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THEORETICALEVALUATION OF ULTRASONIC VELOCITY, ISOTHERMAL COMPRESSIBILITY AND ISENTROPIC COMPRESSIBILITY OF LIQUID MIXTURES AT ELEVATED PRESSURES

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ABSTRACT

Ultrasonic velocity, isothermal compressibility and isentropic compressibility of binary liquid mixtures have been evaluated theoretically using Flory's statistical theory under high pressures. An excellent agreement is obtained between the calculated and experimental values of these properties.

KEYWORDS: Flory's Statistical Theory, Elevated Pressures, Binary Liquid Mixtures, Isothermal Compressibility, Isentropic Compressibility

Ultrasonic study of liquid mixtures is highly useful in understanding the nature of molecular interactions present in the mixtures and ultrasonic propagation parameters are very important in understanding the physico-chemical behaviour of the liquid state.Ultrasonic velocity has been a subject of active interest as it plays an important role in the investigation of structure and intermolecular interactions between components of liquid mixtures and it has been proved to be an important property to test the validity of liquid state models.In addition,there are certain thermodynamic properties of liquid which are not easily accessible by direct experimentation. Measurement of sound velocity provides a convenient method for determining these thermodynamic properties of liquids.

Various successful attempts have been made in the recent past (Baluja and Karia, 2000) (Pandey *et al.*, 1999) (Kalidoss and Srinivasamoorthy, 1997) (Baluja and Parsonia, 1995) on theoretical evaluation of ultrasonic velocity and its correlation with other thermodynamic properties in binary liquid mixtures using statistical and semi-empirical theories. But such studies have been limited to binary liquid mixtures at zero pressures. Due to the lack of experimental data on ultrasonic velocity at elevated pressures, such studies could not be extended at elevated pressures. For the first time, Takagi and Teranishi (1988, 1985, 1984, 1982) have carried out accurate experimental measurements of ultrasonic velocity and related thermodynamic properties of few binary liquid mixtures at elevated pressures over a wide range of temperatures. Although some workon ultrasonic speed and thermodynamic parameters has been carried out, however, comparatively limited work is available at elevated pressures (Pandey *et al.*, 1997) (Lemon *et al.*, 1996) (Garg and Ahluwalia, 1995) (Krahn and Luft, 1994) (Pandey *et al.*, 1992).

Isentropic compressibility (β_S) has been widely used to study the molecular interactions through its excess value (Oswal *et al.*, 1992) (Mehta *et al.*, 1996). On the other hand, it can also be used to deduce other useful thermodynamic properties. Isothermal compressibility (β_T) is more fundamental thermodynamic quantity rather than the isentropic compressibility and is a directly measurable quantity. It is the key parameter in molecular thermodynamics of fluid phase equilibria and has been widely evaluated by many workers (Shukla and Dubey, 1985) (Mansoori *et al.*, 1971) for pure liquids and liquid mixtures.

In the present work, for the theoretical evaluation of ultrasonic velocity, isothermal compressibility and isentropiccompressibility of binary liquid mixtures under investigation,Flory's statistical theory developed for high pressures (Pandey and Singh, 2013) has been employed at pressures (0.1 MPa to 160 MPa) at 303.15K. The two binary liquid mixtures taken for the present investigation are: toluene+ o-xylene and toluene+ aniline.

THEORY

To calculate ultrasonic velocity, isothermal compressibility and isentropic (adiabatic) compressibility of the binary liquid mixtures, the expressions have been derived using Flory's statistical theory (Flory, 1965) developed for high pressures (Pandey and Singh, 2013) and are mentioned below.

Ultrasonic velocity of the mixture,

$$u = \left(\frac{1}{\beta_S \rho}\right)^{1/2} \qquad (1)$$

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The pressure-dependent expression for the isothermal compressibility is

$$\beta_T = \frac{1}{P}\left[\left\{1 + \frac{1}{\tilde{P}\tilde{V}^2}\right\}\left\{\frac{1}{3\left(\tilde{V}^{1/3} - 1\right)} - 1\right\} + 2\right]^{-1} \quad (2)$$

From the well-known thermodynamic relation,

$$\beta_T - \beta_s = \frac{\alpha^2 TV}{C_p}$$

therefore, adiabatic compressibility is given as

$$\beta_s = \beta_T - \frac{\alpha^2 TV}{C_p}$$

(3)

with the help of this equation, we can calculate the value of adiabatic compressibility for the liquid mixtures knowing α, β_T and C_P.

Equations (1), (2) and (3) have been used respectively to obtain ultrasonic velocity, isothermal compressibility and isentropic compressibility of the binary liquid mixtures under consideration with the help of Flory's statistical theory at elevated pressures. The theoretical method employed has already been detailed out earlier (Pandey and Singh, 2013). The reduced and characteristic parameters, which are used in computation, have been deduced by the method discussed there.

RESULTS AND DISCUSSION

The two binary liquid mixtures taken for the present investigation are: toluene + o-xylene and toluene + aniline. The values of ultrasonic velocity, isothermal compressibility and isentropic compressibility of the binary mixtures have been evaluated with the help of Flory's statistical theory at elevated pressures ranging from 0.1 MPa to 160 MPa and at constant temperature of 303.15 K for four different mole fractions.

The calculated values of the properties for both the mixtures are compared with the experimental ones and percentage deviations were calculated.Since the experimental values of isothermal and isentropic compressibilities were not available, we have adopted an indirect method to get these values as

$$\beta_{S(exp.)} = \frac{1}{u_{(exp.)}^2 \rho_{(exp.)}}$$

and (Pandey et al., 1997)

$$\beta_{T\,(exp.)} = \frac{17.1^{*}10^{-4}}{T^{4/9} u_{(exp.)}^2 \rho_{(exp.)}^{4/3}}$$

The experimental values of ultrasonic velocity and density of liquid mixtures have been taken from the literature (Takagi and Teranishi, 1985).

The experimental and calculated values of ultrasonic velocity, isothermal compressibility and isentropic compressibility along with the percentage deviations at different mole fractions for toluene + o-xylene and toluene + aniline are reported in tables 1, 2, 3, 4, 5 and 6 respectively.

A close perusal of tables 1, 2, 3, 4, 5 and 6 reveals that the calculated values of u, β_T and β_S for both the mixtures show the same trend as experimental one. Tables 1 and 2 show that ultrasonic velocity for both the mixtures increases as pressure increases at each mole fraction and at a constant pressure, ultrasonic velocity increases as the concentration of toluene in the mixtures decreases. For both the mixtures, percentage deviation increases as pressure increases with maximum deviation of seventeen percent.

From tables 3 and 4, it is evident that computed values of isothermal compressibility for both the mixtures decrease as the pressure increases and at a constant pressure, it decreases as the mole fraction of toluene in the mixtures decreases. The percentage deviation increases as the pressure increases and maximum percentage deviation for both the mixtures is nearly fifteen percent.

A close observation of tables 5 and 6 reveals that the variation of isentropic compressibility with pressure and also with mole fraction shows the same trend as isothermal compressibility i.e., it decreases as pressure increases or as the concentration of toluene in the mixtures decreases. In this case also, the percentage deviation increases as pressure increases but it is greater than that as observed in the case of isothermal compressibility. Here, the maximum percentage deviation is nearly forty six percent for toluene + o-xylene and is nearly forty three percent for toluene + aniline.

Table 1: Experimental and calculated values of ultrasonic velocity of x-toluene + (1-x) o-xylene at 303.15 K along with percentage deviations

u(m/sec)												
Pressure	**x = 0.8**			**x= 0.6**			**x = 0.4**			**x = 0.2**		
(MPa)	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**
0.1	1291.5	1294.5	-0.23	1302.7	1303.4	-0.05	1310.6	1312.2	-0.12	1320.4	1320.8	-0.03
10	1342.9	1318.6	1.81	1352.0	1327.6	1.81	1359.8	1336.4	1.72	1368.5	1345.0	1.72
20	1388.9	1339.1	3.59	1397.1	1347.5	3.55	1404.5	1355.7	3.47	14120	1363.8	3.41
30	1428.9	1359.5	4.85	1439.2	1367.8	4.96	1445.3	1375.9	4.80	1453.7	1383.7	4.81
40	1471.1	1379.1	6.25	1480.4	1388.5	6.21	1488.1	1397.6	6.08	1496.5	1406.5	6.02
50	1508.3	1396.8	7.39	1515.9	1405.6	7.27	1525.0	1414.3	7.26	1532.8	1422.6	7.19
60	1545.3	1414.3	8.48	1554.3	1432.2	7.86	1562.0	1443.0	7.62	1567.5	1553.5	7.27
70	1576.9	1425.2	9.62	1582.2	1446.2	8.60	1593.4	1455.1	8.68	1601.8	1463.7	8.62
80	1610.7	1440.1	10.59	1616.9	1448.8	10.39	1624.5	1457.2	10.30	1633.3	1465.4	10.28
90	1640.2	1451.2	11.52	1647.7	1459.9	11.40	1655.9	1468.4	11.33	1661.8	1476.5	11.15
100	1667.2	1461.2	12.36	1675.4	1472.7	12.10	1680.6	1479.7	11.95	1684.7	1486.5	11.76
110	1697.4	1473.2	13.21	1706.1	1489.9	12.67	1708.8	1497.6	12.36	1715.5	1505.0	12.27
120	1726.8	1484.2	14.05	1733.2	1492.1	13.91	1739.5	1499.8	13.78	1744.8	1507.1	13.62
130	1752.3	1493.1	14.79	1759.8	1500.7	14.72	1765.1	1508.1	14.56	1770.1	1515.2	14.40
140	1775.1	1503.3	15.31	1782.1	1513.8	15.06	1790.7	1520.3	15.10	1792.4	1526.7	14.82
150	1798.5	1511.8	15.94	1805.6	1518.5	15.90	1810.8	1524.9	15.79	1816.4	1531.0	15.71
160	18230	1519.1	16.67	1829.9	1530.1	16.38	1834.3	1537.8	16.16	1840.8	1545.2	16.06

Table 2: Experimental and calculated values of ultrasonic velocity of x-toluene + (1.x) aniline at 303.15 K along with percentage deviations

u(m/sec)												
Pressure	**x = 0.8**			**x= 0.6**			**x = 0.4**			**x = 0.2**		
(MPa)	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**
0.10	1337.6	1326.0	0.87	1392.0	1352.7	2.82	1459.1	1392.2	4.58	1535.8	1435.4	6.54
10	1380.0	1348.2	2.30	1434.6	1381.6	3.69	1496.4	1422.2	4.96	1569.5	1465.6	6.62
20	1421.4	1367.8	3.77	1475.6	1407.7	4.60	1534.6	1449.9	5.52	1603.3	1494.3	6.80
30	1461.4	1386.9	5.10	1510.0	1424.8	5.64	1566.8	1464.8	6.51	1634.5	1506.8	7.81
40	1501.8	1404.0	6.51	1546.3	1433.5	7.30	1600.4	1468.4	8.25	1661.5	1505.2	9.41
50	1536.5	1420.8	7.53	1580.0	1455.5	7.88	1632.0	1491.9	8.58	1688.8	1529.9	9.41
60	1572.3	1434.8	8.75	1612.3	1459.5	9.48	1660.1	1487.2	10.42	1720.4	1516.6	11.84
70	1602.2	1447.3	9.67	1648.2	1486.5	9.54	1691.8	1518.7	10.23	1747.8	1552.6	11.15
80	1635.1	1461.5	10.62	1671.9	1493.0	10.70	1716.8	1525.8	11.12	1770.9	1559.8	11.92
90	1662.3	1472.0	11.45	1701.0	1495.6	12.07	1739.8	1524.3	12.39	1794.2	1554.2	13.38
100	1692.9	1482.7	12.42	1731.6	1515.5	12.48	1770.0	1546.2	12.65	1821.5	1578.1	13.36
110	1720.7	1493.3	13.22	1756.4	1527.5	13.03	1795.8	1556.2	13.34	1844.7	1586.1	14.02
120	1748.4	1503.6	14.00	1783.0	1532.2	14.07	1822.8	1561.7	14.32	1869.2	1592.2	14.82
130	1770.2	1512.3	14.57	1805.4	1543.0	14.54	1843.6	1573.2	14.67	1888.8	1604.3	15.06
140	1798.2	1522.2	15.35	1826.8	1551.2	15.09	1865.2	1578.2	15.39	1907.7	1606.1	15.81
150	1819.0	1530.3	15.87	1850.4	1556.5	15.98	1886.3	1583.5	16.05	1929.3	1611.3	16.48
160	1842.8	1536.0	16.65	1872.4	1563.5	16.50	1907.8	1589.7	16.68	1949.6	1616.6	17.08

Table 3: Experimental and calculated values of isothermal compressibility of x4oluene + (1-x) o-xylene at 303.15 K along with percentage deviations

$\beta_T(cm^2/dyn)$												
Pressure	**x = 0.8**			**x= 0.6**			**x = 0.4**			**x = 0.2**		
(M Pa)	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**
0.10	98.82	98.28	0.54	97.13	96.54	0.61	95.96	94.86	1.15	94.54	93.25	1.36
10	90.35	91.76	-1.56	89.16	90.19	-1.15	88.16	88.69	-0.59	87.06	87.26	-0.22
20	83.56	86.38	-3.38	82.34	85.03	-3.27	81.15	83.74	-3.19	79.98	82.51	-3.17
30	78.17	81.55	-4.33	76.84	80.34	-4.56	75.91	79.19	-4.32	74.75	78.09	-4.47
40	73.07	77.28	-5.76	71.96	76.06	-5.7	70.97	74.9	-5.54	69.91	73.8	-5.56
50	68.91	73.58	-6.77	68.04	72.5	-6.55	67.01	71.48	-6.67	66.09	70.51	-6.68
60	65.12	70.19	-7.79	64.21	69.01	-7.49	63.37	67.9	-7.15	62.71	66.85	-6.6
70	62.05	67.65	-9.01	61.49	66.69	-8.45	60.44	65.78	-8.82	59.61	64.91	-8.9
80	59.05	64.94	-9.98	58.46	64.08	-9.62	57.44	63.26	-9.57	56.93	62.49	-9.77
90	56.55	62.72	-10.91	55.91	61.9	-10.72	55.2	61.13	-10.76	54.63	60.4	-10.57
100	54.37	60.72	-11.67	53.72	60.04	-11.75	53.24	59.39	-11.55	52.82	58.78	-11.29
110	52.13	58.69	-12.59	51.49	58.01	-12.66	51.18	57.36	-12.06	50.63	56.74	-12.08
120	50.06	56.85	-13.56	49.59	56.21	-13.35	49.1	55.6	-13.24	48.65	55.03	-13.1
130	48.34	55.25	-14.3	47.83	54.65	-14.28	47.42	54.09	-14.08	47.01	53.56	-13.94
140	46.84	53.65	-14.53	46.38	53.13	-14.54	45.82	52.63	-14.86	45.6	52.16	-14.39
150	45.39	52.23	-15.08	44.94	51.75	-15.14	44.58	51.29	-15.06	44.17	50.85	-15.12
160	43.95	50.95	-15.93	43.53	50.42	-15.82	43.22	49.91	-15.49	42.79	49.44	-15.53

Table 4: Experimental and calculated values of isothermal compressibility of x4oluene + (1-x) aniline at 303.15 K along with percentage deviations

$\beta_T(cm^2/dyn)$												
Pressure	**x = 0.8**			**x= 0.6**			**x = 0.4**			**x = 0.2**		
(MPa)	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**
0.1	88.69	88.11	0.65	78.54	80.44	-2.42	68.22	71.62	-4.99	58.85	63.56	-8.01
10	82.46	82.77	-0.38	73.28	74.57	-1.77	64.35	66.40	-3.19	55.95	59.02	-5.5
20	76.97	78..27	-1.69	68.67	69.70	-1.5	60.72	62.01	-2.12	53.25	55.14	-3.56
30	72.15	74.24	-2.9	65.04	66.50	-2.24	57.84	59.52	-2.92	50.90	53.25	-4.62
40	67.74	70.77	-4.48	61.55	64.57	-4.90	55.06	58.44	-6.14	48.95	52.83	-7.94
50	64.19	67.61	-5.33	58.53	61.14	-4.47	52.6	65.27	-5.07	47.09	49.93	-6.03
60	60.83	64.92	-6.72	55.82	60.30	-8.03	50.52	55.29	-9.45	45.12	50.65	-12.26
70	58.16	62.55	-7.54	53.38	56.94	-6.66	48.35	51.8	-7.12	43.49	47.09	-8.27
80	55.46	60.2	-8.55	51.24	55.01	-7.34	46.68	50.24	-7.61	42.11	45.86	-8.91
90	53.31	58.27	-9.3	49.21	54.23	-10.21	45.21	50.01	-10.63	40.81	46.09	-12.92
100	51.08	56.44	-10.5	47.21	51.81	-9.76	43.45	47.53	-9.47	39.40	43.58	-10.63
110	49.14	54.72	-11.35	45.63	50.36	-10.38	41.99	46.34	-10.36	38.22	42.62	-11.49
120	47.32	53.11	-12.23	44.04	49.00	-11.27	40.55	45.19	-11.44	37.05	41.66	-12.43
130	45.91	51.68	-12.59	42.73	47.67	-11.55	39.45	43.99	-11.51	36.12	40.59	-12.37
140	44.25	50.26	-13.58	41.53	46.57	-12.16	38.36	43.14	-12.45	35.25	39.94	-13.31
150	43.02	49.00	-13.91	40.28	45.50	-12.97	37.34	42.22	13.08	34.31	39.17	-14.14
160	41.71	47.93	-14.91	39.15	44.56	-13.81	36.34	41.41	-13.94	33.46	38.46	-14.94

Table 5: Experimental and calculated values of isentropic compressibility of x-toluene (1-x) o-xylene at 303.15 K along with percentage deviations

$\beta_S(cm^2\ /dyn)$												
Pressure	**x = 0.8**			**x= 0.6**			**x = 0.4**			**x = 0.2**		
(M Pa)	**Expt**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**
0.1	69.67	69.32	0.51	68.48	68.16	0.47	67.66	67.04	0.91	66.66	65.97	1.02
10	63.88	66.23	-3.67	63.04	65.14	-3.33	62.33	64.10	-2.83	61.55	63.10	-2.52
20	59.24	63.70	-7.52	58.42	62.72	-7.37	57.64	61.79	-7.21	56.86	60.90	-7.12
30	55.56	61.33	-10.40	54.65	60.43	-10.57	54.04	59.57	-10.23	53.26	58.74	10.28
40	52.05	59.19	-13.71	51.30	58.24	-13.54	50.63	57.34	-13.24	49.93	56.48	13.12
50	49.20	57.33	-16.53	48.61	56.46	-16.16	47.91	55.64	-16.14	47.30	54.86	-15.99
60	46.58	55.57	-19.30	45.96	54.07	-17.63	45.40	53.13	-17.03	44.96	52.25	-16.20
70	44.48	54.42	-22.34	44.10	52.72	-19.54	43.38	51.96	-19.77	42.82	51.24	-19.66
80	42.40	53.00	-25.01	42.00	52.25	-24.39	41.52	51.53	-24.12	40.97	50.85	-24.13
90	51.19	-27.22	39.76	50.50	-27.02	39.38	49.84	-26.57	40.68	51.92	-27.65	40.24
100	39.18	50.97	-30.11	38.73	50.06	-29.26	38.41	49.48	-28.83	38.13	48.93	28.33
110	37.62	49.90	-32.67	37.17	48.68	-30.97	36.98	48.09	-30.03	36.61	47.52	-29.81
120	36.18	48.95	-35.27	35.86	48.32	-34.76	35.53	47.74	-34.36	35.23	47.18	-33.91
130	34.98	48.16	-37.65	34.63	47.57	-37.34	34.36	47.01	-36.82	34.09	46.48	-36.36
140	33.95	47.31	-39.34	33.63	46.56	-38.44	33.25	46.07	-38.56	33.11	45.60	-37.72
150	32.94	46.59	-41.44	32.63	46.09	-41.24	32.39	45.61	-40.84	32.12	45.16	-40.62
160	31.94	45.97	-43.93	31.65	45.22	-42.87	31.44	44.68	-42.10	31.15	44.17	-41.79

Table 6: Experimental and calculated values of isentropic compressibility of x-toluene + (1-x) aniline at 303.15 K along with percentage deviations

Pressure	**x = 0.8**			**x= 0.6**			**x = 0.4**			**x = 0.2**		
(M Pa)	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**	**Expt.**	**Cal.**	**% dev.**
0.10	63.13	62.77	0.56	56.49	57.41	-1.62	49.64	51.43	-3.59	43.31	45.91	-5.99
10	58.84	60.21	-2.32	52.82	54.47	-3.11	46.92	48.77	-3.95	41.25	43.58	-5.66
20	55.06	58.04	-5.42	49.61	51.98	-4.77	44.36	46.48	-4.79	39.33	41.53	-5.60
30	51.73	56.06	-8.36	47.09	50.40	-7.04	42.33	45.27	-6.95	37.65	40.64	-7.92
40	48.67	54.34	-11.65	44.64	49.58	-11.07	40.36	44.92	-11.31	36.27	40.66	-12.12
50	46.22	52.74	-14.11	42.53	47.73	-12.24	38.62	43.18	-11.79	34.95	39.05	-11.73
60	43.88	51.41	-17.16	40.63	47.37	-16.60	37.15	43.44	-16.92	33.53	39.79	-18.67
70	42.03	50.25	-19.56	38.92	45.28	-16.35	35.61	41.27	-15.88	32.37	37.58	-16.11
80	40.15	49.03	-22.11	37.42	44.67	-19.39	34.43	40.70	-18.21	31.38	37.09	-18.18
90	38.66	48.09	-24.40	35.99	44.44	-23.49	33.39	40.78	-22.13	30.45	37.41	-22.83
100	37.09	47.18	-27.18	34.57	42.97	-24.28	32.13	39.30	22.32	29.43	35.95	-22.13
110	35.74	46.30	-29.54	33.46	42.11	-25.85	31.10	38.65	-24.29	28.59	35.46	-24.02
120	34.47	45.47	-31.91	32.34	41.69	-28.90	30.07	38.23	-27.15	27.75	35.07	-26.37
130	33.49	44.77	-33.68	31.42	40.95	-30.32	29.29	37.54	-28.17	27.08	34.42	-27.09
140	32.32	44.01	-36.16	30.57	40.37	-32.02	28.51	37.17	-30.35	26.46	34.23	-29.36
150	31.46	43.38	-37.87	29.69	39.94	-34.53	27.78	36.79	-32.42	25.79	33.90	-31.46
160	30.54	42.90	-40.45	28.90	39.44	-36.51	27.07	36.38	-34.40	25.17	33.57	-33.37

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