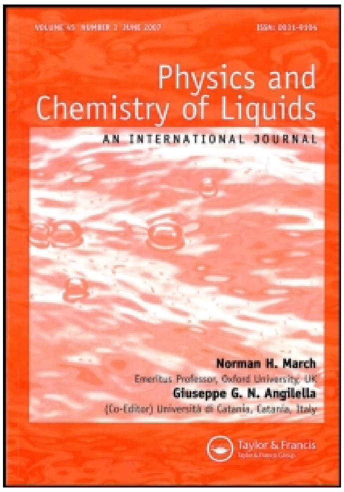

Physics and Chemistry of Liquids
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713646857

Thermodynamics of mixtures containing alkoxyethanols. Part xxiii. Speed of sound predictions and ultrasonic studies of hydroxyether + organic solvent mixtures

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To cite this Article González, J. A. , Mozo, I. , García De la Fuente, I. , Cobos, J. C. and Riesco, N.(2008) 'Thermodynamics of mixtures containing alkoxyethanols. Part xxiii. Speed of sound predictions and ultrasonic studies of hydroxyether + organic solvent mixtures', Physics and Chemistry of Liquids, 46: 4, 390 — 407

To link to this Article: DOI: 10.1080/00319100701434288

URL: http://dx.doi.org/10.1080/00319100701434288

Physics and Chemistry of Liquids
Vol. 46, No. 4, August 2008, 390–407

Thermodynamics of mixtures containing alkoxyethanols. Part xxiii. Speed of sound predictions and ultrasonic studies of hydroxyether + organic solvent mixtures

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(*Received 17 March 2007; in final form 4 May 2007*)

The ability of different models to predict speeds of sound, *u*, of binary mixtures formed by alkoxyethanol and octane, oxaalkane or propylamine has been examined. The models applied are: the free length theory (LFT), the collision factor theory (CFT), and equations such as those proposed by Nomoto, Junjie or Van Dael. Collision factor theory, Nomoto's and Junjie's equations provide similar deviations between experimental and calculated *u*, which is represented quite accurately by these three models. Poorer predictions are obtained when applying the Junjie's equation to propylamine systems, probably due to the existence of strong interactions between unlike molecules in such mixtures. In contrast, slightly better *u* predictions from CFT are obtained for the systems 2-methoxyethanol + polyether, or hydroxyether + propylamine. The good *u* predictions obtained using Nomoto's equation remark the validity of Rao's assumption on additivity of molar sound velocity contributions from atoms, atom groups and chemical bonds of the constituent molecules. Discrepancies between experimental and calculated *u* are larger when using FLT than those obtained from CFT, Nomoto's or Junjie's equations. This has been ascribed to association and size or shape effects. The linear dependence on the molar fractions of the component liquids of the Rao's and Wada's constants suggests that there is no complex formation in the investigated mixtures, and that the interactions present in such systems are of dipolar type.

Keywords: Prediction; Speed of sound; Rao constant; Interactions; Hydroxyethers

1. Introduction

Alkoxyethanols are a very interesting class of substances, because the presence of the O and OH groups in the same molecule allows self-association via inter- and intramolecular hydrogen bonds. Different spectroscopic techniques have been used to investigate the existence of intramolecular hydrogen bonds [1–6], which are present at all conditions, even in vapour phase. In contrast, intermolecular H-bonds appear at

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Physics and Chemistry of Liquids
ISSN 0031-9104 print/ISSN 1029-0451 online
DOI: 10.1080/00319100701434288

higher concentrations of the hydroxyether in liquid state. For molecules of the type CH_3–$(CH_2)_n$–O–$(CH_2)_p$–OH, 5-, 6- and 7-membered rings are formed for $p = 2, 3$ and 4, respectively. Alkoxyethanols with two ether groups and $n = 1$ form 5-membered rings similar to those previously cited, but can also form 8-membered rings of quite different properties [4].

The formation of the intramolecular H-bonds leads to enhanced dipole–dipole interactions in solutions containing alkoxyethanols and alkanes relative to those present in mixtures with homomorphic alkanols [7]. Various evidence supports this conclusion. (a) The Trouton's constant of hydroxyethers, 99.58 $J\,mol^{-1}\,K^{-1}$, is closer to the value of non-self-associated compounds, 92.05 $J\,mol^{-1}\,K^{-1}$, than the value for 1-alkanols, 110.88 $J\,mol^{-1}\,K^{-1}$ [8]. (b) For a given alkane, the upper critical solution temperature (UCST) of the system including an alkoxyethanol is higher than that of the solution with the homomorphic 1-alkanol [7,9,10]. So, UCST(22EEE + n-C_7) = 286.98 K [10], while 1-octanol is miscible with heptane at this temperature, as the molar excess enthalpies, H^E, values of this system indicate, 366 $J\,mol^{-1}$ [11] at equimolar composition and at 288.15 K. (c) H^E (hydroxyether + alkane) > H^E (homomorphic 1-alkanol + alkane) and the corresponding H^E curves for systems with alkoxyethanols are more symmetrical than those for alcoholic solutions, which are skewed towards low concentration of the self-associated compound. For example, at equimolar composition and 298.15 K, H^E (22BEE + heptane) = 1008 $J\,mol^{-1}$ [12] > H^E (1-decanol + heptane) = 421 $J\,mol^{-1}$ [13]. All these features make the theoretical study of mixtures with alkoxyethanols very interesting. We have presented a complete treatment of hydroxyether + alkane systems [7] in terms of DISQUAC (*dispersive-quasichemical*) [14], a purely physical model based on the rigid lattice model developed by Guggemheim [15]. Then, we showed that the proximity effects between the O and OH groups of alkoxyethanols change substantially the interaction parameters of 1-alkanol + $CH_3(CH_2)_uO(CH_2)_vCH_3$ mixtures [16]. We have also shown [17] that the thermodynamic properties of the mentioned solutions can be successfully represented using the so-called ERAS (*extended real associated solution*) model [18], which combines the real association solution model [19] with Flory's equation of state [20]. Moreover, both models, DISQUAC and ERAS, have been used to study mixtures of hydroxyethers with linear monoalkanes or 1-alkanols [21]. In addition, orientational effects in this kind of systems have been examined using the Flory theory [22]. We have also reported experimental data on density, ρ, u and on related quantities such as molar excess volumes, V^E, or isothermal and isentropic compressibilities for alkoxyethanol + dibutylether mixtures [23,24]. As continuation, we present here a detailed comparison between u predictions for binary mixtures formed by hydroxyether and organic solvent (octane, oxaalkanes or propylamine) from the FLT [25,26], CFT [27] and from equations such as Nomoto [28], Junjie [29] or Van Dael [30]. This is important from a practical point of view, as oxygenated compounds are increasingly used as additives to gasoline due their octane-enhancing and pollution-reducing properties [31,32]. In addition, hydroxyethers are non-ionic amphiphile molecules, very effective as surfactants with a large number of applications [33–36]. Finally, in order to gain a deeper insight into the interactions present in alkoxyethanol systems, we investigate such interactions in terms of the the molar sound velocities R_i [37], Wada's constant [38] or the so-called molecular interaction parameter [39].

2. Theories

2.1. *Free length theory (FLT) [25,26]*

Intermolecular free length is defined as [40]:

$$L_f = \frac{2V_a}{Y} \tag{1}$$

where V_a is the available volume and Y is the surface area per mole. These quantities are calculated for pure liquids as follows:

$$V_a = V - V_0 \tag{2}$$

$$Y = (36\pi N_A V_0^2)^{1/3} \tag{3}$$

V is the molar volume, $V_0 = V(1 - T/T_C)^{0.3}$, the molar volume at 0 K and N_A the Avogadro's number. T_C stands for the critical temperature. In the case of a binary mixture, L_f is generalized:

$$L_f = \frac{2[V - \sum x_i V_{0i}]}{\sum x_i Y_i} \tag{4}$$

The value of L_f so obtained is known as thermodynamic L_f. Speeds of sound can be evaluated from the empirical equation obtained by Jacobson [26]:

$$L_f = K\kappa_S^{1/2} \tag{5}$$

where K is a constant dependent on the temperature and k_s is the isentropic compressibility of the mixture. Taking into account the Laplace equation, $k_s = 1/\rho u^2$

$$u = \frac{K}{L_f \rho^{1/2}} \tag{6}$$

2.2. *Collision factor theory (CFT) [27]*

In this theory, the available volume for a pure liquid is determined from the expression:

$$V_a = V - SB \tag{7}$$

where S is the collision factor and B is the actual volume of the molecules per mole, which is calculated as

$$B = \frac{4}{3}\pi N_A r^3 \tag{8}$$

with the molecular radius determined by the Schaaffs relation:

$$r = \sqrt[3]{\frac{M}{\rho N_A}} \sqrt[3]{\frac{3}{16\pi}\left[1 - \frac{\gamma RT}{Mu^2}\left(\sqrt{1 + \frac{Mu^2}{3\gamma RT}} - 1\right)\right]} \tag{9}$$

Here, M is the molecular weight and γ the ratio of heat capacities, C_P/C_V. It has been shown that this equation is a reliable method to estimate the molecular radius of liquids [41]. The collision factor is estimated from the expression:

$$S = \frac{uV}{1600B} \tag{10}$$

Equations (9) and (10) are obtained starting from the van der Waals equation. The latter is generalized to predict u of liquid mixtures:

$$u = 1600\frac{(\sum x_i S_i)(\sum x_i B_i)}{V} \tag{11}$$

It is possible to combine equations (2) and (10) and calculate u as

$$u = \frac{1600V_0}{V} \tag{12}$$

2.3. *Nomoto's equation [28]*

Nomoto [28], under the assumptions that the molar sound velocities are additive and that there is no volume change on mixing, derived the following expression:

$$u = \left[\frac{x_1R_1 + x_2R_2}{x_1V_1 + x_2V_2}\right]^{1/3} \tag{13}$$

where V_i is the molar volume of component i and R_i a quantity characteristic of each compound, which was found experimentally independent of the temperature [38]. R_i is called Rao's constant or molar sound velocity, and is defined by:

$$R_i = V_i u_i^{1/3} \tag{14}$$

It is remarkable that R_i is additive, i.e., it can be calculated as a sum of increments from the atoms or atoms groups in the molecule, and from chemical bonds. Another quantity of similar nature of R_i was introduced by Wada under the name of molecular compressibility [38], W_i, defined as:

$$W_i = V_i \kappa_{si}^{-1/7} \tag{15}$$

2.4. *Junjie's and Van Dael's equations*

These equations have widely used to provide predictions on u of liquid mixtures. They are based on the ideal mixture concept, assuming that $\kappa_S^{id} = \phi_1\kappa_{S1} + \phi_2\kappa_{S2}$ (ϕ_i is the volume fraction), which directly leads to the Junjie's equation [29]:

$$u = \frac{x_1V_1 + x_2V_2}{(x_1M_1 + x_2M_2)^{1/2}}\left[\frac{x_1V_1^2}{M_1u_1^2} + \frac{x_2V_2^2}{M_2u_2^2}\right]^{-1/2} \tag{16}$$

Table 1. Source of excess molar volumes, V^E, speed of sounds, u, and isentropic compressibilities, κ_s, used in this work for hydroxyether + organic solvent mixtures at temperature T.

System	T(K)	V^E(cm^3 mol^{-1})	Ref. u, κ_s
2-Ethoxyethanol (2EE) + *n*-octane	298.15	0.753 [64]	[64]
2-Propoxyethanol (2PE) + *n*-octane	298.15	0.588 [64]	[64]
2-Isopropoxyethanol (2IPE) + *n*-octane	298.15	0.774 [64]	[64]
2-Butoxyethanol (2BE) + *n*-octane	298.15	0.444 [64]	[64]
2-Isobutoxyethanol (2IBE) + *n*-octane	298.15	0.566 [64]	[64]
2-Methoxyethanol (2ME) + dibutylether (4O4)	298.15	0.312 [23]	[23]
2-Ethoxyethanol + dibutylether	298.15	0.226 [23]	[23]
2-Butoxyethanol + dibutylether	298.15	−0.054 [23]	[23]
2-Methoxyethanol + dibutylether	303.15	0.332 [23]	[23]
2-Ethoxyethanol + dibutylether	303.15	0.247 [23]	[23]
2-Butoxyethanol + dibutylether	303.15	−0.044 [23]	[23]
2-(2-Methoxyethoxy)ethanol (22ME) + dibutylether	298.15	0.158 [24]	[24]
2-(2-Ethoxyethoxy)ethanol (22EE) + dibutylether	298.15	0.142 [24]	[24]
2-(2-Butoxyethoxy)ethanol (22BE) + dibutylether	298.15	−0.1057 [24]	[24]
2-(2-Methoxyethoxy)ethanol + dibutylether	303.15	0.166 [24]	[24]
2-(2-Ethoxyethoxy)ethanol + dibutylether	303.15	0.153 [24]	[24]
2-(2-Butoxyethoxy)ethanol + dibutylether	303.15	−0.103 [24]	[24]
2-{2-(2-Methoxyethoxy)ethoxy}ethanol (222MEEE) + dibutylether	298.15	0.0887 [65]	[66]
2-{2-(2-Ethoxyethoxy)ethoxy}ethanol (222EEEE) + dibutylether	298.15	0.0981 [65]	[66]
2-{2-(2-Butoxyethoxy)ethoxy}ethanol (222BEEE) + dibutylether	298.15	−0.1254 [65]	[66]
2-Methoxyethanol + 2,5-dioxahexane (1O2O1)	298.15	−0.2242 [67]	[68]
2-Methoxyethanol + 2,5,8-trioxanonane (1O2O2O1)	298.15	−0.0944 [67]	[68]
2-Methoxyethanol + 2,5,8,11-tetraoxadodecane (1O2OO2O1)	298.15	−0.0943 [68]	[68]
2-Methoxyethanol + 3,6,9-trioxaundecane (2O2O2O2)	298.15	−0.1867 [67]	[68]
2-Methoxyethanol + 5,8,11-trioxapentadecane (4O2O2O4)	298.15	0.0854 [67]	[68]
2-Methoxyethanol + propylamine	298.15	−1.2214 [69]	[70]
2-(2-Methoxyethoxy)ethanol + propylamine	298.15	−1.3131 [69]	[70]
2-(2-Ethoxyethoxy)ethanol + propylamine	298.15	−1.5984 [69]	[70]
2-(2-Butoxyethoxy)ethanol + propylamine	298.15	−1.4169 [69]	[70]
2-{2-(2-Methoxyethoxy)ethoxy}ethanol + propylamine	298.15	−1.4559 [69]	[70]

The Van Dael's equation is derived taking into account that $V_1 = V_2$ [30]:

$$\frac{1}{u^2} = \left(\frac{x_1}{M_1 u_1^2} + \frac{x_2}{M_2 u_2^2}\right)(x_1 M_1 + x_2 M_2) \tag{17}$$

3. Results

Table 1 lists the source of the experimental data on V^E, u and k_s for the systems studied. The physical constants of pure compounds needed to obtain u predictions from the different theories applied in this work are collected in table 2. In the case of FLT, calculations were developed assuming constant values of K (equation (5)) along each homologous series. The values used are collected in table 3. K values dependent on the concentration ($K = x_1 K_1 + x_2 K_2$) have been considered in the literature [42,43]. Table 4 shows a comparison between the u predictions obtained from the different models considered. This table records relative standard deviations for u defined as

$$\sigma_r(u) = \left\{\frac{1}{N}\sum\left[\frac{u_{\text{exp}} - u_{\text{calc}}}{u_{\text{exp}}}\right]^2\right\}^{1/2} \tag{18}$$

Table 2. Physical constants of pure compounds at 298.15 K: molar volume, V, isentropic compressibility, κ_s, molar heats capacity at constant pressure, C_p and constant volume, C_V, speed of sound, u and critical temperature, T_C.

Compound^{a}	V(cm^3 mol^{-1})	α(10^{-4} K^{-1})	k_s(Tpa^{-1})	C_p(J mol^{-1} K^{-1})	C_V(J mol^{-1} K^{-1})	u(m s^{-1})	T_C(K)
2ME	79.26^{b}	9.560^{b}	578.77^{b}	172.43^{b}	141.75^{b}	1341.5^{b}	597.6^{c}
	79.64^{b,d}			172.43^{b,e}	141.75^{b,e}	1324.2^{b,d}	597.6^{e}
2EE	97.43^{b}	9.833^{b}	636.96^{b}	207.21^{b}	170.90^{b}	1301.7^{b}	586.3^{f}
	97.91^{b,d}			207.21^{b,e}	170.90^{b,e}	1284.1^{b,d}	586.3^{e}
2PE	114.76^{g}	10.03^{g}	652.40^{g}	246.53^{g}	203.1^{g}	1299.6^{g}	615.2^{h}
2IPE	115.73^{g}	11.20^{g}	691.18^{g}	239.56^{g}	189.9^{g}	1267.9^{g}	593.2^{f}
2BE	131.90^{b}	9.346^{b}	655.33^{b}	268.9^{b}	225.1^{b}	1305.0^{b}	633.9^{h}
	132.52^{b,d}			268.9^{b,e}	225.1^{b,e}	1288.1^{b,d}	633.9^{e}
2IBE	133.33^{g}	9.44^{g}	710.72^{g}	278.6^{g}	235.3^{g}	1259.9^{g}	615^{f}
22MEE	118.32^{i}	8.493^{i}	491.72^{i}	262.1^{i}	216.6^{i}	1415.2^{i}	672^{c}
	118.85^{i,d}			262.1^{e}	216.6^{e}	1397.9^{i,d}	672^{e}
22EEE	136.45^{i}	9.242^{i}	538.40^{i}	297.2^{i}	244.2^{i}	1374.3^{i}	670^{c}
	137.08^{i,d}			297.2^{e}	244.2^{e}	1356.6^{i,d}	670^{e}
22BEE	171.11^{i}	8.918^{i}	572.80^{i}	354.9^{i}	295.8^{i}	1356.9^{i}	692.3^{h}
	171.90^{i,d}			354.9^{e}	295.8^{e}	1339.9^{i,d}	692.3^{e}
222MEEE	157.43^{j}	8.28^{j}	450.5^{j}	358^{j}	298.5^{j}	1458.9^{j}	679.6^{f}
222EEEE	175.40^{j}	8.55^{j}	490^{j}	389^{j}	324^{j}	1417.3^{j}	701.9^{f}
222BEEE	209.03^{j}	8.53^{j}	517.9^{j}	450^{j}	376.7^{j}	1398.8^{j}	747^{f}
Octane	160.69^{g}	11.64^{g}	1040.16^{g}	254.2^{g}	207.3^{g}	1173.4^{g}	568.8^{k}
4O4	170.41^{b}	11.336^{b}	971.12^{b}	278^{b}	224^{b}	1160.9^{b}	588.1^{k}
	171.43^{b,d}			278^{e}	224^{e}	1141.8^{b,d}	588.1^{e}
1O2O1	104.47^{l}	12.8^{l}	854.1^{l}	193.3^{l}	147.7^{l}	1165.1^{l}	537
1O2O2O1	142.90^{l}	10.6^{l}	651.5^{l}	279.6^{l}	221.5^{l}	1278.6^{l}	569.7^{f}
1O2O2O2O1	181.74^{l}	9.7^{l}	567.11	367.6^{l}	294.5^{l}	1340.9^{l}	636.4^{f}
2O2O2O2	179.56^{l}	11.1^{l}	720^{l}	347.4^{l}	274.9^{l}	1239.8^{l}	614^{f}
4O2O2O4	248.64^{l}	12.7^{l}	707^{l}	479.6^{l}	354.6^{l}	1269^{l}	702.3^{f}
propylamine	82.99^{m}	13.78^{m}	950.8^{m}	161^{m}	123.2^{m}	1215.2^{m}	497^{n}

[a] For symbols, see table 1; [b] [23]; [c] [71]; [d] value at 303.15 K; [e] value used at 303.15 K; [f] calculated using the Joback's method [72]; [g] [64]; [h] [73]; [i] [24]; [j] [66]; [k] [74]; [l] [68]; [m] [70]; [n] [75].

Table 3. Values of Jacobson's constant (equation (5)) at temperature T used in this work.

System	T(K)	K(kg$^{1/2}$ m$^{1/2}$ s^{-1})
2-Alkoxyethanol + octane	298.15	1.92×10^{-6}
2-Alkoxyethanol + dibutylether	298.15	1.92×10^{-6}
	303.15	1.96×10^{-6}
2-(2-Alkoxyethoxy)ethanol + dibutylether	298.15	1.92×10^{-6}
	303.15	1.96×10^{-6}
2-{2-(2-Alkoxyethoxy)ethoxy}ethanol + dibutylether	298.15	2×10^{-6}
2-Methoxyethanol + polyoxaalkane	298.15	2.1×10^{-6}
Hydroxyether + propylamine	298.15	2×10^{-6}

For the sake of clarity, a summary of results is shown in table 5. Table 6 contains values of the interaction molecular parameter, X [44,45] and of ΔL_f defined, respectively, as

$$\chi = \left(\frac{u_{exp}}{u_{id}}\right)^2 - 1 \tag{19}$$

$$\Delta L_f = L_f - \phi_1 L_{f1} + \phi_2 L_{f2} \tag{20}$$

being L_f calculated accordingly to equation (4) (thermodynamic intermolecular free length).

Table 4. Speeds of sound at equimolar composition and temperature T for hydroxyether + organic solvent mixtures. Comparison of experimental (exp.) results with those provided by equations applied in this work. Also included are the relative standard deviations defined by equation (18).

System[a]	T(K)	u(m s^{-1}) exp.[b]	u(m s^{-1}) FLT	u(m s^{-1}) equation (12)	u(m s^{-1}) CFT	u(m s^{-1}) Nomoto	u(m s^{-1}) Junjie	u(m s^{-1}) Van Dael
2EE + n-C_8	298.15	1186.3	1177.4	1277.4	1214.7	1220.8	1198.4	1232.2
			0.012	0.071	0.018	0.022	0.008	0.029
2PE + n-C_8	298.15	1197.9	1199.1	1287.9	1221	1224.9	1203.7	1233.1
			0.002	0.071	0.014	0.017	0.004	0.021
2IPE + n-C_8	298.15	1183.6	1166.7	1280.4	1206.1	1212.2	1194.4	1218.8
			0.016	0.076	0.014	0.018	0.007	0.022
2BE + n-C_8	298.15	1208.9	1209.4	1295.4	1292.2	1231.6	1210.5	1231.8
			0.001	0.069	0.012	0.014	0.002	0.014
2IBE + n-C_8	298.15	1189.7	1184.1	1289.5	1208.1	1212	1196.9	1212.7
			0.005	0.079	0.011	0.014	0.005	0.014
2ME + 4O4	298.15	1194.1	1215.3	1292.9	1214.1	1216.4	1195.7	1221.2
			0.018	0.081	0.013	0.014	0.001	0.018
2EE + 4O4	298.15	1195.8	1191.5	1291.5	1209.7	1210.9	1195.9	1217.6
			0.016	0.079	0.009	0.000	0.0006	0.013
2BE + 4O4	298.15	1215.9	1222.7	1306.9	1224.4	1222.4	1208.6	1228.7
			0.004	0.077	0.005	0.004	0.005	0.007
22MEE + 4O4	298.15	1233.6	1233.2	1312.3	1264.2	1261.2	1220.2	1273.4
			0.008	0.072	0.017	0.016	0.009	0.023
22EEE + 4O4	298.15	1232.3	1221.8	1313.7	1254.7	1252.9	1220.9	1252.5
			0.014	0.072	0.013	0.012	0.007	0.012
22BEE + 4O4	298.15	1245.5	1231.5	1323.6	1259.3	1252.6	1230.3	1229.7
			0.015	0.070	0.008	0.006	0.009	0.009
222MEEE + 4O4	298.15	1265.5	1304.5	1318.1	1302.6	1298.4	1242.9	1260.1
			0.052	0.068	0.021	0.019	0.014	0.004
222EEEE + 4O4	298.15	1263.6	1319.4	1324.5	1289.1	1286.7	1241.9	1235.9
			0.053	0.066	0.014	0.013	0.013	0.017
222BEEE + 4O4	298.15	1273.4	1359.2	1338.6	1290.6	1288.3	1250.3	1205.9
			0.070	0.065	0.010	0.008	0.013	0.039
2ME + 4O4	303.15	1175.6	1210.4	1286.1	1195.5	1197.7	1176.8	1203.6
			0.029	0.091	0.013	0.014	0.001	0.019
2EE + 4O4	303.15	1177.2	1186.5	1284.6	1191	1192.3	1177.1	1199.4
			0.010	0.089	0.009	0.009	0.0008	0.014
2BE + 4O4	303.15	1197.8	1218.2	1300.4	1206.1	1204.1	1189.9	1210.5
			0.015	0.087	0.005	0.004	0.005	0.007
22MEE + 4O4	303.15	1215.5	1229.2	1306	1245.7	1242.5	1201.2	1254.6
			0.010	0.081	0.017	0.015	0.010	0.023
22EEE + 4O4	303.15	1214.4	1217.7	1307.5	1236.2	1234.2	1201.9	1233.7
			0.009	0.081	0.013	0.012	0.008	0.011
22BEE + 4O4	303.15	1227.8	1227.9	1317.6	1241.1	1238.4	1211.6	1211.2
			0.009	0.079	0.008	0.006	0.009	0.010
2ME + 1O2O1	298.15	1244.1	1346.9	1277.6	1244.5	1239.5	1226.1	1247
			0.085	0.041	0.0003	0.003	0.011	0.001
2ME + 1O2O2O1	298.15	1304.4	1280.4	1289.4	1300.3	1300.8	1299.6	1266.4
			0.064	0.016	0.003	0.002	0.003	0.021
2ME + 1O2O2O2O1	298.15	1345	1322.7	1317.6	1337.	1341.1	1341	1228.4
			0.058	0.022	0.005	0.003	0.003	0.063
2ME + 2O2O2O2	298.15	1272.7	1322.1	1309.5	1269.7	1270.4	1266.7	1218.1
			0.059	0.034	0.003	0.002	0.004	0.031
2ME + 4O2O2O4	298.15	1280.1	1390.4	1341.6	1279.6	1286.3	1283.4	1157.2
			0.090	0.047	0.001	0.004	0.002	0.070
2ME + propylamine	298.15	1296.4	1385.6	1276.2	1296.2	1275.8	1249.2	1255.8
			0.058	0.018	0.001	0.010	0.025	0.021
22MEE + propylamine	298.15	1347.3	1392	1306.8	1345.8	1330.3	1284.2	1195.6
			0.028	0.032	0.003	0.008	0.032	0.081
22EE + propylamine	298.15	1330	1385.8	1312.6	1327.9	1312.6	1278.7	1159.2
			0.031	0.015	0.003	0.008	0.027	0.093
22BE + propylamine	298.15	1328	1317.6	1316.9	1314.2	1302.1	1241.1	1104.9
			0.012	0.006	0.007	0.013	0.047	0.122
222MEE + propylamine	298.15	1389.8	1323.8	1316.6	1382.1	1371.4	1314.4	1118.5
			0.052	0.046	0.010	0.005	0.028	0.134

[a] For symbols, see table 1;

[b] for the source of data, see table 1.

Table 5. Mean standard relative deviations[a] for u of hydroxyether + organic solvent mixtures obtained in this work from the different models applied.

System[b]	FLT	Equation (12)	CFT	Nomoto	Junjie	Van Dael
2-Alkoxyethanol + octane[c]	0.0072	0.0732	0.0138	0.017	0.0048	0.020
2-Alkoxyethanol + 4O4[c]	0.0127	0.0522	0.009	0.009	0.0022	0.010
2-(2-Alkoxyethoxyethanol) + 4O4[c]	0.0123	0.0713	0.0127	0.0113	0.0083	0.0147
2-{2-(2-Alkoxyethoxy)ethoxy}ethanol + 4O4[c]	0.0583	0.066	0.015	0.0133	0.0133	0.020
2-Alkoxyethanol + 4O4[d]	0.0180	0.089	0.009	0.009	0.0023	0.0133
2-(2-Alkoxyethoxyethanol) + 4O4[d]	0.0093	0.0803	0.0127	0.0103	0.009	0.0143
2ME + polyether[c]	0.0712	0.032	0.0025	0.0028	0.0045	0.0372
alkoxyethanol + propylamine[c]	0.0362	0.0234	0.0048	0.0088	0.032	0.0902

[a] Calculated as $1/N_s \sum \sigma_r(u)$, N_s, number of systems;
[b] for symbols, see table 1;
[c] $T = 298.15\,\mathrm{K}$;
[d] $T = 303.15\,\mathrm{K}$.

Table 6. Molecular interaction parameter, χ and deviations from the ideal mixing of intermolecular free length, ΔL_f, for hydroxyether + organic solvent mixtures at 298.15 K and equimolar composition.

System[a]	$\chi = (u_{\mathrm{exp}}/u_{\mathrm{id}})^2 - 1$	$\Delta L_f/10^{-11}\,\mathrm{m}$
2EE + C_8	-9.0×10^{-3}	0.119
2PE + C_8	1.3×10^{-3}	0.076
2BE + C_8	2.3×10^{-3}	0.063
2ME + 4O4	7.2×10^{-3}	−0.013
2EE + 4O4	8.2×10^{-3}	0.005
2BE + 4O4	2.1×10^{-2}	−0.037
22MEE + 4O4	3.9×10^{-2}	−0.025
22EEE + 4O4	3.2×10^{-2}	−0.006
22BEE + 4O4	3.8×10^{-2}	−0.028
222MEEE + 4O4	5.9×10^{-2}	0.0001
222EEEE + 4O4	5.4×10^{-2}	0.011
222BEEE + 4O4	5.5×10^{-2}	−0.011
2ME + 1O2O1	4.3×10^{-2}	0.10
2ME + 1O2O2O1	9.7×10^{-3}	−0.090
2ME + 1O2O2O2O1	6.3×10^{-3}	−0.074
2ME + 2O2O2O2	1.3×10^{-2}	−0.112
2ME + 4O2O2O4	2.8×10^{-3}	−0.039
2ME + propylamine	9.3×10^{-2}	−0.399
22MEE + propylamine	0.124	−0.322
22EEE + propylamine	0.10	−0.359
22BEE + propylamine	0.1354	−0.265
222MEEE + propylamine	0.148	−0.292

[a] For symbols, see table 1.

4. Discussion

We note that CFT, Nomoto's and Junjie's equations provide the same average results on speeds of sound (tables 4 and 5), magnitude represented quite accurately by these three models. The poorer predictions are obtained when applying the Junjie's equation to propylamine systems, probably due to the existence of strong interactions between unlike molecules in such mixtures. The following evidence supports this conclusion. (i) Very negative V^E values, which are of the same order than those of 1-alkanol + amine systems, characterized by strong hydrogen bonds of the O–H–N type [46,47]. For example, at 298.15 K and equimolar composition, V^E (methanol + butylamine) = −1.677 [48]; V^E(1-propanol + propylamine) = −1.315 [49]; V^E(1-propanol + triethylamine) = −1.997 [50], all values in $\text{cm}^3\,\text{mol}^{-1}$. (ii) Negative ΔL_f values and u values which are higher than those of the other mixtures investigated in the present work. This is in agreement with the fact that u increases with the decrease of L_f and vice versa [51,52]. (iii) Positive χ values [44]. The discrepancies between experimental and calculated speeds of sound may be explained if one takes into account that the Junjie's equation is based on the ideal mixing concept. This also may explain results from Van Dael's equation, where, in addition, it is assumed that $V_1 = V_2$. Note the large deviations encountered for those systems with components of very different size, e.g. 2-methoxyethanol + 5,8,11-trioxapentadecane or 2-{2-(2-methoxyethoxy)ethoxy}ethanol + propylamine (table 4).

The good u predictions obtained using Nomoto's equation remark the validity of Rao's assumption on additivity of molar sound velocity contributions (Rao's constant) from atoms, atom groups and chemical bonds of the constituent molecules (see further) [53].

On the other hand, results from CFT support the generalization of equation (10) for liquid mixtures ($S = \sum x_i S_i$ and $B = \sum x_i B_i$). Moreover, it is also a test of equation (9). The validity of this expression to calculate the molecular radius has been discussed elsewhere [41]. We note that slightly better u predictions from this model are obtained for the systems 2-methoxyethanol + polyether, or hydroxyether + propylamine (tables 4 and 5). That is, for those mixtures where rather strong interactions between unlike molecules are expected. In the case of 2-methoxyethanol + polyether systems, interactions can be considered to be of dipole–dipole type as the decrease of H^E with the number of oxygen atoms in the oxaalkane suggests. At equimolar composition and 298.15 K, H^E (2-ethoxyethanol + 2,5-dioxahexane) = 338 J mol^{-1} [54] < H^E (2-ethoxyethanol + dibutylether) = 1008 J mol^{-1} [55] and H^E (2-butoxyethanol + 2,5,8-trioxanonane) = 592 J mol^{-1} < H^E (2-butoxyethanol + dibutylether) = 680 J mol^{-1} [56]. For ester (methylacetate or ethylacetate) + hexane systems, characterized by large positive H^E, V^E and Δk_s values, CFT yields deviations for u which are of 3.3% (methylacetate) and 2.1% (ethylacetate) [42]. This maybe explained taking into account the destruction of the dipolar order in the ester molecules by adding nonpolar hexane molecules [42]. It seems that the model provides better results for ordered systems.

Free length theory yields a larger average $\sigma_r(u)$ value than those from CFT, Nomoto's or Junjie's equations (table 5). This might be explained in terms of two important shortcomings of the model: association and shape or size effects [57,58]. For example, it should be kept in mind that equation (3) is valid for spherical molecules only. So, we note that the largest $\sigma_r(u)$ value is encountered for the system containing 5,8,11-trioxapentadecane, the longest molecule considered in this work (table 4). Shape effects could also explain the observed increase in $\sigma_r(u)$

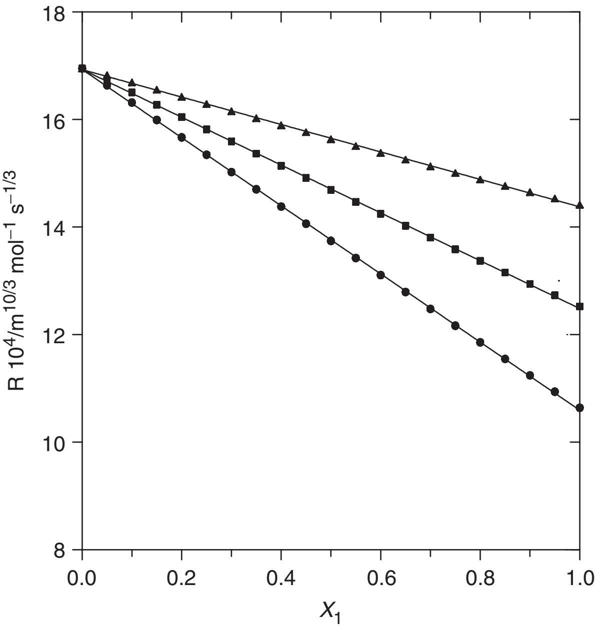

Figure 1. Molar sound velocity, R, at 298.15 K for 2-alkoxyethanol(1) + octane(2) mixtures. Points, experimental results using data from the literature [64]: (●), 2EE; (■), 2PE; (▲), 2BE. Solid lines, linear regressions.

when replacing, in mixtures with dibutylether, the 2-alkoxyethanol or 2-(2-alkoxyethoxy)ethanol by a hydroxyether with three etheric atoms. The good results obtained for the systems 2-butoxyethanol + octane, or + dibutylether, or 2-(2-methoxyethoxy)ethanol + dibutylether, formed by compounds of similar size are remarkable. On the other hand, the rather large $\sigma_r(u)$ values for mixtures such as 2-methoxyethanol + 2,5-dioxahexane, or + propylamine maybe due to association effects (see above).

Finally, we note that equation (12) gives the poorer u predictions, as it depends too directly on V_0.

It is known that the molar sound velocity in binary liquid mixtures is a linearly dependent magnitude on the molar fractions of the component liquids ($R = x_1R_1 + x_2R_2$) in the case there is no association, or in the case that the degree of association does not change with concentration, as long as the density of the component liquids does not differ largely [28,59,60]. Deviations from this behavior are also encountered for mixtures with a component containing halogen atom [42,61]. For the investigated mixtures, the Rao's constant varies linearly with the concentration (figures 1–6), and this indicates the absence of complex formation [59,60]. Such conclusion is also supported by the linear dependence of the Wada's constant with mole fraction (figures 7 and 8). This newly remarks the importance of dipole-dipole interactions in the studied systems. Here it is pertinent to remark that

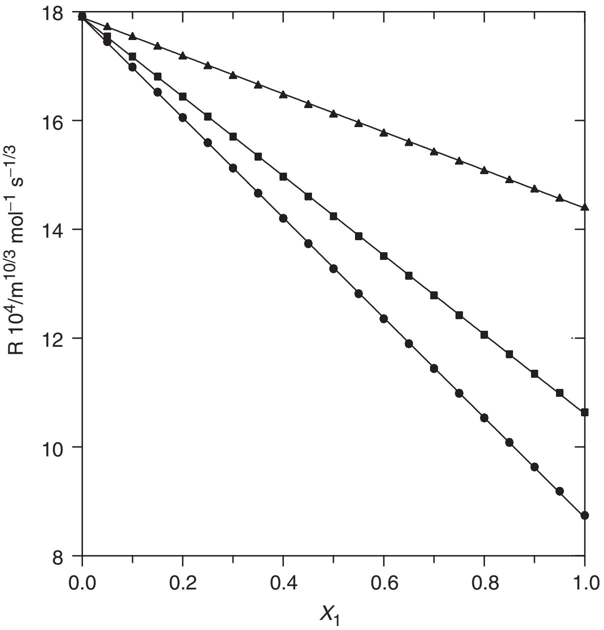

Figure 2. Molar sound velocity, R, at 298.15 K for 2-alkoxyethanol(1) + dibutylether(2) mixtures. Points, experimental results using data from the literature [23]: (●), 2ME; (■), 2EE; (▲), 2BE. Solid lines, linear regressions.

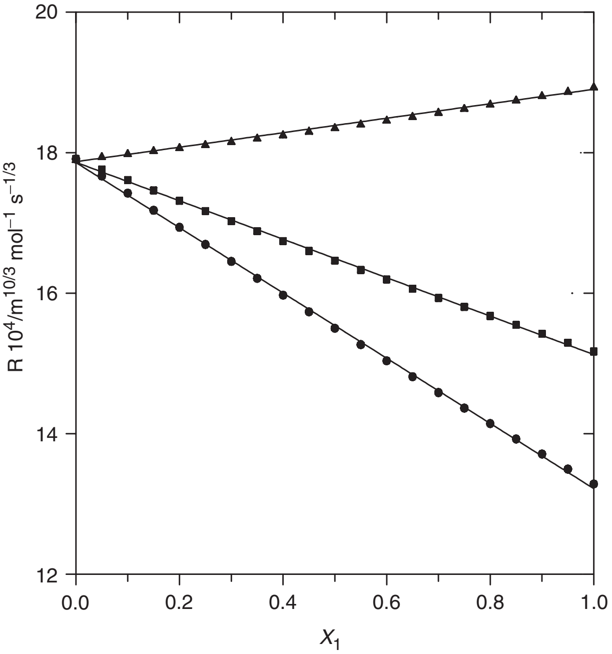

Figure 3. Molar sound velocity, R, at 298.15 K for 2-(2-alkoxyethoxy)ethanol(1) + dibutylether(2) mixtures. Points, experimental results using data from the literature [24]: (●), 22ME; (■), 22EE; (▲), 22BE. Solid lines, linear regressions.

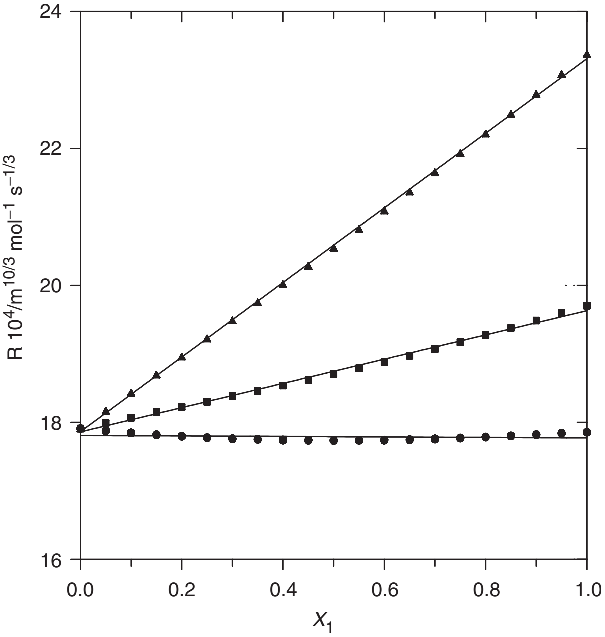

Figure 4. Molar sound velocity, R, at 298.15 K for 2-{2-(2-alkoxyethoxy)ethoxy}ethanol(1) + dibutylether(2) mixtures. Points, experimental results using data from the literature [65,66]: (●), 222MEEE; (■), 222EEEE; (▲), 222BEEE. Solid lines, linear regressions.

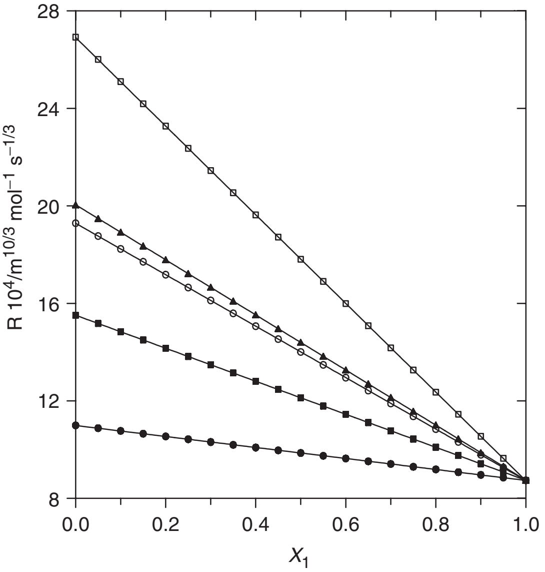

Figure 5. Molar sound velocity, R, at 298.15 K for 2-methoxyethanol(1) + oxaalkane(2) mixtures. Points, experimental results using data from the literature [67, 68]: (●), 2,5-dioxahexane; (■), 2,5,8-trioxanonane; (▲), 2,5,8,11-tetraoxadodecane; (●), 3,6,9-trioxaundecane; (□), 5,8,11-trioxapentadecane. Solid lines, linear regressions.

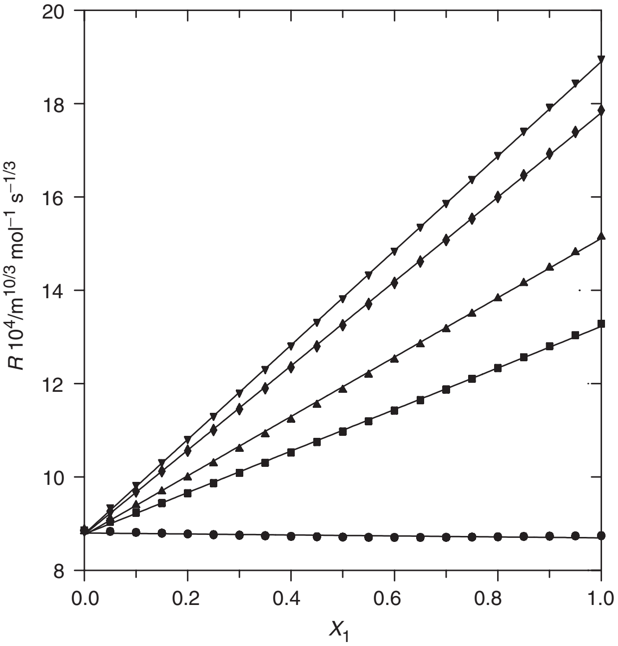

Figure 6. Molar sound velocity, R, at 298.15 K for hydroxyether(1) + propylamine(2) mixtures. Points, experimental results using data from the literature [69,70]: (●), 2ME; (■), 22MEE; (▲), 22EE; (▼), 22BE; (◆), 222MEEE. Solid lines, linear regressions.

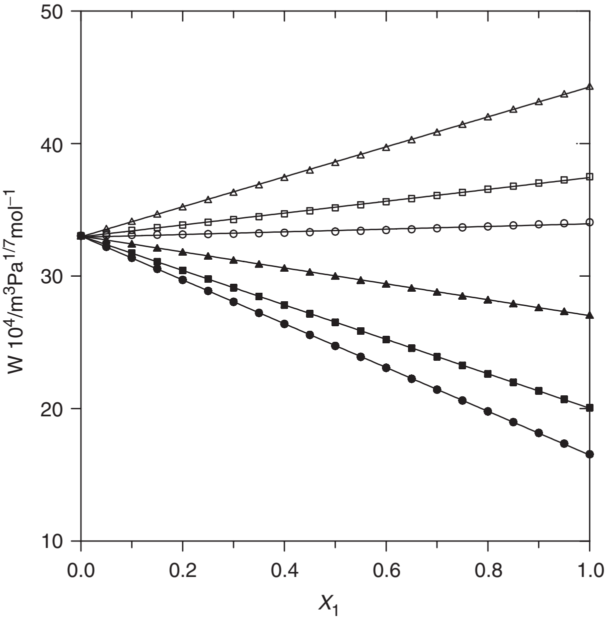

Figure 7. Molecular compressibility, W, at 298.15 K for hydroxyether(1) + dibutylether(2) mixtures. Points, experimental results using data from the literature [23,65,66]: (●), 2ME; (■), 2EE; (▲), 2BE; (●), 222MEEE; (□), 222EEEE; (△), 222BEEE. Solid lines, linear regressions.

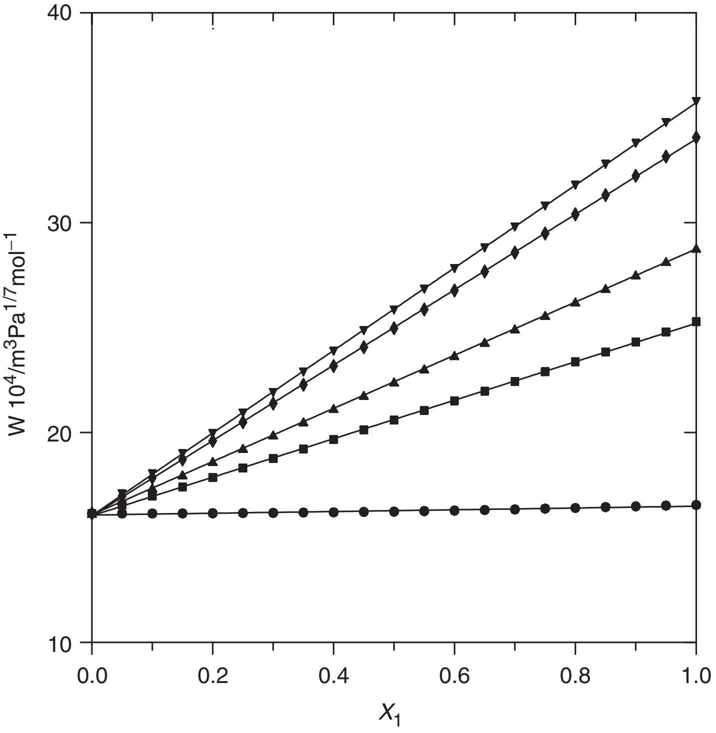

Figure 8. Molecular compressibility, W, at 298.15 K for hydroxyether(1) + propylamine(2) mixtures. Points, experimental results using data from the literature [69,70]: (●), 2ME; (■), 22MEE; (▲), 22EE; (▼), 22BE; (◆), 222MEEE. Solid lines, linear regressions.

the Rao's and Wada's constant have been theoretically developed on the basis of an interaction potential of the Lennard-Jones type [62,63].

The χ parameter (or a parameter defined merely as $(u_{\mathrm{exp}}/u_{\mathrm{id}})^2$ is widely used to estimate the non-ideality of a system, as systems with stronger deviations from the ideal behavior are characterized by high χ values. For example, for mixtures involving 2-pyrrolidone, χ (methanol) ≈ 0.8 and χ (ethanol) ≈ 0.35 [39]. The low χ values listed in table 6 indicate that the investigated mixtures are, in terms of the speed of sound, close to the ideal behavior. The good u predictions obtained using the Junjie's equation support such conclusion. The concentration dependence of χ is shown, for a few examples, in figures 9 and 10. For propylamine mixtures, the curves become shifted to lower mole fractions of hydroxyether when its size increases. That is, interactions between unlike molecules are more probable in that region. In the case of dibutylether systems, the corresponding curves are skewed towards higher mole fractions of the alkoxyethanol. This reveals that the oxaalkane is an active breaker of the hydroxyether structure at low concentrations of this component. Finally, it should be noted that for, a given solvent, propylamine or dibutylether, χ increases by increasing the number of oxyethylene groups in the hydroxyether.

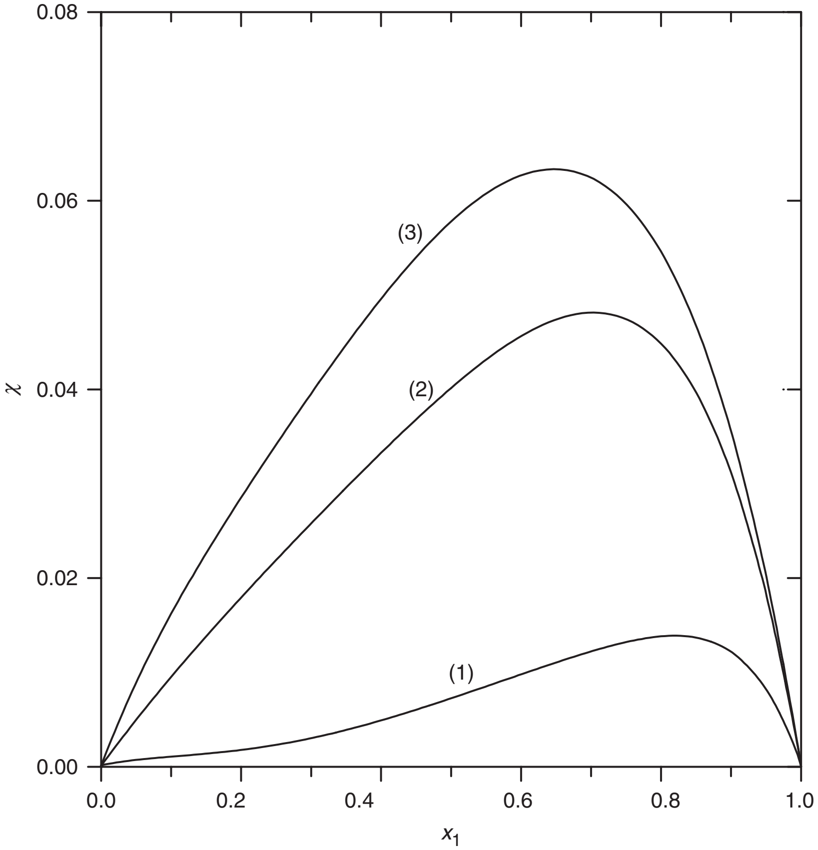

Figure 9. Interaction molecular parameter, χ, at 298.15 K for hydroxyether(1) + dibutylether(2) mixtures: (1), 2ME; (2), 22MEE; (3), 222MEEE.

5. Conclusions

Collision factor theory, Nomoto's and Junjie's equations provide similar deviations between experimental and calculated *u*, which is represented quite accurately by these three models. Poorer predictions are obtained when applying the Junjie's equation to propylamine systems, probably due to the existence of strong interactions between unlike molecules in such mixtures. Slightly better *u* predictions from CFT are obtained for the systems 2-methoxyethanol + polyether, or hydroxyether + propylamine. The good *u* predictions obtained using Nomoto's equation remark the validity of Rao's assumption on additivity of molar sound velocity contributions from atoms, atom groups and chemical bonds of the constituent molecules. The larger discrepancies between experimental and calculated *u* from FLT are explained in terms of association and shape or size effects. Interactions in the investigated mixtures are of dipolar type. This is supported by the linear dependence on the molar fractions of the component liquids of the Rao's and Wada's constants.

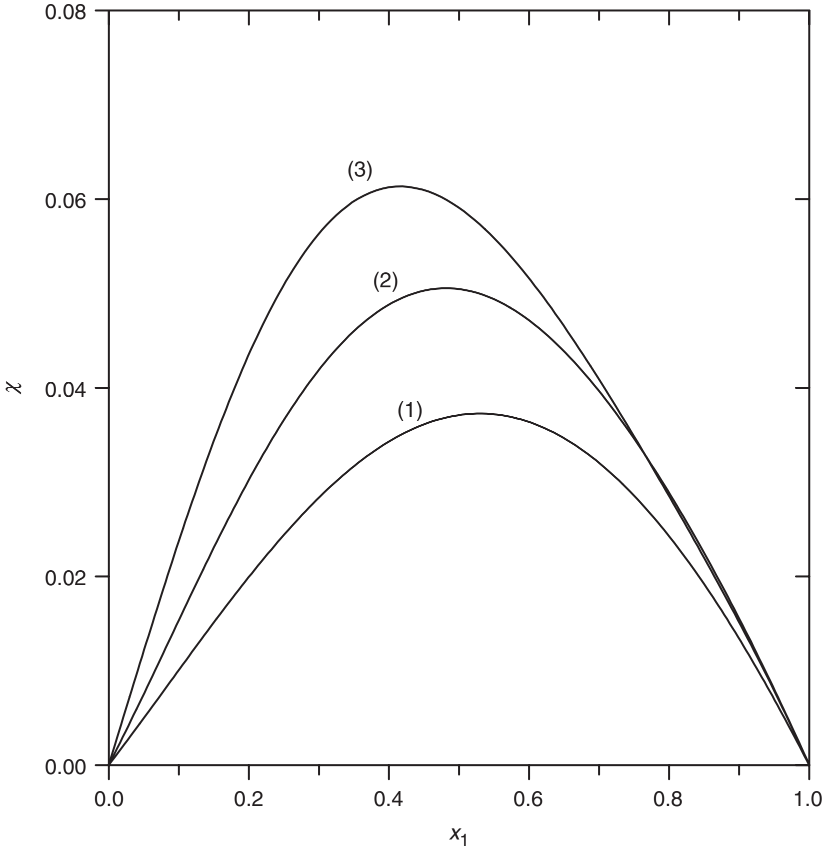

Figure 10. Interaction molecular parameter, χ, at 298.15 K for hydroxyether(1) + propylamine(2) mixtures: (1), 2ME; (2), 22MEE; (3), 222MEEE.

Acknowledgements

The authors gratefully acknowledge the financial support received from the Consejería de Educación y Cultura of Junta de Castilla y León, under Project VA080A06.

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