Open Access Library Journal
2019, Volume 6, e5448
ISSN Online: 2333-9721
ISSN Print: 2333-9705

Determination of Adulteration of Honey Syrup Using Open Ended Coaxial Probe Sensor at Microwave Frequency

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How to cite this paper: Yakubu, A., Abbas, Z. and Muhammed, A. (2019) Determination of Adulteration of Honey Syrup Using Open Ended Coaxial Probe Sensor at Microwave Frequency. *Open Access Library Journal*, **6**: e5448.
https://doi.org/10.4236/oalib.1105448

Received: May 7, 2019
Accepted: July 21, 2019
Published: July 24, 2019

Abstract

Food quality assurance is becoming increasingly important in food processing industry as expectations from the consumers and competitions among food manufacturers continue to grow. For this singular reason, this research work is primarily aimed at studying the dielectric characteristics of pure honey and adulterated ones with a view to determine honey that has been adulterated. In this work, permittivity of pure honey, distilled water and honey—distilled water syrup mixtures with water content from 0% to 80% was studied from 1 to 20 GHz with open-ended coaxial-line sensor and a network analyzer at room temperature. The input reflection coefficient obtained from the vector network analyzer (Agilent 85,071) is then used to calculate and correlate the complex permittivity measured for the water and mixtures. Results showed that the dielectric constants of all samples decreased with increasing frequency, while the pure honey had lower dielectric constant than distilled water but the mixture of water and honey increased with increases in water content. The maximum loss factor decreased with increasing water content.

Subject Areas

Food Science & Technology

Keywords

Open Ended Coaxial Probe, Dielectric Constant, Honey, Industries

1. Introduction

Food quality assurance is becoming increasingly important in food processing industry as expectations from the consumers and competitions among food manufacturers continue to grow. Both environmental and human factors contribute towards quality degradation of food and food products especially during ripening, harvesting, storage, processing, packaging and transportation. Thus there is now an increasing interest in the development of a variety of modern instruments, sensors and probes to assess food quality rapidly and efficiently. The basic underlying principle of these new technologies is essentially based on the interaction between properties of food and physical parameters that can be detected and measured by specific instruments [1].

In the honey and beverages industry, the price of honey and associated products depends on the percentage of quality raw honey. Adulteration or mixture of honey with sucrose by some business outlets and individuals is common in our today markets. Moisture content is the quantity of water contained in a material on a volumetric or gravimetric. Honey is the only naturally sweet product produced by bees using the nectar of plants or honey dew. According to [2], commercially sold adulterated honey shall not be added to any food ingredient or food additives, nor shall any other additions be made other than honey. Unfortunately, for economic gain, honey adulteration is a major problem in the world.

Honey contains more than 180 constituents [3]. The main compositions are fructose and glucose, followed by water, sucrose, minerals, vitamins, proteins and amino acids, etc. [4]. Although the amount of each ingredient varies with bee species, floral sources, geographical origins and climatic conditions [5]. It is stipulated in [2] that the content of fructose and glucose together in honey should not be less than 60 g/100 g (60% in mass ratio), and sucrose content should be not more than 5% on mass basis.

In many countries, such as China, the great majority of honey is collected by honey processing factories from beekeepers, then packed and marketed. A minority of honey is sold by beekeepers themselves. Hand-held refractometer is usually used to check both sugar and water content in honey. In addition, adulterated honey is difficult to identify by visual observation or tasting. These challenges influence both beekeepers, honey processers and factories to produce adulterated honey using both water and sugar.

A number of techniques have been used to measure sugar and water content in honey. Some of these techniques are high performance liquid chromatography (HPLC), isotope ratio mass spectrometry, anthrone spectrophotometer, and silver nitrate and a-naphthol methods [6]. These techniques are associated with expensive measuring instruments, complicated procedures, lengthy processing time and high operation ability. Most of the techniques for detecting food qualities are based on specific requirement for food analysis physical properties which could be acoustic, optical, magnetic, mechanical, thermal and fluid depending on application.

In the dielectric study of food materials, the end target is energy storage and dissipation of electromagnetic energy. In [7], they reported dielectric characterization measuring technique via open ended coaxial probe to be simple, fast, non-destructive and sensitive. It is in the light of this advantage that efforts are geared towards using dielectric characterization in detecting food qualities and adulteration (Honey). In [7], they also reported that dielectric study was used in detecting olive oil adulterated with vegetable oils using dielectric spectroscopy. Many researches have shown that the dielectric properties are influenced by food compositions, such as moisture content, ash content, salt content and fat content, etc. [8].

Research has shown that moisture content in honey significantly changes the dielectric relaxation of honey samples when measured [9]. According to [10], they reported that dielectric properties of honey were affected by ash content.

Based on the literatures and reports available, the adulteration of honey with water has not been fully exploited. It is based on this gap that this research is intended to explore and seek more knowledge about the effect of water content on honey complex permittivity in microwave frequencies range using open ended coaxial probe method (OECP).

In this work, complex permittivity of pure honey, distilled water and honey—distilled water mixtures with water content from 0% to 80% were studied from 1 to 20 GHz with open-ended coaxial-line sensor and a network analyzer at room temperature.

The result obtained from this measurement would serve as a guide in selecting the best honey for both the food processing industries and human alike and it will also provide engineers with relevant information for designing a cost effective sensor for dielectric property measurement to predict honey adulterated with water easily.

2. Microwave Theory

One of the properties of microwaves is the ability to travel through nonconductive materials. In materials with bipolar molecular structure (water), the electric field of microwaves can induce oscillations whilst traveling through the medium. During this process, the microwaves lose some of their energy. This loss in energy increase with the amount of water that medium contains, with the result that as the water concentration increase, less energy will reach the other side of the medium. Water not only absorbs but also reflects some of the microwave energy

Measurement with microwave is dominated by the basic properties of microwaves. Since their penetration in good conducting materials is minimal, they are mainly used to test the non-conducting materials. In lossless or lossy dielectrics, material composition, uniformity of the material, moisture and contamination content, complex permittivity and such diverse properties as porosity are some of the properties that can be measured. Complex permittivity can be mathematically represented as:

$$\varepsilon^{*} = \varepsilon' - j\varepsilon'' \quad (1)$$

where ε' is the dielectric constant and ε'' is the loss factor or the dissipating component. The loss tangent ($\tan\delta$) is particularly important because it explains the wave absorption of materials. The equation for $\tan\delta$ is given as:

$$\tan\delta = \frac{\varepsilon''}{\varepsilon'} \tag{2}$$

The dimension and picture for the OECP HP85071 are shown in Figure 1 and Figure 2, respectively.

For TEM mode propagation, from Misra admittance theory, the input admittance of the probe is given as;

$$\gamma = j\frac{k^2}{\pi k_c \ln\left(\frac{b}{a}\right)}\int_a^b\int_a^b\int_0^\pi \cos(\varnothing)\frac{\exp(-jkR)}{R}\mathrm{d}\varnothing \mathrm{d}r \mathrm{d}r \tag{3}$$

where,

$$k = \omega\sqrt{\varepsilon^*\varepsilon_0\mu_0}$$

$$k_c = \omega\sqrt{\varepsilon_c\varepsilon_0\mu_0}$$

$$R = \sqrt{r^2 + r'^2 - 2rr'\cos(\varnothing')}$$

w is the angular frequency, a and b are the inner radius and outer radius diameter, ε_c is the dielectric constant of the material inside the probe and ε^* is the complex permittivity of the material under test.

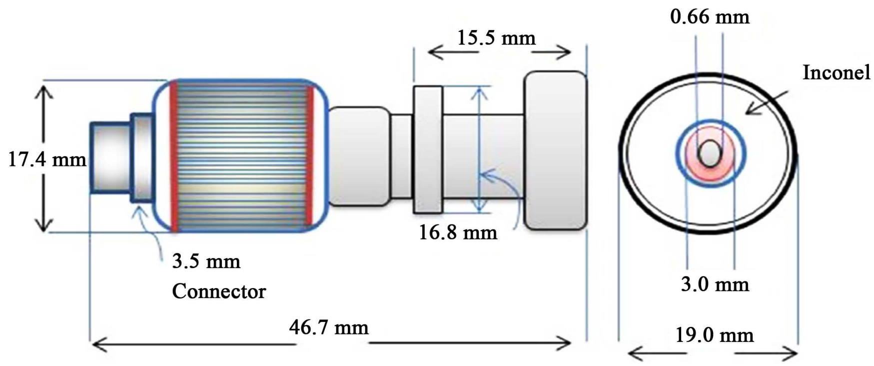

Figure 1. Geometry of open ended coaxial probe (OECP).

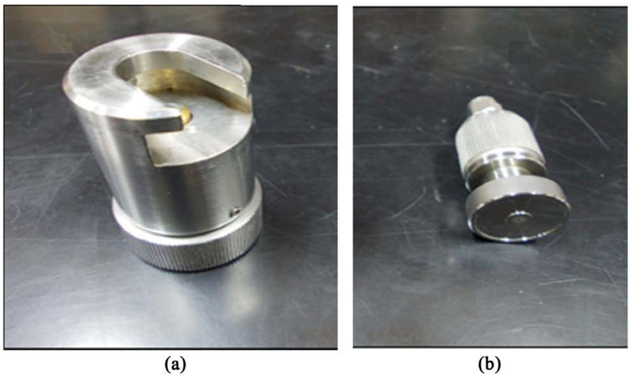

Figure 2. Agilent HP 85071 (a) open ended probe sensor (b) shorting block.

Assuming the coaxial cable opening is electrically very small, then Equation (3) can be approximated by the first few terms of the series expansion. The characteristics admittance γ_0 of the coaxial line can be obtained by;

$$\gamma_0 = \frac{2\pi}{\left[\sqrt{\frac{\mu_0}{\varepsilon_0 \varepsilon_c}} \ln\left(\frac{b}{a}\right)\right]} \tag{4}$$

$$\gamma = \frac{2\omega\varepsilon^*}{\left[\ln\left(\frac{b}{a}\right)\right]^2}\left[I_1 - \frac{k^2 I_3}{2}\right] + \frac{k^3 \pi\omega\varepsilon^*}{12}\left[\frac{b^2 - a^2}{\ln\left(\frac{b}{a}\right)}\right]^2 \tag{5}$$

$$I_1 = \int_a^b\int_a^b\int_0^\pi \frac{\cos\varnothing'}{\sqrt{r^2 + r'^2 - 2rr'\cos\varnothing'}} \mathrm{d}\varnothing \mathrm{d}r \mathrm{d}r' \tag{6}$$

$$I_3 = \int_a^b\int_a^b\int_0^\pi \cos\varnothing'\left[\sqrt{r^2 + r^2 - 2rr'\cos\varnothing'}\right] \mathrm{d}\varnothing \mathrm{d}r \mathrm{d}r' \tag{7}$$

The second term of the series expansion goes to zero on integration of over $\varnothing'$ and the fourth term is reduced to the last term of Equation (5). Under this assumption, Equation (5), reduces to;

$$\gamma = \frac{2\omega\varepsilon^*}{\left[\ln\left(\frac{b}{a}\right)\right]^2}\left[I_1 - \frac{k^2 I_3}{2}\right] \tag{8}$$

I_1 and I_3 are dependent on the physical dimensions of the probe aperture. In general, the measured reflection coefficient can be transformed into effective admittance observed at the aperture of the probe using the circuit theory model.

3. Experimental Set up

The experimental set-up consists of an Agilent 85071B Vector Network Analyser (VNA) and the probe sensor as shown in Figure 3.

All the microwave measurements were carried out at 26˚C. The measurement frequency range was set from 1 GHz to 20 GHz. The accuracy of the VNA

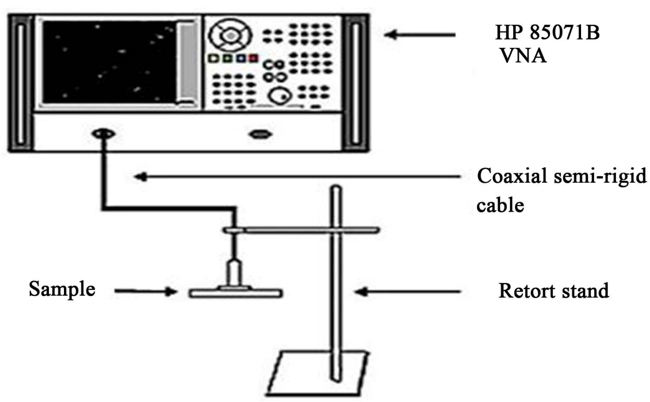

Figure 3. Measurement set-up using Agilent VNA and probe kit.

depends on the quality of the calibration standards. The VNA was calibrated by implementing a full one port calibration procedure at the end of the coaxial cable.

Open, Short and Load standards used in the calibration were automatically measured with the Agilent Electronic Calibration Module (N4691-60004). The whole Agilent 85,071 dielectric probe kit includes a sensor probe, a mounting bracket, a cable, a 3.5 inch high density shorting block for calibration, adapters and a software for data collection and plotting. Distilled water as standard material was measured so as to test the suitability of test instrument. After complete measurement of standard known material, the measurement of the pure and adulterated honey was then carried out. Shown in **Figure 4** is the picture of different percentages of adulteration.

In our work, we used 25 cm^3 for all sample cases. Accepted percentage error after calibration was 5%. The samples were then prepared in different volume percentage. **Table 1** shows the ratios of the samples measured.

The honey-water mixture at each percentage level was mixed evenly with a glass stick, and transferred to 50 ml beakers. Each beaker was placed on a flat support platform allowing the OECP to be immerged easily in each sample for permittivity measurements. Air bubbles between the probe and sample were carefully avoided since they significantly interfere with dielectric property measurement results. All measurements were repeated five times at room temperature (26˚C). This was done to avoid error due to both human and equipment's.

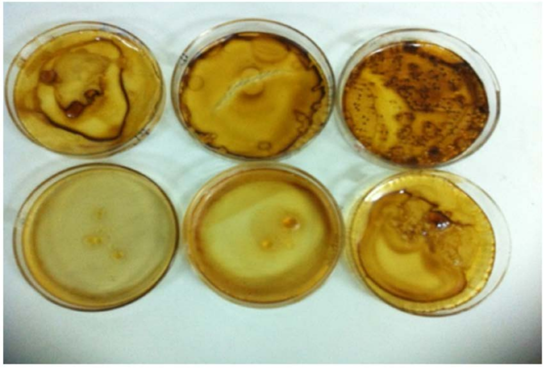

Figure 4. Honey at different percentages of adulteration.

Table 1. Composition of samples used in measurement.

Honey (cm^3)	Water (cm^3)	Water (%)	Total (cm^3)
25.0	0.0	0.0	25.0
20.0	5.0	20.0	25.0
15.0	10.0	40.0	25.0
10.0	15.0	60.0	25.0
5.0	20.0	80.0	25.0
0.0	25.0	100.0	25.0

4. Results and Discussion

4.1. Complex Permittivity of Water and Honey

The complex permittivity of distilled water and pure honey over the frequency range of 1 to 20 GHz at room temperature are shown in Figure 5 and Figure 6, respectively.

The results reveal that both pure honey and distilled water dielectric constants decreased as frequency increases. This result is in correlation with the findings for distilled water and pure honey at microwave wave frequency [1] [10] [11]. The result showed that the dielectric constant for water decreased from 79.2 to 40.1 from 1 to 20 GHz. While it had an increasing loss factor from 4.0 to 36.4 at 1 and 20 GHz, respectively.

The pure honey has a dielectric constant of 40.6 at 1 GHz and 2.51 at 20 GHz while loss factor at the same frequency range was −14.3 and 18.09 GHz, respectively.

The dielectric constant of pure honey was lower at measured frequency range. However, the pattern of honey relaxation is similar to that of distilled water content. Both samples obey the principles of the Debye theory [11]. This assertion might be correct since a report showed that vitamins content in water is 0.55%, while for pure honey it is 60.07% [12]. In the dielectric behaviors of selected Indian honey containing different moisture and ash contents from 900 to 2500 MHz, results showed that honey samples with same moisture content but different ash contents had significantly different permittivity values [12].

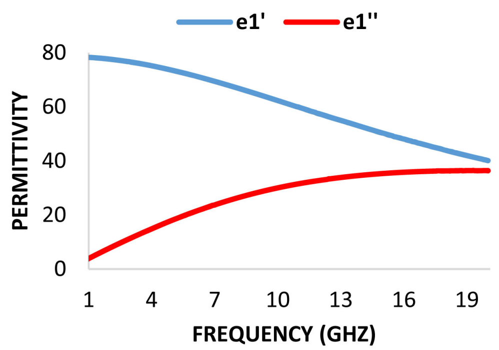

Figure 5. Permittivity of distilled water.

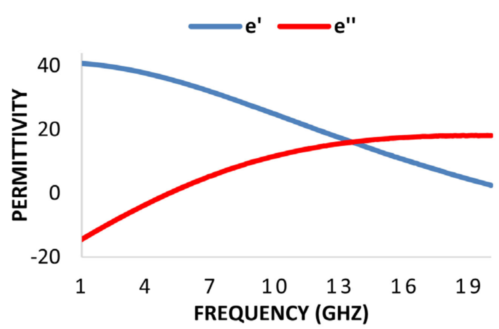

Figure 6. Permittivity of pure honey.

Based on measurement results, it is postulated that the difference in the dielectric constant values between the water and pure honey could be attributed to the different amount of minerals content existing in the samples.

4.2. Dielectric Constant of Water/Honey Mixture

Careful observation on **Figure 7** shows that water content significantly influenced the change of dielectric constant at all frequency range. Previously published reports have demonstrated that moisture content significantly influenced dielectric properties of honey [9]. The relationship between the dielectric constant and water content was sequential as shown in the results. In addition, the honey–water mixture had higher dielectric constant values than pure honey syrup at investigated frequency range.

This behavior is attributed to the high dielectric constant of distilled water, hence their mixture was influenced by the amount of distilled water present in the mixture. The changes in the dielectric constants as frequency increases may also be caused by the different vitamins content in the honey compositions.

The decrease in dielectric constant as frequency increases might be attributed to the gradual decrease in the orientation change or dipole movement at high frequency [1]. In [13], they reported that decrease in dielectric constant as frequency increases is caused by material polarization due to continuous divergent electric field.

4.3. Loss Factor of Water and Honey Mixture

The relationship between frequency and loss factor, ε'' of pure honey and water/honey mixture from 1 GHz to 20 GHz at various percentages of water content is shown in **Figure 8**.

Careful observation shows that the loss factors are almost constant at low moisture content except at higher moisture content (80% and 60%). This behavior might be attributed to small dispersion at low moisture content. The loss

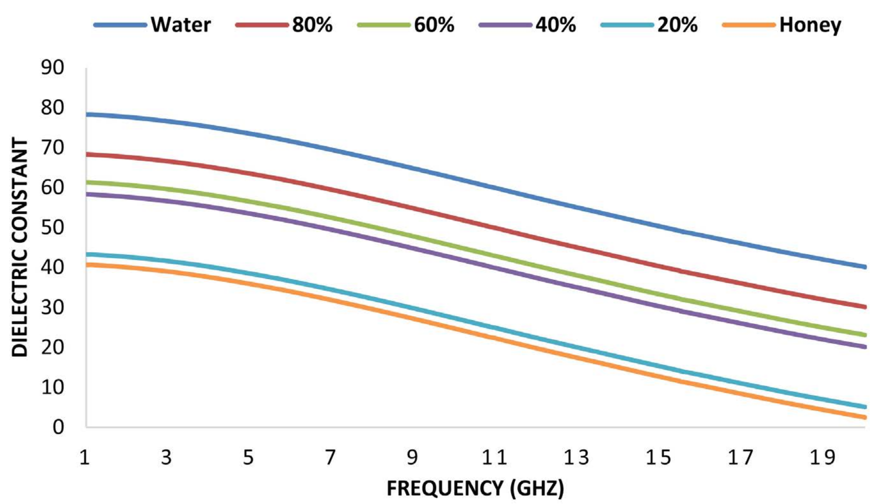

Figure 7. Comparison of dielectric constant of pure honey and honey-water mixture.

factor also increased with decreasing water content but however, it increased with increasing frequency at all levels. This is probably influenced by high polarization of water in the mixture. The low loss at lower frequency might be attributed to the ionic conductivity of the mixture since dielectric properties agricultural produce are mainly affected by their ionic conductivity in their structure [14].

4.4. Loss Tangent of Water and Honey Mixture

The loss tangent was calculated from raw value obtained for both dielectric constant and loss factor as shown in Equation (2). The results obtained were then used to plot the graph shown in **Figure 9**. Observation clearly shows that the value of the loss tangent strongly depends on the loss factor. This dependence is responsible for the sequential values of results obtained for all mixture percentages.

The results also confirmed that the sample with the lowest water percentage had the highest loss at 20 GHz. The loss tangent were all observed to increases with increasing frequency. This result is in agreement with the result obtained for the dielectric constant and which is further confirmed by the orientation polarisation theorem where decrease in the orientation polarisation at high frequency causes a drop in dielectric constant [15]. A summary of the variations at 10 GHz for $\varepsilon', \varepsilon''$ and $\tan\delta$ are tabulated in **Table 2**.

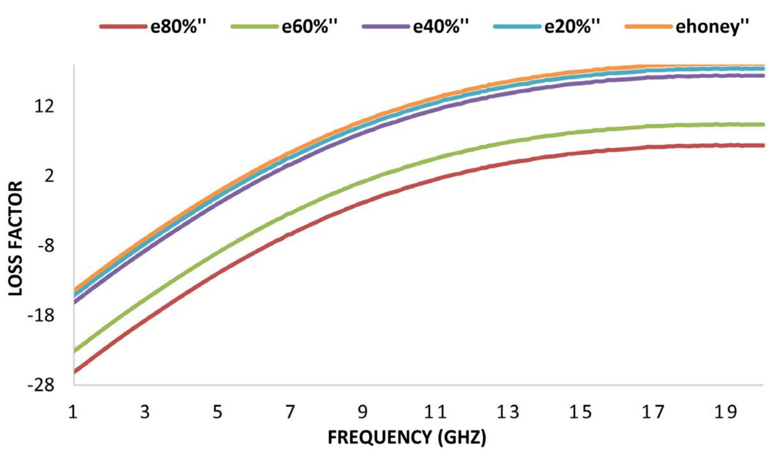

Figure 8. Comparison of Loss Factor of pure honey and honey-water mixture.

Table 2. Summary of Complex permittivity at 10 GHz.

Sample	10 GHz			
	ε'	ε''	$\tan\delta$	Complex Permittivity
Pure Honey	24.0	12.10	0.48	24-j12.10
20%	24.8	12.04	0.43	24.8-j12.04
40%	42.3	12.02	0.24	42.3-j12.02
60%	45.2	3.13	0.20	45.2-j3.13
80%	52.2	0.13	0.20	52.2-j0.13

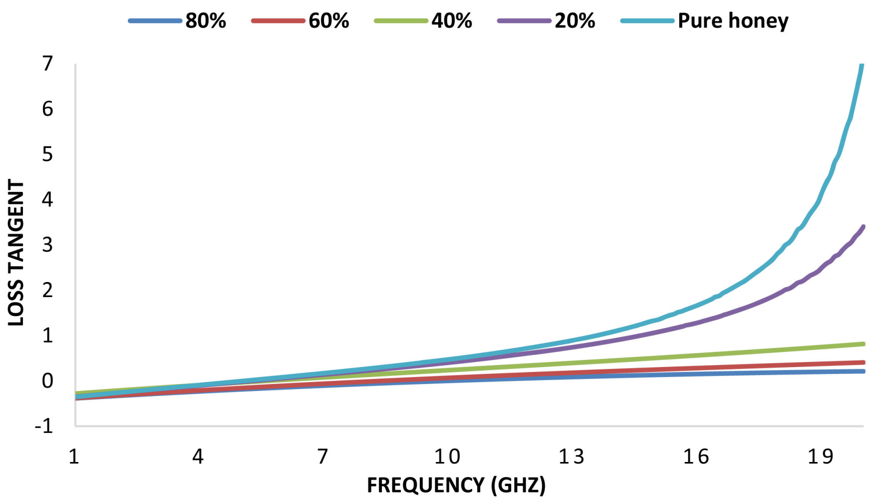

Figure 9. Comparison of Loss Tangent of pure honey and honey-water mixture.

For food industries and domestic applications, the loss tangent of honey is recommended to be in the range of 1.0 to 0.4 [16]. Based on the analysis presented in **Table 2**, it can be seen that honey adulterated with water will loss most of its energy. Energy stored in the honey decreases with increasing water content (see $\tan\delta$).

5. Conclusion

The dielectric constant of pure honey syrup and water-honey mixture decreased with increasing frequency over 1 to 20 GHz range at room temperature. Dielectric constant of pure honey was lower than that of water at all frequency range. Results confirmed that water content significantly influenced the complex permittivity of honey. Dielectric relaxation was detected in pure honey and ionic conductivity played a major role in determining the dielectric properties of both pure honey and mixtures. The loss factor of honey-water mixture continued to drop as water content increased. This drop in loss factor leads to a decline in mineral nutrients or vitamins of pure honey. Generally, this study suggests that dielectric property could be used to detect adulteration of honey mixed with water or any moist substance.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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