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Ultrasound wave as a tool for assessing molecular interactions of the ternary liquid mixtures of p-anisaldehyde with dimethylamine in n-hexane medium at different temperatures through the concept: Correlation of experimental velocities with calculated velocities based on certain theories

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Abstract
The acoustical/physical parameters like density (ρ), ultrasonic velocity (U) and viscosity (η) have been experimentally measured for the ternary liquid mixture of p-anisaldehyde (PA) +dimethylamine (DMA)+ n- Hexane at various temperatures such as 303 K, 308 K and 313 K and at fixed frequency of 2 M Hz. The variation of experimental ultrasonic velocities with respect to different concentrations ranging from 0.001 M to 0.01 M at different temperatures have been discussed with reference to molecular interactions. The theoretical studies like Van-Dael Ideal Mixing Relation (IMR), Impedance Dependant Relation (IMP), Junjie's (JUN) and Nomoto's (NOM) relations are validated with respect to experimental velocities at 303 K, 308 K and 313 K for all the different concentrated solutions of p-anisaldehyde+dimethylamine + n- hexane. The least deviation in experimental velocity with respect to each temperature is determined while comparing with computed theoretical velocities which are based on those theories and the Chi-square test for the goodness of fit is also applied to check the validity of such theories.

Keywords: junjie's relation; nomoto's relation; molecular interactions; chi-square test

1. Introduction
The varieties of various research techniques are undertaken by the scientists particularly physicists or chemists or research scholars all over the world to get insight into the molecular behaviour or molecular interactions of the liquid mixtures of organic compounds in particular. The ultrasonic technique is one such technique which is extensively utilized nowadays to enunciate the mechanism or structure of the molecules. The velocity of sounds plays a vital role in the determination of elastic and thermodynamic properties of all states of substances. The molecular interaction owes the responsible factor in the study of molecular theory of liquids or liquid mixtures. Thus, the present study provides ample scope in the domain of ultrasonic sound wave which may aggravate the molecular interactions in the liquids or liquid mixtures while the sound wave is passed over the liquids or the system under investigation. The theoretical evaluation of ultrasonic velocities based on theories such as Van Dael Ideal Mixing Relation (IMR), Impedance Dependant Relation (IMP), Junjie's (JUN) and Nomoto's (NOM) relation, Rao's velocity method, collision factor theory, free length theory, scaled particle theory and Flory's statistical theory is correlated with experimental velocities that paves the way for building comprehensive theoretical models for liquids or liquid mixtures. Several researchers [1-7] evaluated ultrasonic velocities based on some theories and correlated with their corresponding experimental velocities for the binary and ternary systems. Hence, in this present probe, the author has undertaken one such study which focuses on not only correlating experimental velocities with computed velocities based on certain theoretical models like Van Dael Ideal Mixing Relation (IMR), Impedance Dependant Relation (IMP), Junjie's (JUN) and Nomoto's (NOM) relation; this investigation will find the least deviation of experimental velocity from among the above said four theories and is best suited with values of chi-square test analysis and validated one of the theories. The literature survey reveals that no such study is considered for the ternary mixture of PA+DMA+n-hexane at various temperatures till date, the author imbibed the ideas and aims to provide a theoretical model which is more suitable with the corresponding experimental velocities.

2. Experimental
2.1 Materials
The ternary mixtures of various equimolar concentrations ranging from 0.001 M to 0.01 M of the system PA+DMA +n Hexane were prepared by taking analytical reagent grade and spectroscopic reagent grade chemicals with minimum assay of 99.9%. All the liquids were purified by the standard methods [8-9]. The procurement details are provided in Table 1.

Table 1: CAS Registry Number of the chemicals and its procurement particulars

Sl. No.	Name of the component	CAS RN	Suppliers Name	Brand Name
1	n-Hexane	110-54-3	Chandanmal & Co., Chennai	E. Merck Ltd (India)
2	Dimethylamine	124-40-3		
3	para- Anisaldehyde or 4 methoxy benzaldehyde	123-11-5		

2.2 Apparatus and techniques

The density, viscosity, and ultrasonic velocity were measured for various concentrations viz. 0.001-0.01 M of the system under investigation keeping fixed frequency of 2 M Hz. Ultrasonic velocity measurements were taken using an ultrasonic interferometer (Model F-81, supplied by M/S Mittal Enterprises, New Delhi, India) with an accuracy of ±0.1ms^{-1}. Water at desired temperature 303 K/ 308 K/ 313 K is circulated through the outer jacket of the double-walled measuring cell of the interferometer containing the experimental ternary liquid mixture. The interferometer was calibrated using the speed of sound of water at 298.15 K. A cell with 2 M Hz. frequency was used to measure the speed of sound. The cell was filled with 8 to 10 ml solution and was allowed for 30 minutes before taking the readings. Average of 10 readings was taken as a final value. The measured speed of sound value is accurate up to ± 0.05%. The precession of sound speed based on 10 readings was calculated as ± 0.02%. Separate temperature control water bath procured from Ragas Industries, Chennai, India is attached with Interferometer and it may also be calibrated. The densities of the mixture were measured using a 10 ml specific gravity bottle by relative measurement method. The specific gravity bottle was kept for about 30 minutes in a thermo-stated water bath for minimizing the error in density with an accuracy of ±0.01 kgm^{-3}[10]. An Ostwald viscometer (10 ml) with an accuracy of ±0.001 Nsm^{-2} was used for the viscosity measurement. The flow time was determined using a digital racer stopwatch with an accuracy of ±0.1s. The viscometer was kept for about 30 minutes in a thermo-stated water bath to minimize the thermal fluctuation in viscosity.

3. Theory and calculations

3.1 Theoretical Studies

Based on the following theoretical relations (Equation numbers 1-4), the theoretical velocities were calculated and compared with the experimental values of ultrasonic velocity.

3.1.1 Nomoto's Equation [11]

$$U_{NR} = \sum_{i=1}^{n} \frac{xiRi}{xiVmi} \quad (1)$$

3.1.2 Van Dael Ideal Mixing Relation [12]

$$U_{IMR} = (\sum_{i=1}^{n} xiMi)^{1/2}\left(\sum_{i=1}^{n} \frac{xi}{MiV_{Mi}^{2}}\right)^{1/2} \quad (2)$$

3.1.3 Junjie's Relation [13]

$$U_{JR} = (\sum_{i=1}^{n} xiVmi)(\sum_{i=1}^{n} xiMi)^{1/2}\left[\frac{\sum_{i=1}^{n} xiVmi}{\sum_{i=1}^{n} \rho iU_i^2}\right]^{1/2} \quad (3)$$

3.1.4 Impedance Dependant Relation [14]

$$U_{IMP} = \frac{\sum_{i=0}^{n} xi\rho iUi}{\sum_{i=0}^{n} xi\rho i} \quad (4)$$

Where xi is Mole fraction, Ri is molar sound velocity, ρi is the density and Vmi is the molar volume

3.2 AAPD

$$\text{Absolute Average Percentage Deviation (AAPD)} = \frac{1}{n}\sum\left[\frac{(Uexp-Uthe)}{Uexp}\right] \times 100 \quad (5)$$

3.3 Percentage Error

$$\%\ \text{Error (PE)} = \{(U_{exp}-U_{the})/U_{exp}\}\ X100 \quad (6)$$

Where U_{exp} is experimental ultrasonic velocity and U_{the} is theoretical ultrasonic velocity

3.4 Ideality of the systems

The ideal or non- ideal behaviour of the systems can be determined by the following relation:

$$NI = (U^2_{exp} / U^2_{ideal}) \quad (7)$$

3.5 Molecular interaction parameter

$$\text{Interaction Parameter } \alpha = (U^2_{exp} / U^2_{ideal}) - 1 \quad (8)$$

Where U_{ideal} is ideal mixing velocity and U_{exp} is experimental velocity

3.6 Statistical Analysis

According to Pearson [15], the Chi-square test for the goodness of fit is given by

$$\chi^2 \text{Chi-square} = \sum_{i=1}^{n}\left[\frac{(Uexp-Uthe)(Uexp-Uthe)}{Uthe}\right] \quad (9)$$

Where U_{exp} is experimental ultrasonic velocity and U_{the} is theoretical ultrasonic velocity.

4. Results and Discussions

The experimentally observed values of velocity, viscosity and Density for p-anisaldehyde +Dimethyl amine +n hexane at various temperatures are presented in table 2. Further, computed ultrasonic velocity values based on certain theoretical models are tabulated in Table 3 along with experimental values. In table 4, percentage deviation (in comparison with experimentally observed ultrasonic velocity values) values of theoretical models such as Nomoto's relation, Ideal mixing relation, Impedance dependant relation and Junjie's relations are provided along with absolute average percentage deviation (AAPD). The values of U^2_{Exp}/U^2_{IMX} and degree of molecular interactions (α) are shown in table 5 along with average degree of molecular interactions (ADMI). The AAPD values at 303 K, 308 K and 313 K are represented in table 6 with corresponding Chi-square test values. The comparative study with respect to velocity, viscosity and density for the pure components present in the system at given temperatures, in between literature values and observed values are given in table 7. In addition, the plot between U^2_{Exp}/U^2_{IMX} against concentration is illustrated in figure 1, also, the plots of experimental and theoretical ultrasonic velocity values against concentration at 303 K, 308 K and 313 K is shown in figures 2, 3 and 4 respectively.

It is observed that the values of IMP at 303 K & 308 K provides least deviation with the corresponding experimental values of ultrasonic velocities followed by NOM at the same temperatures. This shows the presence of molecular interactions in between the components present in the ternary system. These interactions are due to the

presence of dipole-dipole, dipole-induced dipole and in particular hydrogen bonded interactions. This result is in accordance with the statement provided by Nagabalasubramanian *et al*., [16] who observed that the most hydrogen bond acceptor in the molecule PA is carbonyl oxygen but within the same, methoxy group also acts as acceptor in addition to aromatic π electron system and the spectral studies like $_1$H, Raman and FTIR support the hydrogen bonded dimers in PA.

The trend in deviation of theoretical based velocities at respective temperatures from experimental velocities may be given as follows.

IMP<NOM<IMR<JUN at 303 K & 308 K
NOM<IMR<IMP<JUN at 313 K

The computed values of velocities based on models like IMR and JUN recorded comparatively high deviation. According to IMR, the ratio of specific heats of ideal mixture and the volume are not equal which once again does not consider the interactions between the molecules which will be existed normally between the molecules of the system because of which its values deviates more which is strongly supported by the present study. Silmilarly, JUN also recorded high deviation. But at 313 K, NOM relation possess nearer agreement with corresponding experimental values because of dissociation between associated molecules that happened at elevated temperature due to increase in kinetic energy of the molecules that may obviously breaks the hydrogen bonding and hence NOM relation shows lesser deviation among other theories at 313 K. The value of U^2_{exp} / U^2_{ideal} provides the nature of ideality or non-ideality of the system. In the present investigation, the values of U^2_{exp} / U^2_{ideal} found to be positive at all the working temperatures which indicate the existence of strong interactions. The degree of molecular interactions α provides information regarding the nature of interactions. Since in the present study, the values of α are positive, the interactions are said to be strong at 303 K, 308 K and 313 K [17-21].

Table 2: Experimental values of velocity, viscosity and Density for p-anisaldehyde +Dimethyl amine +n hexane

Sl. No.	Conc. (M) X10^{-3}	Velocity (U) ms^{-1}			Viscosity (η)Nsm^{-2} X10^{-4}			Density (ρ) Kgm^{-3}		
		303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
1	1	1623	1612	1608	1.8017	1.5675	1.2978	1.1270	1.0987	0.9387
2	2	1632	1621	1615	1.9261	1.6580	1.3429	1.1180	1.0876	0.9872
3	3	1635	1626	1618	2.0651	1.7275	1.4876	1.0967	1.0546	0.9148
4	4	1638	1633	1621	2.1924	1.8233	1.5344	1.0786	1.0324	0.9073
5	5	1651	1642	1624	2.2411	1.9857	1.6987	1.0187	1.0184	0.9011
6	6	1653	1648	1627	2.3912	2.0967	1.6432	0.9867	0.9256	0.8936
7	7	1656	1654	1629	2.4205	2.1379	1.7654	0.9634	0.9187	0.8830
8	8	1662	1649	1631	2.5672	2.2977	1.8976	0.9419	0.8974	0.8735
9	9	1669	1640	1634	2.6888	2.3653	1.9345	0.9197	0.8629	0.8672
10	10	1676	1636	1626	2.8016	2.4588	2.0987	0.8976	0.8718	0.8543

Table 3: Experimental and Theoretical values of Ultrasonic velocities of Ternary liquid mixture containing PA+DMA+n-hexane at different temperatures

CONC.(M) X 10^{-3}	U_{Exp} ms^{-1}	U_{NOM} ms^{-1}	U_{Ideal} ms^{-1}	U_{IMP} ms^{-1}	U_{Jun} ms^{-1}
T=303 K					
1	1623	1572	1556	1557	1559
2	1632	1547	1521	1526	1522
3	1635	1522	1492	1501	1489
4	1638	1498	1466	1481	1457
5	1651	1473	1443	1464	1427
6	1653	1449	1423	1449	1399
7	1656	1426	1406	1437	1373
8	1662	1402	1390	1427	1348
9	1669	1379	1376	1417	1324
10	1676	1356	1363	1409	1301
T=308 K					
1	1612	1412	1402	1404	1401
2	1621	1397	1381	1386	1379
3	1626	1382	1362	1372	1358
4	1633	1368	1346	1361	1338
5	1642	1354	1331	1351	1321
6	1648	1339	1318	1343	1304
7	1654	1325	1307	1337	1289
8	1649	1311	1297	1331	1275
9	1640	1297	1287	1325	1263
10	1636	1283	1278	1321	1251
T=313 K					
1	1608	1286	1297	1295	1279
2	1615	1279	1284	1281	1267
3	1618	1273	1272	1268	1256
4	1621	1266	1261	1257	1246

5	1624	1259	1251	1247	1237
6	1627	1252	1241	1238	1232
7	1629	1246	1232	1230	1221
8	1631	1239	1223	1222	1215
9	1634	1233	1215	1215	1209
10	1626	1226	1207	1208	1205

Table 4: Percentage deviation between Experimental and Theoretical values of Ultrasonic velocities of Ternary liquid mixture containing PA+DMA+n-hexane at different temperatures

CONC.(M) X 10^{-3}	U_{Exp} ms^{-1}	U_{NOM} ms^{-1}	U_{Ideal} ms^{-1}	U_{IMP} ms^{-1}	U_{Jun} ms^{-1}
T=303 K					
1	1623	3.14	4.13	4.07	3.94
2	1632	5.21	6.80	6.50	6.74
3	1635	6.91	8.75	8.20	8.93
4	1638	8.55	10.50	9.58	11.05
5	1651	10.78	12.60	11.33	13.57
6	1653	12.34	13.91	12.34	15.37
7	1656	13.89	15.10	13.22	17.09
8	1662	15.64	16.37	14.14	18.89
9	1669	17.38	17.56	15.10	20.67
10	1676	19.09	18.68	15.93	22.37
AAPD (Absolute Average Percentage Deviation)		11.29	12.44	11.04	13.86
T=308 K					
1	1	12.41	13.03	12.90	13.09
2	2	13.82	14.81	14.50	14.93
3	3	15.01	16.24	15.62	16.48
4	4	16.23	17.58	16.66	18.06
5	5	17.54	18.94	17.72	19.55
6	6	18.75	20.02	18.51	20.87
7	7	19.89	20.98	19.17	22.07
8	8	20.50	21.35	19.28	22.68
9	1	20.91	21.52	19.21	22.99
10	2	21.58	21.88	19.25	23.53
AAPD (Absolute Average Percentage Deviation)		17.66	18.63	17.28	19.43
T=313 K					
1	1	20.02	19.34	19.47	20.46
2	2	20.80	20.50	20.68	21.55
3	3	21.32	21.38	21.63	22.37
4	4	21.90	22.21	22.46	23.13
5	5	22.48	22.97	23.21	23.83
6	6	23.05	23.72	23.91	24.28
7	7	23.51	24.37	24.49	25.05
8	8	24.03	25.02	25.08	25.51
9	9	24.54	25.64	25.64	26.01
10	10	24.60	25.77	25.71	25.89
AAPD (Absolute Average Percentage Deviation)		22.63	23.09	23.23	23.81

Table 5: Values of U^2_{EXP}/U^2_{IMX} and Degree of Molecular Interaction (α) of Ternary liquid mixture containing PA+DMA+n-hexane at different temperatures

CONC.(M) X 10^{-3}	U^2_{EXP}/U^2_{IMX}	Degree of Molecular Interaction(α)
T=303 K		
1	1.0866	0.0866
2	1.1438	0.1438
3	1.1865	0.1865
4	1.2233	0.2233
5	1.2718	0.2718
6	1.3014	0.3014
7	1.3280	0.3280
8	1.3565	0.3565
9	1.3873	0.3873
10	1.4149	0.4149
ADMI (Average Degree of Molecular Interaction) α_{AVG}		0.2700
T=308 K		
1	1.3363	0.3363
2	1.3865	0.3865
3	1.4201	0.4201
4	1.4485	0.4485
5	1.4934	0.4934
6	1.5149	0.5149
7	1.5341	0.5341

8	1.5592	0.5592
9	1.5866	0.5866
10	1.6097	0.6097
ADMI (Average Degree of Molecular Interaction) α_{AVG}		0.4889
T=313 K		
1	1.5928	0.5928
2	1.6282	0.6282
3	1.6496	0.6496
4	1.6740	0.6740
5	1.7197	0.7197
6	1.7432	0.7432
7	1.7664	0.7664
8	1.7994	0.7994
9	1.8323	0.8323
10	1.8688	0.8688
ADMI (Average Degree of Molecular Interaction) α_{AVG}		0.7274

Table 6: Comparative study between theoretical (AAPD) and chi-square test values

System	Temp.	AAPD				Values of Chi-Square test			
		NOM	IMR	IMP	JUN	NOM	IMR	IMP	JUN
PA + DMA+ n-hexane	303 K	11.29	12.44	11.04	13.86	29.98	34.40	25.89	45.81
	308 K	17.66	18.63	17.28	19.43	64.79	72.56	60.51	80.50
	313 K	22.63	23.09	23.23	23.81	108.27	114.21	115.64	122.04

From the table 6, it is understood that IMP relation shows least deviation while comparing with corresponding experimental velocities at 303 K and 308 K among rest of the theories and the same values are substantiated by the values which are obtained through Chi-Square test analysis. Also, at 313 K, NOM relation recorded least value, likewise the minimum value arrived by the Chi-Square test analysis and hence those theories are validated.

Table 7: Comparative study between Literature values and Observed values of Velocity, Viscosity and Density for the pure components present in the system at working temperatures

Sl. No.	Name of the pure component in the mixtures		Velocity (U) ms^{-1}			Viscosity (η)Nsm^{-2} X 10^{-4}			Density (ρ) Kgm^{-3}		
			303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
1.	n- Hexane	Literature	1058.3 [22]	NA	NA	0.296 [23]	NA	NA	0.65 [24]	NA	NA
		Observed	1076.1	1054.4	1032.4	0.320	0.317	0.301	0.651	0.648	0.647
2.	DMA	Literature	1433 @	NA	NA	0.340#	NA	NA	0.827@	NA	NA
		Observed	1678	1527	1412	1.81	1.78	1.74	0.707	0.699	0.707
3.	PA	Literature	NA	NA	NA	NA	NA	NA	NA	NA	NA
		Observed	1079	1068	1057	1.802	1.744	1.693	1.12	1.008	0.997

NA: no data available, # source: www.inchem.org>documents>icsc>eics>1485, @ source: flexim news update 17, *a new perspective*, 17/2017 p5

Table 7 represents the Comparative study between Literature values and Observed values of velocity, viscosity and density for the pure components namely PA, DMA, and n- hexane present in the ternary system at working temperatures such as 303 K, 308 K and 313 K. It is noticed that most of the literature values are in good agreement with their corresponding experimental/observed values except for PA whose literature values are not yet available/ acknowledged.

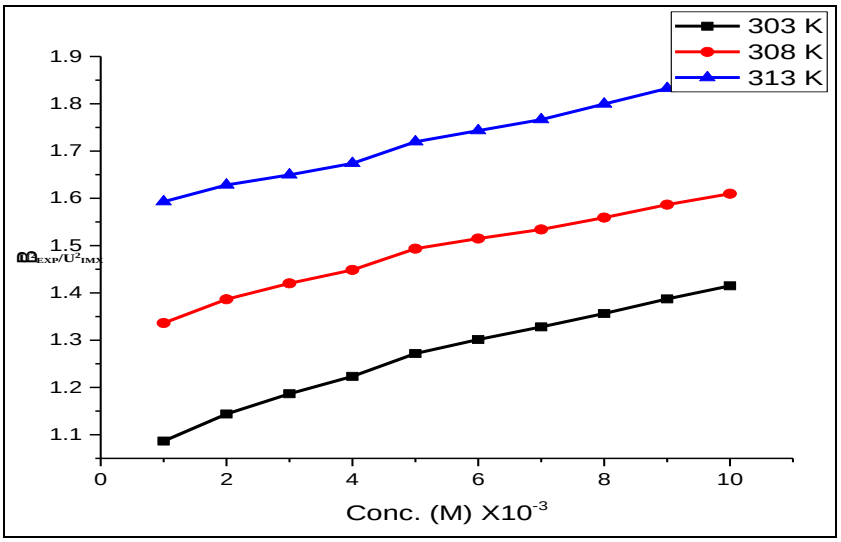

Fig 1: Plot between Conc. Vs U^2_{EXP}/U^2_{IMX} for p-anisaldehyde +Dimethyl amine +n hexane at different temperatures

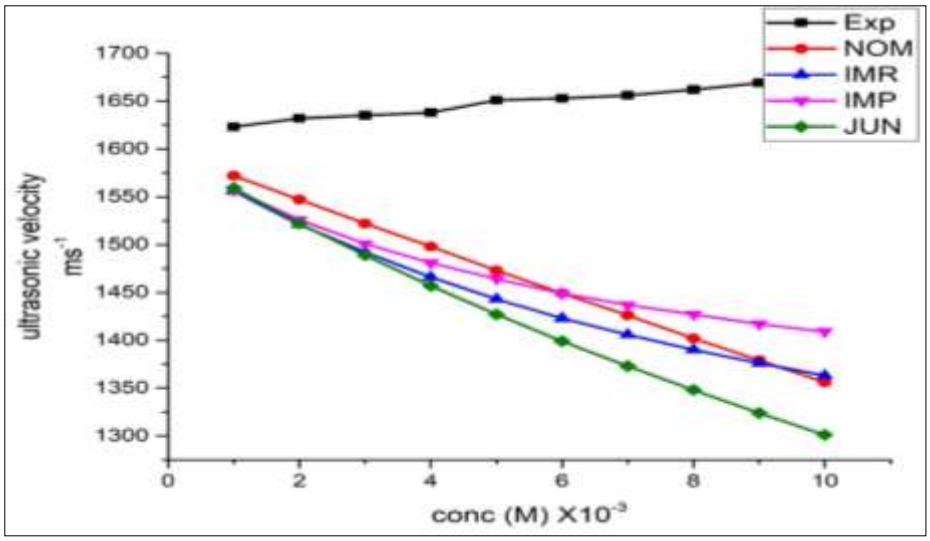

Fig 2: Plot between Conc. Vs Experimental and Theoretical Ultrasonic Velocities for p-anisaldehyde +Dimethyl amine +n hexane at 303 K

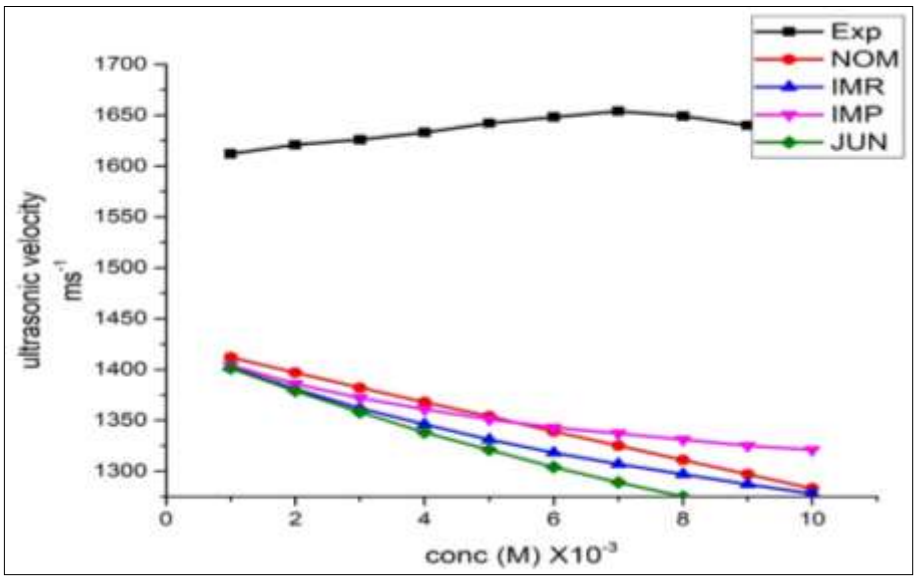

Fig 3: Plot between Conc. Vs Experimental and Theoretical Ultrasonic Velocities for p-anisaldehyde +Dimethyl amine +n hexane at 308 K

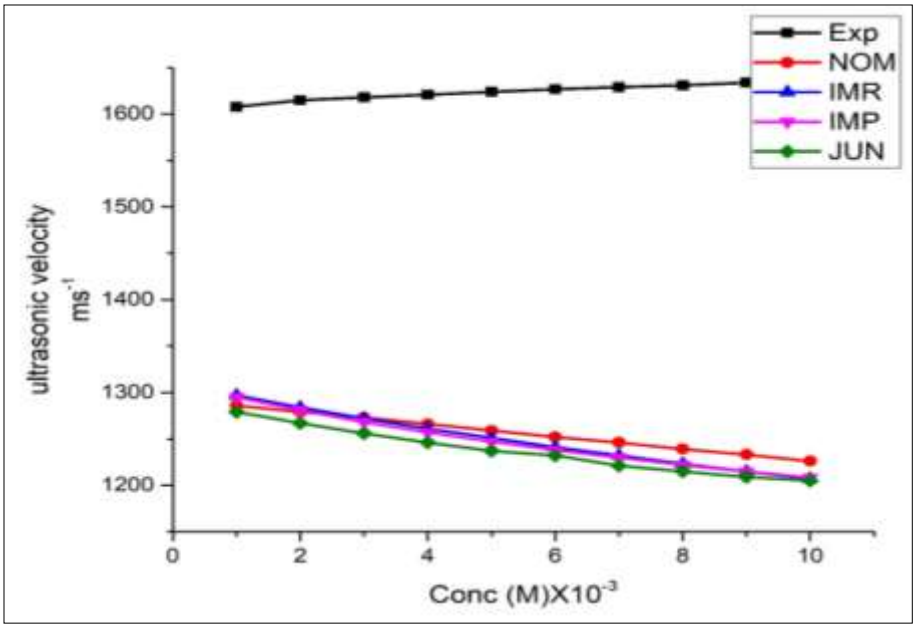

Fig 4: Plot between Conc. Vs Experimental and Theoretical Ultrasonic Velocities for p-anisaldehyde +Dimethyl amine +n hexane 313 K

5. Conclusion

It may be concluded for the system which is under investigation, at all the different temperatures viz. 303 K,308 K and 313 K, it is found that IMP is best suited or having least deviation when comparing with its corresponding observed values at 303 K and 308 K. But at 313 K, Nomoto's relations possessed least deviation among all other theoretical values. The positive α value is observed and hence the presence of strong interactions is expected in the system owing to the interactions involving hydrogen bonded, dipole-dipole, dipole-induced dipole and formation of charge transfer complexes. At the same time, at 313 K, presence of weak interactions is noted which is due to thermal agitation of the molecules because of which kinetic energy of the molecules get increases that paves the way for rupture of hydrogen bonding as well as dissociation of associated molecules. With respect to Chi square analysis test, the system under probe, whose AAPD values vary one another among the theories of consideration at respective temperatures. At 303 K and 308 K, Chi square values are found to be least for the IMP and at 313 K which predicts in support of Nomoto's relation. Similar to AAPD values, the Chi square values are observed to be least for IMP relation at 303 K and 308 K and for Nomoto's relation at 313 K. Thus some extent, the Chi square values are complementary and are best suited to IMP relation as arrived in this study at 303 K and 308 K; at 313 K, NOM relation is supported by Chi square values.

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