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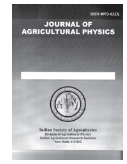

Research Article

Intercorrelations of Ultrasonic Velocity with Density and Viscosity in Adulterated Mustard Oil

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ABSTRACT

The purpose of this study was to analyse the effect of palm oil adulteration in mustard oil on ultrasonic velocity, density and viscosity. The study has been undertaken to establish intercorrelations between them for applicability of analysis techniques. The ultrasonic velocity, viscosity and density were measured at various temperatures (30, 40, 50, 60 and 70°C) for pure mustard oil, pure palm oil and different concentrations of palm oil (10, 20, 30 and 40%) in mustard oil. Various thermodynamic parameters such as surface tension, adiabatic compressibility, bulk modulus, intermolecular free length and acoustic impedance were also calculated. The ultrasonic velocity, viscosity, density, surface tension, bulk modulus and acoustic impedance were observed to decrease with increase in temperature and concentration of palm oil. The intermolecular free length and adiabatic compressibility increased by increasing temperature and concentration of palm oil. All the measured parameters of mustard oil except density were significantly varied with temperature and concentration of palm oil and were significant at the level of 5%. The best correlations of ultrasonic velocity with density and viscosity along with correlation constants, consistent in the temperature range of 30-70 °C were introduced.

Key words: Density, Mustard oil, Palm oil, Ultrasonic velocity, Viscosity

Introduction

Edible oils have crucial role in food product formulations. These are viscous liquids having important role in human diet. The compositions of edible oils consist with fatty acids and triglycerides. Fatty acids are classified into two categories, namely saturated and unsaturated fatty acids. Edible oils having good composition of polyunsaturated and monounsaturated fats are better for human well-being as compared to saturated fats. Kachchi ghani oil is mustard seed oil (Brassica Juncea) extracted using cold press technique. It is frequently used cooking oil due to its natural composition and antioxidant properties which good for human health due to its high nutritional values. Also, it contains good ratio of polyunsaturated fatty acids and monounsaturated fatty acids and superior preservative components.

The consumption of mustard oil has been increased and hence, it's adulteration is increased for economic perspective. Edible oils are simply mixed with cheap quality oils because of their fluidic nature. The adulterant substance has similar physical appearance as that of pure edible oil but differs from natural nutritional and medicinal properties of pure edible oil. Therefore, there is urgent need of more efficient methods for determining the adulteration in mustard oil.

The non-destructive techniques (NDT) are used for determining the quality of mustard oil so

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as to retain its physical and chemical properties. The ultrasonic technique is predominant as NDT in which ultrasonic waves having known frequency require medium to propagate through food. Ultrasonic waves are the mechanical waves having frequency higher than 20 KHz. The adulterants change the physiochemical properties of natural mustard oil and this affects the ultrasonic velocity of waves through the sample. Ultrasonic wave velocity depends on temperature and can be employed for qualitative and quantitative assessment of adulteration at different temperatures. The ultrasonic velocity can be employed to evaluate thermodynamic parameters.

Viscosity is an imperative parameter for fluid processing. It is a physical property and highly affected by the amount of adulterants. The viscosity of adulterated mustard oil differs from natural mustard oil. The measurement of density is important for quantifying adulteration as it is unique for each substance. The dependence of viscosity and density on temperature is also useful for analysis of adulteration over a range of temperature.

In the present work, the effect of temperature and adulteration of palm oil in mustard oil on ultrasonic velocity, viscosity and density has been done.

Materials and Methods

In this study, kachchi ghani mustard oil and refined palm oil were procured from a local departmental store. The raw mustard oil was adulterated with four different concentrations (10%, 20%, 30% and 40%) of palm oil on volume by volume basis. All the measurements were made for pure mustard oil, pure palm oil and four different concentrations of palm oil in mustard oil in the temperature range 30 to 70°C.

The ultrasonic velocity was measured using ultrasonic interferometer having dual frequency of 1 and 3 MHz (Batra Trading Company, Timber Market, Ambala and model 2759/20). The measurements were made at frequency of 3 MHz. Viscosity of mustard oil was measured using Ostwald viscometer. To measure density, the specific gravity method was used using water as a reference liquid.

Results and Discussion

Variation of ultrasonic velocity with temperature

The variation of ultrasonic velocity with temperature of pure mustard oil, pure palm oil and different concentrations of palm oil in mustard oil is as shown in Fig. 1. The ultrasonic velocity is more in mustard oil than palm oil. It is due to the fact that mustard oil has high viscosity and density as compared to palm oil. The similar ultrasonic velocity of mustard oil was 1440 m/s at 30°C and reported by Kumari *et al.* (2015). The ultrasonic velocity of palm oil was 1419 m/s as observed by Ali and Ali (2014). The ultrasonic velocity was decreased with increase in temperature for pure mustard oil, pure palm oil and all concentrations. The similar trend of ultrasonic velocity of heated and unheated mustard oil was observed by Valantina *et al.* (2016). The heating time was increased from 30 to 120 minutes and the ultrasonic velocity decreased from 1440 to 1320m/s respectively. The ultrasonic velocity of mustard oil samples decreases with increase in temperature due to intermolecular free length. By increasing the temperature, thermal energy increases and thus, intermolecular free length increases (Kumari *et al.,* 2015). The molecules at high temperature

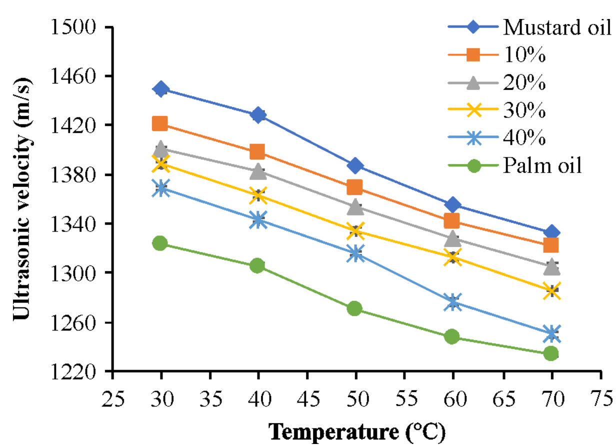

Fig. 1. Variation in ultrasonic velocity with temperature of mustard oil, palm oil and at different concentrations of palm oil in mustard oil

have high energy states and vibrate fast and therefore, ultrasonic waves can travel slower. In case of palm oil, the ultrasonic velocity was decreased with increase in temperature. Similar trend of ultrasonic velocity in palm oil with temperature was reported by Sidek *et al.* (1996). The decrease in velocity with temperature in palm oil might be due to chain length, thermal conduction and degree of unsaturation as explained by Javanaud and Rhalkar (1988). The ultrasonic velocity decreased by increasing palm oil concentration in mustard oil. With increase in quantity of palm oil in mustard oil, attraction between palm oil and mustard oil molecules increase and results in increase in intermolecular free length and hence that reduces ultrasonic velocity.

Variation of viscosity with temperature

The effect of temperature on viscosity for pure mustard oil, pure palm oil and different concentrations of palm oil in mustard oil was plotted as shown in Fig. 2. Mustard oil has higher viscosity than palm oil. The similar value of viscosity observed for mustard oil in units of 56.17 m.Pa.s at 35! as reported by Valantina *et al.* (2016). The value of viscosity of palm oil was 27.00 m.Pa.s and observed by Davies (2016). Viscosity decreased exponentially with increasing temperature in mustard oil, palm oil and all concentrations. The similar variation in viscosity of mustard oil with temperature in the range 25-85°C was studied by Valantina *et al.* (2016). The viscosity of palm oil decreased with temperature as reported by Keshvadi *et al.* (2012). The decrease in viscosity with increase in temperature is due to molecular interaction. The cohesive forces between molecules of the medium decrease with increase in temperature resulting in decrease in viscosity (Ebong *et al.,* 2014). The decrease in viscosity with increase in temperature is also due to intermolecular interactions which decrease by thermal effect (Valantina *et al.,* 2013). With increase in temperature, layers of the liquid pass easily to each other which contributes to decrease in viscosity. Viscosity depends on composition of liquid and decreases by decreasing the unsaturation of fatty acids (Zahir *et al.,* 2017). Viscosity decreased exponentially by increasing the concentration of palm oil in mustard oil. The similar trend was observed by Kumari *et al.* (2015). At high concentration of mustard oil, the intermolecular distance between the components of medium is less and results in strong bonding. This strong bonding attributes to high viscosity of pure mustard oil. With the addition of palm oil, the bond arrangement of constituents of pure mustard oil get disturb decreasing the viscosity of mustard oil.

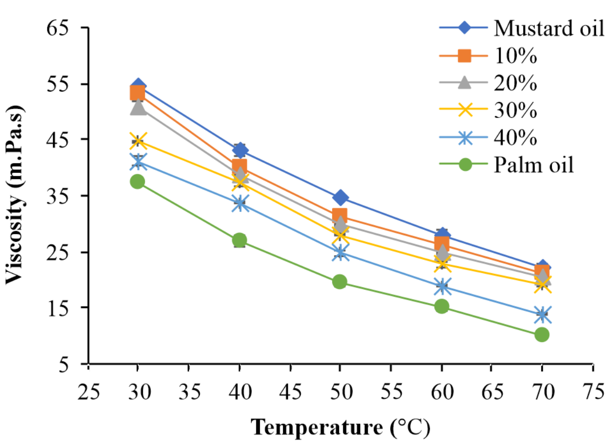

Fig. 2. Variation in viscosity with temperature of mustard oil, palm oil and at different concentrations of palm oil in mustard oil

Variation of density with temperature

The variation of density with temperature for pure mustard oil, pure palm oil and different concentrations of palm oil in mustard oil are expressed graphically as shown in Fig. 3. Pure mustard oil has high density. The density of mustard oil was 904 kg/m^3 at 30°C as observed by Kumari *et al.* (2015). The similar value of density of palm oil was 849.6 Kg/m^3 at 71.1°C as reported by Noureddini (1992). Density was observed to decrease with increase in temperature non-significantly. The similar trend of decrease in density with temperature of different edible oils was reported by Sankarappa *et al.* (2005). The increase in temperature attributes to breaking of bonds in the sample and hence, constituents are loosely packed resulting to decrease in density. Density was found to decrease non-significantly by increasing the concentration of palm oil in mustard oil. The density showed

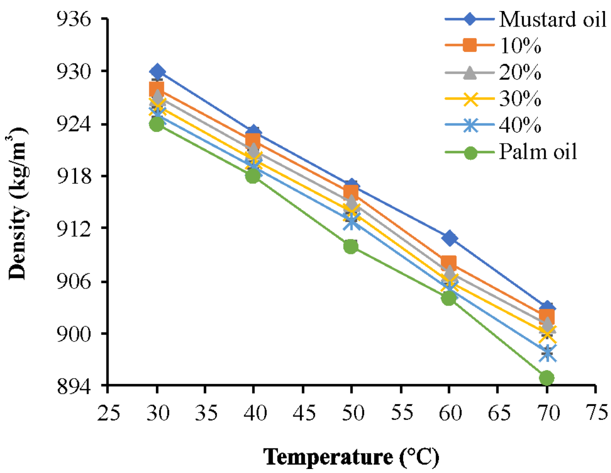

Fig. 3. Variation in density with temperature of mustard oil, palm oil and at different concentrations of palm oil in mustard oil

similar linear trend in mustard oil adulterated with palm oil (10% and 20%) as reported by Kumari *et al.* (2015) at 30°C. With the addition of palm oil, the constituents of mustard oil get disturbed and formation of new bonds occurs and influences its density.

Correlation of ultrasonic velocity with density and viscosity

The ultrasonic velocity was measured using electrical method while density and viscosity were measured using mechanical method. Therefore, measured ultrasonic velocity has more precision than the measured density and viscosity. The correlation of ultrasonic velocity with density and viscosity are plotted in graph for mustard oil, palm oil and different concentrations of palm oil in mustard oil as shown in Fig. 4 and Fig. 5, respectively. The logarithmic velocity and logarithmic density are evaluated to attain a linear relationship between the ultrasonic velocity and density as shown in Fig. 4 while the logarithmic viscosity is evaluated to accomplish a linear relationship between the ultrasonic velocity and viscosity as shown in Fig. 5. These correlations help in determining the quality of mustard oil non-destructively.

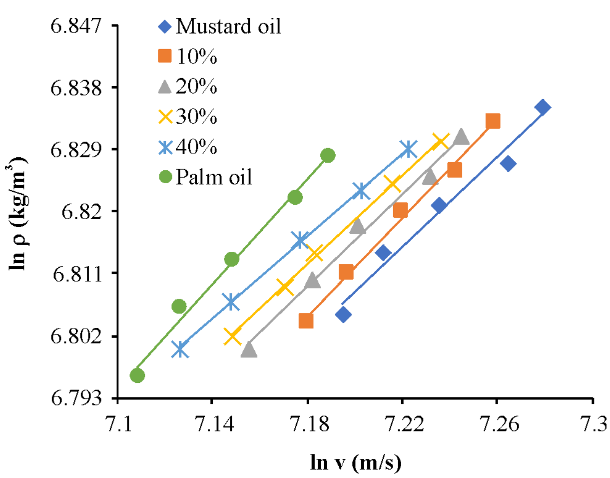

Fig. 4. Correlation between ultrasonic velocity and density of mustard oil, palm oil and at different concentrations of palm oil in mustard oil at different temperatures

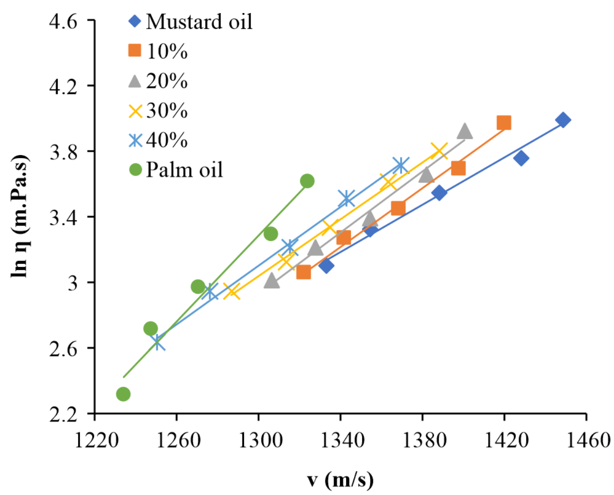

Fig. 5. Correlation between ultrasonic velocity and viscosity of mustard oil, palm oil and at different concentrations of palm oil in mustard oil at different temperatures

Mathematical modelling of ultrasonic velocity, density, viscosity and correlations

The fitting was constructed using the data of ultrasonic velocity, density and viscosity for mustard oil, palm oil and different concentrations of palm oil in mustard oil at different temperatures. The linear model was obtained for ultrasonic velocity and density with temperature and is given below,

$$y = a + b\,T$$

Where, y is the given parameter of mustard oil and T (°C) is the temperature. The values obtained of regression coefficients a, b and the coefficient of determination R^2 for ultrasonic velocity and density are shown in Table 1 and 2, respectively. The fitted correlation of viscosity with temperature is given below,

$$\eta = a\, e^{bT}$$

Where, η is the viscosity at temperature T in °C. The values obtained of regression coefficients a, b and the coefficient of determination R^2 for viscosity are shown in Table 3.

Table 1. The constants and coefficients of variation of ultrasonic velocity for mustard oil, palm oil and different concentrations of palm oil in mustard oil at different temperatures (30-70°C)

Concentrations (w/w, %)	a	b	R^2
Mustard oil	1543.20	-3.05	0.99
10	1493.30	-2.38	0.99
20	1475.00	-2.42	0.99
30	1475.80	-2.83	0.98
40	1467.10	-3.10	0.99
Palm oil	1395.80	-2.30	0.99

Table 2. The constants and coefficients of variation of density for mustard oil, palm oil and different concentrations of palm oil in mustard oil at different temperatures (30-70°C)

Concentration (w/w, %)	a	b	R^2
Mustard oil	949.80	-0.66	0.99
10	948.20	-0.66	0.99
20	947.22	-0.66	0.99
30	946.21	-0.66	0.99
40	946.00	-0.68	0.99
Palm oil	946.20	-0.72	0.99

Table 3. The constants and coefficients of variation of viscosity for mustard oil, palm oil and different concentrations of palm oil in mustard oil at different temperatures (30-70°C)

Concentrations (w/w, %)	a	b	R^2
Mustard oil	105.38	-0.02	0.99
10	100.57	-0.02	0.99
20	97.23	-0.02	0.99
30	86.69	-0.03	0.99
40	97.10	-0.03	0.99
Palm oil	97.34	-0.03	0.99

Table 4. The constants and coefficients of correlation of ultrasonic velocity with density for mustard oil, palm oil and different concentrations of palm oil in mustard oil at different temperatures (30-70°C)

Concentrations (w/w, %)	a	b	R^2
Mustard oil	4.45	0.32	0.97
10	4.24	0.35	0.99
20	4.40	0.33	0.99
30	4.51	0.31	0.99
40	4.66	0.29	0.99
Palm oil	4.10	0.37	0.99

The correlation of ultrasonic velocity with density was fitted linearly using the logarithmic data. The R^2 values show the logarithmic ultrasonic velocity depends upon logarithmic density as shown in Table 4. The relation between ultrasonic velocity and density is given below

$$\ln \rho = b \ln v + a$$

Where, v is ultrasonic velocity in m/s and ρ is density in Kg/m^3.

The linear regression equation was developed relating the ultrasonic velocity with viscosity. The R^2 values show the logarithmic viscosity depends upon ultrasonic velocity as given in Table 5. The relation between ultrasonic velocity and logarithmic viscosity is given below

$$\ln \eta = bv + a$$

Where, v is ultrasonic velocity in m/s and η is viscosity in m.Pa.s.

Table 5. The constants and coefficients of correlation of ultrasonic velocity with viscosity for mustard oil, palm oil and different concentrations of palm oil in mustard oil at different temperatures (30-70°C)

Concentrations (w/w, %)	a	b	R^2
Mustard oil	-6.41	0.01	0.99
10	-8.75	0.01	0.99
20	-9.91	0.01	0.98
30	-8.20	0.01	0.99
40	-8.53	0.01	0.99
Palm oil	-13.76	0.01	0.96

Variation of thermodynamic parameters with temperature

Surface tension for mustard oil, palm oil and different concentrations of palm oil in mustard oil has been calculated at different temperatures as given in Table 6. The surface tension decreased linearly with increase in temperature at a considered temperature. As the temperature increases, the molecules are loosely bound and the kinetic energy of molecules increases and cohesive forces between the molecules decrease. The similar variation of surface tension with temperature was observed in the temperature range 40 to 70°C for vegetable oils by Dikko *et al.* (2015). Surface tension get lowered by increasing the concentration of palm oil because intermolecular separation increases on dilution of sample resulting into decrease in interaction of surface molecules.

The adiabatic compressibility has been evaluated at different temperatures for mustard oil, palm oil and different concentrations of palm oil in mustard oil as shown in Table 6. Adiabatic compressibility was observed to increase by

Table 6. Thermodynamic properties for mustard oil, palm oil and different concentrations of palm oil in mustard oil at different temperatures (30-70°C)

Temperature (°C)	Mustard oil	10%	20%	30%	40%	Palm oil
			Surface tension (N/m)×10^{-2}			
30	4.00	3.88	3.80	3.74	3.67	3.48
40	3.91	3.77	3.71	3.63	3.54	3.38
50	3.73	3.69	3.59	3.51	3.45	3.29
60	3.60	3.57	3.48	3.42	3.32	3.18
70	3.51	3.48	3.39	3.25	3.14	3.10
			Adiabatic compressibility (m^2/N) ×10^{-10}			
30	4.15	4.32	4.44	4.53	4.66	4.98
40	4.26	4.48	4.59	4.72	4.87	5.17
50	4.54	4.60	4.78	4.93	5.03	5.35
60	4.77	4.82	4.98	5.10	5.31	5.60
70	4.93	4.99	5.15	5.45	5.70	5.81
			Bulk modulus (N/m^2) × 10^8			
30	24.0	23.1	22.4	22.0	21.4	20.0
40	23.3	22.3	21.8	21.1	20.5	19.3
50	21.9	21.7	20.8	20.2	19.8	18.6
60	20.9	20.7	20.0	19.5	18.8	17.8
70	20.2	20.0	19.3	18.3	17.5	17.2
			Acoustic impedance (Ns/m^3) × 10^5			
30	16.6	16.2	16.0	15.8	15.6	15.1
40	16.3	15.9	15.7	15.5	15.2	14.7
50	15.8	15.7	15.4	15.2	15.0	14.5
60	15.4	15.3	15.1	14.9	14.6	14.2
70	15.2	15.1	14.8	14.4	14.0	13.9
			Intermolecular free length (A^0)			
30	0.40	0.41	0.41	0.42	0.42	0.44
40	0.41	0.42	0.43	0.44	0.44	0.46
50	0.44	0.44	0.45	0.45	0.46	0.47
60	0.46	0.46	0.47	0.47	0.48	0.49
70	0.47	0.48	0.48	0.50	0.51	0.51

increasing the temperature. Similar variation of adiabatic compressibility with temperature of mustard oil adulterated with palm oil was reported by Kumari *et al.* (2015). With increase in temperature, magnitude of interaction between the molecules decreases and attributes to easy movement of molecules. This results to increase in adiabatic compressibility. This increase in adiabatic compressibility by increasing concentrations of palm oil is due to interaction of mustard oil and palm oil molecules.

The calculated values of bulk modulus for mustard oil, palm oil and different concentrations of palm oil in mustard oil at different temperatures are shown in Table 6. Bulk modulus is the measure of resistance offered to the compressibility and was observed to decrease with increase in temperature. The similar trend of bulk modulus of edible oils with temperature was investigated by Ghosh *et al.* (2017). At high temperatures, compressibility of mustard oil samples increases and results into decrease in bulk modulus. Bulk modulus decreased with the addition of palm oil in mustard oil, because interaction between mustard oil molecules decreases. This interaction turns in to increase in compressibility of mustard oil samples resulting into decreasing the bulk modulus.

The acoustic impedance has been calculated for mustard oil, palm oil and different concentrations of palm oil in mustard oil at different temperatures as shown in Table 6. Acoustic impedance decreased with increase in temperature. As it is the measure of resistance offered to the path of ultrasonic waves. The decrease in ultrasonic velocity with temperature gives information about the easy flow of sound waves through the sample, resulting in decrease in acoustic impedance (Singh and Bhatt, 2010). The decrease in acoustic impedance by increasing the concentration of palm oil is due to interactions which increases the intermolecular free length. This increase tends to decrease in ultrasonic velocity, resulting in acoustic impedance.

The evaluated values of intermolecular free length for mustard oil, palm oil and different concentrations of palm oil in mustard oil at various temperatures are shown in Table 6. The intermolecular free length increased with increase in temperature due to interaction. The increase in temperature results to increase in intermolecular free length as it is distance between neighbouring molecules. Intermolecular free length was obtained to increase linearly with increase in concentration of palm oil as the interaction between mustard oil molecules decreases. This decrease attributes to weakening of intermolecular forces between the constituents of mustard oil and results to increase in intermolecular free length. The similar trend of variation with concentration was evaluated in mustard oil adulterated with palm oil having concentrations of 10% and 20% at 30°C by Kumari *et al.* (2015). The similar results of increase of intermolecular free length with decrease in concentration of pure sample were obtained by Gupta *et al.* (2015) in the liquid medium of acetonitrile.

Conclusions

The ultrasonic measurement provides prospects for non-invasive sensing of quality indicators in food products. The ultrasonic properties are correlated with the quality characteristics such as density and viscosity. The regression equation relating the ultrasonic velocity with density and viscosity had a high coefficient ($R^2>0.95$) in the temperature range 30-70°C for mustard oil, palm oil and different concentrations of palm oil in mustard oil. These good correlations can be established with well-designed instruments for calibration and this can help in assessing the required information for better characterisation of food products.

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