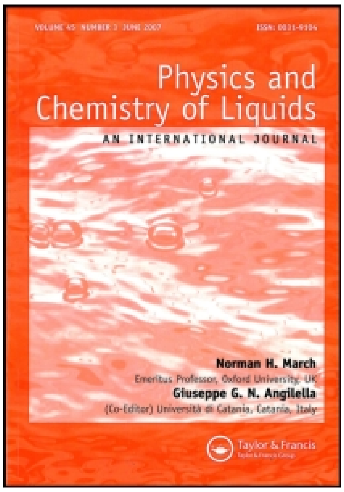

Physics and Chemistry of Liquids

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

Study of the molecular interactions, excess molar volumes and viscosity deviations of ternary systems of 1-butanol (1) + 2-butanol (2) + 1,2-butanediol (3) at 303.15 K

M. Rezaei-Sameti[a]; H. Iloukhani[b]; M. Rakhshi[b]
[a] Department of Chemistry, Faculty of Science, Malayer University, Malayer, Iran [b] Faculty of Chemistry, Bu-Ali Sina University, Hamadan, Iran

To cite this Article Rezaei-Sameti, M. , Iloukhani, H. and Rakhshi, M.(2010) 'Study of the molecular interactions, excess molar volumes and viscosity deviations of ternary systems of 1-butanol (1) + 2-butanol (2) + 1,2-butanediol (3) at 303.15 K', Physics and Chemistry of Liquids, 48: 5, 608 — 617

To link to this Article: DOI: 10.1080/00319100903296113

URL: http://dx.doi.org/10.1080/00319100903296113

Physics and Chemistry of Liquids
Vol. 48, No. 5, October 2010, 608–617

Study of the molecular interactions, excess molar volumes and viscosity deviations of ternary systems of 1-butanol (1) + 2-butanol (2) + 1,2-butanediol (3) at 303.15 K

M. Rezaei-Sameti[a]*, H. Iloukhani[b] and M. Rakhshi[b]

[a]*Department of Chemistry, Faculty of Science, Malayer University, Malayer, Iran;*
[b]*Faculty of Chemistry, Bu-Ali Sina University, Hamadan, Iran*

(*Received 28 March 2008; final version received 29 August 2009*)

This article reports experimental densities and viscosities of ternary systems of 1-butanol (1) + 2-butanol (2) + 1,2-butanediol (3) and the binary systems of 1-butanol (1) + 2-butanol (2), 1-butanol (1) + 1,2-butanediol (3), and 2-butanol (2) + 1,2-butanediol (3), over the entire range of composition at 303.15 K. Excess molar volume V_{123}^E, viscosity deviations $\Delta\eta_{123}$ for viscous flow were evaluated from the experimental data obtained. These derived properties for binary and ternary systems were fitted to Redlich–Kister and Cibulka's equations, respectively. The V_{123}^E data and $\Delta\eta_{123}$ values were negative over the whole range of composition. Several semi-empirical relations are used to predict the binary and ternary viscosities and excess molar volume, and viscosity deviations from experimental results on the constituted ternaries and analysed to discuss the nature and strength of intermolecular interactions in these mixtures.

Keywords: excess molar volume; viscosity deviations; ternary system; 1-butanol; 2-butanol; 1,2-butanediol

1. Introduction

Thermodynamic and transport property data are of great interest in process design and operation. Viscosity and density, vapour–liquid equilibrium data are required in many chemical engineering calculations involving fluid flow, heat and mass transfer. Moreover, knowledge of the dependence of viscosity and density on mixture composition is of considerable interest in understanding the intermolecular interactions [1–5]. This article, as part of a continuing study in our laboratory [6–10], reports the precise measurement of density and kinematic viscosity for the ternaries system formed by ternary system 1-butanol (1) + 2-butanol (2) + 1,2-butanediol, and constituted binary mixtures. Excess molar volumes and deviation in viscosities were calculated for the studied ternary mixtures on the excess properties because of their importance for inferring which type of interactions predominates in liquid mixtures.

---

*Corresponding author. Email: mrsameti@malayeru.ac.ir

ISSN 0031–9104 print/ISSN 1029–0451 online

DOI: 10.1080/00319100903296113
http://www.informaworld.com

2. Experimental section

2.1. *Materials*

The mole fraction purity of the molecules from Merck were: 1-butanol ($\geq$99%), 2-butanol (>99%) and 1,2-butanediol ($\geq$99%). Densities of the pure liquids and their mixtures at 303.15 K were measured with an Anton Paar digital densimeter (Model DMA 4500) operated in the static mode and capable of a precision of better than $\pm 10^{-2}\,\mathrm{kg\,m^{-3}}$ and automatically thermostated within ±0.01 K. Densities and viscosities of the pure reagents were in good agreement with values found in the literature [11–13], reported in Table 1.

2.2. *Apparatus and procedure*

Viscosities at 303.15 K were measured with an Ubbleohde viscometer. The equation for viscosity is

$$\eta = \rho = \nu = \rho(kt - c/t), \tag{1}$$

where k and c are the viscometer constants and t, η and ν are the efflux time, dynamic viscosity and kinematic viscosity, respectively. The dynamic viscosity was reproducible to within $\pm 2\times 10^{-3}$mPa.s. The viscometer was suspended in a thermostated water bath maintained at ±0.01 K.

3. Results and discussion

3.1. *Binary systems*

In Table 2, we show values of densities, dynamic viscosities for binary systems as well as the molar excess volume, V^E and viscosity deviations and $\Delta\eta$ computed with the following equations.

$$V^E/(\mathrm{cm^3 mol^{-1}}) = \frac{\sum_{i=1}^{c} x_i M_i}{\rho} - \sum_{i=1}^{c} x_i \frac{M_i}{\rho_i} \tag{2}$$

$$\Delta\eta/(\mathrm{mPa\,s}) = \eta - \sum_{i=1}^{c} x_i \eta_i, \tag{3}$$

which are valid for ternary systems, as well. In the above equations the presence of the subindex i refer to the property of the pure component and the quantities without

Table 1. Properties of the pure components at 303.15 K.

	ρ (kg m^{-3})		nD	
Substance	Observed	Literature	Observed	Literature
1-butanol	806.13	805.75[a]	1.3974	1.3973[a]
2-butanol	802.46	802.72[b]	1.3950	1.3953[a]
1,2 butanediol	998.74	998.41[c]	1.4381	1.4381[a]

Note: [a]Reference [11]; [b]Reference [12]; [c]Reference [13].

Table 2. Excess molar volume and viscosity deviations of binary mixtures at 303.15 K.

x	ρ (kg m^{-3})	$\Delta\eta$ (mPa s)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPa s)
x 1-butanol + $(1-x)$ 2-butanol				
0.050	798.53	2.401	−0.007	−0.011
0.100	798.80	2.380	−0.015	−0.018
0.149	799.07	2.361	−0.024	−0.025
0.201	799.35	2.342	−0.032	−0.030
0.248	799.59	2.326	−0.038	−0.033
0.302	799.86	2.309	−0.045	−0.036
0.353	800.10	2.294	−0.049	−0.037
0.387	800.25	2.284	−0.050	−0.038
0.449	800.51	2.267	−0.052	−0.039
0.519	800.80	2.248	−0.052	−0.039
0.550	800.92	2.240	−0.052	−0.039
0.600	801.11	2.227	−0.051	−0.039
0.650	801.28	2.215	−0.047	−0.038
0.700	801.44	2.203	−0.043	−0.037
0.750	801.60	2.192	−0.038	−0.034
0.800	801.75	2.181	−0.032	−0.032
0.851	801.89	2.173	−0.024	−0.027
0.899	802.02	2.167	−0.017	−0.020
x 1-butanol + $(1-x)$1,2-butanediol				
0.050	985.80	26.032	−0.047	−2.580
0.101	976.08	22.947	−0.077	−4.240
0.150	966.63	20.158	−0.097	−5.654
0.200	957.08	17.424	−0.114	−7.005
0.297	938.42	13.138	−0.131	−8.600
0.350	928.12	11.031	−0.136	−9.226
0.400	918.31	9.519	−0.139	−9.326
0.450	908.76	8.158	−0.141	−9.311
0.500	899.18	7.134	−0.143	−8.953
0.550	889.49	6.135	−0.143	−8.553
0.600	879.85	5.321	−0.140	−7.977
0.650	870.10	4.675	−0.136	−7.215
0.699	860.66	4.120	−0.132	−6.407
0.750	850.92	3.650	−0.123	−5.472
0.799	841.32	3.236	−0.108	−4.507
0.849	831.62	2.976	−0.088	−3.376
0.900	821.77	2.589	−0.061	−2.356
x 2-butanol + $(1-x)$1,2-butanediol				
0.0499	985.43	26.560	−0.041	−1.196
0.0998	975.70	24.122	−0.070	−2.302
0.1499	965.84	21.521	−0.089	−3.567
0.1992	956.14	19.126	−0.103	−4.650
0.3009	936.04	14.224	−0.121	−6.840
0.3481	926.70	12.353	−0.127	−7.451
0.3995	916.55	10.631	−0.129	−7.805
0.4504	906.49	9.153	−0.131	−7.925
0.5014	896.45	7.808	−0.133	−7.910
0.5503	886.81	6.738	−0.131	−7.676
0.5995	877.14	5.882	−0.129	−7.222
0.6504	867.14	5.249	−0.126	−6.497

(*continued*)

Table 2. Continued.

x	ρ (kg m^{-3})	$\Delta\eta$ (mPa s)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPa s)
0.6994	857.51	4.641	−0.121	−5.797
0.7497	847.62	4.096	−0.110	−5.003
0.7999	837.75	3.635	−0.098	−4.125
0.8500	827.89	3.230	−0.082	−3.193
0.8994	818.16	2.928	−0.060	−2.180
0.9500	808.18	2.664	−0.034	−1.094

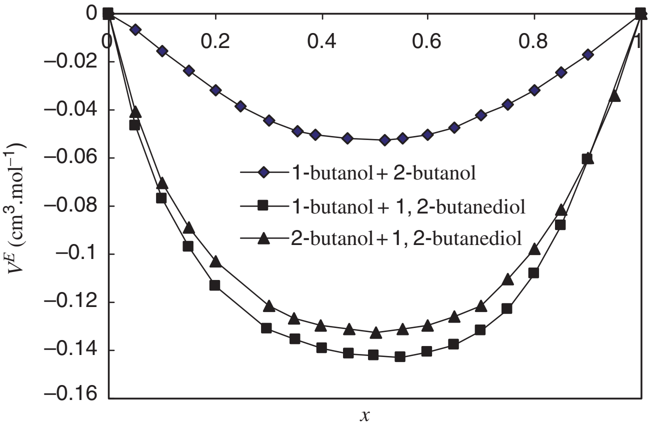

Figure 1. Excess molar volumes of the binary mixtures versus mole fraction x. Experimental results: (◆) x 1-butanol + (1 − x) 2-butanol, (■) x 1-butanol + (1 − x) 1,2 butanediol, (▼) x 2-butanol + (1 − x) 1,2 butanediol. Redlich–Kister fit curves (—) at 303.15 K.

subindex refers to the property of the mixture. M is the molecular weight, x the mole fraction and c refers to the number of components in the mixture.

Figures 1 and 2 show the variation with composition of V^E and $\Delta\eta$ for the binary systems [(x) 1-butanol + (1 − x) 2-butanol, + (1 − x) 1,2-butanediol and (x) 2-butanol + (1 − x) 1,2-butanediol]. As seen in Figure 1 and Table 2, the measured V^E are negative over the entire range of mole fractions for the binary systems. The values of $\Delta\eta$ (Figure 2) were negative over the whole composition range for all of the binary systems studied herein. The sign of $\Delta\eta$ is in agreement with the conclusion by Fort and Moore [14] who proposed that the negative values of this property are characteristic of systems where dispersion forces are predominate.

The V^E and $\Delta\eta$ values were correlated with composition, using the Redlich–Kister equation [15].

$$Y = x(1-x)\sum_{i=0}^{p} A_i(2x-1)^i, \tag{4}$$

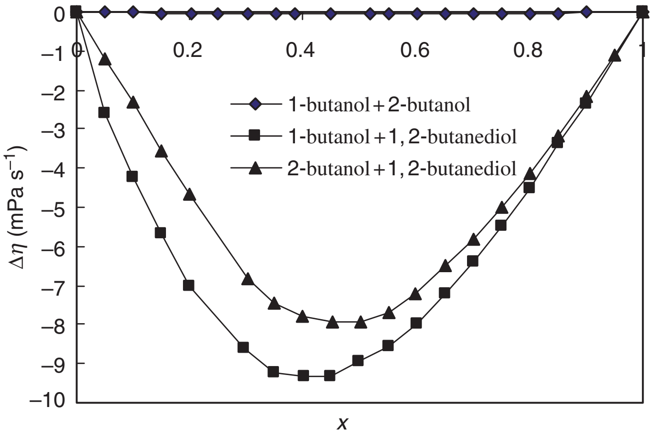

Figure 2. Excess dynamic viscosity of the binary mixtures versus mole fraction x. Experimental results: (◆) x 1-butanol + $(1-x)$ 2-butanol, (■) x 1-butanol + $(1-x)$ 1,2 butanediol, (▼) x 2-butanol + $(1-x)$ 1,2 butanediol. Redlich–Kister fit curves (—) at 303.15 K.

Table 3. Values of adjustable coefficient, A_k in Equation (4) and standard deviation, σ in Equation (5) for excess molar values and viscosity deviation for binary mixtures at 303.15 K.

	A_0	A_1	A_2	A_3	A_4	σ
x 1-butanol + $(1-x)$ 2-butanol						
V^E (cm^3 mol^{-1})	−0.2105	0.0129	0.0111	−0.0334	0.0639	0.0004
$\Delta\eta$ (mPa s)	−0.1572	−0.0101	−0.0862	0.0022	−0.0126	0.0004
x 1-butanol + $(1-x)$ 1,2-butanediol						
V^E (cm^3 mol^{-1})	−0.5721	−0.0662	−0.3199	0.2784		0.0017
$\Delta\eta$ (mPa s)	−36.5310	12.3210	10.3700	2.3402	−18.8070	0.1425
x 2-butanol + $(1-x)$ 1,2-butanediol						
V^E (cm^3 mol^{-1})	−0.5244	−0.0080	−0.3151	0.1133		0.0012
$\Delta\eta$ (mPa s)	−31.7970	5.8739	13.2630	−6.5819	−4.4821	0.0784

where $Y \equiv (V^E$ or $\Delta\eta)$ and x is the mole fraction of the first component. The coefficients A_i were calculated by the unweighted least-square method and the computed values and standard deviation for each of the properties are shown in Table 3.

In each case, the optimum number of coefficients was ascertained from an examination of the variation of standard deviations, σ with

$$\sigma = \left[\frac{\sum (Y_{\mathrm{exp.}} - Y_{\mathrm{cal.}})^2}{(n-p)}\right]^{1/2}, \tag{5}$$

where Y_{exp} and Y_{cal} are the experimental and calculated values of the property Y, respectively, and n and p are the number of experimental points and number of parameters retained in the respective equations. The standard deviations, σ, are also shown in Table 3.

3.2. *Ternary systems*

The results for densities and excess molar volumes and viscosity deviations for the above ternary systems are reported in Table 4.

The excess molar volumes and viscosity deviations were correlated with ternary composition using the equation proposed by Cibulka [16].

$$Y_{123} = Y_{\text{bin}} + x_1 x_2 (1 - x_1 - x_2)(B_0 + B_1 x_1 + B_2 x_2) \tag{6}$$

where $Y \equiv (V^E$ or $\Delta\eta)$ and $Y_{\text{bin}} = Y_{12} + Y_{13} + Y_{23}$ are the so called 'binary contribution'.

Table 4. Excess molar volume and viscosity deviations of ternary mixtures 1-butanol (1) + 2-butanol (2) + 1,2 butandiol (3) at 303.15 K.

X_1	X_2	ρ (kg m^{-3})	$\Delta\eta$ (mPa s)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPa s)
0.100	0.099	955.87	18.089	−0.036	−5.657
0.098	0.201	936.20	13.991	−0.041	−7.096
0.098	0.301	916.16	10.182	−0.040	−8.239
0.100	0.400	896.10	7.054	−0.017	−8.686
0.100	0.499	876.75	4.820	−0.033	−8.272
0.100	0.599	857.11	3.348	−0.031	−7.069
0.100	0.699	838.30	2.540	−0.010	−5.213
0.100	0.799	818.13	2.214	−0.013	−2.868
0.200	0.099	936.96	13.379	−0.085	−7.686
0.202	0.199	916.10	9.684	−0.035	−8.647
0.200	0.300	896.45	6.545	−0.012	−9.164
0.202	0.399	876.71	4.041	−0.019	−8.981
0.200	0.500	857.22	2.447	−0.007	−7.933
0.198	0.602	837.85	1.680	−0.020	−6.030
0.206	0.694	818.55	1.607	−0.028	−3.431
0.298	0.103	916.87	9.405	−0.038	−8.943
0.299	0.201	897.15	6.304	−0.027	−9.397
0.299	0.301	877.44	3.704	−0.028	−9.329
0.300	0.400	857.86	1.924	−0.027	−8.436
0.298	0.501	838.28	1.102	−0.006	−6.606
0.300	0.600	819.52	1.204	−0.081	−3.827
0.397	0.104	897.47	6.249	−0.037	−9.401
0.398	0.202	877.93	3.689	−0.032	−9.319
0.400	0.299	858.41	1.798	−0.025	−8.558
0.400	0.400	839.20	0.836	−0.077	−6.827
0.402	0.498	819.26	0.971	−0.016	−4.025
0.502	0.099	878.26	4.084	−0.031	−8.884
0.500	0.200	858.58	2.077	−0.019	−8.230
0.500	0.300	839.28	0.924	−0.040	−6.716
0.500	0.400	819.68	0.880	−0.013	−4.100
0.599	0.101	859.03	2.784	−0.018	−7.503
0.600	0.200	839.66	1.373	−0.012	−6.263
0.600	0.300	820.07	1.069	−0.010	−3.887
0.700	0.100	839.96	2.134	−0.023	−5.458
0.700	0.199	820.83	1.397	−0.020	−3.572
0.799	0.104	820.43	1.882	−0.020	−2.955

The coefficients, B_p and standard deviations, σ (Equation (5)) obtained by the least-squares method are given in Table 5.

4. Prediction of viscosity of binary and ternary systems

Several semi-empirical relations have been proposed to estimate the viscosity of liquid mixtures in terms of pure component data. We have examined the following equations for binary system:

(1) The single-parameter Grunberg–Nissan [17] equation is

$$\eta = \exp(x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 G_{12}), \tag{7}$$

where G_{12} is a parameter proportional to the interchange energy.

(2) Hind et al. [18] proposed the following equation:

$$\eta = x_1^2 \eta_1 + x_2^2 \eta_2 + 2x_1 x_2 \eta_{12}, \tag{8}$$

where η_{12} is attributed to unlike pair interactions.

(3) The two-parameter McAllister [19] equation based on Eyring's theory of absolute reaction rates and three-body interaction model is

$$\begin{aligned} \ln \nu = {} & x_1^3 \ln \nu_1 + 3x_1^2 x_2 \ln Z_{12} + 3x_1 x_2^2 \ln Z_{21} + x_2^3 \ln \nu_2 - \ln(x_1 + x_2 M_2/M_1) \\ & + 3x_1^2 x_2 \ln(2/3 + M_2/3M_1) + 3x_1 x_2^2 \ln(1/3 + 2M_2/3M_1) + x_2^3 \ln(M_2/M_1), \end{aligned} \tag{9}$$

where Z_{12} and Z_{21} are interaction parameters and M_i and ν_i are the molecular mass and kinematics' viscosity of pure component i, respectively.

(4) The no-parameter equation of Kendall and Monroe [20] is expressed as

$$\eta = (x_1 \eta_1^{1/3} + x_2 \eta_2^{1/3})^3. \tag{10}$$

(5) The Frenkel equation [21]:

$$\eta = x_1^2 \eta_1 + x_2^2 \eta_2. \tag{11}$$

In ternary system the result is fitted by Grunberg–Nissan and Frenkel's equations. The correlating ability of each of Equations (8)–(11) was tested and

Table 5. Fitting parameters for Cibulka equation and standard deviations, σ, for excess molar volume and viscosity the ternary system at $T = 303.15$ K.

	B_0	B_1	B_2	σ
1-butanol (1) + 2-butanol (2) + 1,2-butanediol (3)				
V^E (cm^3 mol^{-1})	5.9922	−3.1647	−3.9491	0.0264
$\Delta\eta$ (mPa s)	56.8258	−143.0790	−132.0460	0.0090

Table 6. Adjustable parameters of Equations (7)–(11) and standard deviations of binary mixture viscosities at 303.15 K.

	x 1-butanol + $(1-x)$ 2-butanol	x 1-butanol + $(1-x)$ 1,2-butanediol	x 2-butanol + $(1-x)$ 1,2-butanediol
Grunberg and Nissan			
G_{12}	−0.0340	0.8106	−0.1091
σ	0.0031	0.4421	0.9421
Kendall and Monroe			
σ	0.0326	0.9543	0.1501
Frenkel			
σ	0.0031	0.7701	0.9421
Hind et al.			
η_{12}	2.2075	−1.1136	0.4754
σ	0.0030	0.2080	0.2260
McAllister three-body			
$\ln(\nu_{12})$	1.0039	1.4758	1.5475
$\ln(\nu_{21})$	1.0473	2.5910	2.8439
σ	0.0003	0.1434	0.2329
1-butanol (1) + 2-butanol (2) + 1,2-butanediol(3)			
Grunberg and Nissan			
G_{123}		−7.6957	
σ		0.6505	
Ferenkel			
$\ln(A_{123})$		3.4211	
σ		0.8305	

Table 7. Standard deviations, σ, in the predictions, V^E_{123} (cm^3 mol^{-1}) and $\Delta\eta_{123}$ (mPa s), with different models for the ternary system at $T = 303.15$ K.

	σ (V^E_{123})	σ ($\Delta\eta_{123}$)
Radojkovic	0.0826	0.5868
Rastogi	0.0822	0.9643
Kohler	0.0444	0.9084
Jacob and Fitzner	0.0878	0.9845
Tsao and Smith[a]	0.1660	0.7254
Tsao and Smith[b]	0.2003	0.7074
Tsao and Smith[c]	0.0783	0.6522
Colinet	0.1517	0.6558
Toop[a]	0.1755	0.6296
Toop[b]	0.0989	0.6069
Toop[c]	0.1820	0.7361
Scatchard[a]	0.2183	0.6224
Scatchard[b]	0.0453	0.6165
Scatchard[c]	0.0628	0.4485

their adjustable parameters and standard deviations (σ):

$$\sigma = \left[\frac{1}{n-k}\sum\left\{\frac{(\eta_{\exp} - \eta_{\mathrm{cal}})}{\eta_{\exp}}\right\}^2\right]^{1/2}, \tag{12}$$

where n represents the number of data points and k is the number of numerical coefficients are given in Table 6. The results show that the two-parameter McAllister equation is the best for calculating the correlation of viscosity of binary mixtures.

Although prediction of the physical properties of multicomponent mixtures from those of their pure components is generally unreliable because of mixing effects, numerous schemes have been put forward for predictions based on the properties of the binary systems formed by pairs of components of the multicomponent system. In this work we applied the following models: Tsao–Smith [22], Jacob–Fitzner [23], Kohler [24], Rastogi et al. [25], Radojkovic et al. [26], Colinet [27], Toop [28] and Scatchard et al. [29]. Standard deviations, σ, presented in Table 7 were determined for all models with Equation (12). The standard deviations are obtained from the Kohler model for excess molar volume and from the Scatchard model for viscosity deviations of small values.

Acknowledgements

The authors are thankful to the authorities of the Bu-Ali Sina and Malayer Universities for providing the necessary facilities to carry out the present work.

References

[1] A.S. Al-Jimaz, J.A. AL-Kandary, and A.M. Abdul-Latif, Fluid Phase Equilibr. **218**, 247 (2004).
[2] A. Rodriguez, J. Canosa, A. Dominguez, and J. Tojo, Fluid Phase Equilibr. **216**, 167 (2004).
[3] F. Giro, M.F. Goncalves, A.G.M. Ferreira, and I.M.A. Fonseca, Fluid Phase Equilibr. **204**, 217 (2003).
[4] U.R. Kapadi, D.G. Hundiwale, N.B. Patil, M.K. Lande, and P.R. Patil, Fluid Phase Equilibr. **192**, 63 (2001).
[5] S.L. Oswal and H.S. Desai, Fluid Phase Equilibr. **149**, 359 (1998).
[6] H. Iloukhani, Z. Rostami, N. Afshari. Phys. Chem. Liq. **47**, 360 (2009).
[7] H. Iloukhani and Z.B. Nojini, Phys. Chem. Liq. **43**, 429 (2006).
[8] H. Iloukhani, M. Rezaei-Sameti, and H.A. Zarei, Thermochim. Acta **438**, 9 (2005).
[9] H. Iloukhani and M. Rezaei-Sameti, J. Chem. Thermodyn. **37**, 1151 (2005).
[10] H. Iloukhani and M. Rezaei-Sameti, J. Chem. Eng. Data **50**, 1928 (2005).
[11] J.A. Riddick, W.B. Bunger, and T.K. Sakano, *Organic Solvents*, 4th ed. (Wiley-Intersience, New York, 1986).
[12] A. Mariano and M. Postigo, Fluid Phase Equilibr. **239**, 146 (2006).
[13] H. Gerey, P. Ulbig, and M. Gorent, J. Chem. Thermodyn. **32**, 1585 (2000).
[14] R. Fort and W.R. Moore, Trans. Faraday. Soc. **62**, 1112 (1966).
[15] O. Redlich and A.T. Kister, Ind. Eng. Chem. **40**, 345 (1948).
[16] I. Cibulka, Collect. Czech. Chem. Commun. **47**, 1414 (1982).
[17] L. Grunberg and A.H. Nissan, Nature **164**, 799 (1949).
[18] R.K. Hind, E. McLaughlin, and A.R. Ubbelohde, Trans. Faraday Soc. **56**, 328 (1960).

[19] R.A. McAllister, AIChE J. **6**, 427 (1960).
[20] J. Kendall and K.P. Monroe, J. Am. Chem. Soc. **39**, 1787 (1917).
[21] J. Frenkel, *Kinetic Theory of Liquids* (Oxford University Press, Oxford, 1946).
[22] C.C. Tsao and J.M. Smith, Chem. Eng. Prog. Symp. Ser. **49**, 107 (1953).
[23] K.T. Jacob and K. Fitzner, Thermochim. Acta **18**, 197 (1977).
[24] F. Kohler, Monatsh. Chem. **91**, 738 (1960).
[25] R.P. Rastogi, J. Nath, and S.S. Das, J. Chem. Eng. Data **22**, 249 (1977).
[26] N. Radojkoric, A. Tasic, D. Grozdanic, B. Djordjevic, and M. Malic, J. Chem. Thermodyn. **9**, 349 (1977).
[27] C. Colinet, Ph.D. thesis, University of Grenoble, France. 1967.
[28] G.W. Toop, Trans. TMS-AIME **223**, 850 (1965).
[29] G. Scatchard, L.B. Ticknor, J.R. Goates, and E.R. McCartney, J. Am. Chem. Soc. **74**, 3721 (1952).