

DIELECTRIC PROPERTIES OF SOME EDIBLE AND MEDICINAL OILS AT MICROWAVE FREQUENCY

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ABSTRACT

Dielectric properties of some edible and medicinal oils have been studied at 455 KHz, 9.1 GHz and optical frequency for the temperature range of 293⁰K to 323⁰K. It has been observed that the dielectric constant at optical frequency (ϵ_{ω}) is almost same for all oil samples where as variation in dielectric permittivity (ϵ') is more prominent at lower frequencies for pure oils. The static dielectric constant (ϵ_0) and dielectric constant at optical frequency (ϵ_{ω}) decrease slightly with increase in temperature for all the oil samples. Dielectric permittivity (ϵ') and loss tangent ($\tan \delta$) of different oil samples at 9.1 GHz show some interesting behaviour at particular temperature. The macroscopic relaxation time (τ_m) and molar free energy of activation (ΔF_e) is also determined which show systematic variation with temperature for most of the oil samples under consideration.

Keywords: Dielectric constant, relaxation time, molecular interaction, gas chromatography.

INTRODUCTION

Dielectric studies of biological substances and agrifood materials in microwave region of electromagnetic (e-m) spectrum are one of the areas of increasing importance. The dependence of dielectric properties of agrifood material on its density, packing fraction, moisture content and frequency of e-m wave has been studied for different grains and food materials (Nelson, 1982; Nelson, 1983^a; Venkatesh *et al.*, 1996). It is important to investigate dielectric properties of agrifood materials to develop microwave based process controls. Further, the dielectric relaxation studies have been widely utilized to investigate the molecular structure and related phenomenon of various liquids in pure and mixture form. However, very less information is available for many agrifood materials. Recently dielectric properties of grains of various packing densities, TyloseTM (a complex food material) and edible oils (Canola, Soya, Sunflower, coconut, groundnut, linseed and mustard oil) were studied at some microwave frequencies (Venkatesh *et al.*, 1996; Bansal *et al.*, 2001; Agrawal and Bhatnagar, 2005). Edible oils are one of the agrifood materials which are highly consumed and the possibility of adulteration is also very high as most of the oils are odorless and colorless. Dielectric studies of pure oils are of importance as any variation from these standard values can be attributed to adulteration or blending with other oils. The dielectric properties of Canola, Soya and Sunflower oils are studied at different temperatures and it was found that all three oils exhibit similar responses at 2.45 GHz, whereas at 915 MHz the values of dielectric permittivity (ϵ') and dielectric loss (ϵ'') responded differently (Venkatesh *et al.*, 1996). Dielectric properties of different varieties of rapeseed-

mustard oil and its mixing with common adulterant (argemone oil) at 8.93 GHz have been studied and the effect of adulterant is also reported (Bansal *et al.*, 2001). Recently the dielectric studies of binary mixture of some edible oils have also been done at microwave frequency (Agrawal and Bhatnagar, 2005).

In the present study, dielectric properties of some oils, under edible and medicinal category were studied at microwave frequency with temperature. The macroscopic relaxation time (τ_m) and the molar free energy of activation (ΔF_e) have been determined for all the oil samples and their variation with temperature is studied. Gas chromatography of various oils (both edible and medicinal) has been done to study the possible correlation of dielectric properties with the different fatty acid compositions present in oil samples. The effect of mixing of various oils on their dielectric constant has also been studied to see the adulteration (or blending) effect. The oils studied are groundnut oil, sunflower oil, sesame oil, cottonseed oil, Soyabean oil (in edible oil category) and Almond oil, Olive Oil, Neem Oil (in medicinal oil category).

MATERIALS AND METHODS

Commercially available double filtered oil samples are used for present investigation. The gas chromatography of oils (both edible and medicinal) is done at Gujarat laboratories, Ahmedabad using Chemito Model No. 1000. The dielectric permittivity (ϵ') and dielectric loss (ϵ'') are determined at microwave frequency of 9.1 GHz with temperature, using the method suggested by Heston *et al.* (1950) adopted for short circuit termination and described elsewhere (Vashishth, 1990). Static permittivity (ϵ_0) of

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oils is measured at 455 KHz for different temperatures by resonance method which uses a tuned oscillator circuit and standard variable capacitor. The permittivity at optical frequency (ϵ_{ω}) is taken as square of refractive indices for sodium D-line measured by Abbe's refractometer. All the measurements were done for a temperature range of 293-323°K. The temperature was electronically controlled within $\pm 0.5^{\circ}\text{C}$ using a constant temperature water bath system. The loss tangent ($\tan\delta$) is determined for various oils at different temperatures. The macroscopic relaxation time is determined for various oils using Debye equation (Frohlich, 1958) and molar free energy of activation (ΔF_c) is calculated using Eyring's rate equation (Glastone *et al.*, 1941).

RESULTS AND DISCUSSION

The measurements of dielectric permittivity (ϵ') at microwave frequency of 9.1 GHz, static dielectric constant (ϵ_0) and dielectric constant at optical frequency of sodium light (ϵ_{ω}) for different oils at temperature of 293K is done and tabulated in table 1. It has been observed that the dielectric constant at optical frequency (ϵ_{ω}) is almost same for all oil samples where as more variation is seen at lower frequency. The gas chromatography (GC) of edible and medicinal oils is done and two typical chromatograms (one for edible and other for medicinal oil) are shown in figures 1 and 2. The fatty acid composition of these oils has been tabulated in table 2. The peak corresponds to different retention time related to various fatty acids present in the oils. Four types of fatty acids are found to be present in almost all the oil samples except the olive oil sample where Linoleic acid is totally absent but none of the oils are showing exactly same fatty acid composition. Tables 1 and 2 shows that the dielectric constant of sunflower and sesame oil comes out to be same although their fatty acid compositions are different. Similar is the case for almond oil and olive oil. So it is observed that although the fatty acids present in oils are in different proportion but their dielectric constant lie very nearby. Similar results are obtained by Bansal *et al.* (2001) for rapeseed mustard oil of different variety having varying proportion of Erucic acid. The Fatty acids contain molecules made up of varying number of carbon, oxygen and hydrogen atoms with very long chain and most of them in the range of 12 to 22 atoms of carbon.

These are molecules of complex nature and hence dielectric behaviour of oil samples will be governed by how the fatty acids present in the samples contribute to dielectric properties according to their own molecular structure individually and also collectively in the presence of other fatty acids. We observe no peculiarity in the value of ϵ' for olive oil where Linoleic acid is totally absent. This suggests that the variation in ϵ' is due to combined effect of the molecular rotations of all the fatty acids present in the triglyceride molecule of oils and the presence or absence of any single fatty acid does not make much difference. So, even if the percentage of a certain fatty acids present in two oils are different, they can show same dielectric properties or vice versa.

Table 1. Values of ϵ_0 , ϵ_{∞} , and ϵ' (at 9.1GHz) for various oils at temperature of 293°K.

Oils	ϵ_0	ϵ' (9.1GHz)	ϵ_{∞}
Groundnut oil	3.09	2.40	2.16
Sunflower oil	3.11	2.42	2.16
Sesame oil	3.11	2.42	2.17
Cottonseed oil	3.15	2.45	2.17
Soyabean oil	3.13	2.46	2.17
Almond oil	3.03	2.40	2.17
Olive oil	3.08	2.40	2.16
Neem oil	3.89	2.57	2.18

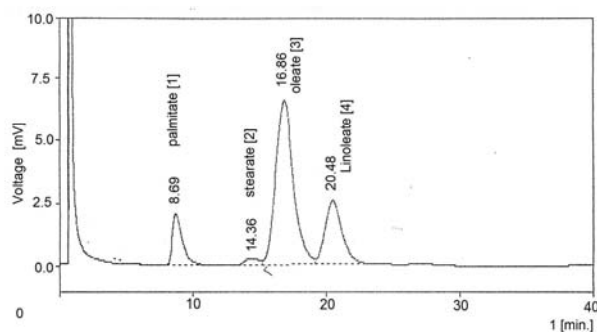


Fig. 1. Gas Chromatogram of Groundnut oil.

Table 3 shows the temperature dependence of static dielectric constant (ϵ_0), dielectric constant at optical frequency of sodium light (ϵ_{ω}), dielectric permittivity

Table 2. Fatty acid composition (%) in different varieties of edible and medicinal oils.

Oils	Palmitic	Stearic	Oleic	Linoleic
Groundnut Oil	10.967	2.005	63.673	23.355
Sunflower Oil	5.480	1.279	42.626	50.615
Sesame Oil	10.116	1.993	32.355	55.536
Olive Oil	12.693	1.079	86.228	-
Almond Oil	7.359	0.635	63.913	28.093

(ϵ') and dielectric loss (ϵ'') at 9.1GHz along with macroscopic relaxation time (τ_m), loss tangent ($\tan \delta$) and molar free energy of activation (ΔF_c) for various oils (edible and medicinal) in the temperature range of 293 to 313K. It is clear from table 3 that ϵ_0 and ϵ_∞ has got similar type of variation with temperature for different oils i.e. both decrease slightly with increase in temperature for all oil samples under consideration. However the variation of ϵ' , ϵ'' and $\tan \delta$ for these oils show little irregularity with temperature except for cottonseed and olive oil where ϵ' increases with temperature in this range. The values of ϵ' and ϵ'' for groundnut oil and sesame oil show a maxima at 313K whereas sunflower oil and neem oil show a minima at this temperature and other oils show still complex behaviour giving maxima and minima both. Similar results were reported by Bansal *et al.* (2001) for different variety of rapeseed-mustard oil. However, Venkatesh *et al.* (1996) reported systematic variation of ϵ' and ϵ'' with temperature in the range of -30°C to 60°C for tylose (a complex food material) and oils (soya and canola) at 2.45 GHz. They observed that frozen Tylose behaved as low loss material whereas at higher temperatures (above 20°C) the reverse effect was observed. Bansal *et al.* (2001) have also observed maxima and minima type of behaviour of ϵ' and ϵ'' at a temperature about 313K and attributed it to molecular resonance/anti resonance occurring between triglyceride molecules of the oil sample used. They suggested that the intermolecular association between two triglyceride molecules is present through their resonating structure which can break up due to thermal motion leading to alignment of molecular dipoles in parallel from anti parallel or vice versa at certain temperature. This will give rise to molecular resonance or anti-resonance giving maxima or minima type behaviour of ϵ' and ϵ'' with temperature. Our results for most of the oils are also in confirmation with what observed by Bansal *et al.* (2001) for rapeseed-mustard oil. As shown in table 3, the macroscopic relaxation time (τ_m) of oils in general decreases with increasing temperature in the range of 293 to 323 K except in the case of sunflower oil. This is general behaviour of organic substances which show the decrement in relaxation time with the increasing temperature due to change in effective dipole length. The molecules are more closely clustered at low temperature and hence take more time to change their orientation giving large relaxation time at low temperatures. A typical plot of relaxation time with temperature for cottonseed oil is shown in figure 3. It is also evident from table 3 that molar free energy of activation (ΔF_c) for each oil sample increases in the temperature range of 293 $^\circ\text{K}$ to 323 $^\circ\text{K}$. This can be understood in terms of increased thermal agitation which makes molecules to require more energy to reach activated state.

The effect of mixing of edible oils on their dielectric properties is also studied at 9.1 GHz. Since at 9.1 GHz,

the dielectric constant of all oils lie very nearby, the mixing is done at 1:1 ratio (i.e. 50-50% by volume) but no significant change is observed in the value of ϵ' and ϵ'' . Recently the dielectric studies of binary mixture of some commercially available edible oils (mustard with coconut, groundnut and linseed) done by Agrawal and Bhatnagar (2005) at 9.42GHz show that the dielectric constant of the oil mixtures takes an average value of the two oils used. They suggested that by dielectric constant measurement, the adulteration in pure oils can be detected if the dielectric constant of pure oil is known. This can only be true when the dielectric constant of the pure oil and the adulterant oil is quite different. In our present study the oils mixed with each other have dielectric constant lying very nearby. Hence the change in dielectric constant at microwave frequency of 9.1GHz is not significantly seen by us. So for detection of adulteration, the dielectric constants of pure oil and the adulterant oil should be quite different giving noticeable change in the dielectric constant when mixed.

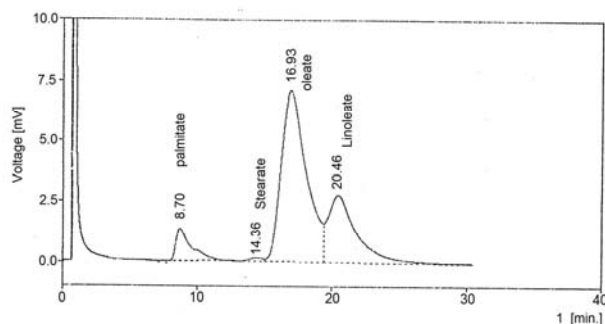


Fig. 2. Gas Chromatogram of Almond oil.

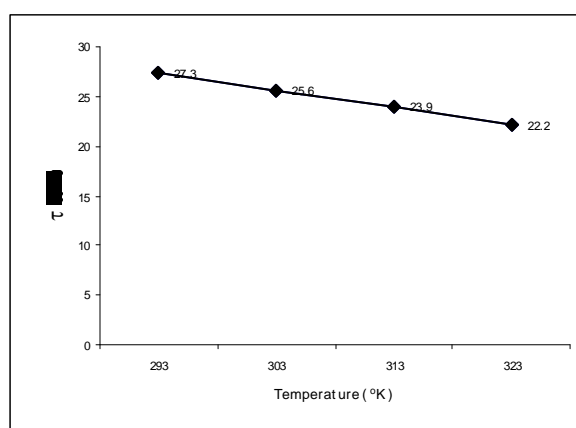


Fig. 3. Plot of Temperature Vs Relaxation time (τ_m) for Cottonseed oil.

CONCLUSION

The variation in the dielectric properties of various oils is understood as the combined effect of the molecular rotations of all the fatty acids present in the triglyceride

Table 3. Values of ϵ_0 , ϵ' , ϵ'' , ϵ_∞ , $\tan \delta$, τ_m and ΔF_ϵ for oils at different temperatures.

Temp. (°K)	ϵ_0 (f=455KHz)	ϵ' (f=9.1GHz)	ϵ''	ϵ_∞ (f=optical)	$\tan \delta$	τ_m (pS)	ΔF_ϵ (kCal/mol)
Edible Oils:							
Groundnut Oil							
293	3.09	2.40	0.09	2.16	0.04	30.0	3031.98
303	3.09	2.41	0.09	2.16	0.04	28.3	3120.55
313	2.95	2.42	0.14	2.15	0.06	24.6	3156.61
323	2.90	2.40	0.12	2.14	0.05	24.8	3282.83
Sunflower Oil							
293	3.11	2.42	0.11	2.16	0.04	28.6	3004.16
303	3.10	2.47	0.11	2.15	0.04	24.9	3043.52
313	3.07	2.43	0.11	2.14	0.05	26.0	3191.02
323	3.03	2.47	0.14	2.12	0.06	28.6	3374.30
Sesame Oil							
293	3.11	2.42	0.08	2.17	0.03	29.0	3012.25
303	3.11	2.44	0.08	2.16	0.03	26.8	3087.77
313	3.08	2.49	0.11	2.14	0.05	23.0	3114.80
323	3.04	2.46	0.13	2.13	0.05	23.1	3237.28
Cottonseed Oil							
293	3.15	2.45	0.07	2.17	0.03	27.3	2977.09
303	3.13	2.46	0.10	2.15	0.04	25.6	3060.20
313	3.12	2.48	0.06	2.13	0.02	23.9	3138.67
323	3.09	2.49	0.15	2.12	0.06	22.2	3211.78
Soyabean Oil							
293	3.13	2.46	0.08	2.17	0.03	26.5	2959.78
303	3.12	2.44	0.12	2.16	0.05	26.9	3090.01
313	3.08	2.45	0.15	2.14	0.06	25.3	3174.06
323	3.04	2.50	0.16	2.13	0.06	21.4	3188.24
Medicinal Oils:							
Almond Oil							
293	3.03	2.40	0.08	2.17	0.03	28.7	3006.19
303	3.02	2.40	0.11	2.15	0.04	28.0	3114.13
313	2.99	2.45	0.09	2.14	0.04	23.4	3125.52
323	2.96	2.45	0.09	2.13	0.04	22.2	3211.78
Olive Oil							
293	3.08	2.40	0.07	2.16	0.03	29.0	3012.25
303	3.04	2.42	0.08	2.14	0.04	26.3	3076.44
313	3.03	2.44	0.14	2.12	0.06	24.0	3141.26
323	3.00	2.45	0.12	2.11	0.05	22.3	3214.67
Neem Oil							
293	3.89	2.57	0.11	2.18	0.04	32.3	3074.97
303	3.89	2.62	0.08	2.17	0.03	29.1	3137.32
313	3.86	2.61	0.10	2.15	0.04	29.1	3261.05
323	3.83	2.63	0.08	2.14	0.03	27.3	3344.45

molecule of oils where the presence or absence of any single fatty acid does not make much difference. The dielectric parameters of oils show systematic temperature dependence except for the dielectric permittivity (ϵ') and loss tangent ($\tan \delta$) which show interesting behaviour at particular temperature suggesting some type of molecular resonance/anti resonance occurring between triglyceride molecules of the oil sample used. It is also observed that for detection of adultration the dielectric constants of pure oil and the adultrant oil should be quiet different giving noticeable change in the dielectric constant when mixed.

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Received: Dec 11, 2008; Revised: June 09, 2009;

Accepted: June 10, 2009