

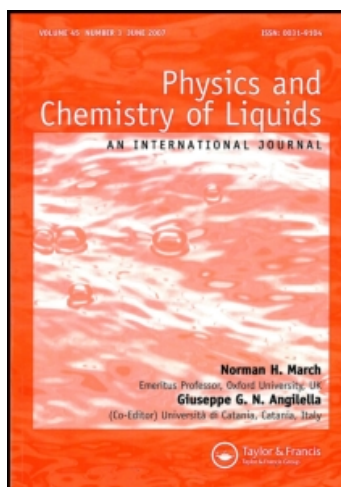
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Ultrasound velocity and density studies in some refined and unrefined edible oils

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Density and ultrasonic velocity at a frequency of 3 MHz and in the temperature range 298–333 K are measured in some of the refined and unrefined edible oils, namely coconut oil, castor oil, sunflower oil, kardi (safflower oil) and groundnut oil, which are predominantly used in south India. Velocity has been observed to be decreasing with temperature nonlinearly in some oils in the temperature range studied. This is in agreement with others' observations made in different oils and fats. Velocity change with temperature is attributed to change in intermolecular distance with temperature and the nature of variation depends on the internal molecular dynamics. Density of all the oils has been found to be decreasing with temperature. Various physical parameters such as specific volume, molar sound velocity, adiabatic compressibility, molar compressibility and intermolecular free length have been estimated using measured data on velocity and density.

Keywords: Ultrasonic velocity; Edible oils

1. Introduction

The study of ultrasonic dispersion in liquids provides valuable information about their physicochemical properties. The greatest amount of information can be obtained by measuring the velocity, v , as a function of frequency and temperature or at a fixed frequency and temperature depending on the information one is looking for, e.g. the degree of crystallization of a fat can be determined by measuring the velocity at a single frequency. Many such studies in a variety of different edible oils [1,2], vegetable oils [3] and insulator oils [4] have been reported. Edible oils are the most important components of the human diet as they provide characteristic nutrition, flavor and textures to foods and therefore, ultrasonic studies in edible oils have been a subject of study of many research scientists for many decades. The chemical constitution of the edible oils is more complex, consists of triglycerides, i.e., mixtures of esters and

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three fatty (saturated and unsaturated) acid molecules [2]. The ultrasonic studies in edible fats and oils are useful to estimate Solid Fat Content (SFC), degree of fat crystallization, molar sound velocity, v_m , adiabatic compressibility, β_{ad} , molar compressibility, β_{mol} , ultrasonic viscosity, η_{ul} , intermolecular free length, L_f , etc. Ultrasonic velocity and density studies are also used to investigate liquid crystal-like phase transitions [2,5]. Indeed, ultrasonic velocities are even used to estimate the composition and adulteration of the oils [6]. In this article, we report our experimental investigations on ultrasonic velocity and density in some of the most common domestic-used (refined and unrefined) oils in south India. Some physical parameters of the systems are also deduced from the data and are reported here.

2. Experimental

The oils selected for the study and their average molecular weights are mentioned in table 1. The saponification value, SV, of each of the oils has been experimentally determined using a chemical volumetric titration method [7]. The mean molecular weight, M , has been calculated as per the relation given in [2]

$$M = 3 \times 56108 \times \frac{1}{SV}. \quad (1)$$

The measured ultrasonic velocities in edible oils as a function of temperature, in the temperature range 298–340 K, are mentioned in table 1. These measurements were

Table 1. Saponification values and average molecular weights of the oils studied.

Name of the oil	Botanical name of the oil	Company/Firm produced	Saponification value	Mean molecular weight (g)
Refined coconut oil	<i>Cocos nucifera</i>	KLF Oil Industries Irinjalakuda, Tamilnadu, India	253.72	663.42
Unrefined coconut oil		Small Scale Industry, Bidar Karnataka, India	246.86	681.86
Refined castor oil	<i>Recinus communis</i>	Standard Laboratories Hyderabad, India	196.42	856.95
Unrefined castor oil		Small Scale Industry, Gulbarga, India	188.00	895.34
Refined sunflower oil	<i>Helianthus annus</i>	Shalimar Agro Tech (P) Ltd. Hyderabad, Andhra state, India	189.23	889.52
Unrefined sunflower oil		Small Scale Industry, Gulbarga India	186.82	901.00
Refined groundnut oil	<i>Arachis hypogea</i>	ROSG Co-op Raichur, India	192.34	875.13
Unrefined Kardi (safflower) oil	<i>Carthamus tinctorius</i>	Small Scale Industry, Gulbarga India	194.66	864.70

carried out at a fixed frequency of 3 MHz using a variable path ultrasonic interferometer (Mittal enterprises, New Delhi) with a least count of 0.0001 cm on its micrometer. The experimental procedure adopted for the determination of velocity has been explained in detail elsewhere [8–10]. The temperature variation in the systems was done by circulating hot water around the measuring cell. The temperature of the circulating water was measured using a thermometer and that was taken to be the temperature of the oil under investigation. The accuracy of temperature measurement was within 0.5°C. The errors on the measured values of velocities, Δv , are estimated to be within 1%. To check the reproducibility and to minimize the errors, the velocity measurements were repeated several times on each of the oils and their average values were taken to be the measured velocities at each temperature.

Density, ρ , measurements as a function of temperature on all the four refined oils have been performed by gravimetric method and the specific volumes, V_{sp} , are estimated as $V_{sp} = 1/\rho$.

Using the specific volume, V_{sp} , and ultrasonic velocity, v , and average molecular weight data the molar sound velocity, v_m , adiabatic compressibility, β_{ad} , molar compressibility, β_{mol} , intermolecular free length, L_f , are estimated using the following relations [2]

$$v_m = MV_{sp}(v)^{1/3}; \quad \beta_{ad} = \frac{V_{sp}}{v^2}; \quad \beta_{mol} = MV_{sp}(\beta_{ad})^{-1/7}; \quad L_f = C[\beta_{ad}]^{1/2} \quad (2)$$

where C is a temperature-dependant parameter due to Jacobson, the value of which in our studied temperature range was taken to be 631×10^{-8} (SI units).

3. Results and discussion

The measured ultrasonic velocity and specific volume data and the estimated values of molar sound velocity, adiabatic compressibility, molar compressibility, inter-molecular free length as a function of temperature of all the oils investigated, are presented in table 2. The variation of ultrasonic velocity, v , with temperature in the range 298–333 K is graphically shown in figure 1. From these it can be noticed that velocity in all the oils decreases with increase of temperature. This is in agreement with the theoretical prediction of velocity variation with temperature in high viscous oils [3] and the experimental observations made in vegetable oils [11]: natural oils of plant origin [2], sperm and seal oils [12] and coconut oil [13]. From figure 1, it is further evident that the velocity variation with temperature in refined and unrefined coconut oils, castor oils and unrefined Kardi (safflower) oil is nonlinear. This kind of nonlinear variation of velocity has been observed [13] in coconut oil in the temperature range 293–298 K. However, the linear variation of velocity with $T^{-5/4}$ as predicted [14] and shown in [13], is not in agreement with our data. It is found that no generalized type of power law can be given for the variation of velocity in all the oils reported in this article. In refined and unrefined sunflower oils and refined groundnut oil, the velocity variation with temperature is approximately linear. These observations clearly indicate that the motion of sound wave largely depends on internal molecular dynamics in each oil. At each of the temperatures considered, the velocity in castor oil is greater than that in other oils. It may be the indication for the existence of stronger intermolecular

Table 2. Ultrasonic data on different refined and unrefined edible oils.

Temperature T (K)	Ultrasonic velocity, v (m s^{-1})	Specific volume, V_{sp} ($\text{m}^3 \text{kg}^{-1}$) $\times 10^{-3}$	Molar sound velocity; $v_m \times 10^3$	Adiabatic compressibility, $\beta_{\text{ad}} \times 10^{-10}$	Molar compressibility $\beta_{\text{mol}} \times 10^{-3}$	Intermolecular free length, L_f (m) $\times 10^{-10}$
1. Refined coconut oil						
298	1420	1.0898	8126	5.4049	15.2421	1.4669
303	1405	1.0916	8111	5.5302	15.2177	1.4838
308	1390	1.0949	8107	5.6672	15.2102	1.5021
313	1384	1.0974	8113	5.7294	15.2209	1.5103
318	1376	1.1001	8117	5.8104	15.2277	1.5210
323	1368	1.1053	8140	5.9066	15.2644	1.5335
328	1360	1.1063	8132	5.9817	15.2508	1.5432
333	1356	1.1090	8143	6.0314	15.2691	1.5496
2. Unrefined coconut oil						
298	1401	1.08474	8276	5.5264	15.5428	1.4833
303	1386	1.08783	8270	5.6628	15.5329	1.5015
308	1371	1.08980	8255	5.7979	15.5087	1.5193
313	1356	1.09311	8249	5.9449	15.5003	1.5385
318	1344	1.09552	8243	6.0648	15.4901	1.5539
323	1338	1.09815	8251	6.1341	15.5022	1.5628
328	1332	1.09996	8252	6.1997	15.5042	1.5711
333	1328	1.10419	8275	6.2610	15.5419	1.5789
3. Refined castor oil						
298	1497	1.0401	10196	4.6411	19.2029	1.3593
303	1476	1.0418	10165	4.7821	19.1527	1.3798
308	1458	1.0431	10136	4.9070	19.1060	1.3977
313	1440	1.0450	10112	5.0396	19.0681	1.4165
318	1428	1.0462	10096	5.1307	19.0417	1.4292
323	1410	1.0475	10066	5.2690	18.9931	1.4484
328	1398	1.0492	10054	5.3688	18.9737	1.4620
333	1389	1.0502	10041	5.4435	18.9535	1.4722
4. Unrefined castor oil						
298	1488	1.06033	10867	4.7889	20.3622	1.3808
303	1467	1.06374	10859	4.9428	20.3355	1.4028
308	1452	1.06678	10839	5.0599	20.3256	1.4193
313	1446	1.06905	10832	5.1128	20.3386	1.4267
318	1440	1.07148	10824	5.1672	20.3540	1.4343
323	1428	1.07218	10834	5.2578	20.3167	1.4468
328	1416	1.07379	10836	5.3554	20.2939	1.4602
333	1410	1.07529	10866	5.4086	20.2936	1.4674
5. Refined sunflower oil						
298	1464	1.0926	11032	5.0977	20.6601	1.4246
303	1446	1.0945	11006	5.2348	20.6189	1.4437
308	1437	1.0971	11009	5.3131	20.6240	1.4544
313	1422	1.0988	10987	5.4342	20.5895	1.4709
318	1407	1.1007	10968	5.5604	20.5580	1.4879
323	1390	1.1036	10953	5.7124	20.5333	1.5081
328	1374	1.1063	10937	5.8604	20.5081	1.5275
333	1353	1.1093	10910	6.0597	20.4643	1.5533
6. Unrefined sunflower oil						
298	1452	1.08801	11100	5.1605	20.8025	1.4334
303	1435	1.09007	11078	5.2936	20.7663	1.4517
308	1425	1.09218	11073	5.3785	20.7592	1.4634
313	1422	1.09525	11097	5.4164	20.7967	1.4685
318	1413	1.09573	11078	5.4880	20.7668	1.4782
323	1398	1.09824	11064	5.6193	20.7441	1.4957
328	1389	1.10055	11063	5.7043	20.7432	1.5070
333	1374	1.10330	11051	5.8441	20.7233	1.5254

(Continued)

Table 2. Continued.

Temperature T (K)	Ultrasonic velocity, v (m s^{-1})	Specific volume, V_{sp} ($\text{m}^3 \text{kg}^{-1}$) $\times 10^{-3}$	Molar sound velocity, $v_m \times 10^3$	Adiabatic compressibility, $\beta_{\text{ad}} \times 10^{-10}$	Molar compressibility $\beta_{\text{mol}} \times 10^{-3}$	Intermolecular free length, L_f (m) $\times 10^{-10}$
7. Refined groundnut oil						
298	1446	1.0933	10820	5.2288	20.2656	1.4428
303	1434	1.0954	10811	5.3271	20.2514	1.4563
308	1428	1.0979	10820	5.3841	20.2663	1.4641
313	1416	1.0999	10809	5.4859	20.2495	1.4779
318	1407	1.1018	10805	5.5659	20.2427	1.4886
323	1401	1.1061	10831	5.6354	20.2848	1.4979
328	1392	1.1064	10811	5.7100	20.2521	1.5078
333	1380	1.1097	10812	5.8271	20.2538	1.5232
8. Unrefined Kardi (safflower oil)						
298	1468	1.0827	10640	5.0244	19.9444	1.4144
303	1449	1.0855	10622	5.1703	19.9143	1.4348
308	1434	1.0872	10601	5.2872	19.8815	1.4509
313	1422	1.0899	10598	5.3901	19.8758	1.4649
318	1412	1.0910	10583	5.4721	19.8525	1.4760
323	1404	1.0937	10590	5.5485	19.8628	1.4863
328	1398	1.0962	10599	5.6093	19.8781	1.4944
333	1395	1.0981	10609	5.6428	19.8941	1.4989

interaction between the triglyceride molecules in castor oil compared to other oils under consideration. Strong intermolecular interactions are expected due to the presence of hydroxyl (OH) groups within the fatty acid chains in castor oil [15]. Molar sound velocity, v_m , also decreases with temperature proportionate to sound velocity, v . In general, the reason for decrease in velocity with temperature may be due to the fact that inter-atomic distance increases with temperature because of thermal expansion and which in turn decreases the velocity. This is further supported by an idea that sound wave travels faster in the close-packed arrangement of atoms than in the loose-packed arrangement of atoms [2].

Density, ρ , values as a function of temperature, T , for all the oils are plotted in figure 2. It can be seen that density decreases monotonically and almost linearly with temperature and this is in agreement with others' observations [2,12]. Within the experimental range of temperature, the densities of refined oils were measured to be smaller than those of unrefined oils except in the case of castor oil where refined oil was found to be denser than unrefined oil. Specific volume, v_m , intermolecular free length, L_f , adiabatic compressibility, β_{ad} , are found to increase with temperature indicating the increase in void space with temperature and which in turn enhances the sound propagation. The molar compressibility, β_{mol} , decreases with temperature.

4. Conclusion

Ultrasonic velocity at a frequency of 3 MHz and density measurements in the temperature range 298–333 K have been carried out in refined and unrefined coconut oils, castor oils, sunflower oils, groundnut oil and Kardi (safflower) oil, which are predominantly used as edible oils in south India. Saponification values of the oils are determined through a chemical volumetric titration method. It has been observed

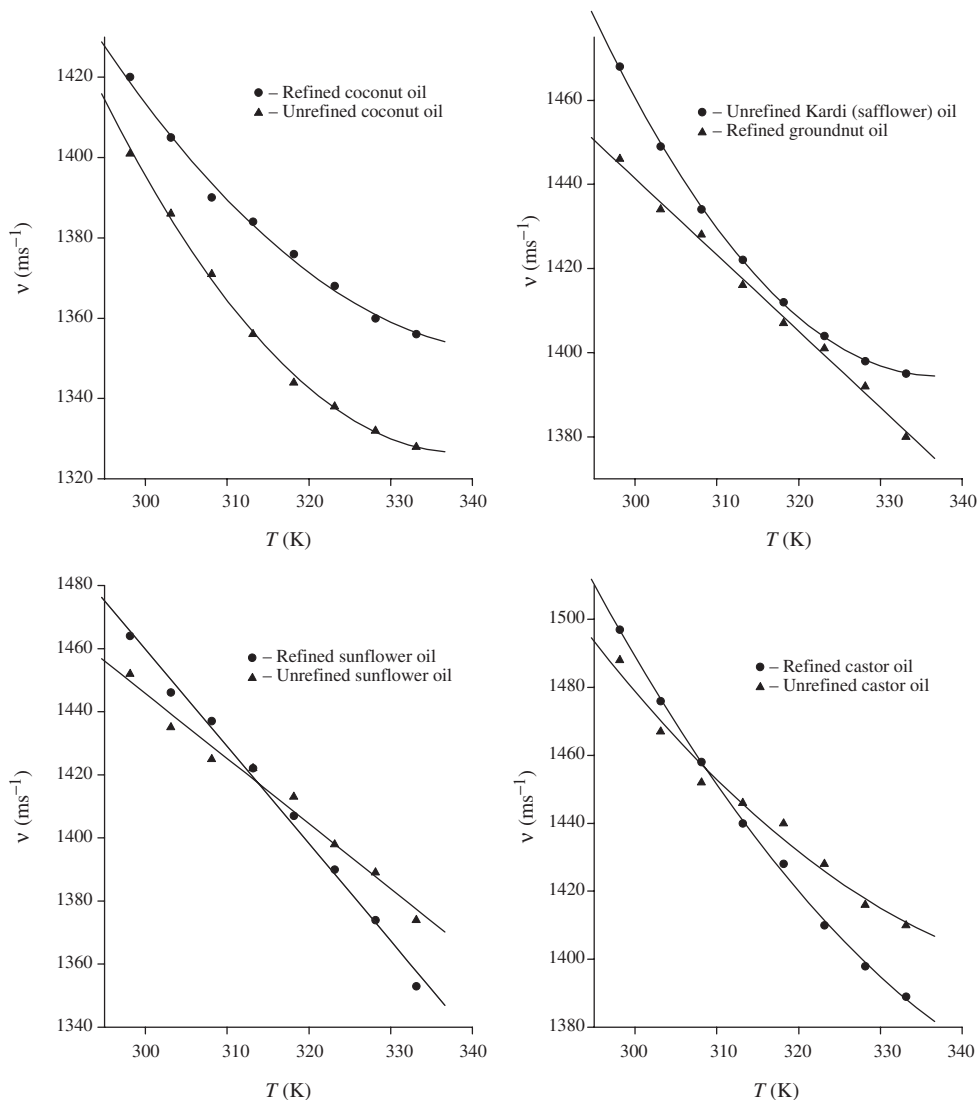


Figure 1. Ultrasonic velocity, v , vs. temperature, T , in edible oils.

that the velocity decreases approximately linearly with temperature in refined and unrefined sunflower oils and refined groundnut oil, and nonlinearly in refined and unrefined coconut oils, castor oils and unrefined Kardi (safflower) oil. Velocity change with temperature is attributed to change in intermolecular distance. Within the considered range of temperature the density of the oils was found to be linearly decreasing with temperature. Various physical parameters such as specific volume, molar sound velocity, adiabatic compressibility, molar compressibility and intermolecular free length are estimated using measured data on velocity, density and saponification values. It is the first time that such an exhaustive ultrasonic study on

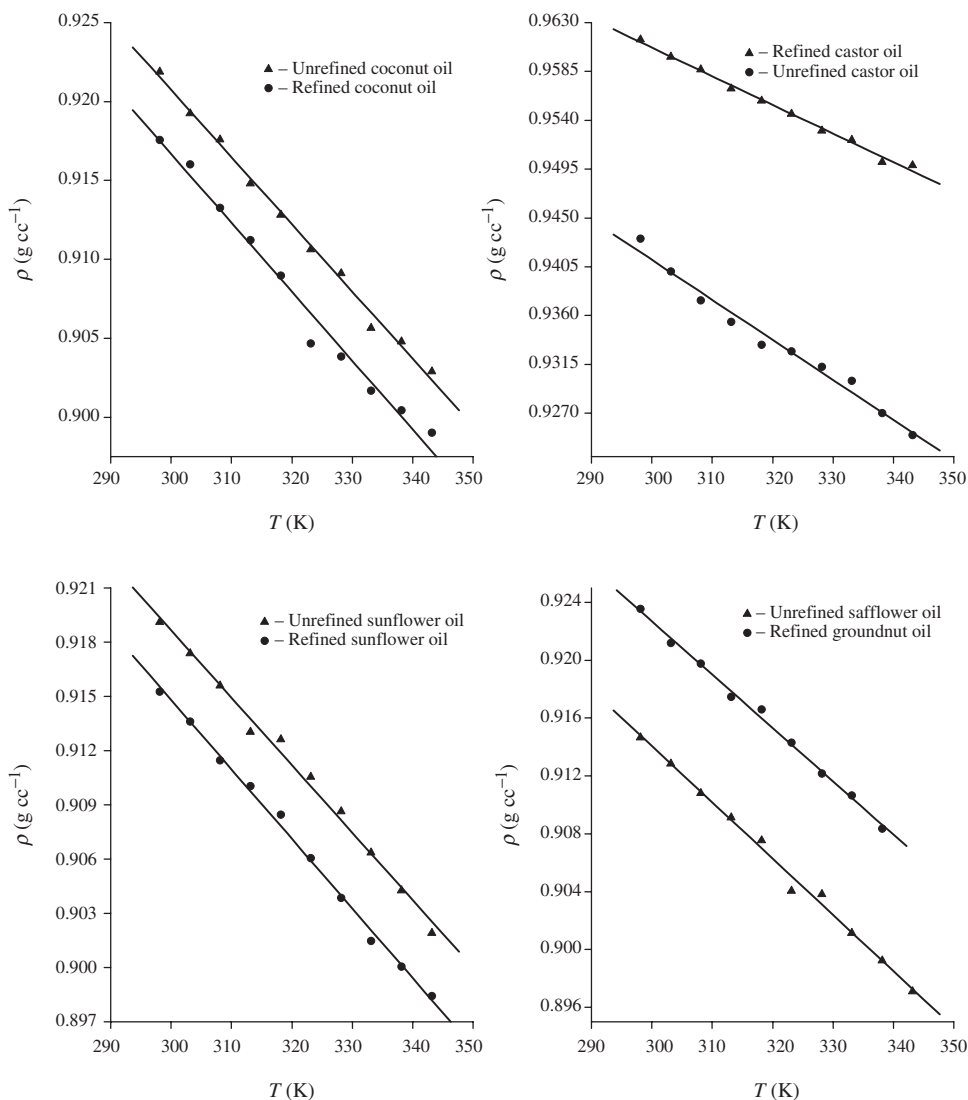


Figure 2. Density, ρ , as a function of temperature, T for edible oils.

refined and unrefined edible oils has been conducted and a good number of useful physical parameters are deduced. The data of the type presented in the article are useful for the estimation of solid fat content, composition etc. as done in [16].

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