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Dielectric properties of honey adulterated with sucrose syrup

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ABSTRACT

Sucrose syrup is a common additive in honey adulteration. To provide information for developing a cheap, simple, convenient and rapid sucrose–adulterated honey detector or sucrose content sensor, the permittivities of pure jujube, yellow-locust and milk-vetch flower honey, pure sucrose syrup and honey–sucrose syrup mixtures with sucrose content from 0% (pure honey) to 80% (pure sucrose syrup) were studied from 10 to 4500 MHz with open-ended coaxial-line technology and a network analyzer at room temperature. The correlations between permittivities and sucrose contents were regressed. The results showed that the dielectric constants of all samples decreased with increasing frequency, while the pure honey had higher dielectric constant than pure sucrose syrup. Dielectric relaxation existed in all samples. The maximum loss factor decreased with increasing sucrose content. The relaxation frequency changed very little with sucrose content. Strong negative linear correlation, $R^2 > 0.98$, was found between loss factor around the relaxation frequency and sucrose content.

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1. Introduction

Honey is the only naturally sweet product produced by bees using the nectar of plants or honeydew. According to International regulations (Codex Alimentarius Commission, 2001) and Chinese standards (GB18796-2005, 2005) for honey, the honey sold as such shall not have added to it any food ingredient, including 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 (Al et al., 2009). The main compositions are fructose and glucose, followed by water, sucrose, minerals, vitamins, proteins and amino acids, etc. (Wang, 2007). Although the amount of each ingredient varies with bee species, floral sources, geographical origins and climatic conditions (Perez-Arquillue et al., 1994), it is stipulated that the content of fructose and glucose together in honey should be not less than 60 g/100 g (60% in mass ratio), and sucrose content should be not more than 5% on mass basis (Codex Alimentarius Commission, 2001; GB18796-2005, 2005). Contrasted with fructose and glucose, sucrose produced from vegetables is much cheaper. Therefore, sucrose syrups of natural origin such as maple, cane sugar, beet sugar, etc., are common sweetener additives in honey for increasing total sugar content.

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 sugar content in honey. But it just provides total soluble solid content. Moreover, sucrose–adulterated honey cannot be identified easily by visually examining or tasting. Therefore, not only beekeepers, but also some honey packers or processing factories, produce sucrose–adulterated honey. High performance liquid chromatography (HPLC) is required to measure sucrose content in honey (GB/T–18932.22–2003, 2003). Besides HPLC method, isotope ratio mass spectrometry, anthrone spectrophotometer, silver nitrate and α -naphthol methods are also used to determine sucrose content in honey (Bogdanov and Martin, 2002; Wang, 2003). However, expensive instruments, complicated procedures, long processing time and/or high operation ability requirement for users make these analytical methods be impractical in the field and market.

Although technologies, developed based on food stuffs' physical properties, such as acoustic, optical, magnetic, mechanical, thermal and fluid properties, have been explored in detecting food qualities, each technique has its application field. Dielectric properties of food materials reflect the storage and dissipation of electromagnetic energy. This technology is not only unlimited to food varieties, but also simple, rapid, non-destructive and sensitive (Lizhi et al., 2010). Therefore, a lot of efforts were made to study dielectric properties in detecting food qualities and adultery. 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. (Içier and Baysal, 2004; Ng et al.,

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2008; Ryynänen, 1995; Sosa-Morales et al., 2010). Based on the dielectric spectroscopy difference, dielectric properties have been used to detect olive oil adulterated with vegetable oils (Lizhi et al., 2010), poultry meat, pork products and fish added with water, etc. (Kent and Anderson, 1996; Kent et al., 2001, 2002).

Several studies also revealed moisture content in honey samples have significant effects on honey dielectric properties (Guo et al., 2010; Puranik et al., 1991). There was strong positive linear relationship between dielectric constant and moisture content in honey (Guo et al. 2010). Dielectric properties could be used to detect honey adulterated with water. Ahmed et al. (2007) noticed that dielectric properties of honey were affected by ash content. However, there is lack of the knowledge about the effect of sucrose content on honey permittivity in radio and microwave frequencies. So the objective of the present study was to measure permittivities of honey samples added with sucrose syrup from 10 to 4500 MHz, and to establish correlations between the permittivities and sucrose content in honey. The results from this study will provide information for developing an affordable, dielectric property-based sensor to rapidly predict sucrose-adulterated honey or sucrose content in honey.

2. Materials and methods

2.1. Honey and sucrose

Jujube honey, yellow-locust honey and milk-vetch honey of Chinese floral sources of Chinese jujube (*Ziziphus jujube*), yellow-locust tree (*Robinia pseudoacacia L.*) and Chinese milk-vetch (*Astragalus sinicus Linn.*), respectively, were used in the study. The honey was collected from beekeepers and packed by Shaanxi Dangdai Honey Industry Co., Ltd. in May, 2010. No crystal was observed in used samples.

The total sugar, sucrose, moisture and ash contents of the samples used are listed in Table 1. The methods that were used to obtain these contents are described in the following sections. The moisture contents of these samples were below 18%, which is the maximum moisture content allowed by Chinese national regulations for honey (GB18796-2005, 2005), and complied with the Codex Alimentarius Standard, which stipulates that honey should have a maximum moisture content of 20% (Codex Alimentarius Commission, 2001). The total sugar contents were 79.8%, 80.3% and 80.8% in jujube, yellow-locust, and milk-vetch honey, respectively.

Sucrose (analytical reagent), met the standard of HG/T3462-1999 (1999) and produced by Tianjin Bodi Chemical Holding Co., Ltd. (Tianjing, China), was used for preparing sucrose syrup.

2.2. Dielectric properties measurement

The dielectric properties are commonly represented by the relative complex permittivity $\varepsilon^* = \varepsilon' - j \varepsilon''$, where the real part ε' is dielectric constant, and the imaginary part ε'' is dielectric loss factor (Guo et al., 2008, 2010). Dielectric properties of samples were measured according to the procedure described in our previous study (Guo et al., 2010) over spectral frequency range from 10 to 4500 MHz using an Agilent Technologies vector network analyzer (E5071C), Agilent Technologies open-ended coaxial-line probe (85070B) and dielectric probe kit software (85070D, Agilent Technologies, Penang, Malaysia). The dielectric constant and loss factor were calculated according to the reflection coefficient of the material in contact with the active tip of the probe. Total 101 measurements were conducted on a logarithmic scale between 10 and 4500 MHz. Before calibrating the coaxial probe with open (air), short circuit and deionized water at 25 °C, the port of the network

Table 1The main compositions of honey samples used in the study.

Honey	Total sugar content (%)	Sucrose content (%)	Moisture content (%)	Ash content (%)
Jujube	79.8	_ ^a	17.5	0.07
Yellow-locust	80.3	_	18.1	0.06
Milk-vetch	80.8	_	17.1	0.04

^a Dashes in the table indicate that sucrose content was not detectable by HPLC method used under our experimental conditions.

analyzer used in the experiment was calibrated with open, short, and matched 50 Ω load in sequence. A measurement was made on 25 °C deionized water to verify that proper permittivity values were being obtained. Otherwise, the probe was calibrated again until measured permittivities values of water matched well with the known data. Typical error of the system was 5% after followed the standard calibration procedure. The entire analysis system was controlled by a computer.

2.3. Total sugar content and moisture content determination

The total sugar content and moisture content were determined by refractometry according to AOAC methods (AOAC 969.38B, 1996) using an Abbe-type refractometer (2WAJ, Shanghai Optical Instrument Factory, Shanghai, China) at 20 °C. Percentages of total sugar content and moisture content in honey samples were obtained from refractive index.

2.4. Sucrose content

The sucrose contents in pure honey samples and sucrose syrups were measured by a high performance liquid chromatography (LC-2010AHT, Shimadzu Corporation, Kyoto, Japan) according to the procedure described by GB/T-18932.22-2003 (2003).

Fig. 1 is chromatograms from the HPLC analysis of sucrose contents for a standard sample according to GB/T-18932.22-2003 (2003) (Fig. 1a), jujube honey (Fig. 1b), yellow-locust honey (Fig. 1c) and milk-vetch honey (Fig. 1d), respectively. The peak of sucrose content appeared at about 9.395 min in Fig. 1a. However, no peak at this time was found in jujube, yellow-locust and milk-vetch honey (Fig. 1c–d). Therefore, the sucrose contents of used jujube, yellow-locust and milk-vetch honey were less than the level (0.2%) that can be detected using the HPLC method under our experimental conditions (Table 1). The honey samples used in the study were regarded as pure honey.

2.5. Ash content

Samples of 5–10 g were weighed into a previously ignited crucible and placed under infrared lamp. The voltage of lamp increased slowly until the samples turned black. Then they were incinerated in a furnace at $600\,^{\circ}\text{C}$ overnight till constant weight before being cooled in desiccators and weighed (AOAC 920.181, 1996).

2.6. Preparing sucrose syrup

The sucrose-water mixture, consisted of 350 g sucrose and 350 g distilled water, were added in a 1000 ml flask with three openings. The masses of sucrose and water were measured with an electronic balance (FA2104A, Shanghai Precision & Scientific Instrument Co., Ltd., Shanghai, China) having precision of 0.0001 g. The flask was placed in an electric jacket (Model 98-1-B, Tianjin Taisite Instrument Co., Ltd., Tianjing, China). A stainless

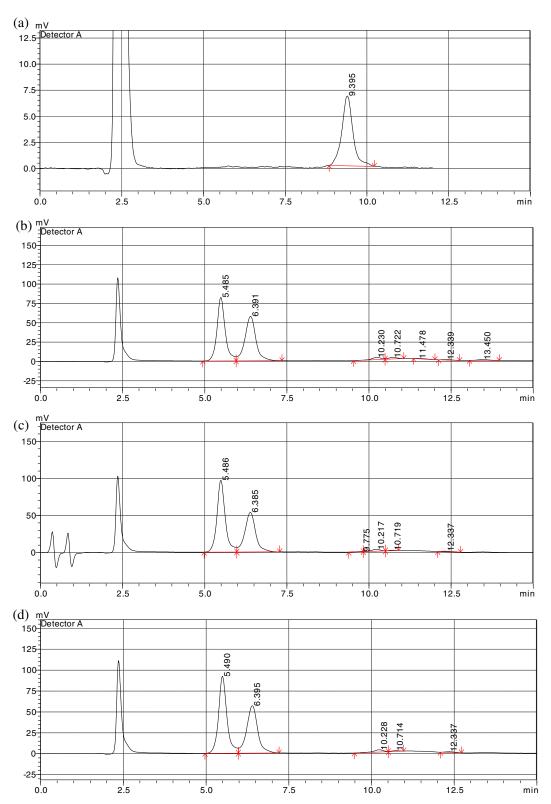


Fig. 1. The chromatogram from HPLC analysis for measuring sucrose content of a standard sample (a), jujube honey (b), yellow-locust honey (c) and milk-vetch honey (d).

steel stick with a laminated end was inserted in solution and the other end was connected to a timing motor stirrer (Model JJ-1, Jiantan Danyangmen Quartz Glass Factory, Jintan, China) through the hole of a rubber plug that was used to block the central opening of flask. During heating, temperature of sucrose–water mixture was kept between 100 and 130 °C, and the mixture was continuously mixed by the timing motor stirrer to make sucrose melt in

water evenly. The other two openings, which were fixed on a steel supporter to prevent the flask from moving during mixing, on each side of flask were open to air for water to evaporate during heating.

About 10 g of distilled water was added to flask with a funnel at 20 min interval to create yellowish decocted sucrose syrup, similar to the color of the honey used in the present study. During mixing and heating, small amount of the sucrose solution was sampled

with a glass stick from the flask (every 10 min) and total sugar content was measured at room temperature until the sucrose content was about 80%. The flask was then taken from electric jacket, covered and cooled at room temperature. According to this method, total three batches of sucrose syrup with total sugar content of 80.5%, 80.8% and 80.8% were made with corresponding moisture contents of 17.3%, 17.0% and 17.0%, respectively. The ash contents of these three batches of sucrose syrup were 0.55%. HPLC analysis showed no detectable other sugars except for sucrose in the final sucrose syrup. These samples were regarded as pure sucrose syrup. The syrup was used for measurement before noticeable crystal appeared.

2.7. Preparation and measurement of honey mixed with sucrose syrup

Pure sucrose syrup were added to 50 g pure jujube, yellow-locust and milk-vetch honey to make honey adulterated with sucrose syrup (honey-sucrose syrup mixture) with different sucrose contents from 0% (pure honey) to about 80% (pure sucrose syrup) at about 8% interval based on w/w ratio at room temperature, respectively. The honey-sucrose syrup mixture at each sucrose content level was mixed evenly with a glass stick, and transferred to three beakers (10 ml each). Each beaker was placed on a 50 mm diameter support platform and raised to make downward open-ended coaxial-line probe immerged in sample for permittivity measurements. Since air bubble between the probe and sample significantly interfere with dielectric property measurement results, it was avoided during permittivity analysis. All measurements were conducted three times at room temperature $(25 \pm 1 \, ^{\circ}\text{C})$. For each sample, total nine measurements were used to calculate the means and standard deviations. Significant differences between mean values for permittivity were determined by analysis of variance and mean separation by Duncan's Mutiple Range Test at the 5% probability level.

3. Results and discussion

3.1. Dielectric constant of honey-sucrose syrup mixture

The dielectric constants of three kinds of pure honey and sucrose syrup (80.5% sucrose) over the frequency range from 10 to 4500 MHz at room temperature (25 \pm 1 °C) are shown in Fig. 2. The results reveal that regardless of the pure honey samples or sucrose syrup, dielectric constants decreased as increasing frequency. This result is consistent with the findings for pure Chinese honey at the same frequency range (Guo et al., 2010, 2011) and Indian honey from 900 to 2500 MHz (Ahmed et al., 2007). The dielectric constant of pure sucrose syrup was lowest among the 4 samples at any given frequency. Previously published reports have demonstrated that moisture content significantly influenced dielectric properties of honey (Guo et al., 2010; Puranik et al., 1991). However, the moisture content (17.3%) of the pure sucrose syrup was similar to that of the pure honey samples (17.1–18.1%). The difference in the dielectric constant values between the pure sucrose syrup and pure honey observed in the present study might result from different amount of minerals or ash content existing in the samples. The ash content in sucrose syrup was 0.55%, while it was $\,$ ≤0.07% for the pure honey. Ahmed et al. (2007) studied the dielectric behaviors of selected Indian honey containing different moisture and ash contents from 900 to 2500 MHz, and showed that honey samples which contained similar moisture content but different ash contents had significantly different permittivity values. Between 915 and 2450 MHz frequencies, the higher the ash content, the lower the average dielectric constant values. This finding is also in agreement with the analysis of salt on dielectric proper-

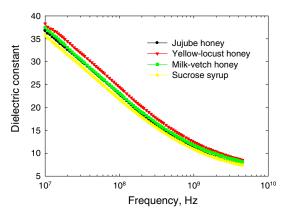


Fig. 2. The dielectric constants of pure jujube, yellow-locust and milk-vetch flower honey samples and pure sucrose syrup (80.5% sucrose content) over the frequency range from 10 to 4500 MHz at 25 °C.

ties of fruits and vegetables (Sipahioglu and Barringer, 2003), meat batter (Zhang et al., 2007), aqueous solutions and simple meat mixtures (Lyng et al., 2005), although no obvious correlation was found in egg album and yolk (Guo et al., 2007). This occurs maybe due to the binding of free water molecules by counter ions of the dissolved salts (Ryynänen, 1995).

The dielectric constants of jujube honey-sucrose syrup mixtures with sucrose contents at six selected levels from 10 to 4500 MHz frequencies at room temperature are shown in Fig. 3. The sucrose content did not influence the change trend of dielectric constant with frequency. Table 2 shows the means and standard deviations of dielectric constants of jujube honey-sucrose syrup mixtures (including 0% sucrose content or pure jujube honey, and pure sucrose syrup 80.5%) with various sucrose contents at four frequencies and room temperature, respectively. The selected frequencies are allocated for ISM (Industrial, Scientific and Medical) applications, i.e. radio frequencies 27 and 41 MHz, and microwave frequencies 915 and 2450 MHz. A few statistical significant differences (P < 0.05) were found for dielectric constant at selected frequencies, but significant trends were not observed. However, the honey-sucrose syrup mixture had higher dielectric constant values than pure sucrose syrup at a given frequency. Because the water content of honey-sucrose syrup mixtures almost kept constant (from 17.3% in pure sucrose syrup to 17.5% in pure jujube honey), the different dielectric constants may be also caused by different ash content. Ash content increased with increasing sucrose content in mixture. The dielectric constant is decreased by

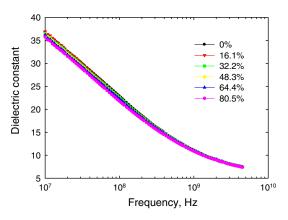


Fig. 3. Dielectric constants of jujube honey–sucrose syrup mixtures at 6 indicated sucrose contents over the frequency range from 10 to 4500 MHz at 25 $^{\circ}$ C.

Table 2The dielectric constant, ε' , and loss factor, ε'' , for pure jujube honey (0%), pure sucrose syrup (80.5%) and jujube honey-sucrose syrup mixtures with different sucrose contents at four selected frequencies and 25 °C

Sucrose content (%)	Frequency (MHz)								
	27		41		915		2450		
	ε'	ε''	ε'	ε''	ε'	ε''	ε'	ε''	
0	30.70 ± 0.04a*	8. 06 ± 0.09b	28.56 ± 0.02a	8.68 ± 0.03a	11.41 ± 0.01ab	5.70 ± 0.00a	8.62 ± 0.01b	3.86 ± 0.00a	
8.1	$30.45 \pm 0.08b$	$8.10 \pm 0.08b$	$28.20 \pm 0.06b$	$8.62 \pm 0.03ab$	$11.30 \pm 0.02c$	5.58 ± 0.02 cd	$8.56 \pm 0.03c$	3.78 ± 0.01cd	
16.1	30.73 ± 0.58a	$8.03 \pm 0.24b$	$28.50 \pm 0.53ab$	8.60 ± 0.16 b	11.48 ± 0.17a	$5.66 \pm 0.10b$	8.68 ± 0.12a	$3.85 \pm 0.06a$	
24.2	30.57 ± 0.15ab	$8.02 \pm 0.07b$	28.36 ± 0.16ab	8.53 ± 0.05bc	11.32 ± 0.05c	5.63 ± 0.03 bc	$8.55 \pm 0.03c$	$3.82 \pm 0.02bc$	
32.2	30.34 ± 0.17bc	8.01 ± 0.15b	28.17 ± 0.11b	8.57 ± 0.03 bc	$11.40 \pm 0.02b$	$5.60 \pm 0.01c$	$8.63 \pm 0.02b$	$3.82 \pm 0.00b$	
40.3	$30.46 \pm 0.06b$	$8.13 \pm 0.09ab$	$28.20 \pm 0.04b$	$8.60 \pm 0.02b$	11.39 ± 0.02bc	5.53 ± 0.01de	8.66 ± 0.01 ab	$3.76 \pm 0.00d$	
48.3	30.13 ± 0.11c	$8.03 \pm 0.08b$	27.94 ± 0.08c	$8.52 \pm 0.03c$	11.32 ± 0.03c	5.51 ± 0.00e	8.58 ± 0.03 bc	3.76 ± 0.00de	
56.4	$30.53 \pm 0.08ab$	8.24 ± 0.08a	$28.29 \pm 0.08b$	8.55 ± 0.06bc	11.48 ± 0.01a	$5.56 \pm 0.00a$	$8.70 \pm 0.01a$	$3.79 \pm 0.00c$	
64.4	29.96 ± 0.09c	7.91 ± 0.12c	27.82 ± 0.06c	$8.41 \pm 0.02d$	11.37 ± 0.01bc	$5.51 \pm 0.00d$	$8.62 \pm 0.02b$	3.78 ± 0.00cd	
72.5	$30.06 \pm 0.09c$	8.13 ± 0.09ab	27.86 ± 0.05c	$8.43 \pm 0.05d$	11.34 ± 0.00bc	$5.46 \pm 0.01 f$	8.61 ± 0.00bc	3.74 ± 0.01e	
80.5	29.63 ± 0.11d	8.15 ± 0.07ab	27.44 ± 0.08d	8.43 ± 0.05d	11.23 ± 0.02d	$5.39 \pm 0.01g$	$8.54 \pm 0.02c$	3.68 ± 0.01 f	

^{*} Means within a column followed by the same letter were not significantly different at the 5% probability level.

the presence of ions, which bind water and reduce its mobility (Bircan and Barringer, 2002).

Similar results were also found in honey–sucrose syrup mixture of yellow-locust and milk-vetch. Tables 3 and 4 list the means and standard deviations of dielectric constants of yellow-locust and milk-vetch–sucrose syrup mixtures with various sucrose contents at four frequencies and room temperature, respectively.

3.2. Dielectric loss factor of honey-sucrose syrup mixture

Dielectric loss factors of pure jujube, yellow-locust and milkvetch honey samples and sucrose syrup (80.5% sucrose) over the frequency range from 10 to 4500 MHz at room temperature are illustrated in Fig. 4. Dielectric relaxations were existed in all tested samples (Fig. 4). These similar results were also reported in our previous honey studies without sucrose treatments (Guo et al., 2010, 2011). The relaxation frequencies of pure honey samples and sucrose syrup were not much different from each other. They were 67, 65, 57 and 63 MHz for pure jujube, yellow-locust, milkvetch honey and sucrose syrup, respectively. However, the maximum average value of loss factor, which appeared at relaxation frequency, of sucrose syrup was much smaller than that of pure honey samples. They were between 9.05 and 9.32 for the pure honey samples as compared to 8.55 for the sucrose syrup. At frequencies above 40 MHz, the loss factor values of sucrose syrup became noticeably smaller than those of pure honey. These differences at higher frequency range might result from the differences in ash contents between pure sucrose syrup and pure honey samples. It has been shown that honey with higher ash content had smaller loss factor at microwave frequencies (Ahmed et al., 2007).

The dielectric loss factors of jujube honey–sucrose syrup mixture with sucrose contents at the 6 selected levels from 10 to 4500 MHz frequencies at room temperature are shown in Fig. 5. Dielectric relaxation was still existed in all samples, and the relaxation frequencies for the honey–sucrose mixture located between 67 and 63 MHz, which were relaxation frequencies of pure jujube honey and pure sucrose syrup, respectively. The loss factor decreased or increased with increasing sucrose content, which could be depended on the temperature and frequency. When equal to or larger than the relaxation frequency and 25 °C, the loss factor of each mixture decreased with increasing sucrose content (Fig. 5). This was probably caused by the low solubility of sucrose, resulting in less hydrogen bonds due to stabilizing by adding more hydroxyl groups of sugars (Liao et al., 2003).

Our previously published study revealed that the relaxation frequency increased as moisture content increased from 18% to 42% in honey (Guo et al., 2010). No obvious change in relaxation frequency in this study may be due to the similar moisture content in jujube honey–sucrose syrup mixture (17.3–17.5%). However, the peak value of loss factor decreased with increasing sucrose content. For example, it was 9.05, 8.95, 8.91, 8.76, 8.68, and 8.55 for the samples with 0%, 16.1%, 32.2%, 48.3%, 64.4% and 80.5% sucrose content, respectively. With increased sucrose content in honey–sucrose syrup mixture, the ash content increased from almost 0% in the pure honey to 0.55% in the pure sucrose syrup. Therefore, when the moisture content was almost constant, the higher ash

Table 3The dielectric constant, ε' , and loss factor, ε'' , for pure yellow-locust honey (0%), pure sucrose syrup (80.8%) and yellow-locust honey–sucrose syrup mixtures with different sucrose contents at four selected frequencies and 25 °C.

Sucrose content (%)	Frequency (MHz)								
	27		41		915		2450		
	ϵ'	$\varepsilon^{\prime\prime}$	ε'	ε''	ε'	ε''	ε'	ε''	
0	32.45 ± 0.05b*	8.74 ± 0.11a	30.25 ± 0.02b	9.12 ± 0.03 a	12.86 ± 0.00e	5.97 ± 0.01g	9.84 ± 0.01d	4.32 ± 0.01g	
8.1	32.68 ± 0.13a	8.62 ± 0.05ab	30.51 ± 0.08a	9.09 ± 0.04 a	13.03 ± 0.04d	6.12 ± 0.01c	$9.91 \pm 0.03c$	4.44 ± 0.01d	
16.2	32.78 ± 0.10a	8.52 ± 0.08bc	30.58 ± 0.08a	$9.03 \pm 0.02 \text{ b}$	13.08 ± 0.03c	$6.08 \pm 0.01d$	$9.97 \pm 0.03b$	4.42 ± 0.01e	
24.2	32.21 ± 0.09c	8.61 ± 0.14b	30.06 ± 0.02c	8.97 ± 0.03 c	12.91 ± 0.00e	$6.04 \pm 0.01e$	$9.81 \pm 0.00d$	4.39 ± 0.01f	
32.3	32.71 ± 0.08a	8.56 ± 0.12bc	30.52 ± 0.06a	8.96 ± 0.06 cd	13.11 ± 0.03c	$6.15 \pm 0.00b$	9.94 ± 0.03bc	$4.49 \pm 0.00b$	
40.4	32.70 ± 0.04a	8.63 ± 0.07ab	30.56 ± 0.07a	8.92 ± 0.03 d	13.26 ± 0.01a	6.21 ± 0.00a	10.02 ± 0.01a	4.55 ± 0.00a	
48.5	31.90 ± 0.14de	8.51 ± 0.04c	29.72 ± 0.14d	8.79 ± 0.04 e	12.89 ± 0.04e	5.91 ± 0.02h	9.84 ± 0.02d	4.33 ± 0.01g	
56.6	31.90 ± 0.19de	8.53 ± 0.11bc	29.72 ± 0.17d	8.78 ± 0.02e	12.90 ± 0.07e	5.89 ± 0.02h	9.85 ± 0.07d	4.32 ± 0.02g	
64.6	32.30 ± 0.09bc	8.45 ± 0.07c	30.19 ± 0.08b	$8.73 \pm 0.06f$	13.22 ± 0.04b	6.05 ± 0.01e	10.04 ± 0.03a	4.46 ± 0.01c	
72.7	31.86 ± 0.19e	$8.34 \pm 0.07d$	29.77 ± 0.14d	$8.61 \pm 0.03g$	13.11 ± 0.05c	$6.03 \pm 0.02f$	$9.95 \pm 0.04b$	4.45 ± 0.01c	
80.8	32.02 ± 0.07d	8.27 ± 0.06d	29.95 ± 0.06c	8.54 ± 0.03h	13.23 ± 0.01ab	$6.02 \pm 0.03 f$	10.05 ± 0.02a	4.46 ± 0.020	

^{*} Means within a column followed by the same letter were not significantly different at the 5% probability level.

Table 4The dielectric constant, ε' , and loss factor, ε'' , for pure milk-vetch honey (0%), pure sucrose syrup (80.8%) and milk-vetch honey–sucrose syrup mixtures with different sucrose contents at four selected frequencies and 25 °C.

Sucrose content (%)	Frequency (MHz)								
	27		41		915		2450		
	ε'	ε''	ε'	ε''	ε'	ε''	ϵ'	ε"	
0	30.98 ± 0.04b*	8.27 ± 0.06a	28.77 ± 0.04b	9.04 ± 0.06a	12.11 ± 0.01e	6.02 ± 0.03c	9.38 ± 0.01f	4.46 ± 0.02e	
8.1	$30.78 \pm 0.06c$	8.71 ± 0.05a	28.57 ± 0.05c	9.01 ± 0.03a	$12.04 \pm 0.01f$	5.55 ± 0.01e	$9.32 \pm 0.01h$	$3.94 \pm 0.01f$	
16.2	31.05 ± 0.09a	$8.71 \pm 0.07b$	$28.88 \pm 0.08a$	$8.89 \pm 0.06b$	12.27 ± 0.05c	$5.49 \pm 0.01b$	$9.47 \pm 0.03d$	$3.89 \pm 0.00c$	
24.2	$30.37 \pm 0.07f$	$8.59 \pm 0.08b$	$28.19 \pm 0.03f$	$8.81 \pm 0.03c$	12.01 ± 0.01fg	$5.61 \pm 0.03g$	$9.34 \pm 0.00g$	$4.00 \pm 0.02h$	
32.3	30.45 ± 0.06e	8.59 ± 0.10bc	28.25 ± 0.03e	$8.78 \pm 0.02c$	12.00 ± 0.00g	$5.40 \pm 0.00f$	9.31 ± 0.00i	$3.85 \pm 0.00g$	
40.4	30.55 ± 0.06d	8.54 ± 0.06d	28.43 ± 0.03d	$8.65 \pm 0.04d$	12.19 ± 0.01d	$5.44 \pm 0.00d$	9.43 ± 0.00e	$3.87 \pm 0.00d$	
48.5	30.19 ± 0.05h	8.33 ± 0.04c	$28.04 \pm 0.04g$	$8.70 \pm 0.04d$	12.03 ± 0.01f	5.52 ± 0.01h	$9.34 \pm 0.01g$	3.96 ± 0.00h	
56.6	30.51 ± 0.03de	8.45 ± 0.09d	28.44 ± 0.04d	$8.50 \pm 0.04 f$	12.39 ± 0.01b	5.38 ± 0.01c	9.55 ± 0.01b	$3.85 \pm 0.00b$	
64.6	30.50 ± 0.04de	8.32 ± 0.06d	28.47 ± 0.00d	8.54 ± 0.03e	12.39 ± 0.00b	5.55 ± 0.01c	$9.55 \pm 0.00b$	4.03 ± 0.01b	
72.7	$30.29 \pm 0.07g$	8.33 ± 0.06e	28.29 ± 0.07e	$8.32 \pm 0.06g$	12.37 ± 0.02b	$5.55 \pm 0.00c$	9.52 ± 0.01c	4.03 ± 0.00b	
80.8	30.41 ± 0.09ef	$8.09 \pm 0.06f$	$28.48 \pm 0.06d$	8.14 ± 0.11h	12.54 ± 0.03a	5.55 ± 0.01a	$9.63 \pm 0.02a$	4.03 ± 0.01a	

Means within a column followed by the same letter were not significantly different at the 5% probability level.

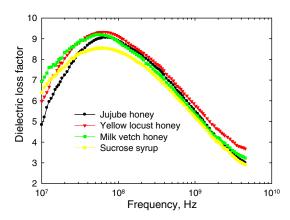


Fig. 4. The dielectric loss factors of pure jujube, yellow-locust and milk-vetch flower honey and pure sucrose syrup (80.5% sucrose content) over the frequency range from 10 to 4500 MHz at 25 °C.

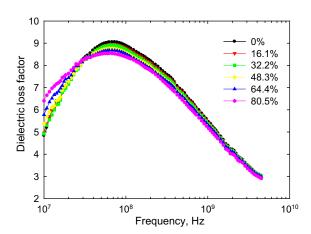


Fig. 5. Dielectric loss factors of jujube honey–sucrose syrup mixtures at 6 indicated sucrose contents over the frequency range from 10 to 4500 MHz at 25 $^{\circ}$ C.

content, the lower loss factor at microwave frequency. The same frequency- and sucrose content-dependent loss factor was also observed in honey-sucrose syrup mixture of yellow locust and milk-vetch honey. This is in good agreement with the similar relation in a model food at 2450 MHz (Sakai et al., 2005).

The means and standard deviations of dielectric loss factors of jujube, yellow-locust and milk-vetch honey-sucrose syrup mix-

tures with various sucrose contents at four frequencies and room temperature are shown in Tables 2–4, respectively. Obvious statistical significant differences (P < 0.05) at each frequency were

3.3. Relationship between dielectric properties and sucrose content

Linear relationships between dielectric constant or loss factor at 101 discrete frequencies from 10 to 4500 MHz and sucrose content in honey–sucrose syrup mixture were regressed. Results presented that there was no significant linear relationship between dielectric constant and sucrose contents in honey samples at any frequency. However, strong negative linear correlation between the dielectric loss factor and sucrose content around relaxation frequency was found. Fig. 6 presents the strongest linear regressions of dielectric loss factors on sucrose content at 104 MHz for jujube honey, 51 MHz for yellow-locust honey, and 63 MHz for milk-vetch honey. The linear relationships can be described as

$$\varepsilon'' = aS + b \tag{1}$$

where *S* represents sucrose content (%); *a* and *b* are slope and intercept of regressed line in Fig. 6, respectively. For jujube honey, a = -0.0061, b = 8.8631 and $R^2 = 0.996$; for yellow-locust honey, a = -0.0083, b = 9.3431 and $R^2 = 0.978$; for milk-vetch honey, a = -0.0103, b = 9.2045 and $R^2 = 0.978$.If the dielectric loss factor of honey sample is known, the sucrose content can be calculated as:

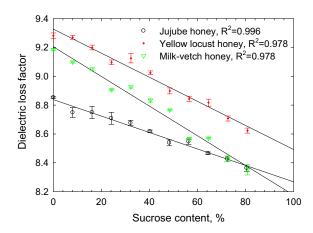


Fig. 6. Linear regressions of dielectric loss factors of jujube at 104 MHz, yellow-locust at 51 MHz, and milk-vetch honey-sucrose syrup mixtures at 63 MHz on sucrose content. at $25\,^{\circ}$ C.

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$$S = \frac{\varepsilon'' - b}{a} \tag{2}$$

The honey could be regarded as adulterated honey with sucrose syrup if the calculated *S* is higher than 5%. The smaller value of *S*, the higher purity of honey. This result makes developing a new sensor not only determining adulterated honey with sucrose syrup, but also predicting sucrose content in adulterated honey be possible.

4. Conclusions

The dielectric constant of pure jujube, yellow-locust and milkvetch honey, sucrose syrup and honey-sucrose syrup mixture decreased with increasing frequency over 10 to 4500 MHz range at room temperature. Dielectric constant of pure sucrose syrup was smaller compared with that of pure honey at any frequency. There was no significant linear correlation between dielectric constant and sucrose content in honey-sucrose syrup mixture. Dielectric relaxation was detected in pure honey, pure sucrose syrup and honey-sucrose syrup mixtures. The relaxation frequency of sucrose syrup was similar to that of pure honey. However, the loss factor of sucrose syrup was smaller than that of pure honey when frequency was higher than 40 MHz, especially at the region nearby its relaxation frequency. The maximum loss factor decreased with increasing sucrose content. Significant and strong negative linear correlation ($R^2 > 0.98$) was found between dielectric loss factor and sucrose content around relaxation frequency in all honey-sucrose syrup mixtures. This study suggests that dielectric property could be used to detect sucrose-adulterated honey or sense sucrose content in honey.

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