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Study of molecular interactions between 2-methyl 1-propanol and m-substituted aniline at various temperatures (thermodynamic and acoustic properties)

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ABSTRACT

Excess molar volumes (V^E), excess acoustic impedance (Z^E), excess isentropic compressibility ($\kappa_s{}^E$), deviation in viscosity ($\Delta\eta$), excess Gibbs energy of activation of viscous flow (G^{*E}), excess free length ($L_f{}^E$), excess enthalpy (H^E) and excess speed of sound (U^E) for binary mixtures of 2-methyl-1-propanol with meta-substituted aniline (3-chloroaniline, 3-methoxyaniline and 3-methylaniline) at selected compositions were determined from the measured values of densities (ρ), viscosities (η) and speeds of sound (u) of pure components and their mixtures in range of 303.15–313.15 K. The results are analysed in terms of interactions arising due to the formation of complex hydrogen bond electron in the binary mixtures. Different theoretical models were applied to check the applicability to the present values of speed of sound and viscosity. Finally, the Prigogine–Flory–Patterson theory is applied to identify the most predominant molecular interaction.

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Density; speed of sound; 2-methyl-1-propanol; meta-substituent aniline

1. Introduction

Volumetric, viscometric and speed of sound investigations of liquid mixtures are of considerable importance to understanding the intermolecular interactions occurring among component molecules, and they find application in several industrial and technological processes [1,2]. Speed of sound investigations of pure liquids and liquid mixtures consisting of polar and non-polar components are also equally important in analysing intermolecular interaction between the component molecules. Speed of sound and related thermodynamic parameters are helpful as well as needed for characterising thermodynamic and physicochemical aspects of binary liquid mixtures such as molecular association and dissociation [3,4].

2-Methyl-1-propanol with m-substituted aniline (3-chloroaniline, 3-methoxyaniline and 3-methylaniline) systems make particularly interesting systems for study because substituents have a marked effect on the association behaviour. A survey of the literature has shown that no excess molar volume measurement for a mixture of 2-methyl-1-propanol with studied m-substituted aniline exists.

In view of the importance of these liquids, binary liquid mixtures of 2-methyl-1-propanol with m-substituted aniline have been prepared to study the molecular interactions through acoustic, volumetric and viscometric investigations at 303.15K, 308.15 K and 313.15 K. For this study, speed of sound, density and viscosity for these mixtures have been measured,

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Table 1. List of chemicals with details of source, CAS number, water content and purity.

Name of the chemical	CAS number	Source	**Water content in mass fraction	Mass fraction purity
2-methyl-1-propanol	78-83-1	S.D. Fine chemicals	0.00044	0.995
3-chloroaniline	108-42-9	S.D. Fine chemicals	0.00043	0.995
3-methylaniline	108-44-1	S.D. Fine chemicals	0.00042	0.996
3-methoxyaniline	536-90-3	Sigma Aldrich, India	0.00042	0.995

Note: ** Karl–Fischer method.

Table 2. Densities, viscosity and speeds of sound data of pure components at various temperatures and 0.1 mPa pressure.

Component (in K)	Density (ρ/g·cm^{-3})		Speed of sound (u/m·s^{-1})		Viscosity (η/mPa·s)	
	Experimental	Literature	Experimental	Literature	Experimental	Literature
2-methyl-1-propanol						
303.15 K	0.79436	0.79437 [5] 0.79434 [8]	1172.4	1172.9 [6] 1172.0 [5]	2.846	2.845 [7] 2.8466 [9]
308.15 K	0.79017	0.7906 [10]	1156.5	1156.2 [6]	2.459	2.4550 [11]
313.15 K	0.78597	0.78596 [12]	1140.6	1138.4 [12]	2.114	2.116 [7] 2.112 [9]
3-chloroaniline						
303.15 K	1.20532	1.20530 [13]	1517.5	1517.0 [13]	4.2554	4.2556 [14]
308.15 K	1.20075	-	1501.2	-	3.8420	
313.15 K	1.19602	1.19600 [13]	1480.0	1484.0 [4]	3.4302	
3-methylaniline						
303.15 K	0.98032	0.9808 [15] 0.98096 [13]	1598.2	1567 [15]	3.018	
308.15 K	0.97611	0.9727 [15]	1530.6	1533 [15]	2.707	
313.15 K	0.97186	0.9646 [15]	1491.7	1492 [15]	2.396	
3-methoxyaniline						
303.15 K	1.09025		1579.9		5.593	
308.15 K	1.08705		1552		5.160	
313.15 K	1.08320		1523.4		4.733	

Note: The standard uncertainties are u(x_1) = 1 × 10^{-4}, u (ρ) ±2 × 10^{-5}g.cm^{-3}, u (u) = ±0.5 m.s^{-1}., u (η) = ±1.12%, u (T) = 0.01 K for density, speed of sound and viscosity, and u (P) = 1 kPa

and these data have been used to evaluate some relevant excess/deviation parameters. The results are analysed to discuss the molecular interactions prevailing in the above binary mixtures.

2 Experimental

2.1 Materials

Meta-substituted aniline and 2-methyl-1-propanol chemicals were purchased from Sigma Aldrich and S.D. Fine Chemical. Table 1 contains data with respect to their source, purity and analysis method. Density, speed of sound and viscosity values are listed in Table 2 and are in good agreement with the data available in the literature [5–15].

2.2 Apparatus and procedure

The details of the density, speed of sound and viscosity methods and their measurement techniques were described elsewhere [16]. The uncertainty in density, speed of sound and viscosity measurement of liquid mixtures were ±2. × 10^{-5} g·cm^{-3}, 0.5 m·s^{-1} and 1.12% respectively. The uncertainty of the mole fraction was ±1 × 10^{-4}.

Table 3. Coefficients of Redlich–Kister equation and standard deviation (σ) values for liquid mixtures of 2-methyl-1-propanol with 3-chloroaniline, 3-methylaniline and 3-methoxyaniline at *T*= (303.15–313.15) K.

T/K	A_0	A_1	A_2	σ
2-methyl-1-propanol (1)+ 3-chloroaniline (2)				
V^E/cm^3·mol^{-1}				
303.15	−0.384	0.107	0.021	0.001
308.15	−0.396	0.112	−0.037	0.001
313.15	−0.408	0.117	−0.094	0.001
κ_s^E/TPa^{-1}				
303.15	−74.16	25.52	−0.774	0.001
308.15	−76.36	23.32	−7.610	0.001
313.15	−78.56	26.82	−14.44	0.001
$\Delta\eta$/mPa• s				
303.15	0.089	−0.002	−0.002	0.001
308.15	0.093	−0.002	0.004	0.001
313.15	0.097	−0.007	0.006	0.001
2-methyl-1-propanol (1)+ 3-methylaniline (2)				
V^E/cm^3·mol^{-1}				
303.15	−0.399	0.108	−0.038	0.001
308.15	−0.407	0.108	−0.076	0.001
313.15	−0.417	0.116	−0.138	0.001
κ_s^E/TPa^{-1}				
303.15	−76.66	24.17	−9.492	0.002
308.15	−78.82	23.71	−17.26	0.001
313.15	−81.85	24.10	−24.12	0.096
$\Delta\eta$/mPa• s				
303.15	0.094	0.002	0.007	0.001
308.15	0.097	0.001	0.015	0.001
313.15	0.102	−0.002	0.020	0.001
2-methyl-1-propanol (1)+ 3-methoxyaniline (2)				
V^E/cm^3·mol^{-1}				
303.15	−0.411	0.106	−0.089	0.001
308.15	−0.418	0.100	−0.124	0.001
313.15	−0.428	0.111	−0.177	0.001
κ_s^E/TPa^{-1}				
303.15	−79.24	25.29	−16.65	0.002
308.15	−82.03	25.16	−26.40	0.001
313.15	−84.83	25.04	−36.16	0.001
$\Delta\eta$/mPa• s				
303.15	0.098	0.002	0.019	0.001
308.15	0.100	0.004	0.033	0.001
313.15	0.106	0.002	0.034	0.001

3 Results and discussion

Table 3 shows the density (ρ), excess molar volumes (V^E), speed of sound (u), excess acoustic impedance (Z^E), excess isentropic compressibility ($\kappa_s{}^E$), viscosity (η), deviation in viscosity ($\Delta\eta$), excess Gibbs energy of activation of viscous flow (G^{*E}), excess free length ($L_f{}^E$), excess enthalpy (H^E) and excess speed of sound (U^E) as a function of mole fraction, x_1 of 2-methyl-1-propanol of binary liquid mixtures at T= (303.15 to 313.15) K and 0.1 mPa pressure.

It could be clearly seen from Figures 1 and 2 that the excess functions (excess molar volume (V^E) and excess isentropic compressibilities ($\kappa_s{}^E$)) of three kinds of binary solutions are negative over the range of the whole component content in this work, and the values of excess functions increase with an increase in temperature. These may suggest that the negative values are attributable to the association between common components and non-common components through hetero-molecular association between the –OH groups in the common component and the nitrogen atom in the non-common components. The strength of the molecular association arising from

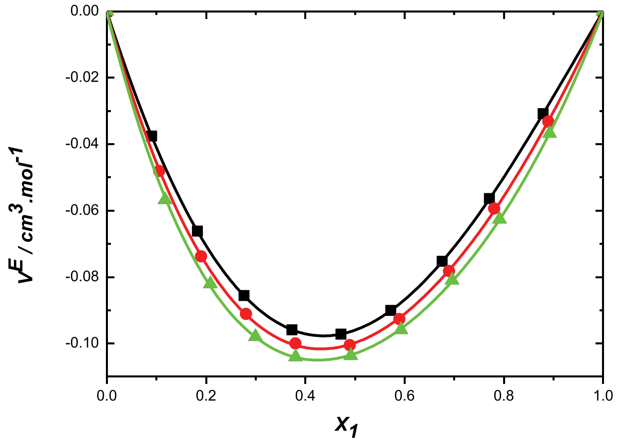

Figure 1. Curves of excess molar volume (V^E) with mole fraction for the binary mixtures of 2-methyl-1-propanol with 3-chloroaniline **(filled square);** 3-methylaniline (**filled circle**) and 3-methoxyaniline (**filled triangle**) at 303.15 K.

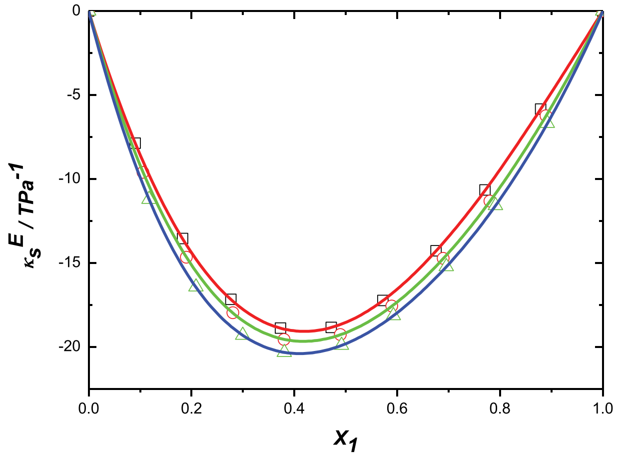

Figure 2. Curves of excess isentropic compressibility with mole fraction for the binary mixtures of 2-methyl-1-propanol with 3-chloroaniline **(hallow square);** 3-methylaniline (**hallow circle**) and 3-methoxyaniline (**hallow triangle**) at 303.15 K.

interactions between the common component and non-common components is stronger than the strength of the molecular association between like molecules [17,18].

The magnitude of the volume contraction follows the sequence of

3-methoxyaniline>3-methylaniline>3-chloroaniline

The excess isentropic compressibility values for all the binary systems are negative at all compositions. The negative values in all of the systems support the formation of complexes through an intermolecular association between common component and non-component molecules [19].

The order of values of excess isentropic compressibility is almost similar to that observed with respect to excess molar volumes.

The L_f^E values have a negative trend similar to what we have observed in the case of the K_s^E at all the temperatures under study. The negative values of L_f^E suggest that specific interactions are present between unlike molecules in these binary systems.

The values of excess functions decrease (more negative) with the rise in temperature for the binary liquid mixtures under study as the interactions between unlike molecules become stronger due to the rise in thermal motions.

The viscosity deviations ($\Delta\eta$) and excess Gibbs energy of activation of viscous flow (ΔG^{*E}) are positive values over the whole mole fraction range at various temperatures. These are shown in Figures 3 and 4. These positive values are due to hetero association of unlike molecules which give rise to the formation of complexes where O-H-N bonds of the mixtures are stronger than O-H-O and N-H-N bonds of the single component solvents.

The positive values of deviation in viscosity for binary liquid mixtures fall in the order:

3-methoxyaniline>3-methylaniline>3-chloroaniline

It is clear that the excess values of enthalpy (H^E) are positive with respect to the mole fraction for the entire composition range and at different temperatures. These values of H^E tend to increase from system 1 to system 3. This arises from the fact that there exist some specific interactions between

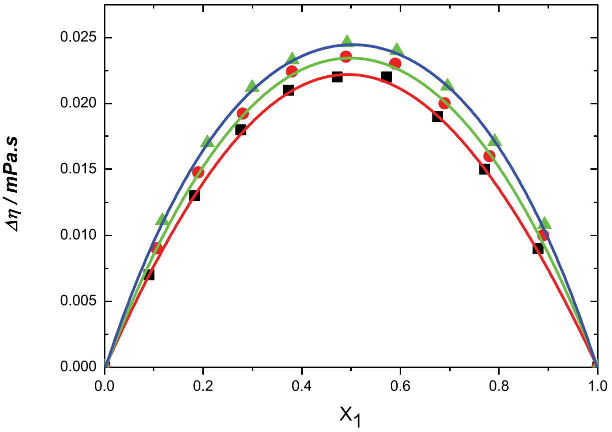

Figure 3. Curves of deviation in viscosity (Δη) with mole fraction for the binary mixtures of 2-methyl-1-propanol with 3-chloroaniline **(filled square);** 3-methylaniline (**filled circle**) and 3-methoxyaniline (**filled triangle**) at 303.15 K.

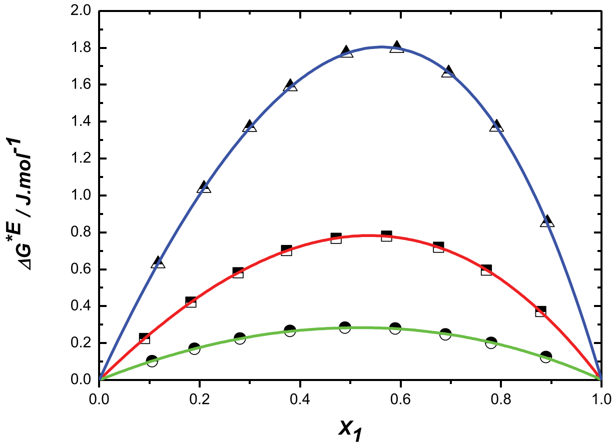

Figure 4. Curves of excess Gibbs energy of activation of viscous flow *(ΔG^{*E})* with mole fraction for the binary mixtures of 2-methyl-1-propanol with 3-chloroaniline **(half filled square);** 3-methylaniline (**half filled circle**) and 3-methoxyaniline (**half filled triangle**) at 303.15 K.

unlike molecules in these liquid mixtures. The positive H^E values also suggest the existence of intermolecular hydrogen bonds and the breaking of associated structures in these cases.

The negative deviations of U^E and Z^E suggest that dispersion forces are operative in the system. The negative values of indicate the decrease in the strength of interaction between the molecules in the mixture. In the present work, these values indicate the presence of weak interaction dispersive forces between the unlike molecules.

Various interaction parameters and the corresponding standard deviations ($\sigma/10^{-3}$ N s m^{-2}) computed from different models for viscosity is tabulated in Table 4.

The interaction terms are the adjustable parameters representing the binary interactions. To test the correlating ability of the applied models we calculated the standard deviation between the experimental and the calculated viscosity values. All the empirical models gave a reasonable fit but there is good agreement between the theoretical and experimental values in these systems. The estimated standard deviations are smaller in all cases indicating that the present mixture viscosities are well correlated by these viscosity models.

Evaluation of the theoretical speed of sound values proves to be useful to verify the applicability of various postulates of these theories of liquid mixtures and to arrive at some useful inferences regarding the strength of molecular interactions between component liquids in some cases. So average percentage error (APE) and chi-square fit values for speed of sound computed from different theoretical models were calculated and are presented in Table 5.

The average percentage error values are small. In comparison, Nomoto's relation and free length relation are found to give some valuable estimates of the experimental values of speed of sound values in these mixtures.

Excess/deviation functions (V^E, $\kappa_s^{E,}$ and $\Delta\eta$) data are fitted to a Redlich–Kister polynomial equation [20]

$$Y^E = x_1 x_2 \sum_{i=0}^{j} A_i (1 - 2x_1)^i, \tag{1}$$

where Y^E is V^E, $\Delta\eta$ and κ_s^E. Values of the coefficients A_i are determined using the method of least squares. The standard deviations σ (Y^E) were calculated using the formula as follows:

Table 4. The values of $\bar{V}^{\circ}_{m,1}$,$V^{*}_{m,1}$,$\bar{V}^{\circ E}_{m,1}$,$\bar{V}^{\circ}_{m,2}$, $V^{*}_{m,2}$and $\bar{V}^{\circ E}_{m,2}$ of the components for 2-methyl-1-propanol with 3-chloroaniline, 3-methylaniline and 3-methoxyaniline at T= (303.15–313.15) K.

T/K	$\bar{V}^{\circ}_{m,1}$	$V^{*}_{m,1}$	·$\bar{V}^{\circ E}_{m,1}$	$\bar{V}^{\circ}_{m,2}$	$V^{*}_{m,2}$	$\bar{V}^{\circ E}_{m,2}$
			(cm^3·mol^{-1})			
2-methyl-1-propanol (1)+ 3-chloroaniline (2)						
303.15	93.06	93.31	−0.255	105.37	105.84	−0.470
308.15	93.48	93.81	−0.320	105.70	106.24	−0.545
313.15	93.92	94.31	−0.385	106.04	106.66	−0.619
2-methyl-1-propanol (1)+ 3-methylaniline (2)						
303.15	92.98	93.31	−0.329	108.76	109.30	−0.544
308.15	93.43	93.81	−0.375	109.18	109.77	−0.590
313.15	93.87	94.31	−0.439	109.58	110.25	−0.670
2-methyl-1-propanol (1)+ 3-methoxyaniline (2)						
303.15	92.92	93.31	−0.394	112.35	112.96	−0.606
308.15	93.36	93.81	−0.442	112.65	113.29	−0.642
313.15	93.81	94.31	−0.494	112.98	113.69	−0.715

Table 5. The values of $\bar{K}^{\circ}_{s,m,1}$, $K^{*}_{s,m,1}$, $\bar{K}^{\circ E}_{s,m,1}$, $\bar{K}^{\circ}_{s,m,2}$, $K^{*}_{s,m,2}$ and $\bar{K}^{\circ E}_{s,m,2}$ of the components 2-methyl-1-propanol with 3-chloroaniline, 3-methylaniline and 3-methoxyaniline at *T*= (303.15–313.15) K.

T/K	$\bar{K}^{\circ}_{s,m,1}$	$\bar{K}^{\circ E}_{s,m,1}$	$K^{*}_{s,m,1}$	$\cdot\bar{K}^{\circ}_{s,m,2}$	$\bar{K}^{\circ E}_{s,m,2}$	$K^{*}_{s,m,2}$
			TPa^{-1}			
2-methyl-1-propanol (1)+ 3-chloroaniline (2)						
303.15	−39.74	−48.28	8.546	−104.46	−108.27	3.813
308.15	−48.49	−57.37	8.876	−115.07	−118.99	3.926
313.15	−57.41	−66.64	9.223	−125.79	−129.86	4.071
2-methyl-1-propanol (1)+ 3-methylaniline (2)						
303.15	−60.68	−69.23	8.546	−125.89	−130.42	4.534
308.15	−63.23	−72.10	8.876	−128.57	−133.37	4.800
313.15	−73.13	−82.35	9.223	−140.46	−145.56	5.098
2-methyl-1-propanol (1)+ 3-methoxyaniline (2)						
303.15	−61.91	−70.45	8.546	−134.31	−138.46	4.151
308.15	−74.70	−83.58	8.876	−148.20	−152.53	4.327
313.15	−87.72	−96.94	9.223	−162.49	−167.01	4.523

Table 6. PFP interaction parameter, χ_{12}, and calculated values of the three contributions from the PFP theory with experimental excess molar volumes at $x_1 = 0.5$ at 303.15K.

		Calculated contributions			V^E ($x = 0.5$) cm^3.mol^{-1}		
Binary mixtures	$\chi_{12}(10^7)$	Interactional (10^{-8})	Free volume	P* effect	EXP	PFP	$\delta/cm^3.mol^{-1}$
2-methyl-1-propanol (1)+ 3-chloroaniline (2)	5.265	1.051	−0.1161	−0.53357	−0.0959	−0.0960	0.0001
2-methyl-1-propanol (1)+ 3-methylaniline (2)	2.728	1.123	−0.0489	−0.3572	−0.0998	−0.0997	− 0.0001
2-methyl-1-propanol (1)+ 3-methoxyaniline (2)	4.327	1.133	−0.4426	0.1503	−0.1030	−0.1028	−0.0002

$$\sigma(Y^E)=\left[\sum\left(Y^E{}_{exp}-Y^E{}_{cal}\right)^2/(m-n)^{1/2}\right], \tag{2}$$

where m is the total number of experimental points and n is the number of parameters. The coefficients, A_i, and corresponding standard deviation values (σ) are presented in Table 6.

3.1 Partial molar properties

The interpretations of excess partial molar properties ($\overline{V}^{E}_{m,1}$,$\overline{V}^{E}_{m,2}$,$\overline{K}^{E}_{s,m,1}$ and $\overline{K}^{E}_{s,m,2}$) and excess partial molar properties at infinite dilution ($\overline{V}^{\circ E}_{m,1}$, $\overline{V}^{\circ E}_{m,2}$$\overline{K}^{\circ E}_{s,m,1}$ and $\overline{K}^{\circ E}_{s,m,2}$) of components have previously been described [16].

From Tables 4 and 5, excess partial molar properties at infinite dilution values are negative over the whole composition range at experimental temperatures. The negative values indicate that the interaction between solute and solvent is stronger than the solvent–solvent/solute–solute interactions of the pure components in the binary mixtures [21].

4. Application of Prigogine–Flory–Patterson theory

The Prigogine–Flory–Patterson (PFP) theory [22–24] was used in the present study to correlate the excess molar volume results for the present mixtures. The details of the PFP theory and their equations are described elsewhere [16]. It is clear from Figure 5 and Table 6 that the free volume contribution is a supporting factor for negative values of excess volumes for binary mixtures.

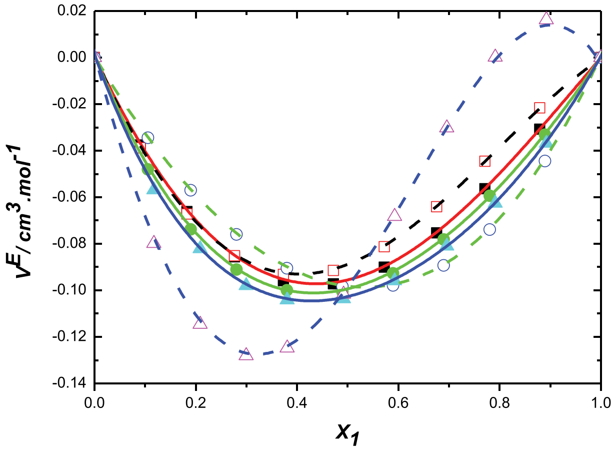

Figure 5. Excess molar volumes with mole fraction (x_1) of 2-methyl-1-propanol in the binary liquid mixtures of 2-methyl-1-propanol with 3-chloroaniline (**filled square**); 3-methyl aniline (**filled circle**) and 3-methoxyaniline (**filled triangle**) at 303.15 K and (—) calculated with PFP theory using parameters.

5. Conclusions

Densities and speeds of the sound of binary mixtures of 2-methyl-1-propanol with m-substituted aniline (3-CA, 3-MA and 3-MOA) have been measured at different temperatures and derived parameters along with their excess values and also excess partial molar properties at infinite dilution are calculated. The results are analysed in terms of the specific interactions through solute–solvent interactions between the components of the mixtures, resulting in the formation of complexes. Moreover, the V^E values were analysed with PFP theory and showed that the free volume contribution is the supporting factor for negative values of excess molar quantities.

Disclosure statement

No potential conflict of interest was reported by the authors.

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