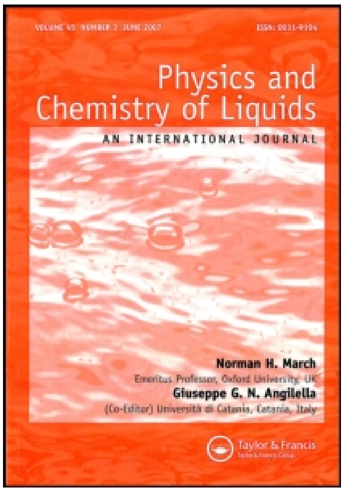

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EXCESS VOLUMES AND EXCESS VISCOSITIES OF MIXTURES HALOHYDROCARBON + 2-BUTANOL AT 298.15 AND 313.15 K

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From density and viscosity measurements excess volumes, V^E, excess viscosities, η^E, and excess free energies of activation for viscous flow, G^{*E}, have been determined at 298.15 and 313.15 K for the binary mixtures of a halogenated (Cl, Br) benzene or cyclohexane with 2-butanol. The results show the influence of both the type of cyclic system and the halogen atom over the self-association of the 2-butanol. V^E presents predominant positive values, only showing negative ones with aromatic derivatives at 298.15 K in a small zone. For η^E and G^{*E}, all mixtures present negative values over the whole composition range at two temperatures.

Keywords: Binary mixtures; 2-butanol; halohydrocarbon; excess properties

INTRODUCTION

Continuing a line of work intended to systematically study the excess properties of binary mixtures of halocompounds with alkanols, our research group has recently reported the data corresponding to systems involving halohydrocarbons and 1-butanol, [1, 2]. Now we present the results for V^E, η^E and G^{*E} obtained for the same halohydrocarbons with another isomer of butanol, 2-butanol, at 298.15 K and 313.15 K in order to show the different effect on the

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association of a secondary alcohol compared with a primary one of both the change from cyclic alkane to the analogous aromatic and the substitution from a halogen atom to another.

EXPERIMENTAL

The liquids used were: chlorobenzene, bromobenzene (both better than 99.5 mol %), and bromocyclohexane (better than 99.0%) provided by Fluka, chlorocyclohexane and 2-butanol (both better than 99.0%) supplied by Aldrich. The purity of chemicals used was checked by GLC and was considered sufficient, so no further purification was attempted. The pure components were stored over activated molecular sieve.

Table I shows the experimental values of density and viscosity for the pure compounds at 298.15 K in comparison with published values [3, 4] The densities of the pure components and mixtures were measured by means of an Anton-Paar DMA-58 vibrating tube densimeter automatically thermostated at ±0.01 K. The viscosity measurements were obtained with an Ubbelhode viscosimeter and a Schott-Geräte automatic measuring unity model AVS-440. The molar fractions of the mixtures were determined by using a Mettler H20T balance. Details of the procedure have been described previously [5].

RESULTS AND DISCUSSION

The excess properties were calculated from our measurements according to the following equations:

TABLE I Densities ρ, and viscosities η, of pure compounds at $T = 298.15$ K compared with literature data [3, 4]

Component	$\rho/g{\cdot}cm^{-3}$		η/cP	
	Lit. [3]	*This paper*	*Lit.*	*This paper*
Chlorobenzene	1.1009	1.10100	0.7530[4]	0.7519
Chlorocyclohexane	-	0.99323	-	1.5600
Bromobenzene	1.48820	1.48818	1.074[4]	1.0715
Bromocyclohexane	-	1.32796	-	2.0441
2-Butanol	0.8026	0.80241	2.998[3]	2.9973

$$V^E = x_I M_1(\rho^{-1} - \rho_1^{-1}) + x_2 M_2(\rho^{-1} - \rho_2^{-1}) \tag{1}$$

$$\eta^E = \eta - (x_1 \eta_1 + x_2 \eta_2) \tag{2}$$

$$G^{*E} = RT\,[\ln \eta V - (x_1 \ln \eta_1 V_1 + x_2 \ln \eta_2 V_2)] \tag{3}$$

where M_i, ρ_i, η_i, x_i and V_i designate molecular weight ($g{\cdot}mol^{-1}$), density ($g{\cdot}cm^{-3}$), absolute viscosity (cP), molar fraction and molar volume ($cm^3{\cdot}mol^{-1}$) of component i in the mixture respectively. Subscript 1 is used for halocompounds; 2 for 2-butanol. Symbols without subscript refer to the mixture.

The experimental values of excess functions for the mixtures at their correspondent temperatures are presented in Tables II–IV, and are graphically shown in Figures 1–6.

The values of the excess properties at each temperature were correlated with a Redlich-Kister polynomial equation by least squares method:

$$Y^E = x_1(1 - x_1)\sum a_i(2x_1 - 1)^i \tag{4}$$

where a_i are adjustable parameters. Table V lists the values of the parameters a_i and the standard deviations $\sigma(Y^E)$.

When examining the excess volume data the main characteristic observed is that values for the mixtures containing aromatic derivatives are clearly smaller than those corresponding to the halocyclohexanes. Although less marked it is also noticeable the lower values obtained for brominated compounds as they are contrasted with the corresponding chlorinated ones. Therefore, at 298.15 K, the curves for chloro- and bromobenzene show slightly negative values in the zone very rich in 2-butanol ($x_I = 0.1$) while the remaining mixtures have positive values of excess volumes at this temperature, being similar the results for chloro- and bromocyclohexane. At 313.15 K, V^E increases and all the systems show positive values in the whole composition range. For both temperatures, the maximum values follow the sequence:

bromobenzene < chlorobenzene < bromocyclohexane < chlorocyclohexane.

TABLE II Excess volumes V^E of binary mixtures halohydrocarbon (1) + 2-butanol (2) at indicated temperature

x_1	$V^E/cm^3{\cdot}mol^{-1}$	x_1	$V^E/cm^3{\cdot}mol^{-1}$	x_1	$V^E/cm^3{\cdot}mol^{-1}$
Chlorobenzene + 2-butanol at 298.15 K					
0.1052	−0.0151	0.4084	0.1200	0.7110	0.1872
0.2035	0.0178	0.4998	0.1515	0.7976	0.1810
0.3064	0.0707	0.6025	0.1784	0.8940	0.1353
Chlorocyclohexane + 2-butanol at 298.15 K					
0.1073	0.0763	0.4095	0.2990	0.6980	0.3236
0.2033	0.1675	0.5104	0.3302	0.7998	0.2862
0.3045	0.2435	0.6086	0.3360	0.9016	0.1982
Bromobenzene + 2-butanol at 298.15 K					
0.1054	−0.0111	0.4001	0.0911	0.6973	0.1563
0.2031	0.0176	0.4974	0.1227	0.7971	0.1452
0.3057	0.0560	0.6006	0.1433	0.8944	0.1080
Bromocyclohexane + 2-butanol at 298.15 K					
0.1024	0.0930	0.4083	0.2628	0.6991	0.3045
0.2086	0.1762	0.5000	0.2843	0.7955	0.2859
0.3112	0.2269	0.6018	0.3029	0.9028	0.2017
Chlorobenzene + 2-butanol at 313.15 K					
0.1068	0.0453	0.4057	0.2208	0.7071	0.2771
0.2042	0.1082	0.5051	0.2521	0.8151	0.2486
0.3013	0.1734	0.6043	0.2738	0.9044	0.1803
Chlorocyclohexane + 2-butanol at 313.15 K					
0.1000	0.1243	0.4105	0.3981	0.6975	0.4222
0.2096	0.2470	0.5019	0.4309	0.7913	0.3669
0.3042	0.3375	0.6019	0.4429	0.8989	0.2414
Bromobenzene + 2-butanol at 313.15 K					
0.1037	0.0453	0.4023	0.1777	0.6971	0.2355
0.2015	0.0943	0.5035	0.2069	0.8020	0.2237
0.3068	0.1403	0.5987	0.2238	0.8960	0.1639
Bromocyclohexane + 2-butanol at 313.15 K					
0.1029	0.1332	0.4050	0.3775	0.6995	0.3805
0.2081	0.2486	0.5014	0.3953	0.7954	0.3412
0.3117	0.3312	0.5881	0.4004	0.8929	0.2405

The maxima for the mixtures containing aromatic derivatives appear when the mole fraction is 0.7 while they are placed in mole fractions richer in alkanol ($x_1 = 0.6$) in the case of cyclohexane

TABLE III Excess viscosities η^E of binary mixtures halohydrocarbon (1) + 2-butanol (2) at indicated temperature

x_1	η^E/cP	x_1	η^E/cP	x_1	η^E/cP
Chlorobenzene + 2-butanol at 298.15 K					
0.1016	−0.6659	0.4055	−0.9997	0.6957	−0.6210
0.2016	−0.9528	0.5007	−0.9124	0.8001	−0.4295
0.3006	−1.0354	0.5972	−0.7811	0.8995	−0.2278
Chlorocyclohexane + 2-butanol at 298.15 K					
0.1063	−0.5970	0.4033	−0.9073	0.7044	−0.6102
0.1999	−0.8274	0.5027	−0.8477	0.7991	−0.4530
0.3075	−0.9113	0.6064	−0.7398	0.8982	−0.2586
Bromobenzene + 2-butanol at 298.15 K					
0.1021	−0.5899	0.4008	−0.9311	0.7001	−0.5862
0.2042	−0.8681	0.4975	−0.8549	0.8033	−0.4136
0.2975	−0.9477	0.5995	−0.7338	0.8997	−0.2323
Bromocyclohexane + 2-butanol at 298.15 K					
0.1007	−0.5391	0.3966	−0.8953	0.6999	−0.6362
0.1983	−0.7896	0.4985	−0.8464	0.8021	−0.4818
0.3022	−0.8899	0.5941	−0.7611	0.8972	−0.3026
Chlorobenzene + 2-butanol at 313.15 K					
0.1016	−0.3296	0.4055	−0.5073	0.6957	−0.3261
0.2016	−0.4730	0.5007	−0.4671	0.8001	−0.2303
0.3006	−0.5190	0.5972	−0.4053	0.8995	−0.1257
Chlorocyclohexane + 2-butanol at 313.15 K					
0.1063	−0.3047	0.4033	−0.4789	0.7044	−0.3322
0.1999	−0.4290	0.5027	−0.4549	0.7991	−0.2490
0.3075	−0.4765	0.6064	−0.4021	0.8982	−0.1411
Bromobenzene + 2-butanol at 313.15 K					
0.1021	−0.2876	0.4008	−0.4666	0.7001	−0.3098
0.2042	−0.4247	0.4975	−0.4348	0.8033	−0.2257
0.2975	−0.4689	0.5995	−0.3797	0.8997	−0.1314
Bromocyclohexane + 2-butanol at 313.15 K					
0.1007	−0.2657	0.3966	−0.4808	0.6999	−0.3689
0.1983	−0.4064	0.4985	−0.4644	0.8021	−0.2910
0.3022	−0.4691	0.5941	−0.4273	0.8972	−0.1906

derivatives. Changes in temperatures does not shift these maximum points.

The results obtained for the properties related to the viscosity (i.e., η^E and G^{*E}) are negative throughout at both temperatures. The excess

TABLE IV Excess energies of activation for viscous flow G^{*E} of binary mixtures halohydrocarbon (1) + 2-butanol (2) at indicated temperature

x_1	$G^{*E}/J{\cdot}mol^{-1}$	x_1	$G^{*E}/J{\cdot}mol^{-1}$	x_1	$G^{*E}/J{\cdot}mol^{-1}$
Chlorobenzene + 2-butanol at 298.15 K					
0.1016	−528.8	0.4055	−1117.1	0.6957	−837.9
0.2016	−874.6	0.5007	−1096.6	0.8001	−615.7
0.3006	−1060.2	0.5972	−996.2	0.8995	−347.2
Chlorocyclohexane + 2-butanol at 298.15 K					
0.1063	−531.1	0.4033	−1018.6	0.7044	−768.1
0.1999	−811.1	0.5027	−997.4	0.7991	−582.1
0.3075	−966.4	0.6064	−903.5	0.8982	−338.7
Bromobenzene + 2-butanol at 298.15 K					
0.1021	−492.2	0.4008	−1051.4	0.7001	−776.3
0.2042	−828.7	0.4975	−1024.0	0.8033	−576.3
0.2975	−989.8	0.5995	−926.9	0.8997	−343.3
Bromocyclohexane + 2-butanol at 298.15 K					
0.1007	−482.4	0.3966	−963.9	0.6999	−723.2
0.1983	−770.4	0.4985	−936.4	0.8021	−550.1
0.3022	−923.1	0.5941	−855.6	0.8972	−345.9
Chlorobenzene + 2-butanol at 313.15 K					
0.1016	−473.3	0.4055	−952.4	0.6957	−713.6
0.2016	−760.0	0.5007	−928.1	0.8001	−532.1
0.3006	−908.4	0.5972	−847.2	0.8995	−307.3
Chlorocyclohexane + 2-butanol at 313.15 K					
0.1063	−477.8	0.4033	−884.6	0.7044	−648.2
0.1999	−724.1	0.5027	−864.5	0.7991	−487.4
0.3075	−849.1	0.6064	−777.9	0.8982	−274.9
Bromobenzene + 2-butanol at 313.15 K					
0.1021	−433.6	0.4008	−880.7	0.7001	−657.6
0.2042	−707.7	0.4975	−857.6	0.8033	−497.4
0.2975	−835.6	0.5995	−778.5	0.8997	−300.9
Bromocyclohexane + 2-butanol at 313.15 K					
0.1007	−410.2	0.3966	−824.5	0.6999	−628.7
0.1983	−664.4	0.4985	−800.7	0.8021	−488.8
0.3022	−792.9	0.5941	−734.5	0.8972	−313.9

viscosity, η^E, shows similar values for the mixtures with the four halogenated derivatives although the results are smaller in the case of aromatic compounds, particularly with chlorobenzene. It seems not to have difference between chlorinated and brominated derivatives. When temperatures rises, the property increases and becomes less

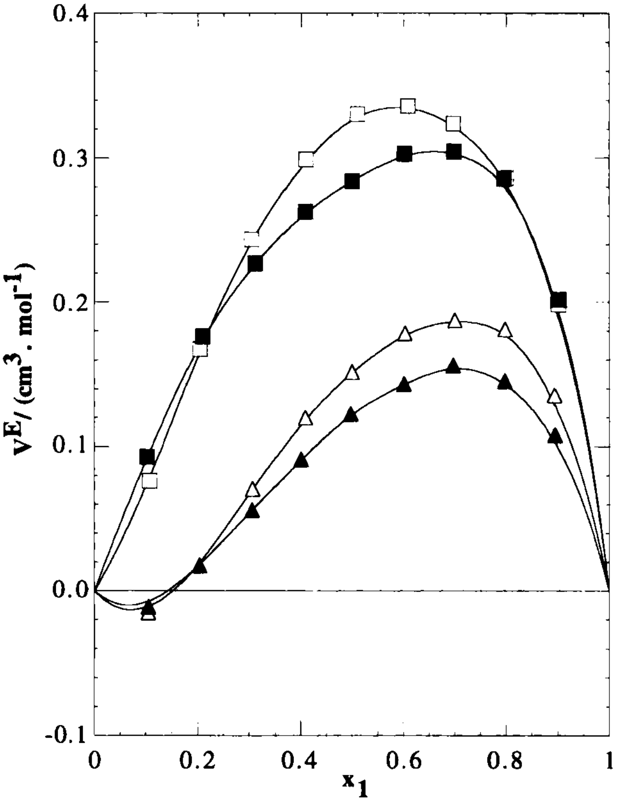

FIGURE 1 Excess volumes, V^E, of halohydrocarbon (1) + 2-butanol (2) at 298.15 K as a function of mole fraction x_l: chlorobenzene (△); chlorocyclohexane (□); bromobenzene (▲); bromocyclohexane (■).

negative in such a way that the minima at 313.15 K are one half of the values obtained at 298.15 K. These minima are placed in mole fractions of 0.3 in the case of the aromatic compounds and of 0.35 in the mixtures with halocyclohexanes.

Referring to free energies of activation for viscous flow (G^{*E}) the smallest values appear also for the mixtures containing the benzenic derivatives. In this property is noticeable a rather more negative value for chlorinated compounds with respect to the brominated ones. As temperature rises, G^{*E} increases but not in a great rate. The order for the minima is

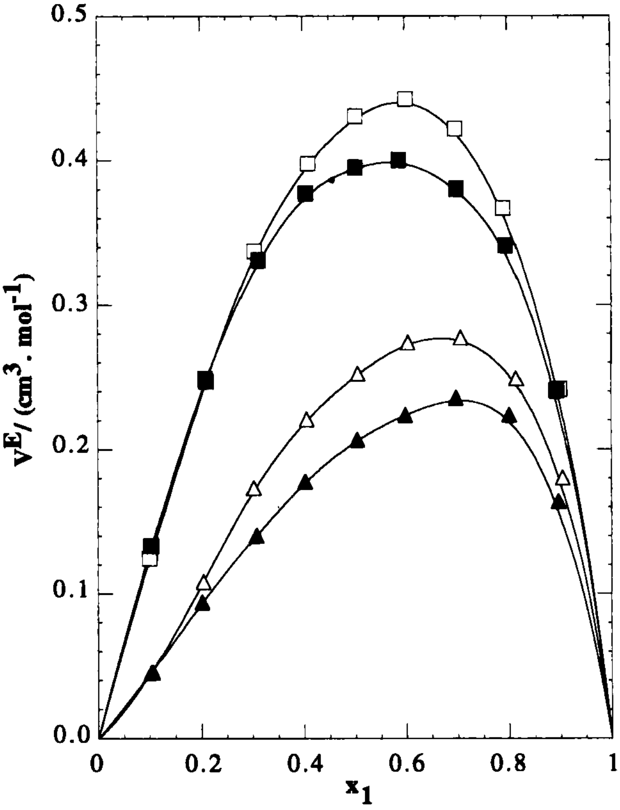

FIGURE 2 Excess volumes, V^E, of halohydrocarbon (1) + 2-butanol (2) at 313.15 K as a function of mole fraction x_l: chlorobenzene (△); chlorocyclohexane (□); bromobenzene (▲); bromocyclohexane (■).

chlorobenzene < bromobenzene ≈ chlorocyclohexane < bromocyclohexane.

The curves corresponding to bromobenzene and chlorocyclohexane superpose each other. The minima are obtained in $x_l = 0.4$ for all systems at both temperatures.

Comparing the results here reported with those obtained when the alkanol isomer is 1-butanol [1] it can be observed that 2-butanol leads to more extreme values: the excess volumes are remarkably more positive and excess viscosities and free energies of activation for viscous flow more negative with the secondary alcohol. For both

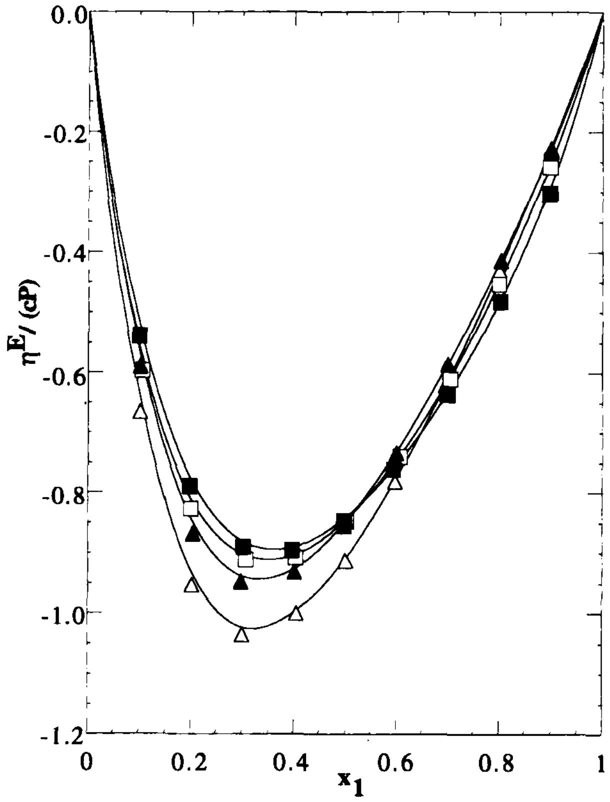

FIGURE 3 Excess viscosities, η^E, of halohydrocarbon (1) + 2-butanol (2) at 298.15 K as a function of mole fraction x_1: chlorobenzene (△); chlorocyclohexane (□); bromobenzene (▲); bromocyclohexane (■).

butanols, the extreme values appear in the regions of high mole fractions in alkanol, but they shift towards richer zones in alcoholic component when 2-butanol is considered.

As happened when 1-butanol was considered, the value of excess properties for the mixtures containing 2-butanol is the sum of both structural effects and intermolecular interactions which go with the mixing process. As some authors have pointed out [6 – 8], the signs for the properties studied are characteristic of systems in which the dispersion forces are predominant. Now is even clearer than in the case of 1-butanol that the prevailing factor is the breaking of the hydrogen

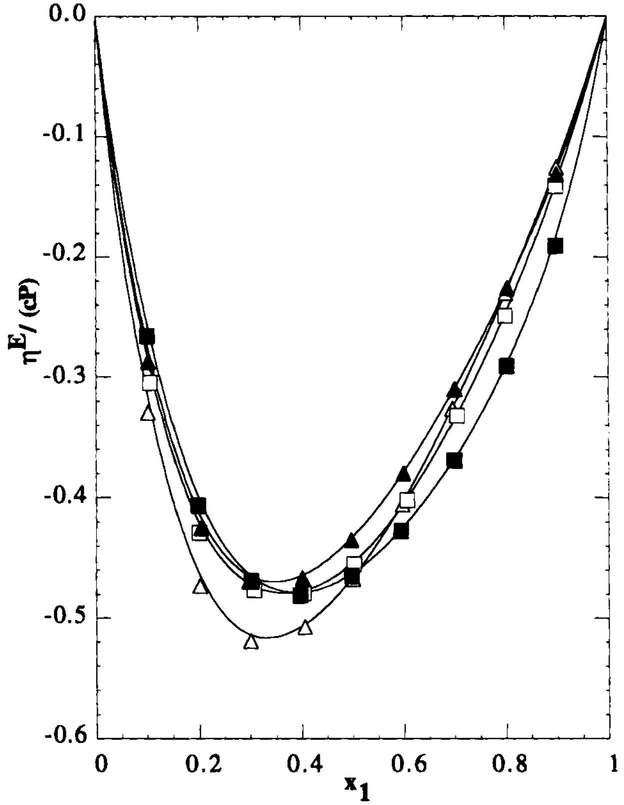

FIGURE 4 Excess viscosities, η^E, of halohydrocarbon (1) + 2-butanol (2) at 313.15 K as a function of mole fraction x_l: chlorobenzene (Δ); chlorocyclohexane (□); bromobenzene (▲); bromocyclohexane (■).

bond association of 2-butanol whose structure of secondary alkanol is more affected by the presence of the halogenated derivatives. This leads to a greater extent of the breaking for 2-butanol than for 1-butanol. The same reason would explain that similar breaking effects in 1-butanol and 2-butanol require smaller amounts of the other component for the secondary alkanol [9–12]. This gives rise to maxima and minima whose mole fractions are richer in 2-butanol than they were in 1-butanol when they both are mixed with the same breaker compounds. On the other hand, the structure of 2-butanol hinders the

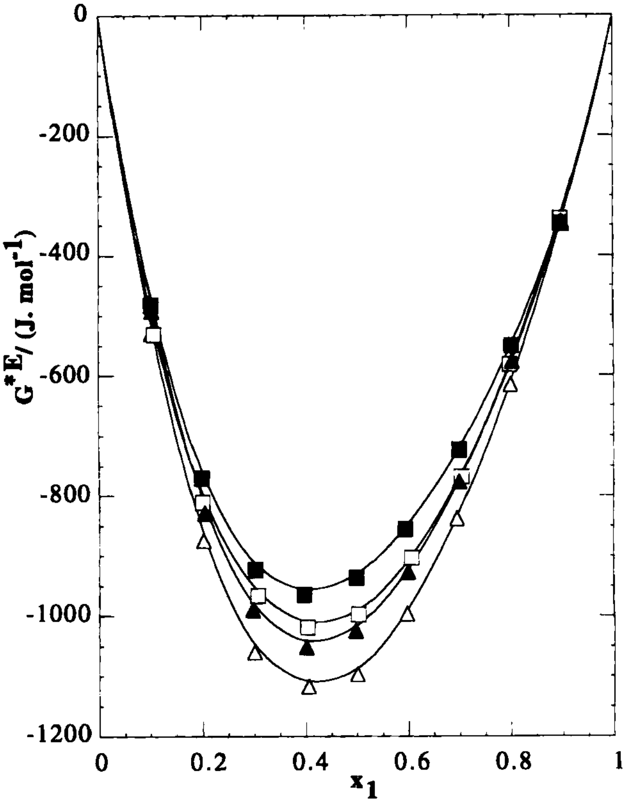

FIGURE 5 Excess energies of activation for viscous flow, G^{*E}, of halohydrocarbon (1) + 2-butanol (2) at 298.15 K as a function of mole fraction x_1: chlorobenzene (△); chlorocyclohexane (□); bromobenzene (▲); bromocyclohexane (■).

interactions OH-halogen atom, interactions that take place in a lower scale than they took with primary alkanol. All of these factors yield the values here reported.

Letcher and Nevines [13] obtained a similar behaviour in mixtures of chlorobenzene with 1-propanol and 2-propanol: the system with 2-propanol (secondary alkanol) presents more positive values with extreme values richer in alkanol than those with primary isomer. They also indicated that chloro-group has a significant effect on lowering the volume excess values.

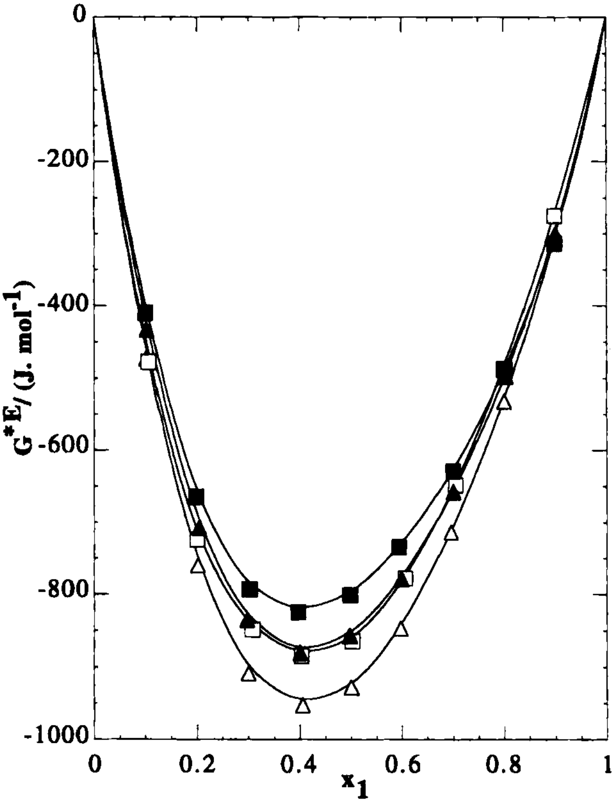

FIGURE 6 Excess energies of activation for viscous flow, G^{*E}, of halohydrocarbon (1) + 2-butanol (2) at 313.15 K as a function of mole fraction x_i: chlorobenzene (△); chlorocyclohexane (□); bromobenzene (▲); bromocyclohexane (■).

At sight of our results, it can be proposed that although the presence of halogen atoms causes a reduction in V^E, the main origin of low values for our systems ought be charged to the aromatic character of some components due to the reasons exposed below. As it has been pointed out, two conclusions are extracted from the curves of excess volume: the aromatic compounds show lower values than the corresponding halocyclohexanes and the brominated derivatives smaller values than the chlorinated ones. The plane structure of benzenic derivatives and the possibility of forming π-OH [14] and X-

TABLE V Coefficients a_i of equation (4) and standard deviations $\sigma(Y^E)$ determined by the least squares method

Function	a_0	a_1	a_2	a_3	σ
Chlorobenzene + 2-butanol at 298.15 K					
$V^E/\mathrm{cm^3\cdot mol^{-1}}$	0.6075	0.6172	0.0276	0.6366	0.0015
η^E/cP	-3.6318	2.3365	-1.9349	1.0396	0.0046
$G^{*E}/\mathrm{J\cdot mol^{-1}}$	-4376.8	1382.3	-721.9	-210.7	4.1
Chlorocyclohexane + 2-butanol at 298.15 K					
$V^E/\mathrm{cm^3\cdot mol^{-1}}$	1.3040	0.3584	0.2898	0.8248	0.0026
η^E/cP	-3.3721	1.6128	-1.7994	0.9421	0.0069
$G^{*E}/\mathrm{J\cdot mol^{-1}}$	-3977.7	1094.9	-1039.7	207.4	4.3
Bromobenzene + 2-butanol at 298.15 K					
$V^E/\mathrm{cm^3\cdot mol^{-1}}$	0.4904	0.5317	0.0387	0.4150	0.0015
η^E/cP	-3.3969	2.0660	-1.7004	0.5935	0.0035
$G^{*E}/\mathrm{J\cdot mol^{-1}}$	-4088.5	1387.0	-793.0	-574.5	3.8
Bromocyclohexane + 2-butanol at 298.15 K					
$V^E/\mathrm{cm^3\cdot mol^{-1}}$	1.1367	0.3897	0.7849	0.6087	0.0021
η^E/cP	-3.3569	1.4518	-1.8529	0.3647	0.0070
$G^{*E}/\mathrm{J\cdot mol^{-1}}$	-3728.7	1246.3	-1215.2	-368.3	4.2
Chlorobenzene + 2-butanol at 313.15 K					
$V^E/\mathrm{cm^3\cdot mol^{-1}}$	1.0077	0.5123	0.3680	0.7554	0.0021
η^E/cP	-1.8598	1.0879	-0.9597	0.4771	0.0029
$G^{*E}/\mathrm{J\cdot mol^{-1}}$	-3706.6	1191.4	-901.3	-890.3	2.7
Chlorocyclohexane + 2-butanol at 313.15 K					
$V^E/\mathrm{cm^3\cdot mol^{-1}}$	1.7234	0.4491	0.4358	0.5368	0.0028
η^E/cP	-1.8074	0.7541	-0.8716	0.4996	0.0034
$G^{*E}/\mathrm{J\cdot mol^{-1}}$	-3442.0	1067.3	-914.2	396.2	4.8
Bromobenzene + 2-butanol at 313.15 K					
$V^E/\mathrm{cm^3\cdot mol^{-1}}$	0.8170	0.4973	0.4830	0.4966	0.0015
η^E/cP	-1.7263	0.9112	-0.8639	0.2376	0.0022
$G^{*E}/\mathrm{J\cdot mol^{-1}}$	-3417.2	1128.9	-949.2	-361.1	2.6
Bromocyclohexane + 2-butanol at 313.15 K					
$V^E/\mathrm{cm^3\cdot mol^{-1}}$	1.5838	0.2247	0.6284	0.7501	0.0091
η^E/cP	-1.8450	0.6105	-0.9864	-0.0834	0.0031
$G^{*E}/\mathrm{J\cdot mol^{-1}}$	-3191.5	1058.6	-1206.6	-508.1	3.5

OH (X = Cl, Br) bonds decrease the excess volumes for the mixtures containing these compounds with respect to the ones for the mixtures with halogenated derivatives of cyclohexane, whose more open and dynamic spatial arrangement provokes a greater breaking and hinders the formation of X-OH (X = Cl or Br) bonds. In the same way can be justified the lesser values for the brominated compounds if it is taken into account that although the greater size of bromine induces a larger breaking in the association of 2-butanol [11], the molecular size of brominated derivatives allows a better interstitial accommodation than that of chlorinated derivatives do.

A lower value for η^E is observed in the mixture with chlorobenzene, result that can be explained by means of the same arguments used in the case of V^E: the plane structures allow an easier flow (with a minor friction) and the same can be stated for a chlorinated derivative with respect to the brominated one, due to the smaller size of the first. The fact that the curves corresponding to chlorocyclohexane and bromobenzene are so similar indicates that the mentioned effects are of the same order and they balanced each other.

When temperature increases, the self association of 2-butanol diminishes and besides it is more difficult that the specific interactions involved in the mixing process take place. Therefore, V^E values go up and η^E and G^{*E} values become less negative because the mixtures flow easier. Similar behaviour has been observed for other mixtures concerning haloalkanes and butanols [11, 12].

Anyway, the resemblance of all of the curves for the different properties points out that the major effect is the breaking of the alkanol association being the other minor effects responsible for the slight differences observed.

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