

Densities and Viscosities of Binary Mixtures of Some Halohydrocarbons with 2-Methyl-1-propanol at 298.15 and 313.15 K

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Densities and viscosities have been measured for the binary mixtures of chlorobenzene, chlorocyclohexane, bromobenzene, and bromocyclohexane with 2-methyl-1-propanol over the entire concentration range at the temperatures 298.15 K and 313.15 K. These values have been used in the determination of the excess volumes, V^E , and the viscosity deviations, $\Delta\eta$. All mixtures present negative values of V^E in a wide region corresponding to the high concentration in alkanol, although this zone becomes smaller at 313.15 K. For $\Delta\eta$, the mixtures show negative values over the whole composition range at both temperatures.

Introduction

This paper is a continuation of the systematic study of thermophysical properties undertaken for our research group, concerning mixtures of halohydrocarbons with isomers of butanol. In this work we present V^E and $\Delta\eta$ obtained for mixtures of 2-methyl-1-propanol and a halohydrocarbon at 298.15 K and 313.15 K. These results illustrate how the self-association of the alcohol is influenced by the halogenated (chloro, bromo) derivatives of benzene and cyclohexane. We also compare the results with those published for mixtures of the same halohydrocarbons with 1-butanol (Artigas et al., 1996a) and 2-butanol (Artigas et al., 1997).

Experimental Section

Materials. The liquids used in this work were chlorobenzene, bromobenzene, 2-methyl-1-propanol (with purity better than 99.5 mol %), bromocyclohexane, and chlorocyclohexane (better than 99.0%). The purity of the liquids was verified by gas chromatography using a semi-capillary methyl silicone column (o.d. 530 μm) and a flame-ionization detector. The chemicals were kept over a molecular sieve and used without further purification.

Methods. Densities of the pure solvents and mixtures were measured by means of a vibrating tube densimeter, in which temperature is controlled automatically at ± 0.01 K. The apparatus was calibrated using deionized doubly distilled water and dry air. The accuracy of density measurements was $\pm 1 \times 10^{-5} \text{ g}\cdot\text{cm}^{-3}$, and the corresponding precision $\pm 3 \times 10^{-5} \text{ g}\cdot\text{cm}^{-3}$.

For the viscosity measurements, an automatic measuring unit has been used together with an Ubbelohde viscosimeter. The constant of the viscosimeter was determined using water as standard liquid. The Hagenbach's correction was taken into account. The temperature was kept constant within ± 0.01 K. The uncertainty of the flow time measurements was ± 0.01 s. At least four flow time measurements (greatest difference allowed 0.05%) were performed for each composition and temperature, and the re-

Table 1. Densities ρ , and Viscosities η , of the Pure Compounds at 298.15 K

compound	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\eta/\text{mPa}\cdot\text{s}$	
	exptl	lit	exptl	lit
chlorobenzene	1.101 00	1.100 9 ^a 1.101 1 ^e	0.7519	—
chlorocyclohexane	0.993 23	0.992 34 ^b	1.5600	1.5807 ^b
bromobenzene	1.488 18	1.488 20 ^a 1.488 58 ^b	1.0715	1.0743 ^c 1.081 ^d
bromocyclohexane	1.327 96	1.324 64 ^h	2.0441	2.2239 ^h
2-methyl-1-propanol	0.797 77	0.797 8 ^a 0.797 8 ^f	3.3328	3.333 ^a 3.338 ^c 3.3330 ^g

^a Riddick et al., 1986. ^b From TRC Source, 1996. ^c Viswanath and Natarajan, 1989. ^d Manjeshwar and Aminabhavi, 1988; from TRC Source 1996. ^e TRC d-7330. ^f TRC d-5030. ^g TRC c-5030. ^h Friend and Hargreaves, 1944; from TRC Source 1996.

sults were averaged. Kinetic energy corrections were applied to the experimental data of kinematic viscosity. From density, ρ , and kinematic viscosity, ν , the absolute viscosity, η , can be obtained by using the following equation:

$$\eta = \rho\nu \quad (1)$$

The estimated uncertainty in the absolute viscosity is $\pm 1 \times 10^{-4} \text{ mPa}\cdot\text{s}$.

The composition of each mixture (molar fraction) was determined by using a mass balance. The possible error in the mole fractions is estimated to be less than ± 0.0001 .

The experimental values of density and viscosity obtained for the pure components and the values taken from the literature are listed in Table 1.

Results and Discussion

The properties have been calculated from our measurements according to the relations

$$V^E = x_1M_1(\rho^{-1} - \rho_1^{-1}) + x_2M_2(\rho^{-1} - \rho_2^{-1}) \quad (2)$$

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \quad (3)$$

where M_i , ρ_i , η_i , x_i , and V_i designate the molecular weight,

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Table 2. Densities and Excess Volumes of Binary Mixtures Halohydrocarbon (1) + 2-Methyl-1-propanol (2)

x_1	$\rho/\text{g}\cdot\text{cm}^{-3}$	$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	x_1	$\rho/\text{g}\cdot\text{cm}^{-3}$	$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	x_1	$\rho/\text{g}\cdot\text{cm}^{-3}$	$V^E/\text{cm}^3\cdot\text{mol}^{-1}$
Chlorobenzene + 2-Methyl-1-propanol at 298.15 K								
0.1058	0.833 57	-0.0949	0.4062	0.928 65	-0.0661	0.7053	1.017 20	0.0349
0.2018	0.864 79	-0.1094	0.5027	0.957 81	-0.0355	0.8025	1.044 93	0.0632
0.3043	0.897 20	-0.0944	0.6005	0.986 79	-0.0039	0.8975	1.071 79	0.0663
Chlorocyclohexane + 2-Methyl-1-propanol at 298.15 K								
0.1047	0.823 71	-0.0443	0.4056	0.889 10	0.0026	0.7014	0.943 86	0.0892
0.2056	0.846 97	-0.0448	0.5042	0.908 26	0.0319	0.7923	0.959 25	0.1040
0.3078	0.869 07	-0.0234	0.6092	0.927 64	0.0625	0.9104	0.978 62	0.0843
Bromobenzene + 2-Methyl-1-propanol at 298.15 K								
0.1000	0.875 97	-0.0871	0.4059	1.100 48	-0.0932	0.7057	1.302 58	0.0170
0.2080	0.957 59	-0.1177	0.5046	1.168 83	-0.0614	0.8037	1.365 39	0.0438
0.3067	1.029 88	-0.1164	0.6064	1.237 34	-0.0196	0.9010	1.426 67	0.0452
Bromocyclohexane + 2-Methyl-1-propanol at 298.15 K								
0.0997	0.866 02	-0.0501	0.4050	1.049 38	-0.0169	0.6998	1.198 08	0.0574
0.2106	0.936 69	-0.0542	0.5045	1.102 32	0.0067	0.7975	1.242 46	0.0757
0.3127	0.997 54	-0.0383	0.6030	1.151 87	0.0322	0.8938	1.284 31	0.0707
Chlorobenzene + 2-Methyl-1-propanol at 313.15 K								
0.1063	0.820 80	-0.0457	0.4106	0.915 28	0.0261	0.6986	0.999 39	0.1182
0.2078	0.853 15	-0.0429	0.5065	0.943 77	0.0626	0.8028	1.029 01	0.1208
0.3051	0.883 31	-0.0139	0.6081	0.973 41	0.0958	0.9021	1.057 05	0.0879
Chlorocyclohexane + 2-Methyl-1-propanol at 313.15 K								
0.1079	0.811 78	-0.0059	0.4005	0.874 15	0.0883	0.6952	0.928 15	0.1750
0.2100	0.834 75	0.0172	0.5016	0.893 51	0.1248	0.7911	0.944 33	0.1767
0.3105	0.856 04	0.0539	0.6022	0.911 88	0.1562	0.8998	0.962 20	0.1317
Bromobenzene + 2-Methyl-1-propanol at 313.15 K								
0.1024	0.864 36	-0.0450	0.4040	1.082 41	-0.0165	0.7016	1.280 70	0.0901
0.2033	0.939 36	-0.0498	0.5000	1.148 05	0.0150	0.8020	1.344 34	0.1169
0.3047	1.012 63	-0.0378	0.6034	1.216 92	0.0531	0.9065	1.409 68	0.0963
Bromocyclohexane + 2-Methyl-1-propanol at 313.15 K								
0.1101	0.859 54	-0.0109	0.4068	1.034 41	0.0720	0.6971	1.179 11	0.1493
0.2086	0.921 11	0.0103	0.5070	1.086 91	0.1064	0.7955	1.223 56	0.1503
0.3082	0.979 67	0.0419	0.6079	1.136 96	0.1334	0.9020	1.269 61	0.1115

density, absolute viscosity, molar fraction, and molar volume of component i in the mixture, respectively. Subscript 1 corresponds to the halocompounds and 2 to 2-methyl-1-propanol. Symbols without subscript refer to the mixture.

The experimental values of density and excess volume are gathered in Table 2, while viscosities and viscosity deviations for the mixtures are presented in Table 3 at their corresponding temperatures. The excess volumes and the viscosity deviations are graphically represented in Figures 1–4.

The values of these properties at each temperature were correlated with a Redlich–Kister polynomial equation

$$V^E \text{ (or } \Delta\eta) = x_1(1 - x_1)\sum A_i(2x_1 - 1)^i \quad (4)$$

where A_i are adjustable parameters determined by the least-squares method. Table 4 lists the values of the parameters A_i together with the standard deviations σ .

All V^E curves are sigmoidal-shape, and negative values appear mainly in the region rich in 2-methyl-1-propanol. When temperature increases, the excess volume becomes greater, but all curves present negative values for the highest alkanol concentrations. The viscosity leads to negative values for $\Delta\eta$ over the entire composition range for all systems at both temperatures. The signs obtained for the different properties are characteristic of systems in which dispersive forces prevail, mainly the breaking of the self-association by hydrogen bonding of alkanol, like other authors have previously stated (Fort and Moore, 1966; Nigam and Mahl, 1971).

The binary systems in which one compound is associated by hydrogen bond show characteristic features in the values of the thermodynamic properties, the excess enthalpies (Brown et al., 1969; Fernández et al., 1989), excess

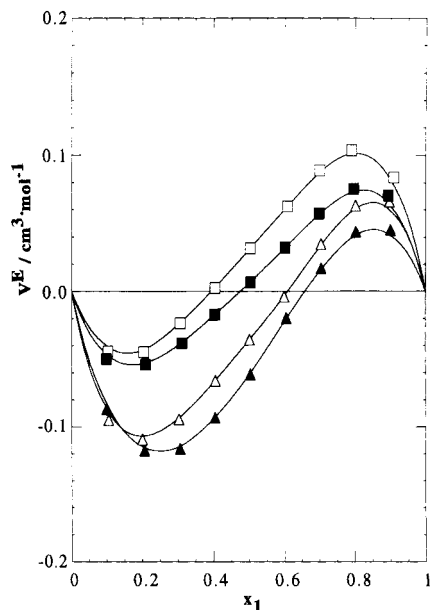
volumes (Nath and Pandey, 1997), and excess Gibbs energies (Oracz, 1986) being clearly positive. 2-Methyl-1-propanol is a liquid associated by hydrogen bonding and in the pure state exhibits an equilibrium between cyclic and linear multimers and monomeric species (Davis et al., 1960; Rytting et al., 1978). In the pure state, halogenated compounds present weak dipole–dipole interactions.

When mixed, the halogenated solvents can interact with the OH group (Letcher et al., 1993; Letcher and Nevine, 1994). The aromatic derivatives set up an interaction between the π electronic cloud and the hydroxyl group (Stokes, 1982). All interactions described are of minor intensity compared with hydrogen bonding, but they can lead to the formation of intermolecular complexes (Prigogine, 1957). This phenomenon explains why mixtures of butanols with benzene (Rodríguez et al., 1993) or cyclohexane (Artigas et al., 1995) show excess volumes considerably higher than those with chloro- or bromobenzene and chloro- or bromocyclohexane, respectively. Finally, the contribution due to structural effects must not be underestimated and is taken into account through the free volume.

The sign of each cited contribution depends on the studied property. With respect to the excess volume, the phenomena that yield the formation of molecular associates and the free volume are negative contributions. In the other hand, the breaking of associated species (like those existing in the pure liquids) leads to positive contributions to V^E . When the viscosity is studied, the interactions that make the flow easier (i.e., the breaking of the intermolecular associates, because the motion of individual species is less difficult than that of great molecular complexes) are negative contributions to $\Delta\eta$, while the interstitial arrangement and interactions that produce the formation of

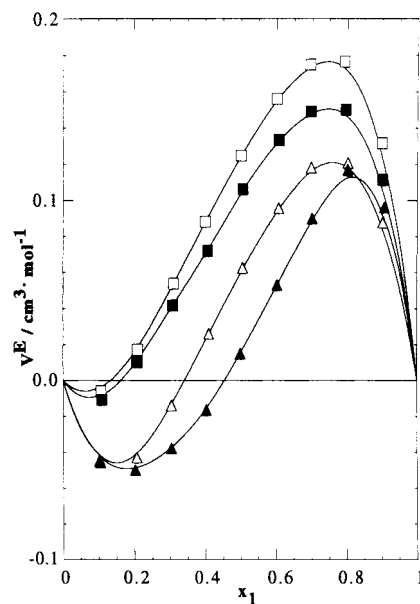
Table 3. Viscosities and Viscosity Deviations of Binary Mixtures Halohydrocarbon (1) + 2-Methyl-1-propanol (2)

x_1	$\eta/\text{mPa}\cdot\text{s}$	$\Delta\eta/\text{mPa}\cdot\text{s}$	x_1	$\eta/\text{mPa}\cdot\text{s}$	$\Delta\eta/\text{mPa}\cdot\text{s}$	x_1	$\eta/\text{mPa}\cdot\text{s}$	$\Delta\eta/\text{mPa}\cdot\text{s}$
Chlorobenzene + 2-Methyl-1-propanol at 298.15 K								
0.1010	2.5578	-0.5145	0.4019	1.3007	-0.9951	0.6965	0.8803	-0.6549
0.2015	1.9752	-0.8377	0.4952	1.1241	-0.9307	0.8009	0.8094	-0.4564
0.2966	1.5934	-0.9740	0.5979	0.9772	-0.8125	0.8977	0.7666	-0.2494
Chlorocyclohexane + 2-Methyl-1-propanol at 298.15 K								
0.1018	2.7714	-0.3811	0.4073	1.8347	-0.7761	0.7053	1.5253	-0.5571
0.2015	2.3424	-0.6332	0.5034	1.6874	-0.7531	0.8003	1.5002	-0.4139
0.3066	2.0349	-0.7544	0.5996	1.5971	-0.6729	0.8949	1.4998	-0.2465
Bromobenzene + 2-Methyl-1-propanol at 298.15 K								
0.1065	2.6494	-0.4427	0.4015	1.5322	-0.8928	0.7006	1.1441	-0.6043
0.1936	2.2049	-0.6903	0.5016	1.3596	-0.8391	0.7967	1.0882	-0.4430
0.2949	1.8307	-0.8353	0.6014	1.2308	-0.7421	0.8995	1.0548	-0.2440
Bromocyclohexane + 2-Methyl-1-propanol at 298.15 K								
0.0998	2.8708	-0.3336	0.3991	2.0947	-0.7245	0.7011	1.8783	-0.5521
0.2024	2.4960	-0.5764	0.4983	1.9815	-0.7100	0.8014	1.8766	-0.4246
0.3028	2.2524	-0.6908	0.5946	1.9152	-0.6523	0.9015	1.9152	-0.2572
Chlorobenzene + 2-Methyl-1-propanol at 313.15 K								
0.1010	1.6627	-0.2996	0.4019	0.9531	-0.5635	0.6965	0.7035	-0.3766
0.2015	1.3360	-0.4774	0.4952	0.8497	-0.5286	0.8009	0.6586	-0.2667
0.2966	1.1276	-0.5449	0.5979	0.7614	-0.4647	0.8977	0.6330	-0.1490
Chlorocyclohexane + 2-Methyl-1-propanol at 313.15 K								
0.1018	1.7837	-0.2363	0.4073	1.2844	-0.4594	0.7053	1.1460	-0.3285
0.2015	1.5553	-0.3746	0.5034	1.2228	-0.4342	0.8003	1.1316	-0.2571
0.3066	1.3927	-0.4421	0.5996	1.1810	-0.3891	0.8949	1.1438	-0.1593
Bromobenzene + 2-Methyl-1-propanol at 313.15 K								
0.1065	1.7269	-0.2548	0.4015	1.1163	-0.5048	0.7006	0.9062	-0.3492
0.1936	1.4828	-0.3925	0.5016	1.0219	-0.4768	0.7967	0.8769	-0.2610
0.2949	1.2805	-0.4709	0.6014	0.9534	-0.4233	0.8995	0.8637	-0.1485
Bromocyclohexane + 2-Methyl-1-propanol at 313.15 K								
0.0998	1.8460	-0.2131	0.3991	1.4705	-0.4301	0.7011	1.3988	-0.3419
0.2024	1.6560	-0.3488	0.4983	1.4204	-0.4277	0.8014	1.4185	-0.2691
0.3028	1.5392	-0.4125	0.5946	1.4008	-0.3963	0.9015	1.4647	-0.1699

**Figure 1.** Excess volumes for halohydrocarbon (1) + 2-methyl-1-propanol (2) at $T = 298.15$ K as a function of mole fraction x_1 : chlorobenzene (Δ); chlorocyclohexane (\square); bromobenzene (\blacktriangle); bromocyclohexane (\blacksquare).

intermolecular associates hinder the flow, so the sign of their contribution to viscosity properties is positive.

Comparing the results reported here with those obtained for the same halohydrocarbons with the linear isomers of 2-methyl-1-propanol, namely, 1-butanol (Artigas et al., 1996a) and 2-butanol (Artigas et al., 1997), it can be pointed out that V^E is very similar to that obtained in the case of 1-butanol considering not only the values but also the po-

**Figure 2.** Excess volumes for halohydrocarbon (1) + 2-methyl-1-propanol (2) at $T = 313.15$ K as a function of mole fraction x_1 : chlorobenzene (Δ); chlorocyclohexane (\square); bromobenzene (\blacktriangle); bromocyclohexane (\blacksquare).

sition of maxima and minima. Again, the systems with aromatic derivatives present excess volumes smaller than those with cyclohexane derivatives. The only difference with mixtures involving 1-butanol is that the mixtures of 2-methyl-1-propanol with brominated compounds show V^E slightly smaller than mixtures with the chlorinated ones.

These results indicate that the breaking of the associated structure of both primary alcohols acts as the most im-

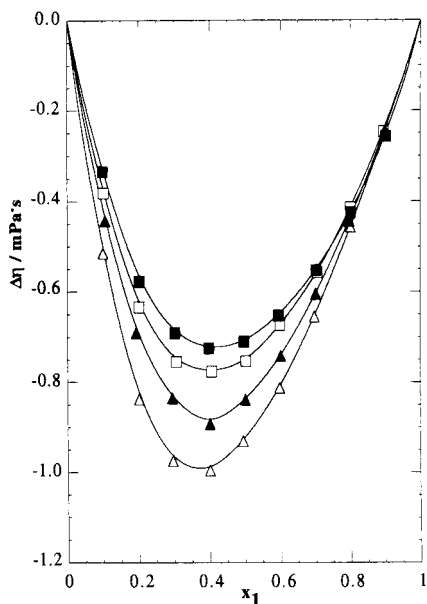


Figure 3. Viscosity deviations for halohydrocarbon (1) + 2-methyl-1-propanol (2) at $T = 298.15$ K as a function of mole fraction x_1 : chlorobenzene (Δ); chlorocyclohexane (\square); bromobenzene (\blacktriangle); bromocyclohexane (\blacksquare).

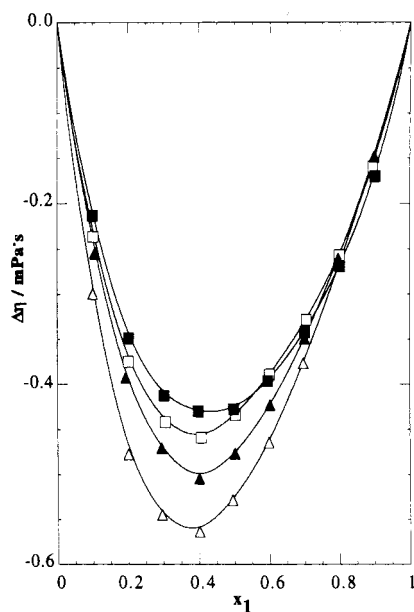


Figure 4. Viscosity deviations for halohydrocarbon (1) + 2-methyl-1-propanol (2) at $T = 313.15$ K as a function of mole fraction x_1 : chlorobenzene (Δ); chlorocyclohexane (\square); bromobenzene (\blacktriangle); bromocyclohexane (\blacksquare).

portant factor in the V^E values. The difference between the two alcohol isomers is that the hydrocarbon chain of 2-methyl-1-propanol is more highly branched, so the monomeric species can be better accommodated than in the completely linear 1-butanol. Thus, the excess volumes are minor for the mixtures containing 2-methyl-1-propanol.

When the results of the viscosimetric study are examined, it can be observed that the negative values of $\Delta\eta$ obtained with the four halohydrocarbons and 2-methyl-1-propanol are very similar to those obtained in mixtures involving 2-butanol. For viscosity and related properties, the branching of the molecules seems to be more important than the OH position because the similar molecular

Table 4. Coefficients, A_i , and Standard Deviations, σ , for Equation 4

function	A_0	A_1	A_2	A_3	σ
Chlorobenzene + 2-Methyl-1-propanol at 298.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-0.1484	0.6460	0.0026	0.7043	0.0006
$\Delta\eta/\text{mPa}\cdot\text{s}$	-3.7271	1.9873	-0.8112	-0.1698	0.0056
Chlorocyclohexane + 2-Methyl-1-propanol at 298.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	0.1196	0.5956	0.1953	0.5132	0.0009
$\Delta\eta/\text{mPa}\cdot\text{s}$	-3.0088	1.1550	-0.6681	-0.2234	0.0060
Bromobenzene + 2-Methyl-1-propanol at 298.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	-0.2479	0.7415	0.0364	0.2713	0.0010
$\Delta\eta/\text{mPa}\cdot\text{s}$	-3.3756	1.5081	-0.4955	-0.4080	0.0050
Bromocyclohexane + 2-Methyl-1-propanol at 298.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	0.0218	0.5005	0.1229	0.5094	0.0005
$\Delta\eta/\text{mPa}\cdot\text{s}$	-2.8340	0.8979	-0.7639	-0.5290	0.0049
Chlorobenzene + 2-Methyl-1-propanol at 313.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	0.2410	0.7501	0.0053	0.2906	0.0009
$\Delta\eta/\text{mPa}\cdot\text{s}$	-2.1098	1.0444	-0.5626	0.0378	0.0029
Chlorocyclohexane + 2-Methyl-1-propanol at 313.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	0.4944	0.6806	0.2874	0.4394	0.0012
$\Delta\eta/\text{mPa}\cdot\text{s}$	-1.7412	0.6946	-0.6295	-0.2353	0.0021
Bromobenzene + 2-Methyl-1-propanol at 313.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	0.0574	0.6917	0.4028	0.4840	0.0010
$\Delta\eta/\text{mPa}\cdot\text{s}$	-1.9117	0.8033	-0.3873	-0.2199	0.0034
Bromocyclohexane + 2-Methyl-1-propanol at 313.15 K					
$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	0.4128	0.5989	0.2267	0.4059	0.0013
$\Delta\eta/\text{mPa}\cdot\text{s}$	-1.6945	0.4390	-0.6642	-0.1933	0.0036

structures of 2-butanol and 2-methyl-1-propanol (both branched in C_2) lead to $\Delta\eta$ of the same order.

Mixtures with chlorinated derivatives present negative values of $\Delta\eta$ lower than those with brominated compounds. This could be explained as due to the greater size of brominated derivatives, which allows a better interstitial accommodation than chlorinated derivatives do, so the flow would become more difficult for the systems containing bromobenzene and bromocyclohexane.

When the temperature rises, the self-association of 2-methyl-1-propanol decreases owing to thermal agitation, and it is more difficult to establish the specific interactions. The major effect is the decrease of self-association, so the V^E and $\Delta\eta$ values increase, maintaining the sequence obtained at 298.15 K. This behavior has been also observed in other systems involving haloalkanes and butanols (Artigas et al., 1996b; Lafuente et al., 1996).

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