



VISCOMETRIC STUDY OF TERNARY LIQUID MIXTURES USING ULTRASONIC VELOCITY AT 298.15K

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Abstract

Different liquid state models [1-7] for viscosity suggested by Bingham, Kendall-Munroe, Eyring and Gambill have been applied to three ternary liquid systems i.e., benzene + cyclohexane + toluene, toluene + cyclohexane + carbon tetrachloride, ethanol + methanol + TAME (2 methoxy + 2 methyl butane) at 298.15 K to calculate viscosity using the properties of pure liquids [20]. Obtained values are compared with the calculated viscosity data from correlation formula [18].

Keywords: Viscosity, Ternary liquid systems, Bingham method, Kendall-Munroe method, Eyring and Gambill method.

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Introduction

Viscometric study of ternary liquid mixtures using ultrasonic velocity is a fascinating area of research that involves investigating the behaviour of three-component liquid mixtures at a specific temperature of 298.15K using ultrasonic techniques. Ultrasonic velocity, which refers to the speed of sound waves propagating through a medium, can provide valuable insights into the physical and chemical properties of liquids, including their viscosity, density, and molecular interactions. By studying the viscometric behaviour of ternary liquid mixtures using ultrasonic velocity, researchers can gain a deeper understanding of the complex interactions among the three components and how they affect the overall properties of the mixture. This type of research has important applications in fields such as chemical engineering, pharmaceuticals, and material science, and can contribute to the development of new industrial processes and formulations. In this study, we will explore the principles of ultrasonic velocity measurement, the theoretical background of viscometry, the experimental setup and methodology, and the potential outcomes and implications of the viscometric study of ternary liquid mixtures at 298.15K.

Thermodynamic and transport properties are very important tool to design liquid systems and to understand their behaviour. These properties provide information about the molecular interactions between the various liquids. As viscosity is a transport property and is very useful to study the physiochemical behaviour and molecular interactions in various liquid systems. A lot of work has been done by various workers [8-17]. In this work we have done a comparative study to compute viscosity for three ternary liquid mixtures namely.

- (i) Benzene (x1) + cyclohexane (x2) + toluene (x3)
- (ii) Toluene (x1) + cyclohexane (x2) + carbon tetra chloride (x3)
- (iii) ethanol (x1) + methanol (x2) + TAME (x3)

Using different liquid state models i.e., Bingham, Kendall-Munroe, Eyring, Gambill and that of a correlation formula.

Theoretical Formulations

The Bingham, Kendall-Munroe, Eyring, and Gambill models are commonly used to describe the viscosity of liquids. These models provide mathematical expressions that can be used to estimate the viscosity of liquid mixtures based on

the composition of the mixture. In the context of the viscometric study of ternary liquid systems, these models can be applied to analyze and predict the viscosity behaviour of the mixtures.

Viscosity: To compute the dynamic viscosity of the liquid mixtures at temperature 298.15K and 323.15K various models using different approaches have been employed Each of them is as follows:

- 1- Bingham Model [1]:** The Bingham model is a simple empirical model that can be expressed as:

$$\eta = x_1 \eta_1 + x_2 \eta_2$$

The above relation is given considering the ideal mixing concept, where x_1 , x_2 are the mole fractions of two components respectively. η_1 , η_2 are the dynamic viscosities of pure substances, η is the dynamic viscosity of liquid mixture formed by mixing them.

- 2- Kendall & Munroe [1-4]:** The Kendall-Munroe model is a two-parameter model that considers both the concentration and temperature dependence of viscosity. The relation is given by taking logarithmic additives:

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2$$

- 3- Eyring's Approach [1, 3]:** The Eyring model is a thermodynamic model that relates viscosity to the activation energy and the temperature. It can be expressed as:

$$\ln(\eta V) = x_1 \ln(\eta_1 V_1) + x_2 \ln(\eta_2 V_2)$$

Where V_1 , V_2 are molar volumes of pure components and V is the molar volume of the mixture.

- 4- Gambill's Approach [1, 6]:** The Gambill model is a semi-empirical model that combines the Bingham and Kendall-Munroe models to account for both yield stress and concentration-temperature dependence of viscosity, pointed out that the error in viscosity is the least when kinematic viscosity of each component is raised to a power of one third.

$$V^{(1/3)} = x_1 v_1^{(1/3)} + x_2 v_2^{(1/3)}$$

$$V = (\eta/p) \text{ where } p \text{ is the density}$$

- 5- Correlation Formula [17]:** Now we have a correlation formula to find the viscosity of the liquid systems: -

$$\eta = N x (u^{3/2} M^{7/12} p^{3/4}) / T^{5/36}$$

The constant N in the above equation was found to vary from 1.2×10^{-6} to 4.0×10^{-6}

Here we consider

$$\eta = 2.2 \times 10^{-6} (u^{3/2} M^{7/12} \rho^{3/4}) / T^{5/36}$$

Where the units η , u , ρ , M and T are in centipoises (cP), ms^{-1} , gcm^{-3} , g and K respectively. The importance of above equation is that to compute the viscosity of liquid system directly, we need only to measure velocity, density and temperature. The value of M will be obtained from mole fraction additively, i.e.,

$$M_{1234} = x_1 M_1 + x_2 M_2 + x_3 M_3 + x_4 M_4$$

The theoretical values of η for all the systems calculated from this relation using density and sound velocity. Data reported in table 2, 3 and 4. For the comparison obtained values of η are also given. All the reported values are at temperature of 298.15 K. The experimental data of density and ultrasonic velocity for the mentioned system were taken from literature [18].

Results and discussion

These models can be applied to three ternary liquid systems to analyze and predict the viscosity behaviour of the mixtures. The choice of the appropriate model depends on the specific characteristics of the ternary liquid systems, such as the nature of the components, the temperature range of interest, and the rheological behaviour observed in the experimental data. Proper parameterization of these models using experimental data can provide valuable insights into the viscosity behaviour of ternary liquid mixtures and aid in the understanding of their rheological properties.

From the present study three ternary liquid mixtures namely.

1. benzene (x_1) + cyclohexane (x_2) + toluene (x_3)
 2. toluene (x_1) + cyclohexane (x_2) + carbon tetra chloride (x_3)
 3. ethanol (x_1) + methanol (x_2) + TAME (x_3)
- have been considered. The experimental data of density and ultrasonic velocity for the mentioned system were taken from literature (19). The parameters of the pure components are recorded in table 1.

Table 1 Estimated values of molecular parameters of pure liquids at 298.15K

Pure liquids	Viscosity(η) mPas	Density(ρ) g/cc	Mol. wt (g)	Molar volume (g/mol)	Kinematic Viscosity ν $10^{-2} \times \text{cm}^2/\text{sec}$
Benzene	0.5985	0.574	78.14	89.405	0.6847
Cyclohexane	0.8799	0.774	84.16	108.734	1.1368
Toluene	0.6035	0.863	92.14	106.767	0.6990
CCl ₄	0.8992	1.585	153.82	97.047	0.5673
Ethanol	1.074	0.785	46.00	58.598	1.368
Methanol	0.594	0.787	32.00	40.660	0.7547
TAME	0.383	0.766	102.00	133.159	0.5
n-Pentane	0.274	0.6262	72.15	115.22	0.4375
n-Heptane	0.387	0.6795	100.205	147.469	.5695
n-Decane	0.833	0.7255	142.2	196.00	1.1481
n-Hexane	0.405	0.6606	86.18	130.454	0.6131

Computation of viscosity for three ternary liquid systems have been carried out at 298.15K by

employing four different approaches reported in table 2, 3 and 4.

Table 2 Computed values of viscosity for three ternary liquid systems by employing four different approaches at 298.15K
BENZENE(X1) + CYCLOHEXANE (X2) + TOULENE (X3)

X1	X2	ρ (g/cc)	u (m/sec)	(correlation) η (mPas)	Bingham η (mPas)	Kendal η (mPas)	Eyring η (mas)	Gambill η (mpPas)
0.133	0.212	0.844	1291	0.5570164	0.6613	0.6529	0.2267	0.65674
0.144	0.212	0.845	1290	0.5562987	0.66125	0.6528	0.2265	0.65738
0.164	0.23	0.843	1269	0.5402908	0.66613	0.6571	0.2267	0.66159
0.185	0.246	0.842	1268	0.5376636	0.67046	0.661	0.227	0.66592
0.23	0.283	0.837	1268	0.5319615	0.68047	0.6701	0.2271	0.67385
0.276	0.32	0.833	1248	0.5142829	0.69049	0.6792	0.2274	0.6826
0.298	0.365	0.83	1249	0.5111749	0.70283	0.6908	0.2297	0.69528
0.339	0.374	0.829	1259	0.5145657	0.70512	0.6929	0.2285	0.697
0.361	0.391	0.827	1265	0.5157226	0.70972	0.6972	0.2286	0.70091
0.382	0.409	0.826	1265	0.5136866	0.7146	0.7018	0.229	0.70603
0.405	0.418	0.823	1272	0.5151227	0.71697	0.7041	0.228	0.70628
0.425	0.425	0.824	1282	0.5204536	0.71882	0.7059	0.2281	0.70931
0.444	0.436	0.823	1278	0.516257	0.72177	0.7087	0.228	0.71204
0.455	0.431	0.823	1271	0.5116078	0.72033	0.7073	0.2277	0.71014

Table 3 Computed values of viscosity for three ternary liquid systems by employing four different approaches at 298.15K

Toluene(x1) + Cyclohexane (x2) + Carbon tetrachloride								
X1	X2	ρ (g/cc)	u (m/sec)	(correlation) η (mPas)	Bingham η (mPas)	Kendal η (mPas)	Eyring η (mPas)	Gambill η (mPas)
0.113	0.204	1.318	984	0.6552593	0.86183	0.8558	0.4921	0.89389
0.127	0.224	1.285	998	0.6501443	0.8573	0.8506	0.4786	0.88693
0.154	0.3	1.206	1021	0.6212821	0.84784	0.8401	0.4487	0.88435
0.173	0.263	1.22	1094	0.6996877	0.84293	0.8344	0.4497	0.87438
0.191	0.282	1.189	1055	0.6425181	0.83724	0.8281	0.4367	0.86703
0.213	0.303	1.158	1064	0.6293747	0.83033	0.8205	0.4233	0.86098
0.232	0.326	1.114	1077	0.6141242	0.82426	0.8139	0.4057	0.8451
0.252	0.344	1.094	1139	0.6506745	0.81799	0.8071	0.3967	0.84387
0.274	0.362	1.061	1146	0.6331776	0.81114	0.7998	0.3828	0.83241
0.297	0.386	1.022	1157	0.6143875	0.80387	0.792	0.3669	0.81902
0.313	0.408	0.996	1168	0.6029304	0.79871	0.7866	0.3566	0.81298
0.336	0.428	0.968	1148	0.5661387	0.79152	0.7791	0.3448	0.80464
0.357	0.441	0.944	1215	0.5973654	0.78505	0.7724	0.3345	0.7949
0.376	0.45	0.921	1235	0.5946928	0.77926	0.7664	0.3247	0.78314
0.396	0.461	0.9	1242	0.5825143	0.77313	0.7601	0.3157	0.77396

Table 4 Computed values of viscosity for three ternary liquid systems by employing four different approaches at 298.15K

Ethanol (x1) + Methanol (x2) + TAME								
X1	X2	ρ (g/cc)	u (m/sec)	(correlation) η (mPas)	Bingham η (mPas)	Kendal η (mPas)	Eyring η (mPas)	Gambill η (mPas)
0.78	0.132	0.783	1139	0.3091904	0.94983	0.9079	0.2368	0.9218
0.704	0.217	0.784	1136	0.3020127	0.91525	0.8713	0.219	0.88643
0.622	0.309	0.784	1133	0.2939171	0.878	0.8336	0.201	0.84819
0.541	0.398	0.784	1130	0.2863477	0.84081	0.7972	0.1848	0.81098
0.448	0.502	0.785	1126	0.2771624	0.79849	0.7581	0.1679	0.77109
0.345	0.617	0.785	1122	0.2667988	0.75158	0.7169	0.1508	0.72735
0.267	0.703	0.785	1118	0.2588399	0.71583	0.6869	0.1391	0.69508
0.19	0.788	0.786	1114	0.2511305	0.68056	0.6585	0.1286	0.66504
0.107	0.881	0.786	1109	0.2418751	0.64283	0.6296	0.1179	0.63307
0.797	0	0.781	1141	0.3388953	0.93373	0.873	0.2617	0.89348
0.698	0.124	0.781	1138	0.3266697	0.89148	0.8322	0.2336	0.851
0.622	0.219	0.782	1135	0.3172258	0.85901	0.802	0.2144	0.82031
0.551	0.308	0.782	1133	0.3082885	0.82873	0.775	0.1976	0.79141
0.488	0.387	0.783	1130	0.3000581	0.80187	0.7517	0.1841	0.76734
0.379	0.524	0.784	1126	0.2857825	0.75545	0.7132	0.1626	0.7262

Obtained viscosity values have a very similar trend for each system (Figure 1, 2 and 3). The lack of smoothness in the deviations are due to the interaction between the component molecules. In the collision factor theory, the molecules are treated as real, non- elastic molecules and the

interactions result due to the collision between the component molecules. Considering these approximation, excellent agreement is achieved by all liquid state models to that of correlation formula. The little deviations are due to the experimental uncertainties in the viscosity values.

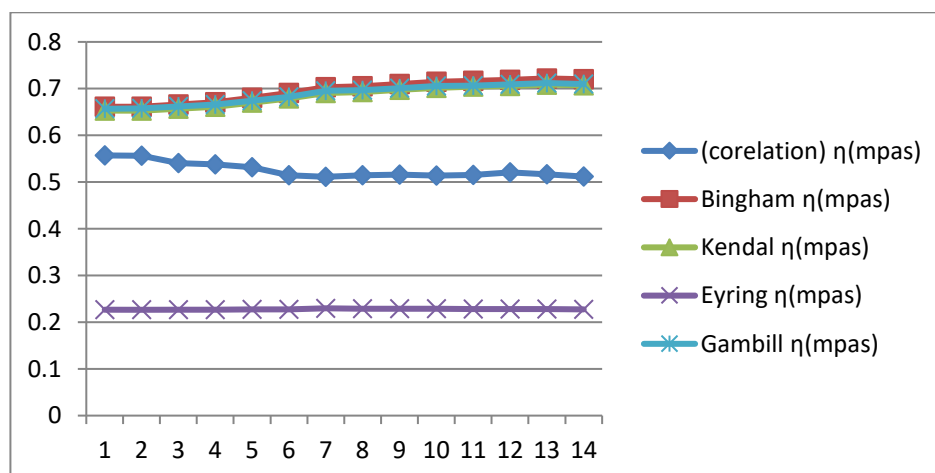


Figure 1- comparison of dynamic viscosity for ternary system Benzene + Cyclohexane + Toluene

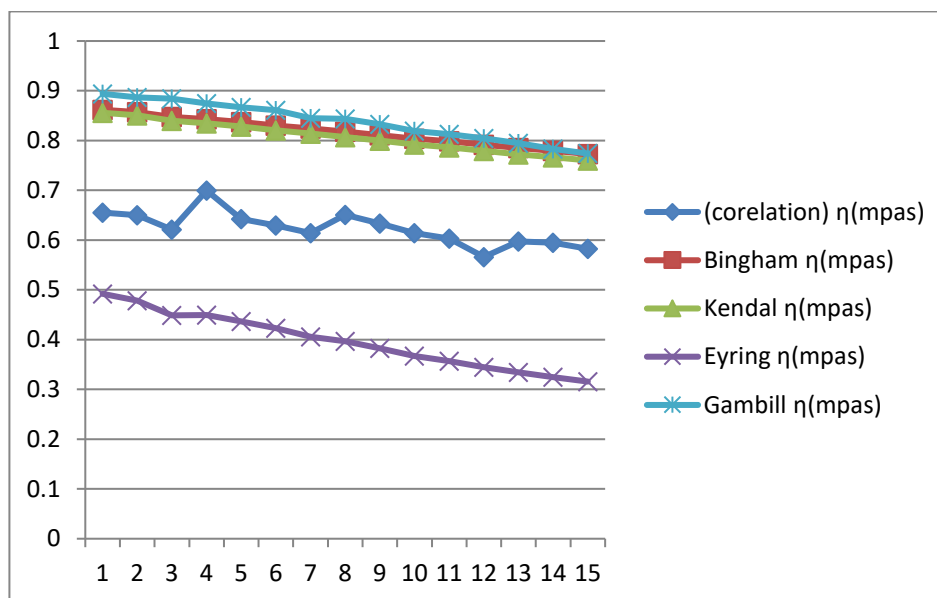


Figure 2- comparison of dynamic viscosity for ternary system Toluene+ Cyclohexane + Carbon tetrachloride

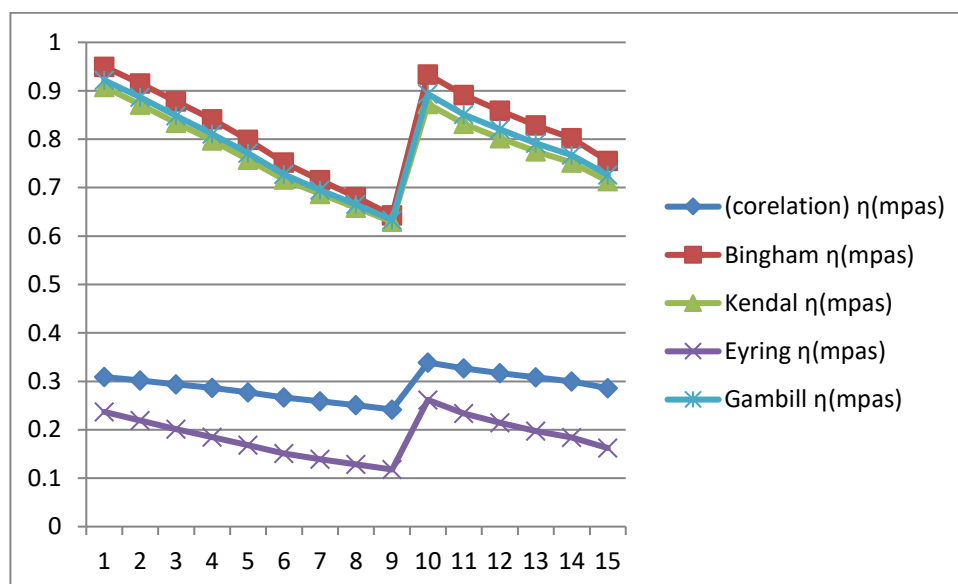


Figure 3- comparison of dynamic viscosity for ternary system Ethanol + Methanol + TAME

The results obtained from the study have revealed several interesting findings. Firstly, the ultrasonic velocity was found to exhibit non-linear behaviour with varying concentrations of the three components in the ternary liquid mixtures. This suggests the presence of significant interactions and intermolecular forces between the components, which influence the overall rheology of the mixture.

Secondly, the viscosity of the ternary liquid mixtures was observed to deviate from the ideal behaviour predicted by various mixing rules. This indicates the existence of synergistic or antagonistic interactions between the components, which affect the flow behaviour of the mixtures. The observed deviations from ideal behaviour

provide insights into the nature and strength of the intermolecular forces operating in the system.

Thirdly, the temperature dependence of the ultrasonic velocity and viscosity was investigated, and it was found that the rheological properties of the ternary mixtures exhibited sensitivity to temperature changes. This indicates that temperature plays a significant role in influencing the flow behaviour of these mixtures, which can have practical implications in various industrial processes where temperature control is crucial.

Conclusion

In conclusion, the viscometric study of ternary liquid mixtures using ultrasonic velocity at 298.15K has provided valuable insights into the

rheological behaviour of these complex systems. The ultrasonic velocity measurements have proven to be a sensitive and reliable tool for characterizing the viscoelastic properties of ternary liquid mixtures.

Overall, the viscometric study of ternary liquid mixtures using ultrasonic velocity at 298.15K has provided valuable information about the rheological behaviour of these complex systems. The findings from this study can contribute to the understanding of the physical and chemical properties of ternary liquid mixtures, and can find applications in diverse areas such as pharmaceuticals, food processing, and chemical engineering. Further research in this area could explore additional variables such as pressure, composition range, and different measurement techniques to gain deeper insights into the rheological behaviour of ternary liquid mixtures.

Conflicts of Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Availability of data

The data that support the findings of this study are available from the corresponding author upon reasonable request.

References

1. R. Dey, A.K. Singh and J.D. Pandey, "A temperature dependent viscometric study of binary liquid mixtures", *J. Mol. Liq.* 137 (2008) 88-91.
2. J.D. Pandey, A.K. Shukla, R. Dey and V. Sanguri, "Viscosity of multicomponent liquid systems-Predictions from binary data and statistical method", *J. Int. Acad. Phys. Sci.* 1-2 (1997) 97.
3. R.J. Fort and W.R. Moore, "Viscosities of binary liquid mixtures", *Trans. Faraday Soc.* 62 (1966) 1112.
4. R. Dey, A. Harshvardhan, S. Verma, Viscometric investigation of binary, ternary and quaternary liquid mixtures: Comparative evaluation of correlative and predictive models, *J. Mol. Liq.* 211 (2015) 686-694. <https://doi.org/10.1016/j.molliq.2015.07.042>
5. W. R. Gambill, How to estimate mixtures viscosities, *Chem. Eng.* 66 (1959) 151-152.
6. F.J. Deive, M.A. Rivas, A. Rodriguez, Study of thermodynamic and transport properties of phosphonium-based ionic liquids, *J. Chem. Thermodyn.* 62 (2013) 98-103. <https://doi.org/10.1016/j.jct.2013.02.012>
7. P. Venkateswara Rao, "Thermodynamic and transport properties of binary liquid mixtures", Department of chemistry, University College of Sciences, Andhra Pradesh, (2017).
8. J.D. Pandey, S. Mukherjee, S.B. Tripathi, N.K. Soni and A.K. Sharma, Viscosity of multi component mixtures of non-electrolyte liquids, *Ind. J. Chem.* 40A (2001) 1214-1217.
9. J.D. Pandey, S. Pandey, S. Gupta and A.K. Shukla, Viscosity of ternary liquid mixtures, *J. Solution Chem.* 23 (1994) 1049-1059. <https://doi.org/10.1007/BF00974102>
10. A.H. Nhaesi and A-F. A. Asfour, Densities and Viscosities of the Regular Quinary System: Toluene (1) + Octane (2) + Ethylbenzene (3) + Tetradecane (4) + Hexadecane (5) and Its Quaternary Subsystems at (308.15 and 313.15) K, *J. Chem. Eng. Data* 50 (2005) 149-153. <https://doi.org/10.1021/je049768f>
11. I.H. Peng and C-H. Tu, Densities and Viscosities of Acetone, Diisopropyl Ether, Ethanol, and Methyl Ethyl Ketone with a Five-Component Hydrocarbon Mixture from 288.15 K to 308.15 K, *J. Chem. Eng. Data* 47 (2002) 1457-1461. <https://doi.org/10.1021/je020077y>
12. D. Gomez-Diaz, J.C. Mejutoi and J.M. Navaza, Physicochemical Properties of Liquid Mixtures. 1.† Viscosity, Density, Surface Tension and Refractive Index of Cyclohexane + 2,2,4-Trimethylpentane Binary Liquid Systems from 25 °C to 50 °C, *J. Chem. Eng. Data* 46(3) (2001) 720-724. <http://dx.doi.org/10.1021/je000310x>
13. Ali and A.K. Nain, Interactions in Binary N,N-Dimethylacetamide + Ethanol and Ternary Lithium Nitrate + N,N-Dimethylacetamide + Ethanol Mixtures, *Phys. Chem. Liq.* 37 (1999) 161-174. <https://doi.org/10.1080/00319109908045123>
14. M.N. Roy, B. Sinha and V.K. Dakua, Excess Molar Volumes and Viscosity Deviations of Binary Liquid Mixtures of 1,3-Dioxolane and 1,4-Dioxane with Butyl Acetate, Butyric Acid, Butylamine, and 2-Butanone at 298.15 K, *J. Chem. Eng. Data* 51(2) (2006) 590-594. <https://doi.org/10.1021/je0504109>

- 15.S.J. Tangeda and S. Nallani, Density and viscosity of binary liquid systems of N-methylacetamide with aromatic ketones at T = 308.15 K, *J. Chem. Thermodyn.* 38(3) (2006) 272-277. <https://doi.org/10.1016/j.jct.2005.05.006>
- 16.J.D. Pandey, S. Mukherjee, M.K. Yadav and R. Dey, Viscosity of multicomponent gas mixtures, *J. Ind. Chem. Soc.* 82 (2005) 39.
- 17.K. Rajagopal, S. Chentilnath and A. K. Nain, Volumetric and Viscometric Studies in Binary Liquid Mixtures of 2-Methyl-2-propanol with Some Ketones at Different Temperatures, *Journal of Solution Chemistry* volume 41 (2012) 1401–1411. <https://doi.org/10.1007/s10953-012-9870-z>
- 18.J.D. Pandey, V. Sanguri, P.K. Guatam, Anjani K. Singh and Sunil,” ultrasonic velocity-viscosity correlation for quaternary liquid systems”, *Journal of International Academy of Physical Sciences*, 18, 2014, 359-365.
- 19.J.D. Pandey, Richa verma and R. Dey, “Excess functions of multicomponent liquid mixtures”, *Acoustic letters* 24(5) (2000) 94.
- 20.Pandey, J.D. Dey, R. and Upadhaya, M., Non-linearity parameter of multicomponent liquid systems, *Acoustics Letters*, 21(6) (1997) 120-125.
- 21.Vikas Singh, K.K. Tiwari and V.K. Singh, “Theoretical predictions of viscosity of Binary and Ternary liquid mixtures at 298.15K.”, *Asian Journal of Chemistry*, 20(6) (2008) 4173-4184.
- 22.J. D. Pandey, A.K. Shukla, Shikha & Neelima Tripathi, Viscosity of ternary liquid systems and the significant structure theory, *Ind. J. Chem.* 33A (1994) 613-615.
- 23.Pandey, J.D. Pandey, S., Gupta, S. and Shukla, A.K., Viscosity of ternary liquid mixtures, *J. Soln. Chem.* 23 (1994) 1049-1059. <https://doi.org/10.1007/BF00974102>
- 24.J. Zielkiewicz, Solvation of amide group by water and alcohols investigated using the Kirkwood–Buff theory of solutions, *J. Chem. Soc. Farad. Trans.* 94 (1998) 1713-1719. <https://doi.org/10.1039/A800943K>