

## Research Article



## Acoustic Response with Theoretical Evaluation of Ultrasonic Velocity in Ternary Mixtures of DMSO, Benzene, and Toluene at Different Temperatures.

Manaswini Mishra<sup>1\*</sup>, Upendra Nath Dash<sup>1</sup>, Nandita Swain<sup>2</sup>

<sup>1</sup>Department of Chemistry, Institute of Technical Education and Research, SOA University, Bhubaneswar, Odisha, India.

<sup>2</sup>Department of Chemistry, College of Basic Science and Humanities, OUAT Bhubaneswar – 751003, Odisha, India.

\*Corresponding author's E-mail: [manaswinimishra15@gmail.com](mailto:manaswinimishra15@gmail.com)

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### ABSTRACT

Ultrasonic velocity (U), density ( $\rho$ ), and viscosity ( $\eta$ ) of ternary mixtures of dimethyl sulphoxide with benzene and toluene were measured at 298.15K, 303.15K, 308.15K, and 313.15K under the atmospheric pressure over the entire range of compositions. The study involves evaluation of different acoustic properties and is interpreted in terms of molecular interactions between the components of the liquid mixtures. The theoretical values of ultrasonic velocity for the ternary mixtures have been evaluated using several theoretical models, such as Nomoto's relation (N), Rao's specific sound velocity relation (R), Impedance dependence relation (IDR), and Van Dael and Vangeel's ideal mixing relation (V) to test their relative applicability.

**Keywords:** Density, ultrasonic velocity, viscosity, ternary mixture, molecular interaction.

### INTRODUCTION

The measurements of ultrasonic velocity, density and viscosity of liquid mixtures are helpful for their applications in characterizing aspects of physico-chemical behavior in solvent extraction process<sup>1,2</sup>. The study of ternary mixture of DMSO is very essential because of its wide range of application in medicine and DMSO is also used as free radical scavenger for various cancer treatment. In continuation of our previous work in ternary system<sup>3</sup> we are reporting here various acoustical parameters viz., Wada's constant (W), Rao's constant (R), ultrasonic attenuation constant ( $\alpha/f^2$ ) and the experimental sound velocities have been compared with theoretical velocities predicted through various empirical relations such as Nomoto's relation, Rao's specific sound velocity, Impedance dependence relation and Van Dael and Vangeel's ideal mixing relation. The validity of these formulations for describing the ultrasonic response in the ternary mixture of DMSO has been examined and the non ideal behavior of the mixtures has been explained in terms of molecular interactions between their constituents.

### MATERIALS AND METHODS

All the chemicals used were of Anala R grades (E.Merk, India) and used as such. The ultrasonic velocities of the pure liquids and the ternary mixtures at 298.15K, 303.15K, 308.15K and 313.15K are determined by using ultrasonic interferometer, F-81 Mittal Enterprises, New Delhi at a fixed frequency of 2 MHz the temperature of the sample is maintained to a precision of  $\pm 0.1K$  by circulating water from the thermostat. The ternary mixtures of DMSO, benzene and toluene are prepared by volume. Nine mixtures are prepared in air tight bottles. Density of pure liquids and their mixtures have been

measured by specific gravity bottle of 25 mL capacity at 298.15K, 303.15K, 308.15K and 313.15K. The maximum error in the density measurement was  $\pm 0.02 \text{ kg m}^{-3}$ . Ostwald's viscometer having a capacity of about 25mL was used for the viscosity measurement of pure liquids and liquid mixtures at 298.15K, 303.15K, 308.15K, and 313.15K.

The Rao's constant and Wada's constant<sup>4,5</sup> have been determined by using the following standard relation.

$$\text{Rao's constant, } R = \left( \frac{M}{\rho} \right) U^{1/3} \quad (1)$$

Where M is effective molecular weight and  $\rho$  is density.

$$\text{Wada's constant, } W = \left( \frac{m}{\rho} \right) K_s^{-1/7} \quad (2)$$

Where W, is molar compressibility or Wada's constant and is independent of temperature and pressure.

Using Stokes theory of absorption coefficient the attenuation constant or absorption coefficient<sup>6</sup> is determined as.

$$\text{Absorption coefficient, } \frac{\alpha}{f^2} = \frac{8\pi^2\eta}{3\rho U^3} \quad (3)$$

And relaxation time,  $\tau = (4/3) K_s \eta$  (4)

Interaction parameter ( $\chi$ ) and relative association ( $R_A$ )<sup>7</sup> are determined using the following standard relations.

$$\text{Interaction parameter, } \chi = (U_{\text{exp}}^2 / U_{\text{ideal}}^2) - 1 \quad (5)$$



Relative association,  $R_A = (\rho/\rho_0) (U_0/U)^{1/3}$  (6)

According to Aurebach relation<sup>8</sup>, surface tension of liquid is given as.

$$\sigma = 6.3 \times 10^{-4} \rho U^{3/2} \quad (7)$$

The theoretical values of ultrasonic velocity such as  $U_N$ ,  $U_R$ ,  $U_{IDR}$  and  $U_V$  were calculated by using Nomoto's relation, Rao's specific sound velocity relation, Impedance dependence relation, Van Dael and Vangeel's ideal mixing relation,<sup>9,10,11,12</sup> respectively.

$$U_N = \left( \frac{\sum x_i R_i}{\sum x_i v_i} \right)^3 \quad (8)$$

$$U_V = \left[ \frac{1}{(x_1 m_1 + x_2 m_2 + x_3 m_3)} \right]^{1/2} \times \left[ \frac{x_1}{m_1 U_1^2} + \frac{x_2}{m_2 U_2^2} + \frac{x_3}{m_3 U_3^2} \right] \quad (11)$$

The percentage of deviation in ultrasonic velocity between experimental and computed values can be calculated as

$$\left( \frac{\Delta U}{U} \right) \% = \left( \frac{U_{\text{exp}} - U_{\text{theo}}}{U_{\text{exp}}} \right) \times 100 \quad (12)$$

## RESULTS AND DISCUSSION

The ultrasonic velocity ( $U$ ), density ( $\rho$ ), viscosity ( $\eta$ ) of pure and liquid mixtures over entire range of compositions of DMSO, benzene and toluene mixtures were measured and reported in Table -1. Dimethyl sulphoxide is used as a polar aprotic solvent. As it is a good solvent, it is used in biology, medicine and many chemical industries. DMSO is less toxic than other members of this class, such as Dimethyl formamide, dimethyl acetamide, N-methyl-2-pyrrolidone, etc. Dimethyl sulphoxide is one of the most widely used and characterised extractants used in liquid-liquid extraction. It is a highly effective extractant for the extraction of valuable metals like Cu, Ni, Co, Mg etc and the extraction efficiency improves with the addition of suitable diluents. In the present investigation attempt has been made to study the physico-chemical behaviour of ternary mixtures of DMSO using benzene and toluene as the diluents. Studies on variation of properties like density, viscosity and ultrasonic velocity are likely to throw light on molecular environment and molecular interactions which may be correlated with the extraction efficiency of DMSO.

In order to explain the physico-chemical behavior of ternary mixtures, Wada's constant (molar compressibility), Rao's constant (molar sound velocity), ultrasonic attenuation constant, relaxation time, interaction parameter, relative association, and surface tension have been evaluated.

Where  $x_i$ ,  $U_i$ , and  $V_i$  are the respective mole fraction, ultrasonic velocity, molar volume and  $R_i = V_i U_i^{1/3}$  the molar sound velocity of the  $i^{\text{th}}$  component of the liquid mixture.

$$U_R = (\sum x_i r_i \rho)^3 \quad (9)$$

Where  $r_i = U_i V_i / \rho_i$  Rao's specific is sound velocity, and  $\rho$  is the density of the  $i^{\text{th}}$  component of the mixture.

$$U_{IPR} = \frac{\sum x_i z_i}{\sum x_i \rho_i} \quad (10)$$

where  $z_i$  is the acoustic impedance of  $i^{\text{th}}$  component of the mixture

From Table-1, it is observed that the ultrasonic velocity ( $U$ ), density ( $\rho$ ) and viscosity ( $\eta$ ) increase with increase in the mole fraction of DMSO. According to Arul and Palaniappan,<sup>13</sup> the increase in the properties suggests to association among molecules of the ternary mixtures. The decrease in Wada's constant ( $W$ ) and Rao's constant ( $R$ ) with increase in concentration of DMSO, indicates that there is weak solute solvent interactions among the liquid mixtures. The ultrasonic attenuation coefficient ( $\alpha/f^2$ ) increases nonlinearly with both concentration and temperature which suggests the existence of molecular interaction between the polar and apolar components. This may be due to formation of weak hydrogen bonds which decreases the inner proton distance between the adjacent hydrogen bonds and strengthening the intermolecular forces of the bulk mixtures<sup>14,15</sup>. The relaxation time is the property of solution which mainly affects the ultrasonic velocity.

Acoustical relaxation time increases with rise in concentration and then decreases in higher concentrations. It shows nonlinear variation with temperature. The relative association ( $R_A$ ) is another property of the liquid which can be studied to understand the ion-ion or ion-solvent interactions<sup>16</sup>. It is influenced by 2 factors.

(1) The breaking up of the associated solvent molecules on addition of the solute to it and (2) The solvation of solute molecules.

**Table 1** : Experimentally determined values of density ( $\rho$ ), ultrasonic velocity ( $U$ ), viscosity ( $\eta$ ) and calculated values of Rao's constant ( $R$ ), Wada's constant ( $W$ ), ultrasonic attenuation constant ( $\alpha/f^2$ ), relaxation time ( $\tau$ ), interaction parameter ( $\chi$ ), relative association ( $R_A$ ), and surface tension ( $\sigma$ ) at temperatures 298.15K, 303.15K, 308.15K and 313.15K.

$x_1$	$x_2$	$x_3$	$\rho_{\text{kgm}^{-3}}$	$U$ m/s	$\eta$ (mpas)	$R[10^{-5}$ $\text{m}^3\text{mol}^{-1}$ ( $\text{ms}^{-1}$ )]	$W[10^{-7}$ $\text{m}^2\text{mol}^{-1}$ ( $\text{Nm}^{-2}$ )]	$\alpha/f^2$ $10^{14}\text{m}^{-1}$ $\text{s}^2$	$\tau$ $10^{-13}\text{s}$	$\chi$	$R_A$	$\sigma 10^{-7}$ $\text{Nm}^{-1}$
298.15K												
0.132	0.472	0.396	895.2	1329.9	0.683	10.27	0.192	0.852	5.75	0.0018	0.849	66.32
0.257	0.404	0.339	921.7	1355.9	0.768	9.95	0.187	0.878	6.04	0.0049	0.869	72.37
0.371	0.341	0.288	948.4	1378.0	0.835	9.63	0.181	0.884	6.18	0.0053	0.889	78.17
0.478	0.283	0.239	984.1	1400.5	0.939	9.26	0.175	0.913	6.48	0.0082	0.917	85.15
0.579	0.227	0.194	1030.7	1419.6	1.025	8.81	0.168	0.913	6.57	0.0078	0.957	92.88
0.677	0.178	0.145	1063.9	1440.9	1.062	8.51	0.162	0.877	6.41	0.0113	0.983	100.25
0.763	0.128	0.109	1108.5	1457.6	1.145	8.14	0.156	0.876	6.48	0.0115	1.021	108.13
0.848	0.084	0.068	1164.7	1475.6	1.199	7.73	0.149	0.842	6.31	0.0138	1.067	117.87
0.926	0.041	0.033	1211.2	1491.3	1.241	7.41	0.144	0.812	6.14	0.0148	1.106	126.53
303.15K												
0.132	0.472	0.396	893.2	1307.9	0.667	10.24	0.192	0.877	5.82	0.0042	0.851	62.94
0.257	0.404	0.339	918.7	1334.4	0.757	9.93	0.186	0.911	6.17	0.0081	0.869	68.76
0.371	0.341	0.288	946.3	1356.4	0.829	9.61	0.181	0.922	6.34	0.0082	0.891	74.38
0.478	0.283	0.239	982.8	1379.2	0.934	9.22	0.175	0.952	6.66	0.0114	0.921	81.21
0.579	0.227	0.194	1027.2	1398.5	1.019	8.80	0.167	0.953	6.76	0.0112	0.957	88.51
0.677	0.178	0.145	1062.2	1420.1	1.046	8.48	0.162	0.904	6.51	0.0151	0.984	95.82
0.763	0.128	0.109	1106.1	1436.4	1.075	8.12	0.156	0.862	6.28	0.0147	1.021	103.25
0.848	0.084	0.068	1162.7	1454.1	1.118	7.70	0.149	0.822	6.06	0.0165	1.069	112.61
0.926	0.041	0.033	1208.1	1469.3	1.141	7.39	0.144	0.782	5.83	0.0167	1.107	120.71
308.15K												
0.132	0.472	0.396	890.3	1289.1	0.645	10.22	0.191	0.889	5.81	0.0077	0.854	60.07
0.257	0.404	0.339	915.9	1316.1	0.725	9.91	0.186	0.912	6.09	0.0107	0.873	65.76
0.371	0.341	0.288	941.2	1338.9	0.793	9.62	0.181	0.922	6.26	0.0105	0.892	71.16
0.478	0.283	0.239	978.8	1363.1	0.879	9.22	0.175	0.932	6.44	0.0146	0.922	78.08
0.579	0.227	0.194	1024.1	1383.4	0.979	8.79	0.167	0.949	6.66	0.0145	0.961	85.41
0.677	0.178	0.145	1059.3	1405.3	1.018	8.47	0.162	0.911	6.48	0.0177	0.987	92.61
0.763	0.128	0.109	1103.9	1422.3	1.053	8.11	0.156	0.871	6.28	0.0172	1.025	100.04
0.848	0.084	0.068	1158.5	1441.1	1.075	7.71	0.149	0.815	5.95	0.0196	1.071	109.21
0.926	0.041	0.033	1456.7	1456.7	1.095	7.39	0.144	0.773	5.71	0.0194	1.111	117.31
313.15K												
0.132	0.472	0.396	884.3	1268.7	0.639	10.24	0.191	0.931	5.98	0.0098	0.853	56.88
0.257	0.404	0.339	907.7	1297.5	0.717	9.95	0.086	0.951	6.25	0.0137	0.869	62.45
0.371	0.341	0.288	935.3	1321.8	0.771	9.64	0.181	0.938	6.29	0.0141	0.891	68.03
0.478	0.283	0.239	973.9	1346.6	0.851	9.23	0.175	0.941	6.42	0.0175	0.921	74.91
0.579	0.227	0.194	1019.5	1368.1	0.937	8.80	0.167	0.943	6.54	0.0177	0.959	82.23
0.677	0.178	0.145	1055.1	1391.1	1.015	8.48	0.162	0.939	6.62	0.0211	0.987	89.47
0.763	0.128	0.109	1099.7	1408.5	1.045	8.11	0.156	0.894	6.38	0.0199	1.025	96.79
0.848	0.084	0.068	1156.3	1428.2	1.055	7.70	0.149	0.823	5.96	0.0224	1.073	106.11
0.926	0.041	0.033	1202.9	1444.2	1.085	7.38	0.143	0.787	5.76	0.0216	1.113	114.13

The former leads to decrease and the latter to the increase in relative association. In the present study,  $R_A$  increases with increase in the concentration of DMSO. It may be due to the H-bond formation in liquid mixtures,

which shows strong solute-solvent interactions. It also suggests that the solvation of the solute may predominant the breaking-up of the solvent structure which supports the increase of interaction parameter ( $\chi$ ).

The value of  $\chi$  increases with increase in concentration as well as temperature. The increasing value of surface tension ( $\sigma$ ) with mole fraction indicates strong solute-

solvent interactions but the decrease in ( $\sigma$ ) value with increase in temperatures reveals that the interactions become weaker with rise in temperature.

**Table 2:** Theoretical values of ultrasonic speed calculated from Nomoto's, Rao's, IDR, and Van Dael and Vangeel's equation along with the experimental values of ultrasonic speed and percentage error for the ternary mixtures at 298.15K, 303.15K, 308.15K and 313.15K.

Mole fraction( $x_1$ )	Ultrasonic Velocity, U				Percentage deviation, ( $\Delta U/U$ ) %				
	U exp m/s	$U_N$ m/s	$U_R$ m/s	$U_{IDR}$ m/s	$U_V$ m/s	$U_N$ m/s	$U_R$ m/s	$U_{IDR}$ m/s	$U_V$ m/s
298.15K									
0.132	1329.9	1321.9	1333.5	1334.1	1321.4	0.59	-0.27	-0.32	0.63
0.257	1355.9	1340.7	1362.4	1361.4	1343.6	1.11	-0.48	-0.41	0.91
0.371	1378.0	1359.3	1395.8	1384.9	1364.6	1.35	-1.29	-0.51	0.97
0.478	1400.5	1378.1	1470.2	1405.8	1384.9	1.59	-4.97	-0.38	1.11
0.579	1419.6	1397.1	1596.8	1424.6	1404.7	1.57	-12.43	-0.35	1.04
0.677	1440.9	1417.1	1659.3	1442.0	1424.5	1.64	-15.15	0.08	1.13
0.763	1457.6	1435.8	1785.5	1456.7	1442.4	1.49	-22.49	0.05	1.03
0.848	1475.6	1455.6	1969.2	1470.7	1460.6	1.35	-33.45	0.33	1.01
0.926	1491.3	1474.9	2113.1	1483.1	1477.8	1.09	-41.69	0.54	0.91
303.15K									
0.132	1307.9	1298.5	1322.8	1310.7	1297.7	0.71	-1.14	-0.21	0.77
0.257	1334.4	1317.3	1347.0	1338.0	1319.9	1.27	-0.94	-0.27	1.08
0.371	1356.4	1335.9	1383.9	1361.6	1340.9	1.51	-2.02	-0.38	1.14
0.478	1379.2	1354.7	1461.1	1382.5	1361.2	1.77	-5.94	-0.24	1.29
0.579	1398.5	1373.8	1575.9	1401.3	1381.1	1.76	-12.68	-0.21	1.24
0.677	1420.1	1393.7	1646.7	1418.8	1400.9	1.85	-15.96	0.09	1.34
0.763	1436.4	1412.4	1768.5	1433.5	1419.0	1.66	-23.12	0.19	1.21
0.848	1454.1	1432.3	1952.5	1447.5	1437.2	1.49	-34.27	0.45	1.15
0.926	1469.3	1451.7	2089.4	1459.9	1454.4	1.19	-42.20	0.63	1.007
308.15K									
0.132	1289.1	1277.2	1310.1	1289.9	1276.4	0.92	-1.63	-0.06	0.97
0.257	1316.1	1296.8	1335.9	1318.5	1299.4	1.46	-1.51	-0.18	1.26
0.371	1338.9	1316.1	1363.9	1343.0	1321.1	1.69	-1.87	-0.31	1.32
0.478	1363.1	1335.7	1446.9	1364.9	1342.3	2.01	-6.14	-0.13	1.52
0.579	1383.4	1355.6	1566.6	1384.5	1363.0	2.01	-13.24	-0.08	1.47
0.677	1405.3	1376.4	1639.7	1402.7	1338.7	2.05	-16.68	0.18	1.53
0.763	1422.3	1395.9	1766.0	1418.1	1402.5	1.85	-24.16	0.29	1.38
0.848	1441.1	1416.7	1941.7	1432.6	1421.6	1.69	-34.73	0.58	1.34
0.926	1456.7	1437.0	2084.6	1445.6	1439.8	1.35	-43.10	0.76	1.15
313.15K									
0.132	1268.7	1255.1	1291.4	1268.7	1254.3	1.06	-1.78	-0.001	1.13
0.257	1297.5	1275.5	1307.3	1298.7	1278.1	1.68	-0.76	-0.09	1.48
0.371	1321.8	1295.7	1344.9	1324.5	1300.8	1.96	-1.75	-0.21	1.58
0.478	1346.6	1316.2	1431.4	1347.4	1322.9	2.25	-6.30	-0.06	1.76
0.579	1368.1	1337.1	1551.6	1368.0	1344.6	2.26	-13.41	0.003	1.71
0.677	1391.1	1358.9	1625.8	1387.0	1366.3	2.31	-16.87	0.28	1.78
0.763	1408.5	1379.4	1751.1	1403.1	1386.2	2.06	-24.32	0.38	1.58
0.848	1428.2	1401.2	1935.5	1418.3	1406.3	1.88	-35.52	0.68	1.52
0.926	1444.2	1422.6	2079.2	1431.8	1425.5	1.49	-43.97	0.85	1.29

The calculated value of ultrasonic speeds are presented in Table-2 over the entire range of compositions of DMSO. The calculated ultrasonic speeds have been compared with the experimental values and the percentage errors in the calculated values from those of the experimental values are presented in Table-2. The results in Table-2 indicate that for all the systems under study, Nomoto, Impedance dependence relation, Van Dael-Vangeel relation predict the experimental data well, while Rao's specific sound velocity relation gives maximum deviation. The percentage of deviation between the experimental and theoretical ultrasonic velocity shows that IDR is best suited for the ternary liquid mixtures. The suitability of these theories based on the percentage deviation value is  $U_{IDR} > U_V > U_N > U_R$ .

## CONCLUSION

The measured values of ultrasonic velocity, density and viscosity with derived values of several acoustic parameters suggest the occurrence of molecular interactions in the ternary mixtures of DMSO with benzene and toluene. The addition of nonpolar diluents, benzene and toluene to the polar DMSO may result in breaking of DMSO multimers thereby releasing several dipoles which may interact with the induced dipoles of non-polar molecules. The measured ultrasonic velocity was compared with the theoretical models, viz Nomoto's relation, Rao's specific sound velocity relation, Impedance dependence relation and Van Dael and Vangeel's ideal mixing relation. The comparison based on these models reveals that, IDR predicts the experimental data well for the ternary mixtures of DMSO, benzene and toluene showing smallest deviations.

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