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Thermo-acoustic and nonlinear properties of Milk in $NaHCO_3$ using Volume expansion coefficient

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ABSTRACT

Thermo-acoustical parameters of liquid mixture of milk+$NaHCO_3$ have been evaluated with different mass concentration of $NaHCO_3$ in milk at different temperatures 283.15K-303.15K. The Moelwyn-Hughes parameter has been utilized to establish some simple relations between the available volume, Bayer's non-linearity parameter, internal pressure and Sharma constant. A relationship among the isobaric, isothermal and isochoric thermo-acoustical parameter have been studied and analyzed in the case of milk+$NaHCO_3$. The Sharma's parameter S_0 remains invariant with the mass concentration over a wide range of temperature and retains its characteristics value i.e. 1.11 ± 0.01 in case of present liquid system. The present treatment has the distinct advantage of volume expansivity, density and ultrasonic velocity measurement in a particular solution of milk to detect chemical additives. Analysis of such type is not found so far in literature.

Keywords: Volume expansivity, density, ultrasonic velocity, Moelwyn-Hughes parameter, available volume.

INTRODUCTION

Over last decade, a great deal of work has been carried out in the evaluation of thermo-acoustical parameters, which are useful quantities in studying the internal structure, molecular order and anharmonicity. Several approaches have been proposed to evaluate thermo-acoustical parameters for a wide variety of liquids, polymers and liquid crystals [1, 2]. Some of these parameters are of fundamental significance in the calculations of equation of state, which serve as an effective guide in determining the mechanism of ultrasonic absorption in liquid and establishing a correlation with the results for detecting chemical additives in milk [3, 4]. They made an attempt to evaluate ultrasonic velocity, density and other related parameters over a wide range of temperature and pressure to analyze interrelationship among various thermo-acoustical parameters. In recent years, Dukhin and Goetz [5-7] acoustic techniques have been applied to food industry. In the acoustic technique, the speed of sound, as compared to attenuation, are more sensitive to the evaluation of chemical compositions and temperature [8]. Recently Pandey et al. [9-11], Tabhane et al. [12] and Reddy et al. [13] used expansivity data to compute various thermo-acoustical parameters. The available volume has been found to be extensively used to understand the intermolecular interactions and other thermodynamic properties in liquid mixtures [14-16]. The variation of density and ultrasonic velocity are related to thermal expansion coefficient and also indicate interactions between the contributing molecules.

The main objective of the present paper is to compute other thermo-acoustical parameters, which are not evaluated by Gokhan et al. [3] and to test the interrelationship between Moelwyn-Hughes parameter, Bayer's non-linearity parameter, available volume, internal pressure, cohesive energy density, molecular constant, etc. These calculations are further extended to compute intermolecular thermal expansion coefficient at different mass concentration. The Moelwyn-Hughes parameter [13, 17-19] is of immense importance because of its close relationship with the Gruneisen parameter and some thermo-acoustical parameters. The ultrasonic velocity and density have been utilized to establish the relationship between volume expansivity, internal pressure, fractional free volume, repulsive exponent of intermolecular potential, Moelwyn-Hughes parameter, Bayer's non-linearity parameter, fractional available volume etc. The treatment has the distinct advantage of calculating different parameters which are interrelated through one and only one parameter, namely Moelwyn-Hughes parameter, that describes the thermoacoustic properties of liquid mixtures. Analysis of such type is not found so far in the literature. The experimental data of density and ultrasonic velocity required for the present estimation of thermo-acoustical parameters have been taken from the literature [3].

Theory

Several authors have been reported for the estimation of thermo-acoustical parameters in the literature [17-24]. As the details of the said approach are reported by various authors [17-24], the results of thermo acoustic parameters in milk with chemical additives of the present work are given in the Table-1.

Sharma [20] had obtained an expression for the parameter α in terms of molar volume (V) as,

$$\alpha = \frac{1}{V}.\left(\frac{dV}{dT}\right) \qquad (1)$$

where molar volume $V = \frac{R}{u^{1/3}}$, where R is Rao's constant given by $R = \frac{u^{1/3}}{\rho}$ & u is ultrasonic velocity.

And also obtained an expression for the Moelwyn-Hughes parameter C_1 in terms of thermal expansion coefficient (α) as,

$$C_1 = \frac{13}{3} + (\alpha \mathrm{T})^{-1} + \frac{4}{3}(\alpha \mathrm{T}) \qquad (2)$$

where T is Temperature

The summary of various interactions by Reddy et al. [13-19] between thermo-acoustical parameters C_1 & others are given as follows:

$$\frac{B}{A} = C_1 - 1 \tag{3}$$

$$\frac{V_a}{V} = \frac{2}{C_1 + 1} = \left[\frac{1}{2}\left(\frac{B}{A}\right) + 1\right]^{-1} \tag{4}$$

where V_a is the available volume

$$V_a = \left[\frac{2}{C_1 + 1}\right] * V = V * \left[\frac{1}{2}\left(\frac{B}{A}\right) + 1\right] \tag{5}$$

The symbols have their usual significance [13-19] and the above equality of the relations has not been tested so far.

Sharma and Reddy [17-25] demonstrated following interaction between n, m and C_1 as:

$$n = 3C_1 - 12 \tag{6}$$

And

$$m = 3C_1 - (n+6) \tag{7}$$

where (B/A) is the Beyer's non linearity parameter for the close packed volume; Va/V is the fractional available (free) volume; m and n are exponents which describing the magnitude of attractive and repulsive forces respectively. Usually m has the value of 6 and n>m as cited by various workers [25-27].

Reduced volume can be expressed in terms of α in zero pressure limits as

$$\overline{V} = 1 + \left[\frac{\alpha T}{3(1 + \alpha T)}\right]^3 \tag{8}$$

Assuming to a first approximation for temperature independence of C_1, showed [19, 25-27].

$$\overline{B} = \left[\overline{V}^{C_1}\right]^{-1} \tag{9}$$

where $\overline{B} = \frac{B'}{B'_{0'}}$ is the reduced bulk modulus, and $\overline{V} = \frac{V'}{V_0}$ is the reduced volume. B' and V' are the bulk modulus and volume at temperature T, and B'_0 and V_0 are the bulk modulus and volume at absolute zero [25-27].

Sharma [25] proposed a theoretical method to establish a relation between the molecular constant r and the isochoric thermo acoustical parameter δ as,

$$\delta = \frac{-\left[2+(\alpha\Gamma)^{-1}\right]\left[\left(-2\alpha\left(\overline{V}^{C_1}\right)^{-1}\right)\right]T}{2} \qquad (10)$$

Where a molecular constant $r = \frac{P_i}{\varepsilon} = (1-\delta)^{-1}$ (11)

Here P_i and ε are internal pressure and cohesive energy density respectively.

Isochoric temperature coefficient of internal pressure (P_i) has also been evaluated using α, V, C_1 and $\overline{V}$ employing the following relation:

$$P_i = \frac{-2(1+2\alpha\mathrm{T})}{\overline{V}^{C_1}} \qquad (12)$$

Huggins parameter [28, 29] as an extension to the liquid state may be expressed as

$$F = 1 + \left(\frac{2}{3}\right)\alpha\mathrm{T} + \left(\frac{d\ln\beta}{d\ln\mathrm{T}}\right)_{\mathrm{P}} + \left(\frac{\alpha\mathrm{T}}{\beta}\right)\left(\frac{d\ln\beta}{dp}\right)_{\mathrm{T}} \qquad (13)$$

$$F = 2 - S^*(2+\alpha\mathrm{T}) + \left(\frac{d\ln\beta}{d\ln\mathrm{T}}\right)_P \qquad (14)$$

$$F = 2 - S^* + S_0(S^*-1)(\alpha\mathrm{T})^{-1} \qquad (15)$$

Sharma [30] and Reddy et al. [13] have introduced two dimensionless parameters S_0 and S* as

S_0=δ (3+4αT) =3δS* (16)

S*= [1+ (4/3) αT] (17)

where δ is the isochoric acoustical parameter given by Eq (10).

Since S_o may be expressed as $S_0 = (1+2\alpha\mathrm{T})(3+4\alpha\mathrm{T})/\left(\overline{V}\right)^{C_1}$

The reduced volume expansivity $\left(\overline{\alpha}\right)$ [30] is defined as,

$$\left(\overline{\alpha}\right) = \left(\frac{\partial\ln\overline{V}}{\partial\overline{T}}\right)_P = \alpha\mathrm{T}^* \qquad (18)$$

Characteristic temperature T* [31] may be expressed as

$$\overline{T} = \frac{T}{T^*} = \frac{\left(\overline{V}^{1/3}-1\right)}{\overline{V}^{4/3}} \qquad (19)$$

where, $\overline{T} = T/T^*$ and $\overline{V} = V/V^*$ are respectively the reduced temperature and specific volume and T*, V* are the corresponding characteristic parameters (constant) of the material.

Assuming the sound velocity u is a function of both volume V and temperature T, the isobaric (K), isothermal (K') and isochoric (K") thermo acoustical parameters are related as [32, 33],

$$K' = K + K'' \qquad (20)$$

in which

$$K = \frac{5}{3} + (2\alpha T)^{-1} + \frac{2\alpha T}{3} \qquad (21)$$

$$K'' = -\frac{1}{\overline{V}^{C_1}}\left[(2\alpha T)^{-1} - \overline{V}^{C_1} + 1\right] \qquad (22)$$

The details of the above relations are available in the literature [32, 33].

RESULTS AND DISCUSSION

The necessary experimental data density (ρ) and ultrasonic velocity (u) for the system under consideration have been taken from the literature [3]. Table 1 gives the calculated values of different thermo-acoustical parameters α, C_1, V, V_a, B/A, $\overline{V}$, f, n, $\overline{B}$, δ, P_i, ε, r, F, S_o, S^*, $\overline{T}$, T^*, $\overline{\alpha}$, V*, K, K', K", etc. using given formulae (1)–(22) for different mass concentration and at temperature 293.15K-303.15K. The present treatment has the great advantage to describe thermo-acoustical parameters of liquid employing only expansivity data alone. This study gives an opportunity to test the relation between C_1, B/A, V_a and other parameters. The validity of (5) is clearly tested in the present study using experimental data.

The density is found to increase nonlinearly with increase in concentration of $NaHCO_3$ in milk. Also, it has been decrease with rise in temperature from 298.15 K to 303.15 K. It is usually due to the destruction of H-bonds & reduction in association among milk species, with increase in concentration and temperature. The u.s. velocity is found to increase with increase in concentration and temperature which is anomalous. The nonlinear variation of u.s. velocity and adiabatic compressibility of the milk + $NaHCO_3$ generally indicates complex formation [34].

Moelwyn – Hughes [20] parameter C_1 shows a non-linear increase or decrease with mass concentration, which signifies the non-linear variation of volume expansivity with mass concentration. This result indicates the associating nature of the mixture.

Bayer's non-linearity parameter B/A is strongly structure dependent quantity and characterizes the lattice behaviour of liquids. It shows nonlinear increase and decrease with rise in mass concentration of $NaHCO_3$ in milk. The increase of this parameter with mass concentration shows the decrease in intramolecular modes of vibrations and anharmonicity in liquid mixture. This shows the associating nature and weak intermolecular forces in the solutions.

Reduced volume ($\overline{V}$) and free available volume (f) are identical in nature. Reduced volume $\overline{V}$ show a nonlinear increase and decrease with mass concentration. The increased values of $\overline{V}$ and f with mass concentration shows the enhancement in the liquids, because of the increased mobility of the molecules. If the available volume has a small value then fractional free volume would also have a small value. This shows the larger size of the molecules in liquid state. The increase in available volume with raising temperature is quite natural phenomenon and it is attributed due to increase intermolecular attraction at higher temperatures.

Table 1: Acoustical parameters for milk + $NaHCO_3$ at T = 283.15–303.15K

Mass concentration(%) w → Temp.		0	0.2011	0.3909	0.5855	0.7844	0.9722	1.1683	1.551
Density (ρ)	283.15 293.15 303.15	1029.55 1027.3 1024.3	1031.16 1028.99 1025.89	1032.999 1030.78 1027.69	1033.479 1031.37 1028.19	1034.875 1032.82 1029.52	1037.041 1034.48 1031.59	1038.482 1036.13 1032.83	1041.544 1039.19 1035.94
Ultrasonic Velocity (u)	283.15	1484.13	1485.31	1487.2	1489.23	1493.34	1495.11	1497.11	1501.54
	293.15	1511.8	1513.09	1515.23	1516.75	1520.52	1521.94	1523.99	1527.90
	303.15	1531.9	1533.83	1536.03	1537.27	1540.51	1541.85	1543.99	1547.39
Molar Volume (V) * x10^{-4}	283.15	9.7130	9.6979	9.6806	9.6760	9.6630	9.6425	9.6296	9.6008
	293.15	9.7330	9.7180	9.7016	9.6957	9.6825	9.6664	9.6517	9.6230
	303.15	9.7620	9.7470	9.7304	9.7260	9.7132	9.6939	9.6825	9.6530
Volume Expansivity (α)x10^{-4}	283.15	3.0061	2.9728	2.9760	3.1324	3.1740	2.8178	3.2006	3.1591
	293.15	2.9630	2.9676	2.9696	3.1261	3.1676	2.8108	3.1932	3.1520
	303.15	2.9910	2.9588	2.9608	3.1164	3.1576	2.8029	3.1831	3.1421
Moelwyn-Hughes Parameter (C_1)	283.15	16.1953	16.3255	16.3128	15.7263	15.5803	16.9732	15.4888	15.6321
	293.15	15.961	15.9443	15.9365	15.3675	15.2263	16.5795	15.1408	15.2791
	303.15	15.482	15.6017	15.5941	15.0443	14.9079	16.2155	14.8253	14.9589
Bayer's non-linearity parameter $(B/A)_O$	283.1	15.195	15.3255	15.3128	14.7263	14.5803	15.9732	14.4888	14.6321
	293.15	14.961	14.9443	14.9365	14.3675	14.2263	15.5795	14.1408	14.2791
	303.15	14.482	14.6017	14.5941	14.0443	13.9079	15.2155	13.8253	13.9589
Availoble Volume (Va)x10^{-4}	283.15	1.1297	1.1195	1.1183	1.1570	1.1656	1.0730	1.1680	1.1545
	293.15	1.1476	1.1471	1.1456	1.1848	1.1934	1.0997	1.1959	1.1823
	303.15	1.1845	1.1742	1.1728	1.2124	1.2212	1.1262	1.2237	1.2097
Fractional free Volume (f)	283.15	0.1163	0.1154	0.1155	0.1196	0.1206	0.1113	0.1213	0.1202
	293.15	0.1179	0.1180	0.1181	0.1222	0.1233	0.1138	0.1239	0.1229
	303.15	0.1213	0.1205	0.1205	0.1247	0.1257	0.1162	0.1264	0.1253
Repulsive exponent of intermolecular Potential (n)	283.15	36.5860	36.9764	36.9384	35.1789	34.7408	38.9197	34.4663	34.8962
	293.15	35.885	35.8328	35.8094	34.1026	33.6790	37.7385	33.4225	33.8373
	303.15	34.448	34.8050	34.7824	33.1330	32.7237	36.6466	32.4758	32.8766
Reduced Volume ($\overline{V}$)	283.15	1.0805	1.0797	1.0797	1.0837	1.0847	1.0757	1.0854	1.0844
	293.15	1.0821	1.0822	1.0822	1.0863	1.0874	1.0781	1.0881	1.0870
	303.15	1.0855	1.0846	1.0846	1.0888	1.0899	1.0804	1.0906	1.0895
Reduced Bulk Modulus $\left(\overline{B}\right)$	283.15	0.2853	0.2861	0.2860	0.2825	0.2816	0.2897	0.2810	0.2819
	293.15	0.2839	0.2838	0.2838	0.2802	0.2792	0.2875	0.2786	0.2796
	303.15	0.2809	0.2817	0.2816	0.2780	0.2770	0.2855	0.2764	0.2774
Anderson Gruneisen parameter (δ)	283.15	0.3339	0.3343	0.3342	0.3326	0.3322	0.3359	0.3319	0.3323
	293.15	0.3333	0.3332	0.3332	0.3315	0.3311	0.3349	0.3308	0.3312
	303.15	0.3319	0.3322	0.3322	0.3305	0.3300	0.3340	0.3297	0.3302
Internal Pressure (Pi)x10^{8}	283.15	1.4002	1.3935	1.4006	1.4575	1.4813	1.3658	1.5028	1.5022
	293.15	1.4711	1.4775	1.4850	1.5440	1.5683	1.4453	1.5894	1.5878
	303.15	1.5526	1.5470	1.5549	1.6151	1.6389	1.5124	1.6608	1.6582
Cohesive energy density (ε) x10^{8}	283.15	0.9327	0.9277	0.9325	0.9727	0.9892	0.9070	1.0040	1.0030
	293.15	0.9808	0.9852	0.9902	1.0321	1.0490	0.9612	1.0636	1.0618
	303.15	1.0374	1.0330	1.0383	1.0813	1.0980	1.0073	1.1132	1.1107
Molecular constant (r)	283.15	1.5013	1.5021	1.5020	1.4984	1.4974	1.5058	1.4968	1.4977
	293.15	1.4999	1.4998	1.4997	1.4959	1.4949	1.5036	1.4943	1.4953
	303.15								
Huggin's Parameter (F)	283.15	2.3738	2.3749	2.3748	2.3695	2.3681	2.3802	2.3671	2.3686

Mass concentration(%) w → **Temp.**		**0**	**0.2011**	**0.3909**	**0.5855**	**0.7844**	**0.9722**	**1.1683**	**1.551**
	293.15	2.3717	2.3715	2.3715	2.3659	2.3644	2.3771	2.3635	2.3650
	303.15	2.3671	2.3683	2.3682	2.3625	2.3610	2.3740	2.3600	2.3615
Sharma's constant (S_0)	283.15	1.1155	1.1154	1.1154	1.1158	1.1159	1.1149	1.1160	1.1159
	293.15	1.1156	1.1156	1.1156	1.1161	1.1162	1.1152	1.1163	1.1161
	303.15	1.1160	1.1159	1.1159	1.1163	1.1164	1.1154	1.1165	1.1164
Sharma's Parameter(S*)	283.15	1.1135	1.1122	1.1124	1.1183	1.1198	1.1064	1.1208	1.1193
	293.15	1.1158	1.1160	1.1161	1.1222	1.1238	1.1099	1.1248	1.1232
	303.15	1.1209	1.1196	1.1197	1.1260	1.1276	1.1133	1.1287	1.1270
Reduced Temperature $(\overline{T})$	283.15	0.0236	0.0234	0.0234	0.0244	0.0247	0.0223	0.0248	0.0246
	293.15	0.0240	0.0240	0.0240	0.0251	0.0253	0.0230	0.0255	0.0252
	303.15	0.0248	0.0246	0.0246	0.0257	0.0260	0.0235	0.0261	0.0259
Characteristic Temperature (T*)	283.15	12007.0	12118.1	12107.36	11606.33	11481.53	12671.17	11403.33	11525.79
	293.15	12224.	12209.0	12202.1	11698.7	11573.7	12770.7	11498.0	11620.4
	303.15	12203.	12312.1	12305.2	11801.9	11677.0	12873.6	11601.3	11723.7
Reduced Volume Expansivity $(\overline{\alpha})$	283.15	3.6094	3.6025	3.6032	3.6356	3.6442	3.5705	3.6497	3.6411
	293.15	3.6222	3.6231	3.6236	3.6572	3.6661	3.5895	3.6716	3.6627
	303.15	3.6501	3.6429	3.6434	3.6779	3.6871	3.6083	3.6928	3.6836
Characteristic volume (V*)x10^{-4}	283.15	8.9893	8.9823	8.9656	8.9287	8.9081	8.9637	8.8718	8.8538
	293.15	8.9948	8.9800	8.9644	8.9253	8.9043	8.9664	8.8705	8.8529
	303.15	8.9934	8.9868	8.9710	8.9326	8.9117	8.9727	8.8781	8.8599
Lattice Gruneisen Parameter (Γ=K)	283.15	7.5977	7.6627	7.6564	7.3632	7.2901	7.9866	7.2444	7.3160
	293.15	7.4809	7.4721	7.4682	7.1838	7.1132	7.7898	7.0704	7.1395
	303.15	7.2414	7.3008	7.2971	7.0222	6.9540	7.6078	6.9126	6.9794
Isothermal Gruneisen parameter (Γ' or K')	283.15	6.6361	6.6772	6.6732	6.4882	6.4421	6.8816	6.4132	6.4584
	293.15	6.5624	6.5569	6.5545	6.3750	6.3305	6.7573	6.3035	6.3471
	303.15	6.4114	6.4488	6.4465	6.2731	6.2301	6.6425	6.2040	6.2461
Isochoric Gruneisen parameter (K")	283.15	-0.9616	-0.9856	-0.9832	-0.8750	-0.8480	-1.1051	-0.8311	-0.8576
	293.15	-0.9184	-0.9152	-0.9138	-0.8088	-0.7827	-1.0324	-0.7669	-0.7924
	303.15	-0.8300	-0.8520	-0.8506	-0.7491	-0.7239	-0.9653	-0.7086	-0.7333

The high value of repulsive exponent (n) shows the bulk nature of molecule. Repulsive exponent is found to show a nonlinear decrease and increase with mass concentration in the mixture of chemical additives. The decrease in n with mass concentration shows the dissociating nature of molecules and the increase in n with mass concentration thus would show associating nature of molecules.

The internal pressure (P_i) is the resultant of attractive and repulsive forces between the molecules. It measures the molecular cohesion and the instantaneous volume derivative of the milk. The increase P_i with increase in mass concentration indicates the orientation of the solvent molecules around the ions. This may be due to the influence of electrostatic field of ions i.e. solutions becomes harder to compress. This also shows associating tendency of the molecules in milk with chemical additives. The reduction in P_i with in mass concentration shows the dissociating tendency of the molecules in the solutions. This study indicates that the internal pressure in Milk + $NaHCO_3$ could also be used for studying the molecular association through hydrogen bonding [35].

The cohesive energy of a material is the energy required to disassemble it into its constituent parts, also known as binding energy. It depends upon the nature of the atoms present in the material. In the present system, the cohesive energy density (ε) increases or decreases with mass concentration. The increase in ε with mass concentration shows the decrease in degree of dissociation in the molecules.

Molecular constant 'r' measures the molecular cohesion. If 'r' is low then liquid state causes a molecular dissociative effective due to increasingly loose packing of the molecules resulting in a very weak intermolecular cohesion. The molecular constant r has the dimension reciprocal of volume. Moreover, the value of r with mass concentration is found in order in the liquid system. The reduction in the value of r shows an increasing molecular order and the increase in r with mass concentration shows decrease in molecular order.

The Sharma's parameter S_0 remains invariant even in the wide range of temperatures and mass concentration and its characteristics value is 1.11±0.01. Also the calculated values of S* are around 1.11+0.1, as compared to 1.15 for polymers [18]. The reduced bulk modulus $(\overline{B})$ is found to be approximately equal to the isochoric thermo acoustic parameter (δ) i.e., $\delta \approx \overline{B} + 0.1$.

The variation of remaining parmeters S*, S_0, F, K, K', K", $\overline{B}$, $\overline{\alpha}$, $\overline{T}$, T^*, V^* exhibits nonlinear increases or decreases with rise in mass concentration of chemical additive in milk. In the present case the thermo-acoustical parameter values of associated polar liquids like milk and chemical additives are found to be slightly deviated. This may be due to greater degree of polarisability or polar interactions & hydrogen bending effects of these molecules.

CONCLUSION

The observed complex formation and molecular interaction in the present solution is used to detect the chemical additives in food materials. Also, the association of molecules of food and chemical additives may be more at higher mass concentration of chemical additives. This indicates the strong Solute-Solvent interaction.

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