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A new ultrasonic method to detect chemical additives in branded milk

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Abstract. A new ultrasonic method – thermoacoustic analysis – is reported for the detection of the added chemical preservatives in branded milk. The nature of variation and shift in the thermal response of the acoustic parameters specific acoustic impedance, adiabatic compressibility and Rao's specific sound velocity for different samples of branded milk as compared to the chemical added pure milk are explained as due to the presence of chemicals in these branded samples.

Keywords. Acoustic parameters; chemical additives; milk.

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Adulteration of foodstuffs is a menace, which saps the vitality of common man. One of the commonly adulterated food is milk and milk products. Recent media reports reveal that many brands of milk commercially available in Kerala contain chemical additives such as sodium carbonate (Na_2CO_3), sodium bicarbonate ($NaHCO_3$), formalin (HCHO) etc. These are added to milk as neutralizers to preserve it for longer time and to prevent curdling. The continuous use of such milk may cause health hazards to the society [1,2]. Since the frequency and quantity of milk consumed by infants and children are much more compared to adults, the health risk is more to them. We are engaged in a systematic study of liquid state using some optoacoustic parameters [3,4]. In the present work, the potentiality of ultrasonics is used in the place of expensive and time consuming physical and chemical methods such as spectroscopic, chromatographic and enzymatic methods [5] to monitor the quality of milk. We are reporting a new, simple and quick method 'thermoacoustic analysis', for detecting chemical additives in branded milk. Though direct parameters such as ultrasonic velocity (U) and density (ρ) can be used for the analysis, studies on different systems of liquids and liquid mixtures reveal that, the derived acoustic parameters are more useful. So the derived parameters such as specific acoustic impedance (Z_A) [6], adiabatic compressibility (β_s) [7] and Rao's specific sound velocity (r) [8–10] are used in our present study.

The density (ρ) and ultrasonic velocity (U) were determined at five different temperatures, viz., 298.15, 303.15, 308.15, 313.15 and 318.15 K for (1) pure milk obtained from a cow, (2) pure milk added with chemicals (sodium carbonate, sodium bicarbonate and formalin) and (3) different brands of milk commercially available. The temperature was maintained constant using a thermostatically-controlled water circulating arrangement with an accuracy of ± 0.1 K. Density measurements were performed using a 12 cm^3 double stem pyknometer. Masses were measured using a single pan electronic balance with an accuracy of ± 0.1 mg. Ultrasonic velocities were measured by a single crystal ultrasonic interferometer (Mittal enterprises – Model No: F 81) at a frequency of 2 MHz with an accuracy of ± 0.1 m/s.

Thermal analysis may be defined as the measurement of physical and chemical properties of materials as a function of temperature [11]. We have extended thermal analysis to include the derived acoustic parameters Z_A, β_s and r which are defined as

$$Z_A = U\rho \tag{1}$$

$$\beta_s = 1/U^2\rho \tag{2}$$

$$r = R/m = U^{1/3}/\rho \tag{3}$$

where m is the molar mass. Although the parameter r is assumed to be temperature independent, the thermal response of this parameter is found to be significant. Thermal motion of molecules depends on temperature and is responsible for the thermal properties of all bodies. Since the molecular configuration is different for different substances depending upon the type of bonds and molecular forces in them, their thermal response is different. Thus, just as enthalpy change in a body on heating forms the basis of differential thermal analysis (DTA), the change in acoustic properties of a sample on heating forms the basis of this new technique called thermoacoustic analysis.

The density and ultrasonic velocity were measured at different temperatures for pure milk, pure milk added with chemicals such as sodium bicarbonate (3 mg/cm^3), sodium carbonate (3 mg/cm^3), formalin (0.1 ml/cm^3) and four samples of branded milk, viz., A, B, C and D available in Thiruvananthapuram. The values of Z_A, β_s and r were calculated using eqs (1)–(3) at temperatures 298.15, 303.15, 308.15, 313.15 and 318.15 K. Graphs were plotted with temperature vs. the above parameters and are shown in figures 1–3.

Figure 1a shows the variation of Z_A with temperature for pure milk and pure milk added with sodium bicarbonate, sodium carbonate and formalin. Addition of the neutralizer causes an upward shift for all the curves. The shift is maximum for HCHO added milk and minimum for $NaHCO_3$ added milk. The shape of the curves for $NaHCO_3$ and Na_2CO_3 added milk is almost similar to that of pure milk, while it is different in the case of HCHO added milk.

An analysis of the variation of Z_A with temperature for different samples reveals an upward shift for samples B and C and a downward shift for sample A (figure 1b). Sample A shows a variation similar in nature to that of pure milk. This similarity in shape, together with the downward shift of the curve, may indicate the absence of chemical additives in it. For samples B and C, the general nature of variation shows a close resemblance to that of pure milk + HCHO. This is an indication of the presence of HCHO in these samples. Sample D shows an entirely different behavior – concave up, which may be due the presence of some other additives.

From the variation of β_s with temperature we observe that chemical additives cause a downward shift. The deviation from pure milk is the greatest for HCHO added milk and the least for $NaHCO_3$ added one and Na_2CO_3 added milk lies in between these two. This is depicted in figure 2a. An analysis of thermal variation of β_s for different samples (figure 2b) shows that there is a downward shift for samples B and C and an upward shift for sample A, when compared to that of pure milk. The nature of variation for sample D is contradictory to the pure and the other samples. The downward shift and the dissimilarity in the shape of the curves indicate the presence of chemical additives in samples B, C and D. The pure milk and sample A has the same compressibility at the lowest temperature. The decrease in compressibility with temperature is smooth for pure milk and sample A.

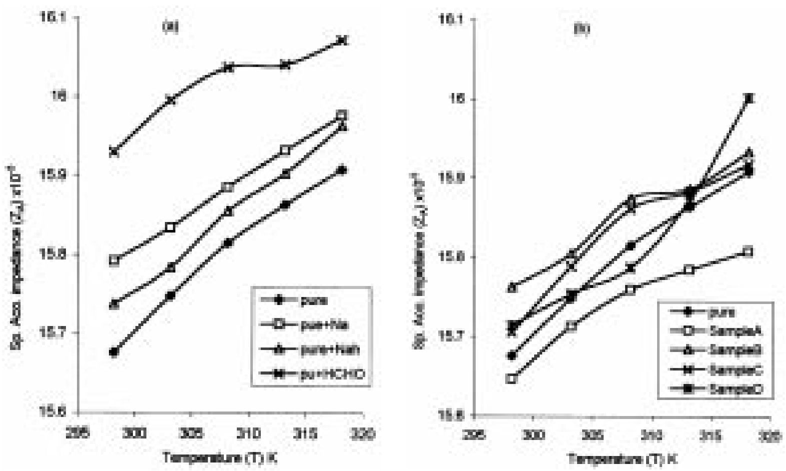

Figure 1. Variation of Z_A with temperature. (**a**) For pure milk and pure milk + chemical additives and (**b**) for pure milk and different samples.

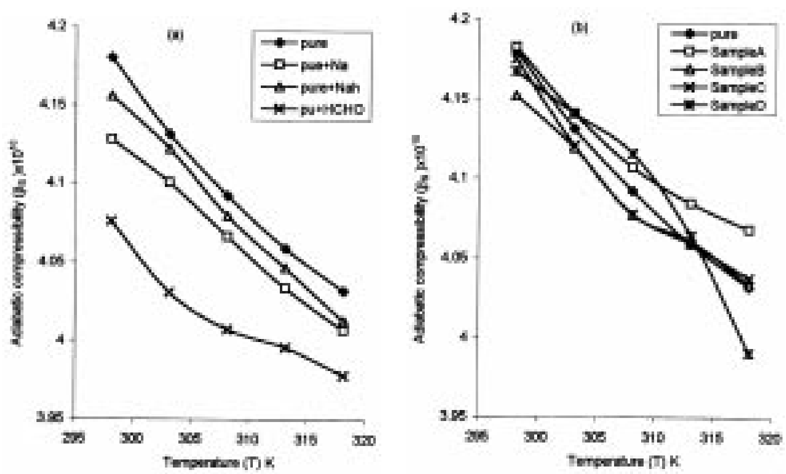

Figure 2. Variation of β_s with temperature. (**a**) For pure milk and pure milk + chemical additives and (**b**) for pure milk and different samples.

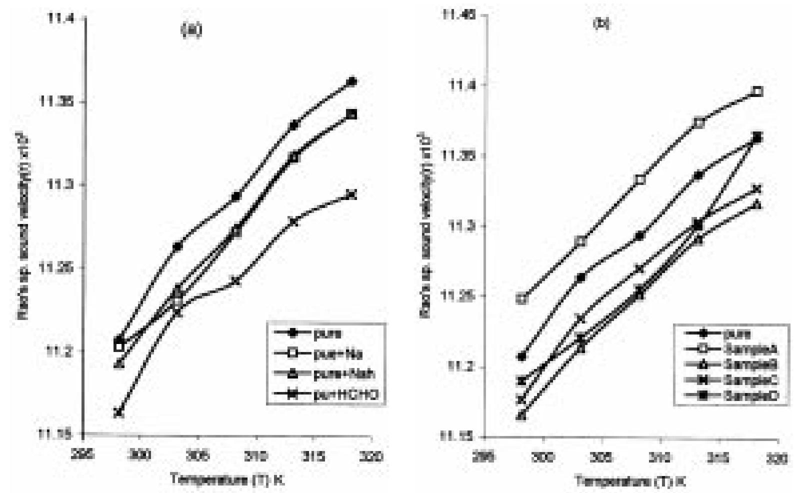

Figure 3. Variation of r with temperature. (**a**) For pure milk and pure milk + chemical additives and (**b**) for pure milk and different samples.

However, the fall in compressibility with temperature for sample A is a little slower than that of pure milk. Such a smooth variation is not seen in the case of samples B and C. Higher β_s values of sample A indicates the absence of chemical additives in it. In sodium carbonate added milk the deviation for β_s is maximum at the lowest temperature and the deviation remains constant for the whole range of temperature studied. For HCHO added milk the deviation is maximum at lower temperature and it decreases with increase in temperature and the deviation is least at the highest temperature studied here. A similar trend can be seen in the cases of samples B and C. This may be due to the fact that the samples B and C contain HCHO as neutralizer, which supports the results from the analysis of Z_A values.

Considering Rao's specific sound velocity (r) at different temperatures, it is observed that the addition of all chemicals produce a downward shift from that of the pure milk, the shift being maximum for HCHO added milk (figure 3a). Figure 3b shows the variation of r with temperature for pure milk and different samples of branded milk. The curves shift downward from that of pure milk for samples B, C and D indicating the presence of chemical additives in them, whereas for sample A the shift is upwards, showing the absence of any contamination. For HCHO added milk the shift in r is minimum at the lowest temperature and is maximum at the highest temperature. A similar trend is seen for samples B and C. This confirms our inference that samples B and C are contaminated with HCHO. The shape is entirely different for sample D, which indicates the presence of some other chemical. The curves for samples A, B and C are concave down as for the pure milk whereas it is concave up and smooth for sample D.

From this analysis it can be seen that sample D exhibits behavior totally different from pure milk and from all the other samples considered. Thus it is doubtful whether this sample can be taken as standard milk. Sample A has been certified by Central Dairy Research

Institute as the one keeping the quality of standard milk. From the present analysis, we also arrive at the conclusion that Sample A is free from chemical additives. The shift for this sample from the pure sample may be due to the presence of required level of fat content for the standard milk, which may be greater than that for the pure sample we have chosen for our present study. It is interesting to note that the nature of variation of all the acoustic parameters (Z_A, β_s and r) used in our study indicates that samples B, C and D are chemical added milk. In samples B and C, HCHO is added as neutralizer. This is apparent from the nature of variation of Z_A β_s and r with temperature. However sample D, which has a contradictory behavior is not even maintaining the standard of milk and contains some other chemical.

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