Vol. 19, No. 2, pp. 282-290 (2019)
Journal of Agricultural Physics
ISSN 0973-032X
http://www.agrophysics.in

Research Article

Effect of Adulteration on Dielectric Properties and Electrical Conductivity of Honey

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ABSTRACT

Dielectric properties and electrical conductivity measurements were made on honey adulterated with different concentrations of sugar syrup. The objective of the measurements was to check the potential of their applicability for checking the quality of honey. These properties were studied in the frequency range from 10-100 kHz and temperatures range 20-80 °C. Pure honey was adulterated with five different concentrations of sugar syrup. It has been observed that dielectric constant decreased and loss tangent increased with temperature. These parameters decreased with increasing frequency. Both parameters decreased in a linear fashion with the addition of sugar syrup. Electrical conductivity increased linearly with temperature and sugar syrup concentration, while the frequency has negligible effect on it. The regression equations developed relating dielectric constant, loss tangent and electrical conductivity had high correlation coefficient (R^2 >0.95).

Key words: Dielectric constant, Electrical conductivity, Honey, Loss tangent, Sugar syrup

Introduction

Honey is a natural sweet and highly viscous food substance produced by honeybees, either from the secretions of flowers called nectar honey or of the other insects called honeydew honey. It is a natural sweetening agent which can be used by humans without any processing. Honey has been used since ancient times due to its medicinal importance. The demand for honey has been increasing ever since and these facts made honey vulnerable to adulteration. Under adulteration, an illicit substitution or addition of a foreign substance is carried out, which leads to the impurity of the substance and often lowers its quality, referred to as sub-standardized substance. This practice is commonly carried out with food products all over the world. Since the composition of adulterants is almost identical to honey, its adulteration is comparatively easy. Honey can be adultered using the adulterants like molasses, glucose, sucrose, water and inverted sugar (Ruoff and Bogdanov, 2004). Thus, the adulteration of honey has become a significant economic problem. Therefore, there is a need, for the development of more efficient methods to detect honey adulteration. The adulteration can be checked by various chemical methods Zebrodska and Vorlova (2015). Although they are highly efficient and productive, there are some shortcomings associated with them. First of all, they require very expensive instrumentation and laboratory setup. In the sense, they generally are not in the reach of the common consumer and other small-scale bee-keepers. They are time-consuming, laborious and require higher analytical skills and technical knowledge to operate. Therefore, some other more efficient

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techniques which overcome these listed shortcomings are used. In recent decades, the measurement of dielectric properties and electrical conductivity of materials has opened new pathways for understanding and explaining certain physiochemical properties and characteristics of the material (Hlavacova, 2003). In the sense, the techniques which are simple to operate, comparatively inexpensive, productive and most importantly, NDTs. Non- destructive testing or NDT can be defined as any analysis technique, used in science and technology, to evaluate the properties of a material or system without any change in any of its physical or chemical property. Dielectric properties and electrical conductivity measurements can be used as non-destructive techniques. The dielectric properties are usually defined in terms of two important parameters; the relative permittivity, ε_r or ε' (also known as dielectric constant) and the loss tangent, tan δ. Electrical conductivity of a substance is defined as its ability to conduct an electrical current (or charges). An abundance of work has been done using chemical methods and the NDT methods in the radio and microwave frequency range, both of which are much expensive and highly technical. In the present work, dielectric properties and electrical conductivity has been analysed using much simpler capacitance method in the much lower frequency range of 10 to 100 kHz.

Material and Methods

In this study, the experiments were performed on raw honey, procured from the Department of Entomology, Punjab Agricultural University. In order to prepare the sugar syrup, the granular processed sugar was procured from a local departmental store. An equal amount of sugar and distilled water at 200 g each were taken in a beaker of capacity 350 ml. The mixture was then heated carefully in the temperature range of 100-110 °C, with a constant stirring for 18 to 20 minutes. A little amount of distilled water (10 ml) was added after fixed intervals to provide a golden-yellowish coloration to the syrup, in order to give it an appearance of honey. The sugar syrup-honey solutions were made on volume by volume basis. Honey was adulterated by adding 10, 20, 30, 40 and 50% sugar syrup. The pure honey was assumed to have 0% impurity level. In order to prepare 10% impurity sample, 10 ml of sugar syrup and 90 ml of honey were taken in a beaker. The solution was mildly warmed between 40 to 45 °C and thoroughly stirred in order to obtain a uniform solution. In the same way, other concentrations were prepared. The experiment for calculating every parameter was repeated thrice for each temperature, frequency and concentration. The mean values of each measurement were taken as the final value. The cylindrical-shaped cell was constructed to carry this study. It consisted of two cylinders of stainless steel: an inner cylinder with radius 1.735 cm and an outer cylinder with radius 2.385 cm acted as the outer cylinder. The electrical insulation was achieved by fixing the inner cylinder on a nylon bush base and welding the outer cylinder on a steel plate, thus forming a piston system. These cylinders acted as electrodes and the sample solution was filled in the space between the two cylinders. A nylon bush ring was fitted on the body of the inner cylinder, which would keep the cylinders parallel. The schematic diagram of sample cell has been shown in Fig. 1. The parameters were measured using the HIOKI LCR HiTESTER (model 3522-50). The dielectric constant of the given honey sample was evaluated by measuring the capacitance of the sample with air as dielectric

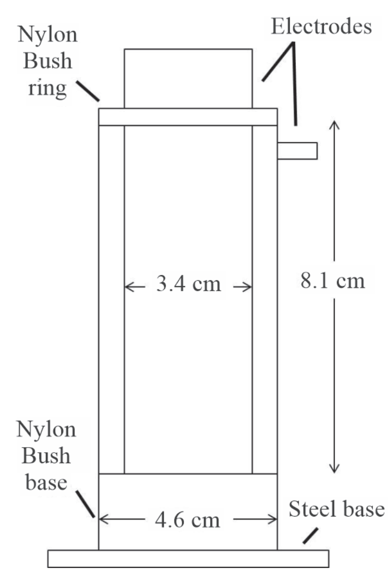

Fig. 1. Simple outline of the sample holder

medium and then that with the honey sample as the dielectric medium. The permittivity of honey sample is then simply evaluated by dividing former with the later.

$$\varepsilon' = \frac{c}{c_o} \tag{i}$$

The loss tangent of honey was evaluated by measuring phase angle è using the relation,

$$\tan\delta = \left|\frac{1}{\tan\theta}\right| \tag{ii}$$

The conductivity was evaluated by measuring the conductance of the sample using relationship.

$$\sigma = Gl/A \tag{iii}$$

where G is conductance, l is the distance between electrodes and A is the common surface area of the electrodes. The instrument is automatically designed and adjusted for coaxial cable test leads; hence the influence of the connecting wires in the measurement process was negligible. The sample holder cell was thoroughly washed and rapidly dried before each measurement, so as to obtain a moisture-free air as a dielectric medium. The data obtained from the experiments were analysed using One-way ANOVA method at 5% level of confidence using software SPSS version 20.0. The post hoc tests for significance were done using Tukey's test.

Results and Discussion

Variation of dielectric properties and electrical conductivity with temperature

The dielectric constant was in general found to decrease with increasing temperature as shown in the Fig. 2. The decrease in dielectric constant with an increase in temperature is due to the decrease in density of the samples, which was directly related to the density of dipoles in the sample. Due to increase in temperature, kinetic energy of molecules also increases, which leads to greater randomness in motion and it decreases the dipole orientation which results in low dielectric constant. The values obtained for pure honey closely approximate to those given by Luczycka (2009), Luczycka *et al.* (2012) and Pentos and Luczycka (2018). Guo *et. al* (2010) observed that the dielectric constant of given honey samples decreased with increasing temperature below 40 MHz frequency and increased when the frequency was above 40 MHz, in the temperature range of 20-80 °C. Loss tangent increased with an increasing temperature since ionic losses dominate over dipolar losses at frequencies below 1000 MHz (Metaxas and Meredith, 1983). The plots have been shown for 50 kHz in Fig. 3. Similar trends were also reported by Luczycka (2009) and Pentos and Luczycka (2018). In these studies, the loss tangent of different varieties of honey increased with an increase in the temperature. The electrical con-

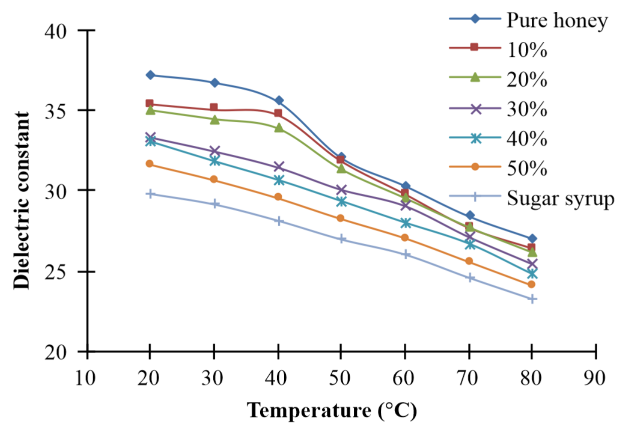

Fig. 2. Variation of dielectric constant with temperature for different concentrations at 50 kHz frequency

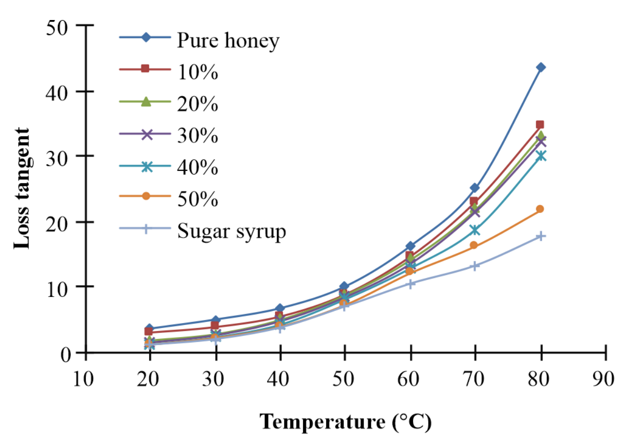

Fig. 3. Variation of loss tangent with temperature for different concentrations at 50 kHz test frequency

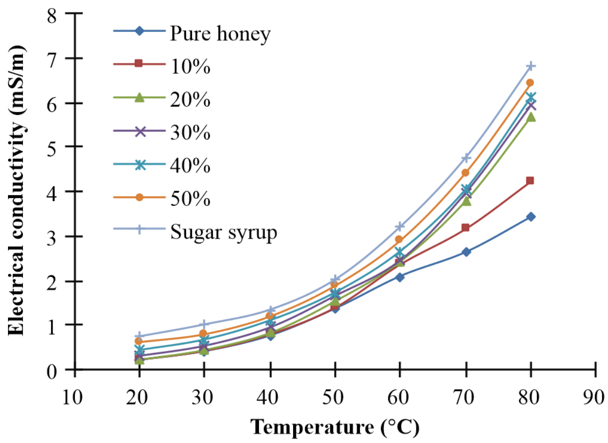

Fig. 4. Variation of the electrical conductivity with the temperature for different concentrations at 50 kHz test frequency

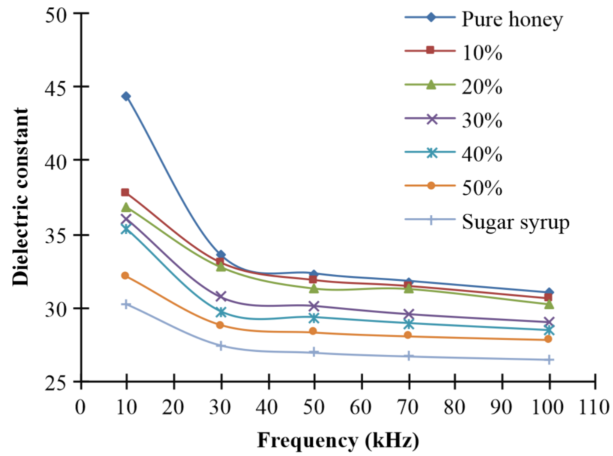

Fig. 5. Variation of dielectric constant with frequency for different concentrations at 50 °C temperature

ductivity also increased with increasing temperature. The increase in electrical conductivity with temperature follows a linear trend as shown in Fig. 4. for 50 kHz. The linear trend has been estimated using regression model between electrical conductivity and temperature for different concentrations at different frequencies. Szczesna and Ryback-Chmielewska (2004) reported the linear correlation between temperature and electrical conductivity of honey. The conductivity was in the range of 0.1-0.4 mS/cm with a temperature range of 15-30 °C. The trends of this study are also in agreement with the study of Luczycka (2009), in which the electrical conductivities of six different varieties of honey were found to increase with increasing temperature in the range 0.2-0.7 mS/m at the frequency 800 Hz.

Variation of dielectric properties and electrical conductivity with frequency

At any given temperature and for any given sample, the dielectric constant as well loss tangent was found to decrease with an increasing frequency. The plot has been shown in Fig. 5 and 6 for 50 °C. The dependence of the dielectric properties on frequency can be understood by the fact that the alignment of molecules of a given material in an external alternating field was mechanical in nature. As the oscillating frequency of the external field goes on increasing, the

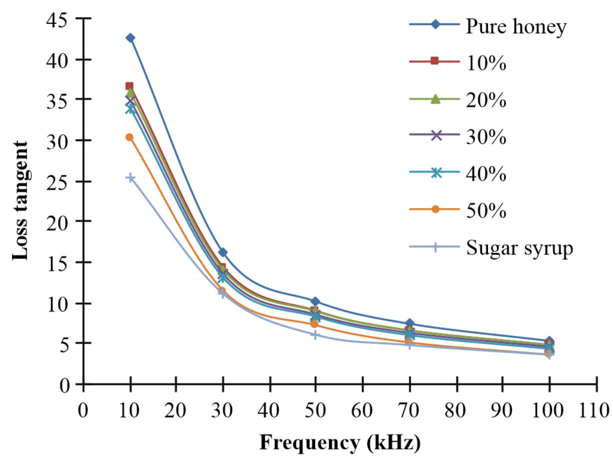

Fig. 6. Variation of loss tangent with temperature for different frequencies at 50 °C temperature

molecules get lesser time to align along the field, and thus, the electrical energy storage capacity of the material goes on decreasing with increasing frequency. It also results in low loss values as molecules don't have enough time to react along the field. The observed trends were in agreement with the study of Luczycka *et al.* (2012), where the dielectric constant and loss tangent of honey decreased with increasing frequency in the range 1-5 kHz at room temperature. Malame *et al.* (2014) observed the decrease in dielectric properties with increasing frequency in the range 300-13500 MHz and the trend was further supported by Yang *et al.* (2018) in the frequency range 1-10 GHz. The frequency has a negligible effect on the conductivity of honey. Luczycka

(2009) reported that the conductivity of honey increased with frequency in the range 0.1-0.6 mS/m in the frequency range 1-5 kHz, however, the effect was practically very little. Hence, electrical conductivity remains constant with frequency.

Variation of dielectric properties and electrical conductivity with sugar syrup adulteration

At any frequency and temperature, dielectric constant and loss tangent were found to decrease with an increase in the impurity concentration in the honey as shown in Fig. 7 and 8 for 50 kHz. The dielectric properties of pure honey have the highest value, while that of the sugar syrup are the lowest. The moisture content significantly influences dielectric properties of honey. However, the moisture content of pure honey and that of the sugar syrup in this study were kept at almost similar values. It was observed by Guo *et al.* (2011) that the difference in the dielectric properties between the pure sugar syrup and pure honey were due to the different amount of ash content in them. The sugar syrup has higher ash content than pure honey. Dielectric properties decreased with increasing sugar content. Similar trends were observed by Ahmed *et al.* (2007), where the dielectric behaviours of selected Indian honey containing different moisture and ash contents were studied in the range 900-2500 MHz. It showed that honey samples which contained similar moisture content but different ash contents had significantly different dielectric properties. Also, higher the ash content, lower the dielectric properties and vice-versa. The Analysis of Variance (ANOVA) method has been applied on the dielectric constant and loss tangent data to determine the significance of effect of sugar syrup adulteration on the dielectric properties of pure honey at 5% confidence level for different temperatures. The change is significant for impurity levels above 10% and below the temperature below 50 °C. On the other hand, electrical conductivity was found to increase with an increase in the impurity concentration in the honey as shown in Fig. 9 for 50. The electrical conductivity of pure honey has the lowest value, while that of the sugar syrup is

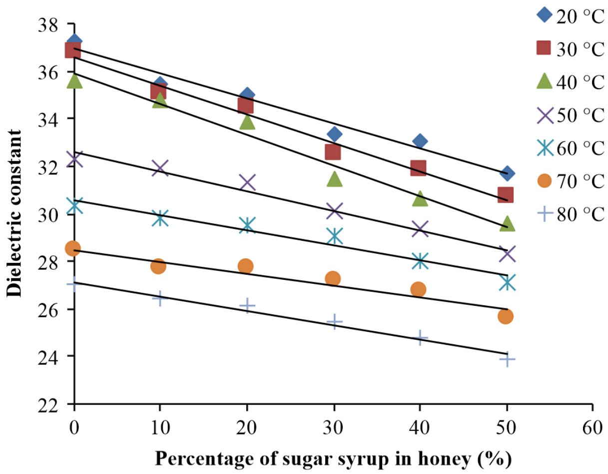

Fig. 7. Variation of dielectric constant with addition of sugar syrup in honey for different temperatures at 50 kHz frequency

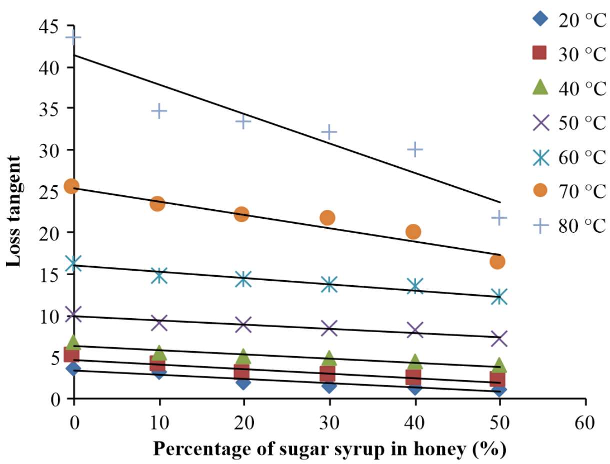

Fig. 8. Variation of the loss tangent with the addition of sugar syrup in honey for different temperatures at 50 kHz frequency

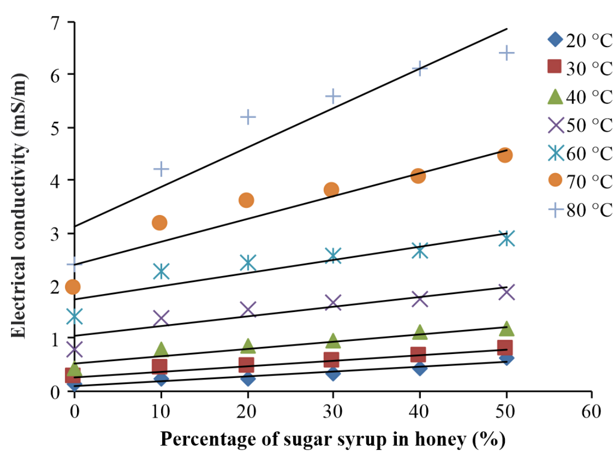

Fig. 9. Variation of the conductivity with the addition of sugar syrup in honey for different temperatures at 50 kHz frequency

the highest. The difference in the electrical conductivity values between the pure sugar syrup and pure honey observed in the present study may be due to the different amount of minerals or ash content in the given samples. Guo *et al.* (2011) has established that the ash content of honey is increased by addition of sugar syrup impurity. Since, the electrical conductivity of honey is directly associated with its ash content, hence the conductivity get increased with increase in sugar syrup impurity. Kropf *et al.* (2008) reported a linear correlation between the ash content and

Table 1. The constants and coefficients of dielectric constant-sugar syrup relationships for honey adulterated at different levels for different temperatures

Frequency (kHz)	Temperature (°C)	Dielectric constant-Sugar syrup (%)	Regression coefficient (R^2)
10	20	$\varepsilon' = -0.3384S + 52.36$	0.963
	30	$\varepsilon' = -0.3030S + 49.91$	0.957
	40	$\varepsilon' = -0.2919S + 49.73$	0.991
	50	$\varepsilon' = -0.1987S + 42.01$	0.831
	60	$\varepsilon' = -0.1533S + 39.54$	0.923
	70	$\varepsilon' = -0.1250S + 38.14$	0.915
	80	$\varepsilon' = -0.1243S + 37.33$	0.981
30	20	$\varepsilon' = -0.1882S + 40.83$	0.995
	30	$\varepsilon' = -0.1845S + 39.65$	0.980
	40	$\varepsilon' = -0.1771S + 38.34$	0.986
	50	$\varepsilon' = -0.1024S + 34.00$	0.949
	60	$\varepsilon' = -0.0711S + 31.85$	0.960
	70	$\varepsilon' = -0.0727S + 30.50$	0.971
	80	$\varepsilon' = -0.0805S + 29.68$	0.963
50	20	$\varepsilon' = -0.1053S + 36.93$	0.971
	30	$\varepsilon' = -0.1207S + 36.57$	0.983
	40	$\varepsilon' = -0.1285S + 35.88$	0.975
	50	$\varepsilon' = -0.0827S + 32.61$	0.975
	60	$\varepsilon' = -0.0633S + 30.56$	0.944
	70	$\varepsilon' = -0.0506S + 28.50$	0.927
	80	$\varepsilon' = -0.0602S + 27.14$	0.976
70	20	$\varepsilon' = -0.0849S + 35.66$	0.972
	30	$\varepsilon' = -0.0965S + 35.20$	0.986
	40	$\varepsilon' = -0.1035S + 34.54$	0.980
	50	$\varepsilon' = -0.0947S + 32.71$	0.974
	60	$\varepsilon' = -0.0521S + 29.90$	0.918
	70	$\varepsilon' = -0.0470S + 27.82$	0.938
	80	$\varepsilon' = -0.0481S + 27.25$	0.928
100	20	$\varepsilon' = -0.0704S + 34.65$	0.976
	30	$\varepsilon' = -0.0904S + 34.07$	0.989
	40	$\varepsilon' = -0.0865S + 33.17$	0.993
	50	$\varepsilon' = -0.0682S + 31.23$	0.966
	60	$\varepsilon' = -0.0505S + 29.13$	0.958
	70	$\varepsilon' = -0.0461S + 27.29$	0.961
	80	$\varepsilon' = -0.0417S + 25.41$	0.876

electrical conductivity of honey. A similar linear dependence was also reported by Sancho *et al.* (1991), Derebasi *et al.* (2014), and Gebremariam and Brhane (2014). The statistical results show that the change in electrical conductivity of honey is significant above the impurity level 20 % and 50 °C.

Mathematical modeling of dielectric properties and electrical conductivity

A mathematical model was constructed using the data dielectric constant, loss tangent and conductivity for different sugar syrup impurity levels in honey at different temperatures and

Table 2. The constants and coefficients of loss tangent-sugar syrup relationships for honey adulterated at different levels for different temperatures

Frequency (kHz)	Temperature (°C)	Loss tangent-Sugar syrup (%)	Regression coefficient (R^2)
10	20	tanδ = -0.2331S + 13.55	0.801
	30	tanδ = -0.2200S + 17.97	0.855
	40	tanδ = -0.2303S + 26.09	0.952
	50	tanδ = -0.2025S + 40.74	0.863
	60	tanδ = -0.4630S + 67.25	0.974
	70	tanδ = -0.817S + 100.51	0.945
	80	tanδ = -1.250S + 138.77	0.953
30	20	tanδ = -0.0863S + 5.43	0.884
	30	tanδ = -0.0906S + 7.21	0.862
	40	tanδ = -0.089S + 10.14	0.911
	50	tanδ = -0.078S + 15.66	0.925
	60	tanδ = -0.145S + 25.97	0.883
	70	tanδ = -0.276S + 40.06	0.979
	80	tanδ = -0.569S + 62.81	0.955
50	20	tanδ = -0.0525S + 3.40	0.894
	30	tanδ = -0.0550S + 4.55	0.871
	40	tanδ = -0.0527S + 6.38	0.934
	50	tanδ = -0.0488S + 9.87	0.917
	60	tanδ = -0.072S + 15.94	0.936
	70	tanδ = -0.158S + 25.32	0.927
	80	tanδ = -0.354S + 41.42	0.881
70	20	tanδ = -0.0387S + 2.50	0.918
	30	tanδ = -0.0396S + 3.35	0.893
	40	tanδ = -0.0370S + 4.72	0.956
	50	tanδ = -0.0382S + 7.27	0.920
	60	tanδ = -0.073S + 12.11	0.993
	70	tanδ = -0.132S + 18.50	0.988
	80	tanδ = -0.261S + 29.71	0.945
100	20	tanδ = -0.0264S + 1.79	0.905
	30	tanδ = -0.0282S + 2.43	0.910
	40	tanδ = -0.0259S + 3.42	0.973
	50	tanδ = -0.0287S + 5.26	0.932
	60	tanδ = -0.0438S + 8.00	0.976
	70	tanδ = -0.093S + 13.61	0.913
	80	tanδ = -0.166S + 31.38	0.870

frequencies. A linear model was obtained for the data of honey samples, given as

$$y(f,T,S) = B(f,T)S + A(f,T) \quad \text{(iv)}$$

where y(f,T,S) represents the given parameter of the honey sample obtained at a specific frequency, f(kHz) for a particular temperature, T (°C) in terms of impurity by percentage volume, S. A (f,T) and B (f,T) are regression coefficients. The linear models for dielectric properties have been given in Table 1 and 2, and such linear dependence is also verified by Guo *et al.* (2011). The linear model for conductivity has been given in Table 3, and such model was verified by Sancho *et al.* (1991).

Table 3. The constants and coefficients of conductivity-sugar syrup relationships for honey adulterated at different levels for different temperatures

Frequency (kHz)	Temperature (°C)	Electrical conductivity-Sugar syrup (%)	Regression coefficient (R^2)
10	20	σ = 0.0086S + 0.0733	0.951
	30	σ = 0.0092S + 0.2433	0.957
	40	σ = 0.0126S + 0.4900	0.906
	50	σ = 0.0177S + 0.9548	0.812
	60	σ = 0.0251S + 1.5681	0.797
	70	σ = 0.0436S + 2.2490	0.851
	80	σ = 0.0738S + 2.8643	0.908
30	20	σ = 0.0093S + 0.0781	0.930
	30	σ = 0.0097S + 0.2548	0.959
	40	σ = 0.0133S + 0.5171	0.905
	50	σ = 0.0184S + 1.0090	0.805
	60	σ = 0.0250S + 1.6800	0.796
	70	σ = 0.0435S + 2.3505	0.859
	80	σ = 0.0748S + 3.0224	0.887
50	20	σ = 0.0089S + 0.1019	0.923
	30	σ = 0.0101S + 0.2695	0.977
	40	σ = 0.0138S + 0.5376	0.926
	50	σ = 0.0187S + 1.0352	0.837
	60	σ = 0.0252S + 1.7400	0.812
	70	σ = 0.0435S + 2.3981	0.884
	80	σ = 0.0749S + 3.1186	0.890
70	20	σ = 0.0083S + 0.1338	0.901
	30	σ = 0.0095S + 0.3129	0.967
	40	σ = 0.0153S + 0.5638	0.937
	50	σ = 0.0186S + 1.0557	0.847
	60	σ = 0.0224S + 1.8024	0.800
	70	σ = 0.0439S + 2.4519	0.884
	80	σ = 0.0747S + 3.2029	0.894
100	20	σ = 0.0080S + 0.1567	0.886
	30	σ = 0.0098S + 0.3257	0.982
	40	σ = 0.0142S + 0.5743	0.949
	50	σ = 0.0193S + 1.0514	0.881
	60	σ = 0.0275S + 1.7148	0.917
	70	σ = 0.0461S + 2.4152	0.923
	80	σ = 0.0754S + 3.2676	0.886

Conclusions

Dielectric properties and electrical conductivity provide good opportunities for non-destructive sensing of quality of honey. From the correlations developed one can easily estimate the percentage of sugar syrup. All measurements were electrical, therefore the accuracy in measurements is very high. The change in dielectric properties is significant above 10% impurity and below 50 °C. The change in conductivity is significant above 20% impurity level and 50 °C.

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Received: September 5, 2019; Accepted: December 7, 2019