2$	0.355	0.356
CCl_3CH_3	0.549	0.549
$CHCl_3$	0.350	0.350
$c-C_6H_{12}$	1.067	1.066
$C_6H_{10}O$	1.925	1.923

^a Values of ν obtained from relation $\nu = \eta/\rho$ using the values of the dynamic viscosities η and densities ρ of the pure liquids available in the literature (Riddick and Bunger, 1970; Timmermans, 1950).

Experimental Section

Materials. Cyclohexanone of AR quality and stated minimum purity of 99.5%, HPLC quality chemicals, dichloromethane and 1,2-dichloroethane, both of stated minimum purity of 99.8% (GLC), and cyclohexane, of stated minimum purity of 99.7% (GLC), and UV spectral grade trichloroethene of minimum purity of 99.5% (GLC) were all obtained from Sisco Research Laboratories, Ltd., Mumbai, India. CH_2Cl_2 , CH_2ClCH_2Cl , $c-C_6H_{12}$, and $CHClCCl_2$ were used without further purification. Cyclohexanone was placed over anhydrous sodium sulfate to remove traces of water and then subjected to fractional distillations. Trichloromethane of AR quality (obtained from Qualigens Fine Chemicals, Ltd., Mumbai, India) was shaken repeatedly with distilled water to remove ethanol present as stabilizer, dried over anhydrous calcium chloride, distilled fractionally, and stored in dark colored bottles. 1,1,1-Trichloroethane (Spectrochem product) of AR quality and stated minimum purity of 99.0% (GLC) was washed with 10% potassium carbonate solution, dried over calcium chloride, and then distilled fractionally.

Table 2. Kinematic Viscosities, ν , of the Various Mixtures of Cyclohexanone at $T = 303.15$ K

x	$\nu/\text{mm}^2\cdot\text{s}^{-1}$	x	$\nu/\text{mm}^2\cdot\text{s}^{-1}$	x	$\nu/\text{mm}^2\cdot\text{s}^{-1}$
$x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CH}_2\text{Cl}_2$					
0.0326	0.325	0.2956	0.564	0.5938	0.980
0.0617	0.350	0.3301	0.596	0.6554	1.090
0.1031	0.380	0.3977	0.682	0.7128	1.198
0.1359	0.411	0.4474	0.747	0.7824	1.355
0.1746	0.442	0.4892	0.811	0.8528	1.520
0.2127	0.477	0.5423	0.887	0.9154	1.685
0.2475	0.510				
$x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CHCl}_3$					
0.0504	0.398	0.3517	0.763	0.6573	1.239
0.0875	0.442	0.3858	0.812	0.7158	1.342
0.1238	0.484	0.4488	0.905	0.7668	1.434
0.1662	0.530	0.5045	0.989	0.8279	1.552
0.2046	0.578	0.5572	1.066	0.8886	1.680
0.2532	0.632	0.6085	1.152	0.9436	1.802
0.2952	0.690				
$x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CH}_2\text{ClCH}_2\text{Cl}$					
0.0415	0.624	0.3282	0.895	0.6511	1.288
0.0731	0.648	0.3802	0.945	0.6990	1.361
0.1164	0.689	0.4438	1.020	0.7535	1.449
0.1576	0.728	0.4921	1.077	0.8192	1.566
0.1964	0.762	0.5430	1.142	0.8754	1.672
0.2392	0.803	0.5943	1.210	0.9410	1.802
0.2880	0.849				
$x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CHClCCl}_2$					
0.0369	0.384	0.3556	0.684	0.6738	1.140
0.0833	0.420	0.3996	0.733	0.7265	1.236
0.1266	0.458	0.4729	0.819	0.7844	1.363
0.1736	0.498	0.5288	0.897	0.8374	1.488
0.2161	0.540	0.5715	0.967	0.8937	1.623
0.2616	0.584	0.6272	1.053	0.9435	1.764
0.3124	0.635				
$x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CCl}_3\text{CH}_3$					
0.0513	0.588	0.3892	0.903	0.7050	1.339
0.0983	0.631	0.4371	0.960	0.7546	1.429
0.1480	0.673	0.4989	1.040	0.8038	1.512
0.2006	0.714	0.5609	1.125	0.8574	1.622
0.2354	0.746	0.6044	1.186	0.9046	1.715
0.2937	0.807	0.6561	1.260	0.9519	1.820
0.3335	0.850				
$x\text{C}_6\text{H}_{10}\text{O} + (1-x)c\text{-C}_6\text{H}_{12}$					
0.0445	1.082	0.3979	1.288	0.7163	1.580
0.1000	1.112	0.4480	1.332	0.7673	1.630
0.1508	1.134	0.5032	1.372	0.8206	1.698
0.2042	1.167	0.5707	1.433	0.8649	1.745
0.2505	1.196	0.6184	1.475	0.9158	1.816
0.3004	1.225	0.6704	1.534	0.9624	1.876
0.3459	1.252				

Methods. (i) Excess molar volumes, V_m^E , were measured with an imprecision of the order of ± 0.002 $\text{cm}^3\cdot\text{mol}^{-1}$, using a two-limbed Pyrex glass dilatometer that was similar to that used in earlier measurements (Nath and Chaudhary, 1992; Nath and Pandey, 1997a). Known amounts of the two liquid components were confined separately over mercury in the absence of air spaces in the two limbs of the dilatometer, which (mounted on a stand) was immersed in water of a thermostat (controlled to ± 0.01 K). The mixing of the components was achieved by rocking the cell back and forth through a definite angle, and the mercury levels in the capillary of the dilatometer were noted with a cathetometer that had an accuracy of ± 0.001 cm.

(ii) The kinematic viscosities, ν , of the various pure liquids and binary mixtures were measured at (303.15 ± 0.01) K by using the kinematic viscometer described by Taun and Fuoss (1963) and by following the procedure described by Nath and Dubey (1981), Nath and Dixit (1984b), and Nath and Pandey (1997b). The measured values of ν of the pure liquid CH_2Cl_2 , $\text{CH}_2\text{ClCH}_2\text{Cl}$, CHClCCl_2 , CCl_3CH_3 , CHCl_3 , $c\text{-C}_6\text{H}_{12}$, and $\text{C}_6\text{H}_{10}\text{O}$ have been compared with the available data in Table 1, which shows that the present values of ν for the various liquids

agree with the literature values within a deviation of the order of ± 0.002 $\text{mm}^2\cdot\text{s}^{-1}$.

Results and Discussion

The values of ν of binary mixtures of $\text{C}_6\text{H}_{10}\text{O}$ with CH_2Cl_2 , CHCl_3 , $\text{CH}_2\text{ClCH}_2\text{Cl}$, CHClCCl_2 , CCl_3CH_3 , and $c\text{-C}_6\text{H}_{12}$ at $T = 303.15$ K are given in Table 2, where x refers to the mole fraction of $\text{C}_6\text{H}_{10}\text{O}$ in the mixture. x has an uncertainty of ± 0.0001 . The values of V_m^E of the present mixtures of $\text{C}_6\text{H}_{10}\text{O}$ at $T = 298.15$ K are reported in Table 3. The values of ν for the various mixtures of $\text{C}_6\text{H}_{10}\text{O}$ have been plotted against x in Figure 1, whereas the values of V_m^E have been plotted against x in Figure 2. The values of ν and V_m^E for the present mixtures have been fitted by the method of least-squares to the equation

$$Y = a \sum_{k=0}^n B_k z^k \quad (1)$$

using $a = 1$, $z = x$ when $Y = \nu$ and $a = x(1-x)$, $z = (2x-1)$ when $Y = V_m^E$. The values of the parameters B_k of eq 1, along with the standard deviations, δ , are given in Table 4.

Table 3. Experimental Values of the Excess Molar Volumes, V_m^E , for $C_6H_{10}O + CH_2Cl_2$, $+ CHCl_3$, $+ CH_2ClCH_2Cl$, $+ CHClCCl_2$, $+ CCl_3CH_3$, and $+ c-C_6H_{12}$ at $T = 298.15$ K

x	$V_m^E/cm^3 \cdot mol^{-1}$	x	$V_m^E/cm^3 \cdot mol^{-1}$	x	$V_m^E/cm^3 \cdot mol^{-1}$	x	$V_m^E/cm^3 \cdot mol^{-1}$
$x C_6H_{10}O + (1-x) CH_2Cl_2$							
0.0342	-0.013	0.2107	-0.068	0.3996	-0.092	0.6405	-0.070
0.0726	-0.022	0.2133	-0.069	0.4605	-0.091	0.7216	-0.055
0.1045	-0.031	0.2366	-0.076	0.4750	-0.090	0.7680	-0.043
0.1413	-0.048	0.3186	-0.087	0.5354	-0.087	0.8402	-0.030
0.1672	-0.053	0.3595	-0.094	0.5878	-0.080	0.8912	-0.021
$x C_6H_{10}O + (1-x) CHCl_3$							
0.0481	-0.130	0.3623	-0.473	0.5876	-0.385	0.8845	-0.115
0.0913	-0.220	0.4022	-0.471	0.6545	-0.334	0.9433	-0.052
0.1967	-0.380	0.4440	-0.460	0.7143	-0.278		
0.2802	-0.449	0.5109	-0.436	0.7593	-0.237		
0.3226	-0.466	0.5585	-0.407	0.8218	-0.176		
$x C_6H_{10}O + (1-x) CH_2ClCH_2Cl$							
0.0521	0.009	0.2464	0.046	0.4513	0.043	0.8113	-0.019
0.0872	0.016	0.2729	0.047	0.5533	0.027	0.8555	-0.021
0.1317	0.027	0.3211	0.052	0.6013	0.017	0.9188	-0.022
0.1627	0.032	0.3623	0.049	0.6444	0.013		
0.2012	0.039	0.4017	0.047	0.7541	-0.008		
$x C_6H_{10}O + (1-x) CHClCCl_2$							
0.0588	-0.088	0.2701	-0.251	0.5349	-0.235	0.7778	-0.126
0.0952	-0.138	0.3044	-0.261	0.5727	-0.222	0.8321	-0.094
0.1428	-0.181	0.3394	-0.270	0.6454	-0.195	0.8788	-0.068
0.1604	-0.195	0.4404	-0.262	0.6848	-0.171	0.9282	-0.040
0.2114	-0.228	0.4943	-0.250	0.7277	-0.151		
$x C_6H_{10}O + (1-x) CCl_3CH_3$							
0.0574	-0.173	0.3325	-0.460	0.6161	-0.457	0.8983	-0.180
0.1114	-0.277	0.4219	-0.473	0.6593	-0.438	0.9205	-0.142
0.1897	-0.378	0.4606	-0.475	0.7129	-0.401		
0.2458	-0.420	0.5167	-0.476	0.7674	-0.351		
0.2871	-0.443	0.5696	-0.472	0.8035	-0.315		
$x C_6H_{10}O + (1-x) c-C_6H_{12}$							
0.0660	0.062	0.2949	0.162	0.5377	0.127	0.7665	0.056
0.1172	0.098	0.3451	0.160	0.5806	0.112	0.8911	0.020
0.1675	0.128	0.3919	0.159	0.6297	0.097	0.9504	0.007
0.1944	0.138	0.4360	0.152	0.6683	0.085		
0.2519	0.156	0.4810	0.142	0.7078	0.072		

Table 4. Values of the Parameters, B_k , of Eq 1 and the Standard Deviations, δ , for the Various Mixtures

property	T/K	B_0	B_1	B_2	B_3	δ
$x C_6H_{10}O + (1-x) CH_2Cl_2$						
$V_m^E/cm^3 \cdot mol^{-1}$	298.15	-0.356 17	0.172 49	0.086 89	-0.125 18	0.0022
$\nu/mm^2 \cdot s^{-1}$	303.15	0.302 82	0.701 83	0.446 46	0.474 92	0.0025
$x C_6H_{10}O + (1-x) CHCl_3$						
$V_m^E/cm^3 \cdot mol^{-1}$	298.15	-1.760 86	0.952 32	-0.162 56	0.072 48	0.0022
$\nu/mm^2 \cdot s^{-1}$	303.15	0.348 07	1.033 68	0.367 22	0.176 53	0.0025
$x C_6H_{10}O + (1-x) CH_2ClCH_2Cl$						
$V_m^E/cm^3 \cdot mol^{-1}$	298.15	0.157 23	-0.299 71	-0.271 26	0.019 22	0.0023
$\nu/mm^2 \cdot s^{-1}$	303.15	0.587 39	0.860 19	0.071 06	0.408 79	0.0020
$x C_6H_{10}O + (1-x) CHClCCl_2$						
$V_m^E/cm^3 \cdot mol^{-1}$	298.15	-0.991 96	0.566 93	-0.147 54	0.019 46	0.0020
$\nu/mm^2 \cdot s^{-1}$	303.15	0.353 23	0.825 59	-0.001 56	0.750 12	0.0028
$x C_6H_{10}O + (1-x) CCl_3CH_3$						
$V_m^E/cm^3 \cdot mol^{-1}$	298.15	-1.911 68	0.071 72	-0.812 11	0.807 65	0.0023
$\nu/mm^2 \cdot s^{-1}$	303.15	0.550 07	0.761 40	0.263 24	0.352 65	0.0029
$x C_6H_{10}O + (1-x) c-C_6H_{12}$						
$V_m^E/cm^3 \cdot mol^{-1}$	298.15	0.552 35	-0.522 93	0.049 31	0.052 16	0.0015
$\nu/mm^2 \cdot s^{-1}$	303.15	1.066 09	0.406 53	0.365 50	0.088 84	0.0029

Table 5. Values of the Parameters ν_{AB} and ν_{BA} of Eq 2, the Standard Deviations, $\delta(\nu)$, Average Percent Deviations $(\Delta\%)_{av}$, and the Maximum Percent Deviations, $(\Delta\%)_{max}$, for the Various Mixtures at $T = 303.15$ K

mixture	$\nu_{AB}/mm^2 \cdot s^{-1}$	$\nu_{BA}/mm^2 \cdot s^{-1}$	$\delta(\nu)/mm^2 \cdot s^{-1}$	$(\Delta\%)_{av}$	$(\Delta\%)_{max}$
$x C_6H_{10}O + (1-x) CH_2Cl_2$	1.1164	0.6579	0.0059	0.59	1.68
$x C_6H_{10}O + (1-x) CHCl_3$	1.2734	0.8687	0.0051	0.49	1.03
$x C_6H_{10}O + (1-x) CH_2ClCH_2Cl$	1.3003	0.9338	0.0042	0.30	0.63
$x C_6H_{10}O + (1-x) CHClCCl_2$	1.1139	0.7098	0.0072	0.54	1.71
$x C_6H_{10}O + (1-x) CCl_3CH_3$	1.2888	0.8673	0.0044	0.34	0.88
$x C_6H_{10}O + (1-x) c-C_6H_{12}$	1.5241	1.2023	0.0034	0.19	0.50

The viscosity data of binary liquid mixtures can be analyzed in light of the various theories of liquid-mixture viscosity. The various theories for prediction of liquid-

mixture viscosities have been discussed by Reid et al. (1977). According to McAllister's approach (McAllister, 1960), the kinematic viscosity, ν , of a binary mixture of

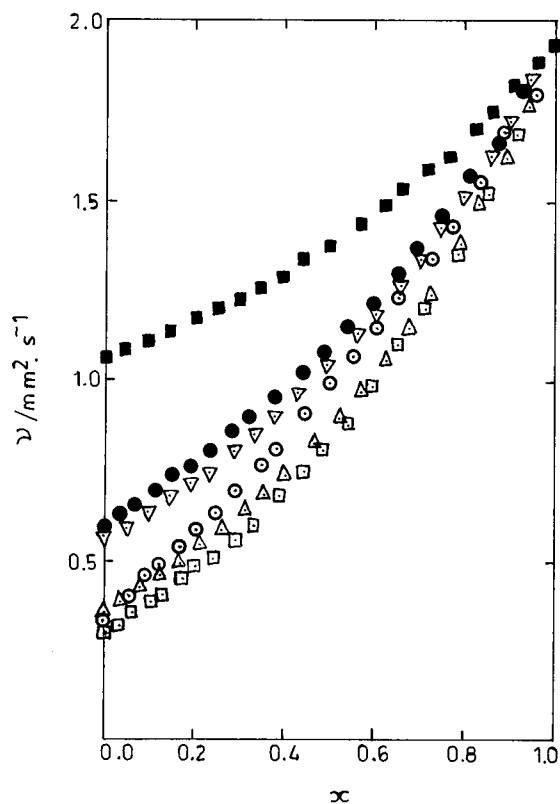


Figure 1. Plot of ν versus x for the various mixtures at $T = 303.15$ K: \square , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CH}_2\text{Cl}_2$; \circ , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CHCl}_3$; \bullet , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CH}_2\text{ClCH}_2\text{Cl}$; \triangle , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CHClCCl}_2$; ∇ , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CCl}_3\text{CH}_3$; \blacksquare , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)c\text{-C}_6\text{H}_{12}$.

components A and B is given by the relation

$$\ln \nu = x_A^3 \ln \nu_A + 3x_A^2 x_B \ln \nu_{AB} + 3x_A x_B^2 \ln \nu_{BA} + x_B^3 \ln \nu_B + R^\circ \quad (2)$$

where

$$R^\circ = x_B^3 \ln \frac{M_B}{M_A} + 3x_A x_B^2 \ln \frac{1 + 2M_B/M_A}{3} + 3x_A^2 x_B \ln \frac{2 + M_B/M_A}{3} - \ln(x_A + x_B M_B/M_A) \quad (3)$$

In eq 2, x_A and x_B are the mole fractions of the components A and B in the mixture, and ν_A and ν_B are the kinematic viscosities of the two pure liquids. ν_{AB} and ν_{BA} are the two undetermined parameters that are characteristic of a system. In eq 3, M_A and M_B are the molecular weights of the components A and B, respectively. Taking A for $\text{C}_6\text{H}_{10}\text{O}$ and B for the other component in the various mixtures, the values of ν_{AB} and ν_{BA} were determined from eq 2, using the viscosity data of the present mixtures. The values of ν_{AB} and ν_{BA} , along with those of the standard deviations, $\delta(\nu)$, are given in Table 5. The values of the average percent deviations, $(\Delta\%)_{av}$, and those of the maximum percent deviations, $(\Delta\%)_{max}$, of the experimental values of kinematic viscosities from those estimated for the various mixtures from eq 2 are given in Table 5, which shows that McAllister's eq 2 correlates the liquid-mixture viscosity to a significantly high degree of accuracy for the present mixtures.

The data show (see Table 3) that V_m^E is negative throughout the entire range of x for mixtures of $\text{C}_6\text{H}_{10}\text{O} + \text{CH}_2\text{Cl}_2$, $+ \text{CHCl}_3$, $+ \text{CHClCCl}_2$, and $+ \text{CCl}_3\text{CH}_3$, and

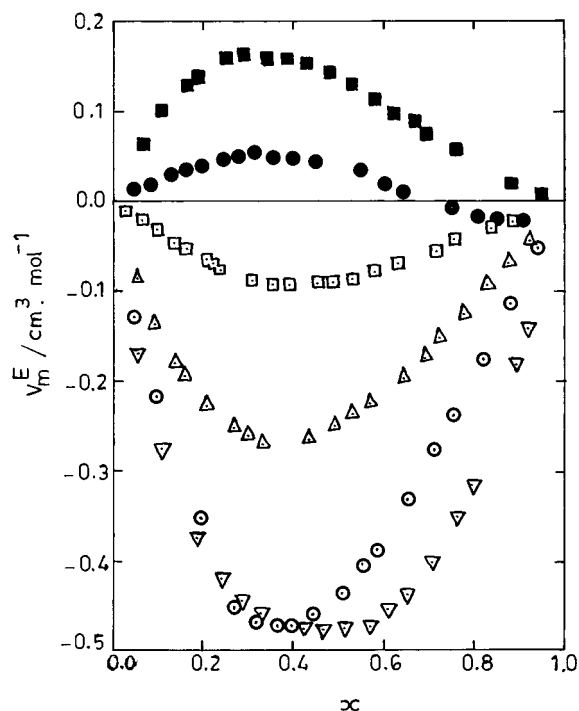
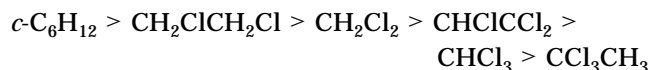


Figure 2. Plot of V_m^E versus x for the various mixtures at $T = 298.15$ K: \square , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CH}_2\text{Cl}_2$; \circ , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CHCl}_3$; \bullet , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CH}_2\text{ClCH}_2\text{Cl}$; \triangle , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CHClCCl}_2$; ∇ , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)\text{CCl}_3\text{CH}_3$; \blacksquare , $x\text{C}_6\text{H}_{10}\text{O} + (1-x)c\text{-C}_6\text{H}_{12}$.

positive for $\text{C}_6\text{H}_{10}\text{O} + c\text{-C}_6\text{H}_{12}$. For $\text{C}_6\text{H}_{10}\text{O} + \text{CH}_2\text{ClCH}_2\text{Cl}$, V_m^E is positive at lower values of x and negative at its higher values. At $x = 0.5$, V_m^E for the various systems of C_6H_{10} has the sequence



The negative values of V_m^E for mixtures of $\text{C}_6\text{H}_{10}\text{O} + \text{CH}_2\text{Cl}_2$, $+ \text{CHCl}_3$, $+ \text{CHClCCl}_2$, and $+ \text{CCl}_3\text{CH}_3$ can be visualized as being due to a closer approach of the unlike molecules in solution, thus indicating the existence of specific interaction between the components of the various mixtures in the liquid state.

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