

Physico-chemical analysis in ternary organic mixtures at various temperatures with an acoustical approach

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Abstract:

Thermo-dynamical analyses of organic mixtures are essential across industries. Ultrasonic sound velocity (U) measurements in the ternary organic mixtures of Toluene, Chloroform, and Cyclohexane have been conducted between 303.15, 308.15 and 313.15 K. The experimental/derived data were utilised to compute deviations for their excess values such as adiabatic compressibility (β^E), free length (L_f^E), free volume (V_f^E), impedance (Z^E), internal pressure (π_i^E) and enthalpy (H^E) to investigate and confirm the types of interactions. Using these derived parameters, excess parameters were calculated to further reaffirm the type, strength, magnitude, and potential interactions such as complex formation, dipole-dipole, and dispersive types. Moreover, both ideal and non-ideal behaviours were analysed. The ultrasonic velocity data were validated against some contemporary and well-known models like Namoto's relation (NR), Ideal mixture relation (IMR), Impedance dependence relation (IDR), Collision factor theory (CFT) and Nutch-Kunkies (NK) theorem. From the IMR and IDR are gives well approach to experimental one. And a comparative study of these models was conducted to ascertain the possible types of interactions within the mixtures through their molecular interaction parameter, percentage deviation, standard percentage error deviation.

Keywords: enthalpy, ideality, model, molecular interaction, organic mixture, physical parameter, theory.

Classification numbers: 1.3, 2.1, 2.2

1. Introduction

In recent years, thermo-acoustical investigations of organic, ionic, and electrolyte liquid mixtures have become crucial in numerous industries. Primarily, the intermolecular interactions in liquid mixtures are of significant interest due to their excess properties. Continuing from earlier reports [1-4], the physico-chemical analysis, along with the excess parameters, was computed using standard and experimental data, observing deviations from the ideal mixtures of any solution [5, 6]. Ultrasonic and thermo-dynamical studies in binary liquid mixtures, involving certain alcohols with aniline [7, 8], were undertaken at varying temperatures. The analysis of non-polar liquids, vital in the extraction

processes of coal for compound analysis, and in the membranes of nylon fibre, nylon moulding, resin, paint, varnish, and oils, as well as its use as a plasticiser, is paramount. Such fluid mixtures are utilised for processing and product formation across numerous industries. For instance, organic solvents might be used in the food industry for the gas-sweetening absorption process and as intermediates in the manufacture of other chemicals. To comprehend the interactions among the molecules, particularly in polar and non-polar molecules [9, 10], physico-chemical analyses are indispensable. Moreover, these analyses provide insights into the bonding of associated molecular complexes and other molecular processes.

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The experimental findings were juxtaposed with theoretical models, correlating and validating these against proposed empirical theories [11-13]. The aim of the present investigation, centred on the molecular interactions of adopted systems, necessitates selecting a particular model and applying a concept from the various available ones [14, 15]. If none of the extant models provide a satisfactory prediction of molecular functioning, it might be prudent to propose new models considering the nature of the molecules. Consequently, the theoretical assessment of ultrasonic velocity in the ternary liquid mixtures of organic solutions at different temperatures has been calculated, employing methods such as NR [16], Van Deal-Vangeal's IMR [17], IDR [18], CFT [19], N-K [20]. Furthermore, some supplementary test parameters have also been calculated [21]. In this study, the authors present excess properties at various temperatures for mixtures of Toluene, Chloroform, and Cyclohexane. The types of molecular interaction are then validated against the theoretical model to reaffirm the precision of interaction. The article is structured as follows: Section 2 details experimental procedures; Section 3 covers some excess parameters with pertinent theories/models of ultrasonic velocity; Section 4 presents results and their comprehensive discussion; and Section 5 concludes with the derived outcomes.

2. Experimental

Analytical Grade chemicals were purified using standard procedures [22, 23]. Their purity was estimated at 99.7%. Sound velocities in liquids were measured using an interferometer (2 MHz), with a precision of ± 0.1 m/s. The densities and viscosities of the solutions were determined using a specific gravity bottle (10 ml) and Oswald's viscometer. Temperatures were maintained consistently with a deviation of 0.1 K. Computations were performed in the Excel environment, deriving parameters from the following expressions [1].

3. Theories and models

The excess parameters such as excess compressibility are given as

$$\beta^E = \beta_{\text{exp}} - (x_1\beta_1 + x_2\beta_2 + x_3\beta_3) \quad (\text{I})$$

and the corresponding excess free length is expressed as

$$L_f^E = L_{f_{\text{exp}}} - (x_1L_{f1} + x_2L_{f2} + x_3L_{f3}) \quad (\text{II})$$

The excess free volume is denoted as

$$V_f^E = V_{f_{\text{exp}}} - (x_1V_{f1} + x_2V_{f2} + x_3V_{f3}) \quad (\text{III})$$

and the excess internal pressure is represented by

$$\pi_i^E = \pi_{i_{\text{exp}}} - (x_1\pi_{i1} + x_2\pi_{i2} + x_3\pi_{i3}) \quad (\text{IV})$$

The excess impedance is given by

$$Z^E = Z_{\text{exp}} - (x_1Z_1 + x_2Z_2 + x_3Z_3) \quad (\text{V})$$

and the excess enthalpy takes the following form:

$$H^E = H_{\text{exp}} - (x_1H_1 + x_2H_2 + x_3H_3) \quad (\text{VI})$$

where all notations have their usual meanings.

NR: O. Nomoto (1958) [16] formulated an assumption based on the linear dependence of sound velocity on molar volume in binary mixtures, expressed as

$$U_{NR} = \left(\frac{X_1R_1 + X_2R_2}{X_1V_1 + X_2V_2} \right)^3 \quad (1)$$

From Eq. (1), its expansion to a ternary system is

$$U_{NR} = \left(\frac{X_1R_1 + X_2R_2 + X_3R_3}{X_1V_1 + X_2V_2 + X_3V_3} \right)^3 \quad (2)$$

Molar volume (V) follows the additive rule, e.g.,

$$V = X_1V_1 + X_2V_2 + X_3V_3 \quad (3)$$

Ideal mixture relation (IMR): In this relation, the adiabatic compressibility (β) of the mixture is [17]:

$$\beta = q_1\beta_1 + q_2\beta_2 \quad (4)$$

where q is the volume fraction.

By adopting specific heat, γ , it can be applied to a homogeneous medium as.

$$\beta_{IM} = q_1 \left[\frac{\gamma_1}{\gamma_{IM}} \right] \beta_1 + q_2 \left[\frac{\gamma_2}{\gamma_{IM}} \right] \beta_2 \quad (5)$$

The following relationship is only applicable to ideal systems:

$$\gamma_1 = -\gamma_2 = \gamma_{IM} \quad (6)$$

Using Eq. (4), Eq. (6) can be written as

$$\beta_{IM} = X_1\beta_1 + X_2\beta_2 \quad (7)$$

In binary systems,

$$U_{IMR} = \left(\frac{1}{X_1m_1 + X_2m_2} \right)^{\frac{1}{2}} \left(\frac{X_1}{m_1U_1^2} + \frac{X_2}{m_2U_2^2} \right)^{-\frac{1}{2}} \quad (8)$$

For ternary systems, Eq. (8) can be extended and expressed as

$$U_{IMR} = \left(\frac{1}{X_1m_1 + X_2m_2 + X_3m_3} \right)^{\frac{1}{2}} \left(\frac{X_1}{m_1U_1^2} + \frac{X_2}{m_2U_2^2} + \frac{X_3}{m_3U_3^2} \right)^{-\frac{1}{2}} \quad (9)$$

The degree to which molecules interact (α) is represented by

$$\alpha = \left(\frac{U_{exp}^2}{U_{IMR}^2} \right) - 1 \quad (10)$$

IDR: The impedance or resistance (Z) of liquid mixtures is contingent on their sound velocity and density. This can be computed from the following relation [18]:

$$U_{IDR} = \sum_{i=1}^3 \frac{X_i Z_i}{X_i \rho_i} \quad (11)$$

CFT: This theory's application to ternary systems is given by the following [19]:

$$U_{CFT} = U_a \left(\frac{(X_1S_1 + X_2S_2 + X_3S_3)(X_1B_1 + X_2B_2 + X_3B_3)}{V_m} \right)^3 \quad (12)$$

where V_m is the molar volume; S_i is the collision factor; B_i is the actual volume; X_i is the mole fraction of the component i .

NK theorem: This theorem is formulated from collision factor theory [20], which can be expressed as:

$$U_{NK} = \frac{U_a (\sum X_i S_i) (\sum X_i B_i)^3}{\sum X_i V_i} \quad (13)$$

where all the notations have their usual meaning.

Percentage deviation: The computed value's deviation from the experiment is determined by

$$\left(\frac{\Delta U}{U} \right) \% = \left(\frac{U_{exp} - U_{the}}{U_{exp}} \right) 100 \% \quad (14)$$

Standard percentage error deviation: This error is calculated from the following expression:

$$e = \left(\left(\sum \frac{\Delta U}{U} \right) \% \right)^2 \quad (15)$$

4. Results and discussion

All excess and theoretical velocity values are presented in Table 1. From Fig. 1A, it can be observed that the values of (β^E) show an increasing trend up to 0.06 m, thereafter a decreasing trend is observed with Toluene at all temperatures, with maximum values at 313.15 K. The positive sign of (β^E) is a result of dispersion forces [23-25]. The trend of excess free length mirrors that of (β^E), as shown in Fig. 1B. The dissociation of like molecules and the positive excess contribution indicate weak interactions; the negative sign of β^E strongly suggests increasing solute-solute molecular interactions. This implies the potential formation of complexes between donors and acceptors, causing molecules to come closer and leading to decreased adiabatic compressibility [26]. For these concentrations, the negative values of free length suggest the possibility of close arrangements due to molecular ordering, thus intensifying interactions amongst molecules [27].

From Fig. 1B, the increasing trends of L_f^E values highlight the structural variations among molecules of different sizes [28, 29]. From Fig. 1C, the (V_f^E) values increase and show an upward trend with Toluene across all molalities and temperatures, with peak values at 303.15 K up to 0.06 m, followed by maximum values at 308.15 K. This suggests the strength of the hydrogen bond. A positive V_f^E indicates the formation of complexes and the prevalence of interactions between dissimilar molecules [30, 31]. The values of (π_i^E) are negative and increase with Toluene at all molalities and temperatures. However, the larger values observed at 303.15 K are depicted in Fig. 1D. These suggest weaker interactions between dissimilar molecules [32].

Table 1. The measured and calculated values of ultrasonic velocity (U), their percentage deviation ($\Delta U/U$), standard percentage error (e), and molecular interaction parameter (α) for various molarity (m) of Toluene + Chloroform + Cyclohexane at 303.15, 308.15, and 313.15 K.

(m)	(U) – (m/s)						(ΔU/U) – (m/s)						(a)
	U_{exp}	U_{NR}	U_{IMR}	U_{IDR}	U_{NK}	U_{CFT}	U_{NR}	U_{IMR}	U_{IDR}	U_{NK}	U_{CFT}		
303.15 K													
0.01	1151.2	1178.2	1156.7	1131.3	1168.2	1173.8	-1.2441	-0.2148	0.1246	-1.4321	-3.3765	-0.0316	
0.02	1144.1	1180.3	1158.9	1138.0	1171.2	1195.9	-3.1731	-1.3024	0.5245	-2.3776	-4.5367	-0.0527	
0.03	1136.3	1195.5	1162.2	1142.7	1175.1	1200.9	-5.2377	-2.3063	-0.5897	-3.4419	-5.7130	-0.0700	
0.04	1120.0	1211.6	1165.5	1147.2	1179.2	1222.3	-8.1786	-4.0625	-2.4285	-5.2857	-9.1339	-0.1263	
0.05	1112.6	1228.6	1168.9	1151.7	1183.1	1227.1	-10.486	-5.1169	-3.5701	-6.3938	-10.350	-0.1047	
0.06	1104.7	1246.6	1172.3	1156.0	1187.2	1230.5	-12.917	-6.1865	-4.7101	-7.5362	-11.458	-0.0832	
0.07	1096.2	1265.7	1175.7	1160.3	1191.1	1233.2	-15.484	-7.2719	-5.8667	-8.6770	-12.518	-0.0485	
0.08	1112.4	1285.9	1179.2	1164.5	1195.2	1232.8	-15.638	-6.0431	-4.7212	-7.4820	-10.863	-0.0404	
0.09	1136.9	1307.5	1182.7	1168.6	1199.1	1240.6	-15.097	-4.1102	-2.8697	-5.5545	-9.2077	0.0213	
0.10	1152.1	1330.4	1186.2	1172.7	1203.1	1239.3	-15.486	-2.9687	-1.7968	-4.4357	-7.5781	0.0288	
e	----	0.0379	0.0293	0.0020	0.0349	0.0438							
308.15 K													
0.01	1142.3	1169.2	1139.8	1121.5	1159.2	1189.9	-2.0142	-0.9827	0.6623	-1.3233	-31298	-0.0813	
0.02	1136.3	1170.7	1148.7	1127.3	1161.6	1191.3	-3.0546	-1.1180	0.7659	-2.2535	-4.8680	-0.0763	
0.03	1126.7	1172.9	1151.6	1131.7	1165.1	1198.3	-4.1652	-2.2735	-0.5062	-3.4725	-6.4210	-0.0668	
0.04	1112.3	1175.2	1154.6	1135.9	1168.8	1213.4	-5.6835	-3.8309	-2.1493	-5.1079	-9.1187	-0.1237	
0.05	1104.8	1177.7	1157.6	1140.1	1172.4	1218.3	-6.6757	-4.8551	-3.2699	-6.1957	-10.353	-0.1148	
0.06	1096.1	1180.2	1160.6	1144.1	1175.9	1222.7	-7.6825	-5.8942	-4.3887	-7.2901	-11.560	-0.0931	
0.07	1072.7	1182.8	1163.6	1148.1	1179.6	1222.1	-10.336	-8.5448	-7.0989	-10.037	-14.002	-0.0847	
0.08	1104.5	1185.6	1166.7	1152.0	1183.1	1222.0	-7.3913	-5.6793	-4.3478	-7.1649	-10.688	-0.0500	
0.09	1128.6	1188.5	1169.9	1155.8	1186.7	1232.8	-5.3635	-3.7145	-2.4645	-5.2039	-9.2908	-0.0015	
0.10	1136.3	1191.5	1173.0	1159.6	1190.4	1229.7	-4.8856	-3.2570	-2.0775	-4.7887	-8.2482	0.0060	
e	----	0.0413	0.0330	0.0026	0.0384	0.0484							
313.15 K													
0.01	1141.2	1159.2	1127.3	1109.2	1148.2	1187.2	-0.3165	-0.0021	0.0032	-0.0198	-3.2432	-0.0821	
0.02	1128.0	1160.9	1138.4	1117.3	1151.9	1189.5	-2.9166	-0.9219	0.9486	-2.1188	-5.4521	-0.0731	
0.03	1096.4	1163.3	1141.5	1121.8	1155.7	1190.6	-6.1405	-4.1514	-2.3540	-5.4471	-8.6314	-0.0635	
0.04	1088.2	1165.8	1144.6	1126.3	1159.5	1205.2	-7.1507	-5.2022	-3.5202	-6.5717	-10.772	-0.1209	
0.05	1072.1	1168.3	1147.8	1130.6	1163.3	1216.1	-8.9832	-7.0709	-5.4664	-8.5168	-13.442	-0.1119	
0.06	1056.9	1171.0	1151.0	1134.8	1167.1	1220.1	-10.890	-8.9962	-7.4621	-10.521	-15.540	-0.1031	
0.07	1042.8	1173.8	1154.3	1139.0	1170.9	1221.1	-12.648	-10.777	-9.3090	-12.37	-17.188	-0.0946	
0.08	1088.8	1176.7	1157.5	1143.1	1174.7	1217.6	-8.1525	-6.3878	-5.0643	-7.9688	-11.912	-0.0730	
0.09	1112.2	1179.8	1160.8	1147.1	1178.5	1231.0	-6.0971	-4.3884	-3.1564	-5.9802	-10.701	-0.0245	
0.10	1120.1	1183.2	1164.2	1151.2	1182.3	1229.8	-5.6250	-3.9464	-2.7678	-5.5625	-9.8036	-0.0030	
e	----	0.0429	0.0346	0.0028	0.0402	0.0503							

The excess acoustical impedance (Z^E) generally shows a negative value and an increasing trend up to 0.06 m, after which a decrease is noted with Toluene. The maximum values are observed at 313.15 K (Fig. 1E). Similar patterns have been observed by other researchers [33, 34]. Positive excess values suggest interactions among dissimilar molecules [35]. There exists the potential for additional interactions between molecule constituents, and the formation of charge transfers in complexes is indicated by the positive deviations in acoustic impedance. Thus, deviations in acoustic impedance clearly suggest the presence of more charge transfer complexes.

The value of excess enthalpy shows an increasing trend with concentration and decreases at higher concentrations with negative values (Fig. 1F). These patterns suggest the possibility of molecular attraction during the mixing process, and another potential for molecular attraction due to polarisation between dissimilar molecules. This becomes dominant over the attraction between like molecules, and the effects of temperature lead to variations in these values, especially at low temperatures. Therefore, the deviations diminish at higher temperatures [36].

The experimental and computed ultrasonic velocities, along with their respective parameters for the mixture of toluene, chloroform, and cyclohexane, are presented in Table 1. From Table 1, the observed trend for ultrasonic velocity consistently increases with concentration up to 0.07 m, and a decreasing trend is noted across all temperatures.

From Table 1, IDR closely aligns with the experimental values at all temperatures, while IMR moderately does so. This suggests both a good and exceptional compatibility with the experimental results.

The molecular interactions between the components of a liquid combination may account for the percentage differences between observed and estimated sound velocities. In the context of relevant theories, such interactions have not been fully considered [37-39]. Moreover, they do not comprehensively account for the experimental manifestation of molecular interaction in various ultrasonic processes. The assumptions made in these theories regarding non-polar-non-polar and non-polar-polar interactions between molecules might explain the variation in velocities.

Of the various theories considered, IDR provides an outstanding prediction of sound velocity for all temperatures in this study. With respect to the combination of toluene, cyclohexane, and chloroform, the predictions from the IDR model closely match experimental results

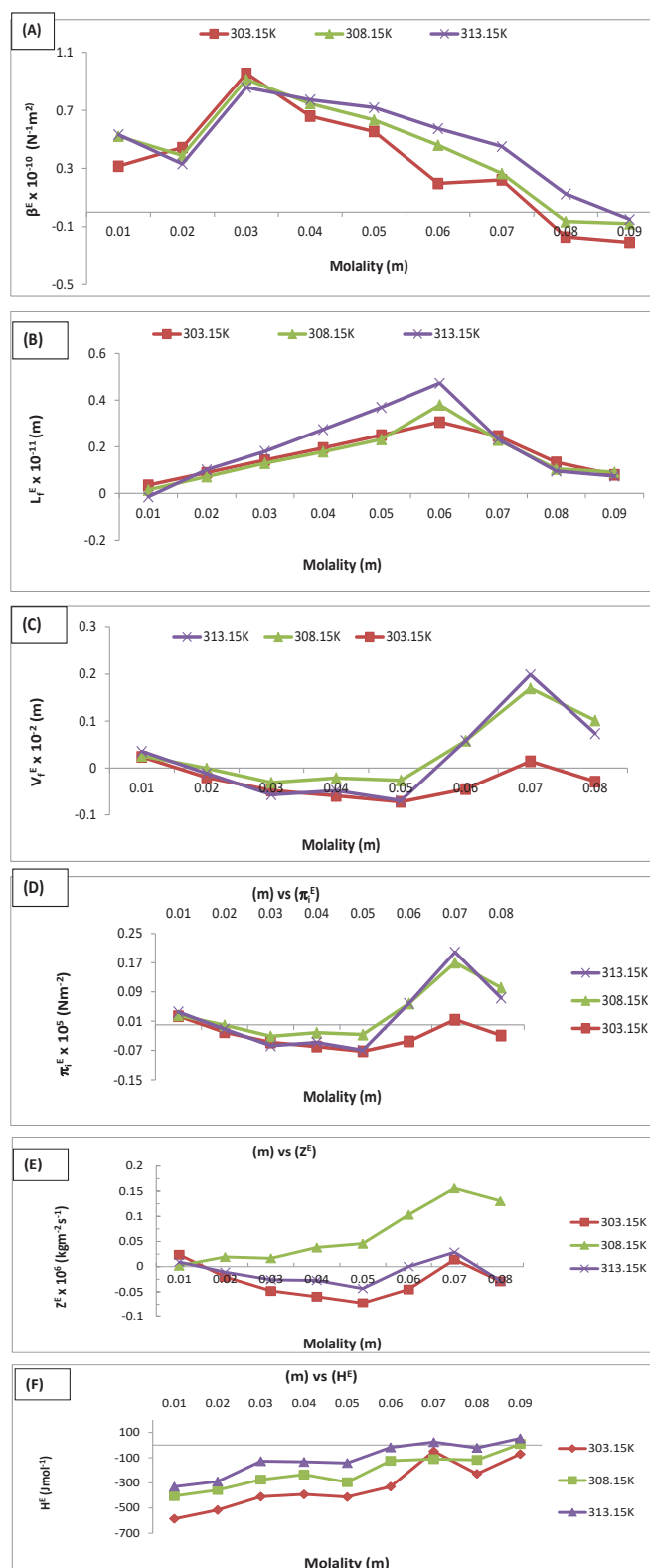


Fig. 1. Excess compressibility (A), excess free length (B), excess free volume (C), excess internal pressure (D), excess impedance (E), and excess enthalpy for various molalities (m) of Toluene + Chloroform + Cyclohexane (F) at various temperatures.

for most molarities in Table 1. Some alignment is also noted for several molarities in IMR. The percentage deviation being close to zero for both IDR and IMR reinforces the reliability of predictions for both in the ternary systems.

Molecular interaction largely depends on the strength of the molecules and is influenced by other factors such as dispersion forces [40]. Furthermore, the values of (α), computed from the IMR values and listed in Table 3, show that α is negative for ternary mixtures across all considered molarities. These negative values suggest the potential formation of intra-molecular hydrogen bonds [2] and further indicate weak dipolar and dispersive interactions. The maximum α value recorded is -0.1850 at 0.07 m, while the minimum is -0.02 and 0.05 at 0.01 and 0.10 m, respectively. This re-emphasises that cyclohexane exhibits much lower interactions compared to the other constituents of the mixture.

The overall molecular interactivity in a liquid mixture is influenced by multiple factors. Given its size, cyclohexane predominantly offers dispersive intra-molecular interactions. This confirms the dispersive interactions between similar molecules and weak dipolar interactions between dissimilar ones [41].

5. Conclusions

1. The in-depth analysis affirms the formation of hydrogen bonds between the molecules.
2. The introduction of toluene results in solute-solvent interactions at lower concentrations, and solvent-solvent interactions at higher ones.
3. Distinct types of interactions among the components are evident.
4. Among the theories considered in this study, IDR aligns exceptionally well with experimental values, while IMR does so to a lesser degree.
5. The mixtures approach ideal behaviour at higher concentrations.

CRedit author statement

S. Nithiyanantham: Methodology, Idea, Editing; V. Vanathi: Data analysis, Drawing figures; S. Mullainathan: Investigation and Writing; S. Mahalakshmi: Support to Editing and Investigation; L. Palaniappan: Validation and Writing.

COMPETING INTERESTS

The authors declare that there is no conflict of interest regarding the publication of this article.

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