



THEORETICAL EVALUATION OF SOUND VELOCITY, VISCOSITY AND DENSITY OF BINARY LIQUID SYSTEM

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ABSTRACT

Ultrasonic velocity, density and viscosity have been measured using the standard techniques in the binary mixtures of acetone with carbon tetrachloride. The study of ultrasonic waves through the solution is used for knowing the nature and strength of the intermolecular forces and their interactions in pure liquids and their mixtures. The theoretical ultrasonic velocity in binary mixtures computed using theoretical models of liquid mixtures such as Nomoto's relation, Ideal mixture relation, Impedance relation, Rao's specific velocity relation, Junjie's relation and Danusso model. The validity of the theories was interpreted in terms of average percentage error (APE). Further, various theoretical models estimate the liquid viscosity and density of the binary mixtures at atmospheric temperature and pressure, over the whole concentration range. The theoretical models studied in this work predict with accuracy of the liquid density of pure compounds. The relative merits of these relations were discussed.

Key words: Binary liquid mixtures, Ultrasonic velocity, Theoretical models, Viscosity empirical relations, Density, APE.

INTRODUCTION

In the recent years much important has been given to the behavior of mixed components rather than the single component because of their widespread range of applications. The practical need for thermodynamic data for investigation and research as well as for design and set up of industrial processes continue to drive research in the study of multicomponent systems. Thermodynamic properties derived from the measurement of ultrasonic velocities, densities and viscosities for binary mixtures are useful in understanding the nature and type of intermolecular interactions present between the constituent molecules. In chemical process industries, materials are normally handled in fluid form and as a

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consequence, the physical, chemical and transport properties of fluids, assume importance. Thus, data on some of the properties associated with the liquids and liquid mixtures like ultrasonic velocity, viscosity and density invention extensive application in solution theory models and molecular dynamics. The classification of mixtures through their thermodynamic and transport properties is substantial from the foremost viewpoint of understand their mixing behavior.

The impetus of the present investigation is to compare the experimentally determined ultrasonic velocity, viscosity and density with the computed values using different analytical models and empirical relations.

EXPERIMENTAL

Material and methods

The chemicals acetone used in present work are procured from Fisher scientific, Bombay with assay of 99.5% and Carbon tetra chloride from Sdfine Chemical limited, Bombay with assay of 99.8%. All compounds were of AR/GR grade, used without further purification. The ultrasonic velocities in pure and binary mixtures were measured using the single-crystal variable-path ultrasonic interferometer operating at 2MHz. The densities of pure and binary mixture were measured using the pre-calibrated standard 10ml specific gravity bottle with Teflon stopper and digital weighing balance with accuracy of ± 0.1 mg. Viscosity measurements were made by a pre-calibrated Ostwald's viscometer, using the method based on the measurement of flow time of the liquid. An electronic digital stop watch with reliability of ± 0.01 sec was used for flow time measurements. The measurement of flow time of the solution between the two points on the viscometer was performed at least five times for each solution and the result was averaged.

Theory and calculations

The various acoustical parameters computed using the following relations,

$$\text{Adiabatic compressibility } \beta_s = 1/U^2 \rho$$

$$\text{Linear free length } L_f = K/\sqrt{\rho} U$$

Where K is the temperature independent Jacobson's constant

$$\text{Acoustic impedance } Z = \rho U$$

$$\text{Molar volume } V_m = M_{eff}/\rho$$

Where $M_{eff} = \sum_i x_i M_i$

Available volume $V_a = V_m \left(1 - \frac{U_{exp}}{U_\infty}\right)$

Where $U_\infty = 1600$ m/s

The evaluation of theoretical values of ultrasonic velocity, viscosity and density in binary mixture those obtained from the experimentally in the binary mixture is expected to interpretation the nature of interaction between components molecules. Such theoretical study is useful in the wide-ranging theoretical model for the liquid mixtures.

Van Dael & Vangel Ideal Mix relation⁴

$$U_{VV} = \sum_i x_i / m_i u_i^2 \times \sum_i 1 / x_i m_i$$

Where x_i is mole fraction, m_i is molecular weight, and u_i is ultrasonic velocity of i^{th} components respectively.

Impedance relation⁵

$$U_{IMP} = \sum_i x_i z_i / \sum_i x_i \rho_i$$

Where x_i is mole fraction, z_i is the acoustic impedance and ρ_i is the density of the i^{th} components, respectively.

Nomoto's relation⁶

$$U_{NOM} = \left(\sum_i R_i / \sum_i V_i \right)^3$$

$$R_i = m_i / \rho_i \times u_i^{1/3}$$

$$V_i = m_i / \rho_i$$

Where m_i is molecular weight, ρ_i is the density and u_i is ultrasonic velocity of i^{th} components, respectively.

Jungie's relation⁷

$$U_{JR} = \left[\frac{\sum_i x_i V_i}{\sum_i x_i m_i^{1/2}} \right] \times \left[\frac{\sum_i x_i V_i}{\sum_i \rho_i U_i^2} \right]^{-1/2}$$

Where x_i is mole fraction, V_i is molar volume, m_i is molecular weight, ρ_i is the density and u_i is ultrasonic velocity of i^{th} components respectively.

Rao's specific velocity⁷

$$U_R = \sum_i (x_i r_i \rho_i)^3$$

Where x_i is mole fraction, ρ_i is the density of i^{th} component and r_i is the Rao's specific sound velocity $r_i = U_i^{1/3}/Z_i$ and Z_i is acoustic impedance.

Danusso model⁸

$$U_D = (1/\rho_{mix}) \left(1/M_{eff} \times \sum_i x_i m_i / \rho_i^2 U_i^2 \right)$$

$$M_{eff} = x_i M_i$$

Where M_{eff} is the effective molecular weight of the solute and solvent, x_i is mole fraction, m_i is molecular weight of i^{th} component.

Molecular association

The degree of intermolecular interaction or molecular association is given by

$$\alpha = (U_{exp} - U_{comp}) - 1$$

Viscosity deviation

The viscosity deviation evaluated from following relation,

$$\Delta\eta = \eta_m - \sum_i x_i \eta_i$$

Where η_m is the viscosity of the mixture and x_i is mole fraction, η_i is viscosity of i^{th} component.

The viscosity of binary liquid mixtures based on zero adjustable parameter computed using following relations

Kendall and monroe⁹

$$\eta_m = (x_1 \eta_1^{1/3} + x_2 \eta_2^{1/3})^3$$

Modified Kendall-Monroe equation⁹

$$E\eta_m = x_1x_2(x_1\eta_1^{1/3} + x_2\eta_2^{1/3})^3$$

Arrhenius⁹

$$\log \eta_m = x_1 \log \eta_1 + x_2 \log \eta_2$$

Where x_1 , x_2 and η_1 , η_2 are the mole fraction and viscosity of pure component respectively.

The viscosity of binary liquid mixtures based on adjustable parameter computed using following relations

Grunberg–Nissan¹⁰

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1x_2d_{12}$$

Where d_{12} is the Grunberg-Nissan interaction parameter.

Katti and Chaudhri¹¹

$$\ln \eta V = x_1 \ln V_1\eta_1 + x_2 \ln V_2\eta_2 + x_1x_2 W_{vis}/RT$$

Where W_{vis} represents the interaction energy between the components, V is the molar volume and others were usual meaning.

Hind-Ubbelohde¹²

$$\eta = x_1^2\eta_1 + x_2^2\eta_2 + 2x_1x_2H_{12}$$

Where H_{12} is Hind interaction parameter, it is attributed to unlike pair interaction and others were usual meaning.

Wijk¹³

$$\lg \eta = x_1^2 \lg \eta_1 + x_2^2 \lg \eta_2 + 2x_1x_2 \lg \eta_{12}$$

Where η_{12} interaction parameter and others were usual meaning.

Mchaweh–Nasrifar-Mashfeghian (MNM) model¹⁹

$$\rho_{MNM} = \rho_{cmix}\rho_{0mix}$$

Where ρ_{mix} is the solution density and ρ_{cmix} is the critical density of mixture.

The critical density of the mixture is calculated with the following equation.

$$\rho_{cmix} = \left[\sum_{i=1}^N x_i \rho_{ci}^{-3/4} \right]^{-4/3}$$

Where x_i is the mole fraction and ρ_{ci} is the critical density of the i^{th} component.

The parameter ρ_{0mix} is the reference density of the mixture and calculated with the following equation.

$$\rho_{0mix} = 1 + 1.169 \tau_{mix}^{1/3} + 1.818 \tau_{mix}^{2/3} - 2.658 \tau_{mix}^{3/3} + 2.161 \tau_{mix}^{4/3}$$

Where the temperature dependent variable τ_{mix} is calculated by the following expression

$$\tau_{mix} = 1 - \frac{T_{mix}}{\alpha_{SRK}}$$

In above equation T_{mix} is the reduced temperature of the mixture α_{SRK} is the term from the original Soave-Redlich-Kwong equation of state.

The reduced temperature is defined as

$$T_{mix} = \frac{T}{\sum_{i=1}^N x_i T_{ci}}$$

Where T_{ci} is the critical temperature of the i^{th} component.

The parameter α_{SRK} is defined in term of reduced temperature T_r

$$\alpha_{SRK} = [1 + m(1 - \sqrt{T_{mix}})]^2$$

Where m is calculated with the following equation

$$m = 0.480 + 1.571\omega_{mix} - 0.176\omega_{mix}^2$$

The acentric factor of the solution ω_{mix} is calculated with the following expression

$$\omega_{mix} = \sum_{i=1}^N x_i \omega_i$$

Where ω_i is the acentric factor of the i^{th} component. The acentric factor is a measure of the complexity of the molecule as form in relation to a molecule with spherical symmetry of the simple fluid for which $\omega = 0$.

Hankinson and Thomson (HT) model¹⁹

Based on the corresponding state principle and is valid of $0.25 < T_r < 0.95$. The density of the compound is defined by

$$\rho_{HT} = \frac{\rho_c}{[V^{(0)}(1 - \omega V^{(1)})]}$$

$$V^{(0)} = 1 - 1.5281(1 - T_r)^{1/3} + 1.4390(1 - T_r)^{2/3} - 0.8144(1 - T_r) + 0.19045(1 - T_r)^{4/3}$$

$$V^{(1)} = (-0.296123 + 0.386914 T_r - 0.0427258 T_r^2 - 0.0480645 T_r^3)/(T_r - 1.00001)$$

The reduced temperature of the component is defined as

$$T_r = \frac{T}{T_c}$$

Yamada and Gunn (YG) model¹⁹

Yamada - Gunn model extended the Racket equation and requires the molecular weight M the critical density ρ_c , the reduced temperature T_r and the acentric factor ω .

$$\rho_{YG} = \rho_c(0.29056 - 0.08775 \omega)^{-(1-T_r)^{2/7}}$$

Reid et al. (RR) model¹⁹

Reid et al. model proposed an equation based also on the molecular weight, critical density, reduced temperature and acentric factor.

$$\rho_{RR} = \rho_c [1 + 0.85(1 - T_r) + (1.6916 + 0.984\omega)(1 - T_r)^{1/3}]$$

In order to extend the Hankinson-Thomson, Yamada-Gunn and Reid et. al models to multicomponent systems,

$$\rho_{mix} = \sum_{i=1}^N x_i \rho_i$$

$$T_{mix} = \frac{T}{\sum_{i=1}^N x_i T_{ci}}$$

$$\omega_{mix} = \sum_{i=1}^N x_i \omega_i$$

Percentage error

The percentage of deviation between experimental and computed values can be calculated as –

$$\text{Percentage deviation } \Delta U/U\% = [(D_{exp} - D_{comp})/D_{exp}] \times 100\%$$

The measured physical parameters such as ultrasonic velocity, density, viscosities data are recorded sequential in Microsoft Excel 2010 and the various acoustical, thermodynamical, viscosity parameters, densities are computed using the user friendly developed package in VB.NET language running under 64-bit Windows 8 platform.

RESULTS AND DISCUSSION

The experimental values of ultrasonic velocity U , density ρ and viscosity η of binary mixtures of acetone with carbon tetrachloride and over the entire composition range (0.0 to 1.0) expressed in terms of mole fraction x_1 are listed in Table 1.

Table 1: Experimental value of ultrasonic velocity U , density ρ and viscosity η of acetone with carbon tetra chloride

Mole fraction acetone x_1	Velocity U m/s	Density ρ Kg/m ³	Viscosity η x 10 ⁻⁴ Ns/m ²
0.0000	992.00	1592	9.1241
0.1295	952.00	1525	8.0020
0.2508	852.80	1554	7.8030
0.3646	990.40	1382	6.7205
0.4716	1081.60	1281	6.0481
0.5725	1100.80	1201	5.3894

Cont...

Mole fraction acetone x_1	Velocity U	Density ρ	Viscosity η
	m/s	Kg/m ³	$\times 10^{-4}$ Ns/m ²
0.6676	1054.40	1122	5.1407
0.7575	1003.20	1036	4.7559
0.8426	1120.00	952	4.0891
0.9233	1236.80	872	3.8471
1.0000	860.80	805	3.5043

The computed acoustical, thermodynamical parameters listed in Table 2. The change in ultrasonic velocity shows the growing of molecular interactions between solute and solvent molecules. The decrease in viscosity and density with increase in mole fraction of component x_1 indicates formation of intermolecular forces between the binary mixtures.

Table 2: Computed acoustical thermodynamical parameters of acetone with carbon tetra chloride

Mole fraction acetone x_1	Compressibility β	Linear free length L_f	Acoustic impedance Z	Molar volume V_m	Available volume V_a
	$\times 10^{-10}$ kg ⁻¹ ms ⁻²	$\times 10^{-10}$ m	$\times 10^5$ kg/m ² s	$\times 10^{-2}$ m ³	$\times 10^{-3}$ m ³
0.0000	6.38313	5.2425	15.7926	9.6621	3.6716
0.1295	7.23530	5.5814	14.5180	9.2734	3.7557
0.2508	8.84819	6.1723	13.2525	8.3530	3.9009
0.3646	7.37685	5.6358	13.6873	8.6041	3.2782
0.4716	6.67294	5.3601	13.8552	8.4826	2.7483
0.5725	6.87132	5.4392	13.2206	8.2439	2.5721
0.6676	8.01671	5.8751	11.8303	8.0125	2.7323
0.7575	9.59103	6.4261	10.3931	7.8466	2.9268
0.8426	8.37389	6.0046	10.6624	7.6829	2.3049
0.9233	7.49695	5.6815	10.7848	7.5017	1.7029
1.0000	16.76485	8.4961	6.92944	7.2149	3.3333

The variation of compressibility β for acetone with CCl_4 indicates the molecular association and hydrogen bond formation between unlike molecules. The deviation of intermolecular free length L_f with increasing in mole fraction of component x_1 proposes that the molecular interaction between the constituents of two system binary mixture molecules. According to Eyring and Kincaid¹⁴, the ultrasonic velocity increases, if the L_f decreases and vice-versa in a result of mixing components. The decrease or increase in values of L_f can be explained on the basis of interactions between the solute and solvent molecules. The nonlinear variation of acoustic impedance Z exposes that presence of specific interaction and cluster formation between the mixing elements. The molar volume V_m changes in decreasing trend and available volume V_a changes non-linearly with concentration of solutions.

From the various computed acoustical, thermodynamical parameters for the binary mixtures suggest that the specific chemical interactions directed towards the molecular interaction prevailing in the present systems of the binary mixtures. Such interactions are due to dipole-dipole and dipole-induced dipole forces. Dispersive forces are also found to exist between the components of the mixtures¹⁵⁻¹⁸.

The computed theoretical ultrasonic theoretical velocities are listed in Table 3 for the binary system acetone with carbon tetra chloride, the corresponding percentage of deviation and interaction parameters (α) are listed in Table 4.

Table 3: Experimental velocities and computed theoretical velocities for acetone with carbon tetra chloride binary system in m/s

Mole fraction acetone x_1	U_{EXP}	U_{NOM}	U_{JR}	U_{VV}	U_R	U_{IMP}	U_D
0.1295	952.00	978.31	943.62	898.43	974.30	982.81	936.38
0.2508	852.80	964.76	907.74	845.45	957.92	973.00	837.99
0.3646	990.40	951.33	881.20	814.69	942.71	962.48	864.56
0.4716	1081.60	938.02	862.05	798.03	928.57	951.19	859.50
0.5725	1100.80	924.85	849.06	791.38	915.37	939.02	847.29
0.6676	1054.40	911.79	841.46	792.53	903.03	925.88	839.81
0.7575	1003.20	898.86	838.84	800.34	891.47	911.65	842.97
0.8426	1120.00	886.05	841.07	814.27	880.62	896.17	850.26
0.9233	1236.80	873.36	848.26	834.30	870.41	879.29	859.64

Table 4: Percentage deviation for acetone with carbon tetra chloride binary system.

Mole fraction acetone x_1	% U_{NOM}	% U_{JR}	% U_{VV}	% U_R	% U_{IMP}	% U_D	α
0.1295	-2.7645	0.8802	5.6272	-2.3428	-3.2373	1.6406	0.12281
0.2508	-13.1289	-6.4427	0.8609	-12.3267	-14.0953	1.7357	0.01744
0.3646	3.9445	11.0251	17.7405	4.8144	2.8182	12.7055	0.47784
0.4716	13.2739	20.2979	26.2171	14.1485	12.0569	20.5343	0.83691
0.5725	15.9839	22.8686	28.1086	16.8449	14.6959	23.0292	0.93485
0.6676	13.5249	20.1951	24.8352	14.3557	12.1882	20.3516	0.76999
0.7575	10.4005	16.3830	20.2211	11.1370	9.1256	15.9714	0.57117
0.8426	20.8882	24.9042	27.2968	21.3731	19.9842	24.0839	0.89188
0.9233	29.3851	31.4149	32.5436	29.6236	28.9058	30.4947	1.19763

The observed deviations of the theoretical ultrasonic velocity from the experimental values are attributed to the presence of intermolecular interactions between the component molecules of the mixture. The observed deviations following all models illustrate molecular interaction between constituent molecules in the liquid mixtures. The suitability of these theories based on percentage deviation. The overall comparison of percentage deviation of this system shows the following trend

$$U_{VV} < U_D < U_{JR} < U_D \ll U_R < U_{NOM} < U_{IMP}$$

The viscometric information includes viscosity as a function of composition on the bases of weight, volume, and mole fraction, comparison of experimental viscosities with those calculated with several different equations. Viscosity itself is consequently not a simple additive property. The computed viscosity interaction parameters and Computed theoretical viscosities are listed in Tables 5, 6 and 7, respectively.

The values of $\Delta\eta$ were negative, indicating that dispersion forces are primarily responsible for interaction. The viscosity of a mixture⁹⁻¹³ depends on the molecular interactions between the components mixture with strong interactions between different molecules show positive viscosity deviations, while for mixtures absence of specific interactions, viscosity deviations are negative. The negative viscosity deviation in the

present investigation suggests that without specific interactions between component molecules over the entire composition range in all the binary mixtures. The experimental viscosity excellent agreement with various analytical models such as with Grunberg Nissan, Katti and Chaudhri, Hind and Wijk theoretical approaches, whereas, Kendall-Monroe, Modified Kendall-Monroe, Arrhenius shows significant deviations between theoretical and experimental values of viscosities.

Table 5: Computed viscosity interaction parameters for acetone with carbon tetra chloride binary system

Mole fraction acetone x_1	Grunberg Nissan d_{12}	Katti and Chaudhri W_{vis}	Hind $H_{12} \times 10^{-4}$	Wijk $\eta_{12} \times 10^{-4}$
0.1295	-0.0646	-234.8451	4.5661	5.4748
0.2508	0.4449	151.2426	6.5496	7.0634
0.3646	0.1864	366.6239	5.5495	6.2069
0.4716	0.1613	482.7251	5.4609	6.1293
0.5725	0.0873	306.9742	5.2572	5.9067
0.6676	0.2936	828.0870	5.7928	6.5487
0.7575	0.3996	1186.8913	6.0124	6.9052
0.8426	0.0286	393.3705	5.1854	5.7361
0.9233	0.2829	1304.1763	5.6940	6.5138

Table 6: Experimental viscosity, viscosity deviation and computed theoretical viscosities for acetone with carbon tetra chloride binary system.

Mole fraction acetone x_1	Experimental viscosity η $\times 10^{-4}$ Ns/m ²	Viscosity deviation $\Delta\eta$ $\times 10^{-5}$	Grunberg Nissan $GN\eta$ $\times 10^{-4}$ Ns/m ²	Katti and Chaudhri $KC\eta$ $\times 10^{-4}$ Ns/m ²	Hind $H\eta$ $\times 10^{-4}$ Ns/m ²	Wijk $W\eta$ $\times 10^{-4}$ Ns/m ²
0.1295	8.0020	-3.9420	8.0020	8.0020	8.0020	8.0020
0.2508	7.8030	0.8846	7.8030	7.8030	7.8030	7.8030
0.3646	6.7205	-3.5434	6.7205	6.7205	6.7205	6.7205
0.4716	6.0481	-4.2526	6.0481	6.0481	6.0481	6.0481

Cont...

Mole fraction acetone x_1	Experimental viscosity η $\times 10^{-4}$ Ns/m ²	Viscosity deviation $\Delta\eta$ $\times 10^{-5}$	Grunberg Nissan $GN\eta$ $\times 10^{-4}$ Ns/m ²	Katti and Chaudhri $KC\eta$ $\times 10^{-4}$ Ns/m ²	Hind $H\eta$ $\times 10^{-4}$ Ns/m ²	Wijk $W\eta$ $\times 10^{-4}$ Ns/m ²
0.5725	5.3894	-5.1736	5.3894	5.3894	5.3894	5.3894
0.6676	5.1407	-2.3141	5.1407	5.1407	5.1407	5.1407
0.7575	4.7559	-1.1086	4.7559	4.7559	4.7559	4.7559
0.8426	4.0891	-2.9927	4.0891	4.0891	4.0891	4.0891
0.9233	3.8471	-0.8775	3.8471	3.8471	3.8471	3.8471

Table 7: Computed theoretical viscosities for acetone with carbon tetra chloride binary system

Kendall -Monroe $KM\eta$ $\times 10^{-3}$ Ns/m ²	Modified Kendall-Monroe $MKM\eta$ $\times 10^{-4}$ Ns/m ²	Arrhenius $Ar\eta$ $\times 10^{-4}$ Ns/m ²
2.1109	2.3800	8.0605
2.3289	4.3763	7.1771
2.3926	5.5433	6.4364
2.3680	5.9010	5.8099
2.2794	5.5787	5.2755
2.1369	4.7417	4.8165
1.9413	3.5655	4.4193
1.6831	2.2312	4.0736
1.3255	0.9376	3.7709

Table 8 show the values deviation of densities between the experimental and computed values. The overall comparison of percentage deviation of this system shows the following trend

$$\rho_{MNM} > \rho_{YG} > \rho_{RR} > \rho_{HT}$$

Table 8: Percentage of deviation in density for acetone with carbon tetra chloride binary system

Mole fraction acetone x_1	MNM model	HT model	YG model	RR model
0.1295	-83.63	11.97	89.54	16.50
0.2508	-173.02	19.41	90.48	24.07
0.3646	-315.47	15.57	90.08	20.97
0.4716	-475.94	15.26	90.09	21.18
0.5725	-673.99	16.07	90.24	22.41
0.6676	-951.51	16.75	90.36	23.49
0.7575	-1409.56	16.66	90.39	23.83
0.8426	-2349.16	16.39	90.39	24.00
0.9233	-5574.24	16.13	90.40	24.16

CONCLUSION

In the present investigational work, the experimental ultrasonic velocities densities, and viscosities of binary mixtures for the acetone with carbon tetra chloride determined as function of composition at temperature 303.15K. Using these data, various acoustical and thermodynamical parameters were computed with the standard relations obtained from the literature survey. All these parameters indicate the presence of precise interactions between the subjected system components of constituent molecules. Again, the computed ultrasonic velocities, viscosities and densities for binary mixture from different theories and various analytical models have been interrelated with the experimentally measured values.

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