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Dielectric properties of *Camellia oleifera* seed kernels related to microwave and radio frequency drying

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Abstract

Understanding the dielectric properties plays an important role in the efficient use of microwave and radio frequency for dielectric heating and drying *Camellia oleifera* seed kernels. The dielectric properties of *C. oleifera* seed kernels were studied with the moisture content ranging from 5.50% to 44.22% (w.b.) over a frequency range from 10 to 3,000 MHz and a temperature range from 20 to 90°C. The results showed that both dielectric constant and dielectric loss factor decreased with increasing frequency, but increased with increasing moisture content and temperature, and the increase rate was greater at higher temperature and moisture content than that at lower levels. The penetration depth decreased with increasing frequency, moisture content and temperature. And the maximum RF heating rates can be estimated by the dielectric property parameters. The present work may provide an effective guide to develop microwave and radio frequency drying protocols of *C. oleifera* seed kernels in the future.

Introduction

Camellia oleifera is a unique woody oil tree species in China, being one of the four major oil trees, together with oil palm, olive and coconut (Lin *et al.*, 2018). *Camellia oleifera* is mainly distributed in the hilly region of southern China where the planting area had reached 4 million hectares in 2015. *Camellia oleifera* oil is the main product of *C. oleifera* seed kernels, and the annual output is more than 2.2 million tons, which accounts for half of the total weight of the dry kernels. It is considered as one of the highest qualities oils and is called "Oriental olive oil" (Liang *et al.*, 2011; Yao *et al.*, 2011). *Camellia oleifera* oil is receiving more and more attention because of its rich unsaturated fatty acids that are good for human health. The unsaturated fatty acids account for approximately 90% of all fatty acids, including monounsaturated fatty acids, oleic acids, polyunsaturated fatty acids and linoleic acids (Ma *et al.*, 2011; Yang *et al.*, 2016). Now it has been widely applied in cosmetics, skin care products and lubricants.

Drying is very important to the processing and storage of *C. oleifera* seed kernels and to the quality of *C. oleifera* oil (Bellagha *et al.*, 2007; Wang *et al.*, 2007). Due to the economic conditions, natural drying method is still used in drying *C. oleifera* seed kernels in many areas. However, fresh *C. oleifera* seed kernels with high moisture content are susceptible to weather conditions and prone to mildew on rainy days, thus increasing the complexities of the subsequent refining processes of *C. oleifera* oil (Xing *et al.*, 2012; Yuan *et al.*, 2013; Zhou *et al.*, 2017). It took 98 h to dry fresh *C. oleifera* seed kernels to safe moisture content (9%) by natural drying (Long *et al.*, 2016), and 20, 15 and 13 h were needed by hot air drying at 40, 55 and 70°C, respectively (Zhang *et al.*, 2010). The longer the drying time and the higher the hot air temperature, the greater the concentration of benzopyrene that is a carcinogen in *C. oleifera* oil (Wang *et al.*, 2016). Therefore, it is necessary to explore more efficient ways to dry *C. oleifera* seed kernels.

Due to quick heating, microwave (MW) and radio frequency (RF) drying are usually used for drying agricultural products such as wheat and corn (Jiao *et al.*, 2016), wheat flour (Ozturk *et al.*, 2017), soybeans (Huang *et al.*, 2015) and celery (Soysal *et al.*, 2006). However, the composition of *C. oleifera* seed kernels is different from that of cereals, thus understanding its dielectric properties is necessary to better use MW and RF to dry *C. oleifera* seed kernels (Tiwari *et al.*, 2011; Ştefănoiu *et al.*, 2016).

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The dielectric properties of the material determine its ability of heating in MW and RF. The complex relative permittivity is denoted by $\varepsilon = \varepsilon' - j\varepsilon''$; the real part ε' is referred to as the dielectric constant and represents the ability of material to store electromagnetic energy, while the dielectric loss factor ε'', which is the imaginary part, reflects the ability of material to convert electromagnetic energy into internal energy (Wang *et al.*, 2003).

The dielectric properties of many agricultural products have been studied at different temperatures, moisture contents and frequencies. They are mainly used for drying (Silva *et al.*, 2006; Zhang *et al.*, 2016b; Jia *et al.*, 2016), sterilisation (Zheng *et al.*, 2017; Li *et al.*, 2017b), pest control (Wang *et al.*, 2001; 2003; 2006) and non-destructive detection (Trabelsi *et al.*, 2013). Bansal *et al.* (2015) measured the dielectric property parameters of corn flour under different compaction densities and moisture contents using open-ended coaxial-line probe over a frequency range from 0.2 to 10 GHz and a temperature range from 25 to 75°C. Torrealba-Meléndez *et al.* (2015) measured the dielectric properties of barley, corn (white and yellow), sorghum and wheat with the free space transmission method at 915, 2,450, 5,800 MHz and the temperature ranging from 20 to 60°C, and found that the dielectric properties of these cereals decreased with increasing frequency. Ling *et al.* (2015) found that a quadratic polynomial can be used to describe the relationship between moisture contents, temperatures and dielectric property parameters of pistachio kernels at four specific frequencies (27.12, 40.68, 915, 2,450 MHz). The dielectric properties of chickpea decreased with increasing frequency but increased with increasing moisture content and temperature (Guo *et al.*, 2008). Moreover, Guo *et al.* (2010) compared the dielectric properties of four common legumes (chickpeas, mung beans, lentils and soybeans) at 27 MHz and 20°C and found that there was no obvious difference of dielectric property among the four legumes with low moisture content. When the moisture content rose, soybean had the largest dielectric loss factor, followed by lentils, mung beans and chickpeas.

Presently, the dielectric properties of *C. oleifera* seed kernels have not been reported. So, the present work aims to measure the dielectric property parameters of *C. oleifera* seed kernels, to determine the penetration depth and the RF heating rates to provide basic data for RF drying.

Materials and methods

Materials

Camellia oleifera seed kernels (Ganyou) were harvested from October to early November in Ganzhou, Jiangxi province, China, sealed in plastic bags and stored at 4°C until analyses. The initial moisture content of fresh *C. oleifera* seed kernels is about 51.64%.

Sample preparation

In order to obtain *C. oleifera* seed kernels with different moisture contents, 3 kg fresh seed kernels were dried at 60°C in a DHG-9140 oven (Shanghai Jinghong Experimental Equipment Co., Ltd., Shanghai, China). Sampling at the moisture content of 40%, 30%, 20%, 10%, 5% respectively, cooling in a desiccator, then sealing in a plastic bag and stored at 4°C until analyses.

To keep the open-ended coaxial probe contact closely with sample surface during measuring the dielectric property parameters by an impedance analyser (E4991B, Keysight Technologies Co. LTD., Palo Alto, California, USA), *C. oleifera* seed kernels with irregular shape were ground into powder, and the density of compacted powder equals to the density of *C. oleifera* seed kernels. This method is often used for measuring dielectric properties of various dried fruit (raisins, dried jujube, apricots, dried figs) (Alfaifi *et al.*, 2013) and legumes (chickpeas, lentils, mung beans, soybeans) (Guo *et al.*, 2010). But the highest moisture content of sample powder would be lower than that of fresh material, since some water would be pressed out from sample powder with high moisture content.

Moisture content and density measurement

The moisture content of *C. oleifera* seed kernels was determined by the AOAC Official Method 925.40 (AOAC, 1998). *Camellia oleifera* seed kernels were ground into powder. About 2-3 g powder was dried in an aluminium disk to a constant weight at 95 - 100°C in a vacuum oven, then cooled in a desiccator for 30 min. The average moisture content of powder was calculated by the triplicate experimental results.

The true density of *C. oleifera* seed kernels at different moisture contents was determined by the liquid displacement method. Toluene (C_7H_8) was used as the displaced liquid to reduce the measurement error caused by liquid absorption (Ling *et al.*, 2015). The *C. oleifera* seed kernels were immersed in toluene in a 100 mL tube, and the volume change before and after immersion were calculated. The average density of *C. oleifera* seed kernels was obtained from the mass and the average volume by three replicates.

Measurement of dielectric property parameters

The open-ended coaxial probe technique is a popular method for measuring dielectric property parameters of liquid or semi-solid materials over a broad frequency range due to easy handling and high accuracy (Li *et al.*, 2017b). The dielectric property parameters of ground *C. oleifera* seed kernels were measured in three replicates at 20, 30, 40, 50, 60, 70, 80, and 90°C between 10 and 3,000 MHz by an impedance analyser. Before the measurement, the impedance analyser was turned on and kept in a standby mode for at least 30 min. Then a series of calibrations were performed. The circulating oil bath system was connected with the jacket to control the sample temperature by controlling the oil temperature. The sample temperatures increased in turn from 20°C to 90°C with a temperature interval of 10°C, monitored by a thermocouple (E5cc, Omron Corporation, Kyoto, Japan). At each moisture content and temperature, the dielectric constants and dielectric loss factors at specific frequency (27.12, 40.68, 915, 2,450 MHz) were recorded. After each measurement, the sample holder and the probe were washed with deionised water and wiped dry for the next measurement.

Penetration depth

The penetration depth of the electromagnetic wave in the material affects the uniformity of the electromagnetic heating and the dry depth of material. The penetration depth is usually expressed by Equation (1):

$$d_p = \frac{c}{2\pi f\sqrt{2\varepsilon'\left[\sqrt{1+\left(\frac{\varepsilon''}{\varepsilon'}\right)^2}-1\right]}}$$

(Equation 1)

where d_p = penetration depth in m, f = frequency of RF or MW in Hz, c = speed of light in free space (3.0×10^8 m s^{-1}).

RF heating system and temperature measurement

In order to further explain the relationship between the dielectric properties and the heating rates of *C. oleifera* seed kernels, heating rates were obtained by experiment. A 6 kW, 27.12 MHz free-running oscillator RF system (Combi 6-S, Strayfield International Limited, Wokingham, U.K.) was used to heat 3 kg *C. oleifera* seed kernels in a polypropylene mesh rectangular container (350 × 250 × 30 mm). *Camellia oleifera* seed kernels with a moisture content of 10%, 20%, 30% w.b. were heated for 3 min by the RF system with an electrode gap of 110 mm. The temperature inside the *C. oleifera* seed kernels were measured by the eight-channel fibre optical temperature sensor system (Umi8, Fiso Technologies Inc., Quebec, Canada).

Statistical analysis

Analysis of variance (ANOVA) was carried out to determine whether the dielectric properties of *C. oleifera* seed kernels were significantly influenced by the temperature and moisture content.

Results and discussion

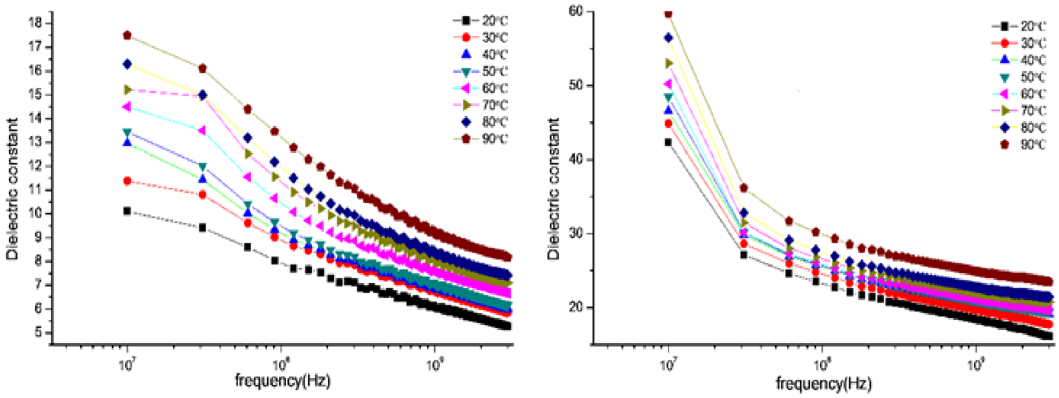
Effect of moisture content on true density of Camellia oleifera seed kernels

The true density of *C. oleifera* seed kernels were 0.98, 1.03, 1.08, 1.12, and 1.17 g cm^{-3} with the moisture content of 5.50 ± 0.05%, 14.04 ± 0.00%, 21.1 ± 0.10%, 34.26 ± 0.10%, and 44.22 ± 0.20%, respectively. It increased with the moisture content, and the similar phenomenon was also found in peanut with a moisture content of 10% to 30% (Zhang *et al.*, 2016a) and almond with a moisture content of 4.2% to 19.6% (Gao *et al.*, 2012; Li *et al.*, 2017a).

Dielectric constant

Frequency-dependent dielectric constant

Figure 1 shows the dielectric constants of *C. oleifera* seed kernels with a moisture content of 14.04% w.b. (Figure 1a) and 44.22% w.b. (Figure 1b) when the frequency ranging from 10 and 3,000 MHz, and the temperature ranging from 20°C to 90°C. The dielectric constant increased with increasing temperature and moisture content, but decreased with increasing frequency, especially at high moisture content. The dielectric constant of *C. oleifera* seed kernels with the moisture content of 14.04% decreased from 11.39, 17.49 to 5.88, 8.17 at 30°C and 90°C, respectively. That might be caused by the lower ionic conduction and greater bound water relaxation of samples at MW frequencies, and higher temperature reduced the viscosity of biomaterials, which may raise ionic conductivity (Wang *et al.*, 2013). Figure 1 also illustrates that the dielectric constant increased with increasing temperature under a specific frequency. This frequency-dependent trend may be caused by complex interaction among frequency, temperature, moisture and fat content (Nelson, 1996). The dielectric constants also decreased with increasing frequency at other moisture contents. When the frequency increased from 10 to 3,000 MHz, the dielectric constant decreased from 6.11, 34.50, 41.91 to 4.08, 16.85, 22.71 at 60°C and the moisture content of 5.50%, 22.11%, 34.26%, respectively.



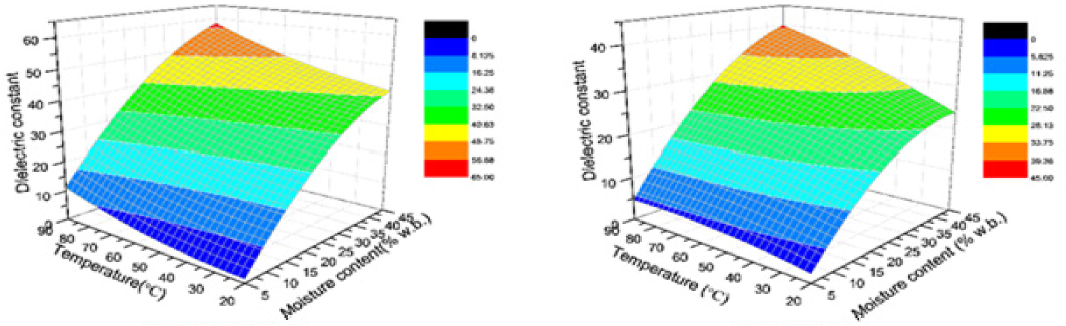
(a) 14.04%

(b) 44.22%

Figure 1. The dielectric constants of Camellia oleifera seed kernels.

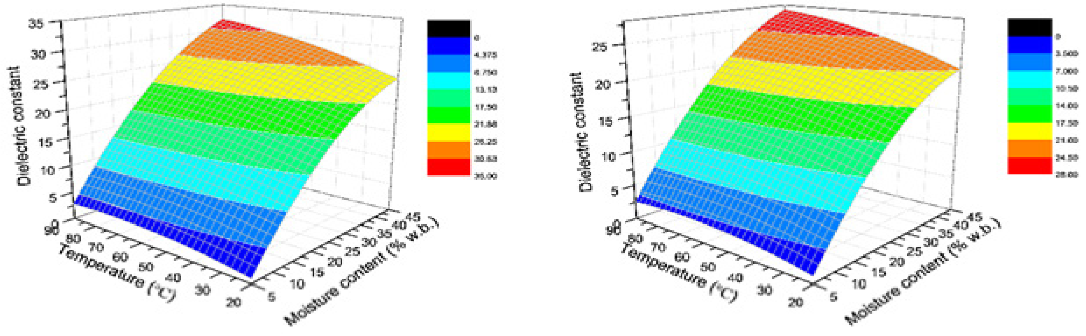
Moisture content- and temperature-dependent dielectric constant

Figure 2 shows the dielectric constants of *C. oleifera* seed kernels as a function of moisture content and temperature at 27.12 MHz (Figure 2a), 40.68 MHz (Figure 2b), 915 MHz (Figure 2c), 2450 MHz (Figure 2d). The dielectric constant increased with increasing moisture content and temperature. At 27.12 MHz, the dielectric constant of *C. oleifera* seed kernels with a moisture content of 14.04%, 22.11% and 44.22% increased from 11.11, 23.94 and 43.75 to 33.49, 38.46 and 59.54, respectively, when the temperature increased from 20°C to 90°C (Figure 2a). At 915 MHz, when the moisture content increased from 5.50% to 44.22% and the temperature at 20, 40, 60 and 80°C, the dielectric constant increased from 2.9, 3.99, 4.42, 5.09 to 22.63, 27.98, 28.29, 29.92, respectively (Figure 2c). This may be caused by the dominant role of dipolar rotations on the dielectric properties at higher moisture content, and the lower viscosity and higher ionic conductivity of biomaterials at higher temperature (Wang *et al.*, 2014). The dielectric constant increasing with moisture content and temperature at a specific frequency was also reported on almonds (Gao *et al.*, 2012) and chestnuts (Guo *et al.*, 2011) in the frequency range from 10 to 4,500 MHz.



(a) 27.12 MHz

(b) 40.68 MHz



(c) 915 MHz

(d) 2450MHz

Figure 2. The dielectric constant of Camellia oleifera seed kernels at different moisture contents and temperatures.

The data in Figure 2 was analysed with Design-expert 8.0 to obtain the regression models reflecting the relationships among the dielectric constant of *C. oleifera* seed kernels with the temperature and the moisture content at 27.12, 40.68, 915 and 2,450 MHz, respectively, which are given in Equations 2-5. All equations were quadratic polynomials.

$$\varepsilon'_{27.12} = -7.17160 + 1.82424M - 0.061202T + 2.42312\times10^{-3}MT - 0.017225M^2 + 1.73976\times10^{-3}T^2 \quad \text{(Equation 2)}$$

$$\varepsilon'_{40.68} = -5.72637 + 1.15831M + 0.10460T + 4.79143\times10^{-3}MT - 0.014172M^2 - 8.16369\times10^{-4}T^2 \quad \text{(Equation 3)}$$

$$\varepsilon'_{915} = -6.48556 + 1.10372M + 0.10684T + 1.98061\times10^{-3}MT - 0.010976M^2 - 7.99286\times10^{-4}T^2 \quad \text{(Equation 4)}$$

$$\varepsilon'_{2450} = -5.45440 + 0.93930M + 0.10396T + 1.63322\times10^{-3}MT - 9.37498\times10^{-3}M^2 - 8.0667\times10^{-4}T^2 \quad \text{(Equation 5)}$$

where M = moisture content in % w.b., $5.50 \leq M \leq 44.22$, T = temperature of *C. oleifera* seed kernels in °C, $20 \leq T \leq 90$.

The ANOVA results showed that the significant probability of each model was less than 0.0001 and R^2 was above 0.9557. So, these models can accurately predict the dielectric constant of *Ca. oleifera* seed kernels at any given moisture content between 5.50% - 44.22% w.b., at a temperature ranging from 20°C to 90°C and the interested frequencies (27.12, 40.68, 915 and 2,450 MHz).

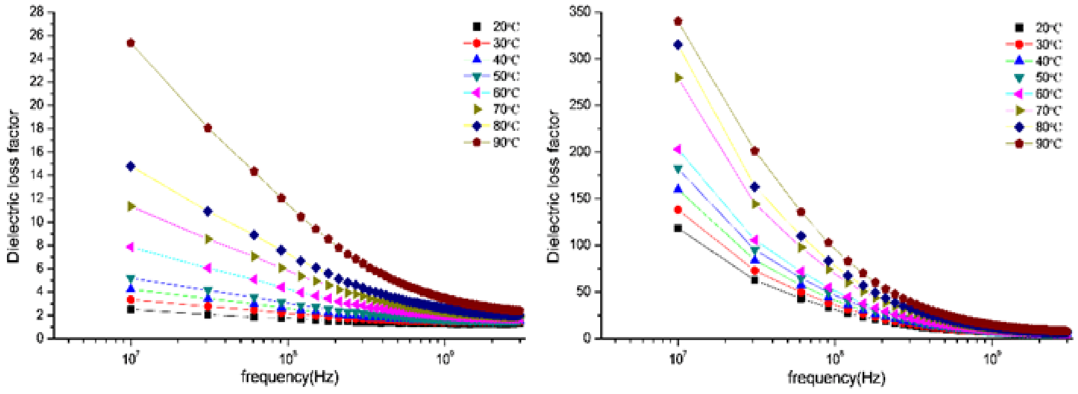
Dielectric loss factor

Frequency-dependent dielectric loss factor

Figure 3 shows the dielectric loss factor of *C. oleifera* seed kernels with a moisture content of 14.04% w.b. (Figure 3a) and 44.22% w.b. (Figure 3b) at the indicated temperature over a frequency range from 10 to 3,000 MHz. The dielectric loss factors decreased more sharply with frequency increasing in lower frequency range than in higher frequency range. At 60°C, the loss factor of *C, oleifera* seed kernels with a moisture content of 14.04% and 44.22% w.b. decreased from 14.04, 203.09 to 10.01, 102.88, respectively when the frequency increased from 10 to 100 MHz, then decreased to 6.67, 5.92 accordingly with the frequency increasing to 3,000 MHz. This is because ionic conduction plays a dominant effect on dielectric loss factor at low frequencies (Guo and Zhu, 2014). In addition, the loss factor decreased more rapidly at higher temperature than at lower temperature. At 10 MHz, the dielectric loss factors of *C. oleifera* seed kernels with the moisture content of 44.22% were 118.49 and 203.09 at the temperature of 20°C and 60°C, respectively, but reduced to 6.70 and 7.34 accordingly when the frequency increased to 2,450 MHz. Similar trends have also been found in pistachio, chickpea and corn flour (Guo *et al.*, 2008; Ling *et al.*, 2015; Bansal *et al.*, 2015).

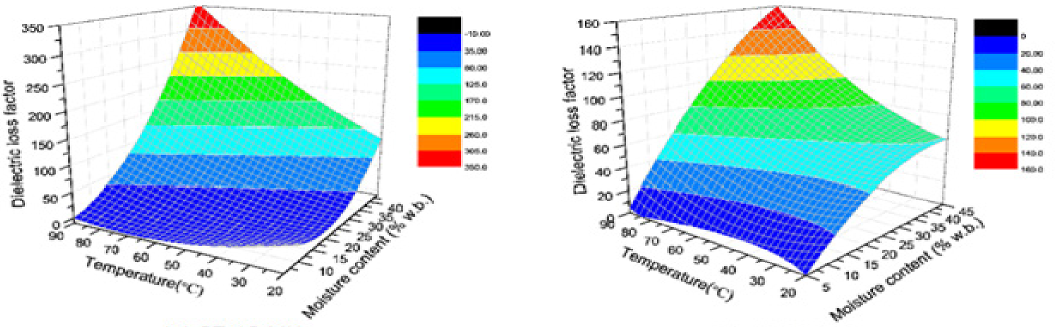
Moisture content- and temperature-dependent dielectric loss factor

Figure 4 shows the dielectric loss factors of *C. oleifera* seed kernels at 27.12 MHz (Figure 4a), 40.68 MHz (Figure 4b), 915 MHz (Figure 4c), and 2,450 MHz (Figure 4d) over a moisture content range from 5.50% to 44.22% w.b. and a temperature range from 20 to 90°C. The dielectric loss factor is a function of

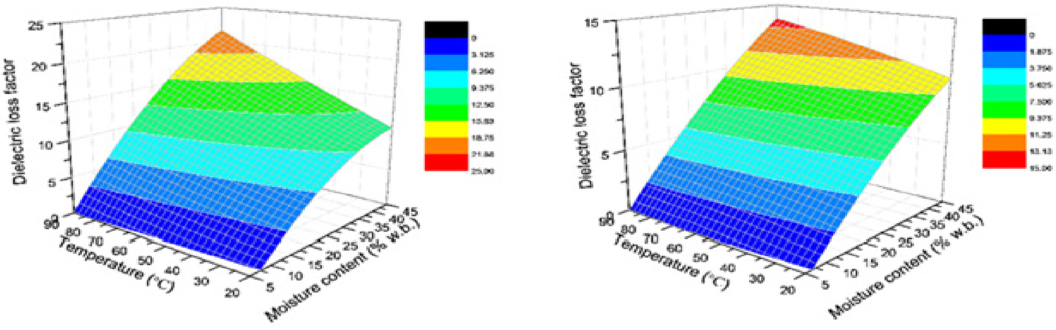


(a) 14.04% (b) 44.22%

Figure 3. The dielectric loss factors of Camellia oleifera seed kernels.



(a) 27.12 MHz (b) 40.68 MHz



(c) 915 MHz (d) 2450 MHz

Figure 4. The dielectric loss factor of Camellia oleifera seed kernels at different moisture contents and temperatures.

moisture content and temperature, which increased with increasing moisture content and temperature.

The dielectric loss factor is to characterise the ability of a material to convert electromagnetic energy into internal energy; the higher the dielectric loss factor, the more electromagnetic energy can be absorbed by material (Huang *et al.*, 2016). Figure 4 shows that the dielectric loss factor of *C. oleifera* seed kernels increased with temperature, so that more electromagnetic energy will be absorbed by *C. oleifera* seed kernels, and easily lead to partial overheating and non-uniformity drying during MW or RF drying. The dielectric loss factor also increased with moisture content; the higher the moisture content, the more electromagnetic energy was absorbed by material, and the faster the water evaporated, which is beneficial to improve the drying uniformity during MW or RF drying *C. oleifera* seed kernels with different moisture contents (Zhu *et al.*, 2012).

Regression equations of dielectric loss factor are the function of temperature and moisture content, which are quadratic polynomials as shown in Equations 6-9 at 27.12, 40.68, 915 and 2,450 MHz, respectively.

$$\varepsilon''_{27.12} = 124.89005 - 8.97102M - 3.05043T + 0.10604MT + 0.17512M^2 + 0.01741T^2 \qquad \text{(Equation 6)}$$

$$\varepsilon''_{40.68} = 9.64099 + 2.90276M - 1.60498T + 0.034377MT - 0.042159M^2 + 0.013664T^2 \qquad \text{(Equation 7)}$$

$$\varepsilon''_{915} = -4.00765 + 0.54698M - 0.015519T + 3.28353\times10^{-3}MT - 6.13405\times10^{-3}M^2 + 1.87307\times10^{-4}T^2 \qquad \text{(Equation 8)}$$

$$\varepsilon''_{2450} = -2.78370 + 0.36233M + 0.018598T + 9.96625\times10^{-4}MT - 2.16914\times10^{-3}M^2 - 1.28751\times10^{-4}T^2 \qquad \text{(Equation 9)}$$

where M = moisture content in % w.b., $5.50 \leq M \leq 44.33$; T = temperature in °C, $20 \leq T \leq 90$.

The significant probability of each model was less than 0.0001 and R^2 was above 0.9528. These models can be used to precisely estimate the dielectric loss factor value of *C. oleifera* seed kernels at any given moisture content (5.50% - 44.22% w.b.) and temperature (20 - 90°C) at indicated frequencies (27.12, 40.68, 915 and 2,450 MHz).

Penetration depth

The penetration depths (mm) can be obtained by Equation (1) from the measured dielectric property parameters of *C. oleifera* seed kernels. Penetration depth decreased with increasing frequency, moisture content and temperature. At the moisture content of 14.04% and the temperature of 50°C, the penetration depth decreased from 591.59 mm at 27.12 MHz to 31.42 mm at 2,450 MHz. With the frequency and temperature remaining at 27.12 MHz and 60°C, the penetration depth decreased from 407.40 mm to 107.76 mm when the moisture content increased from 14.04% to 44.22%. At 27.12 MHz, the penetration depth of *C. oleifera* seed kernels with the moisture content of 21.11% w.b. decreased from 803.94 mm to 92.15 mm when the temperature increased from 20°C to 90°C. This trend of penetration depth in *C. oleifera* seed kernels is similar to chestnut (Zhu *et al.*, 2012) and almond (Li *et al.*, 2017a). It is obvious that the penetration depth of RF heating is much greater than that of MW heating. So, RF has better heating uniformity and can be used to dry deep layer materials, while MW is suitable for drying thin layer materials.

RF heating rates

Figure 5 shows the experimental temperature-time histories of *C. oleifera* seed kernels with a moisture content of 10%, 20% and 30% w.b. subjected to RF heating for 3 min with an electrode gap of 110 mm. It was obvious that the heating rate of *C. oleifera* seed kernels with the moisture content of 30% w.b. was much slower than that of 10% and 20% w.b. The results show that the sample temperatures increased almost linearly with the RF heating time, and the heating rates were in the following order: 20% > 10% > 30% w.b. RF heating rate increased initially then decreased with increasing loss factor. Similar phenomenon has already been found in RF drying peanuts (Zhang *et al.*, 2016a). The maximum RF heating rate exists when $\varepsilon'' = \varepsilon' + d_m \cdot d_0^{-1}$($d_m$ is the thickness of material, d_0 is the air gap between the top electrode and the upper surface of material) (Jiao et al, 2014). In the present work, the maximum RF heating rate existed at the moisture content of 21.22% w.b. and temperature of 50°C based on the measured dielectric property parameters.

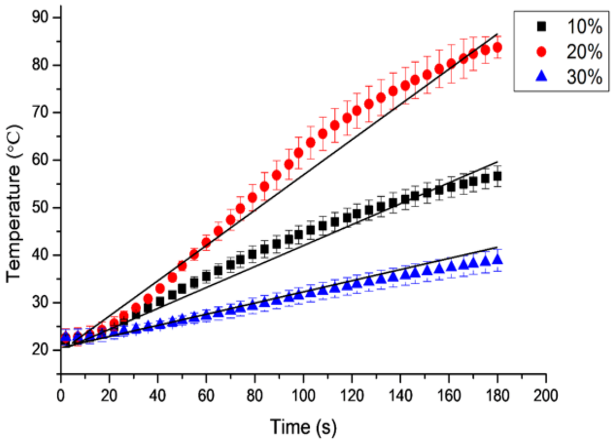


Figure 5. The experimental temperature-time histories of Camellia oleifera seed kernels.

Conclusion

Dielectric properties of *C. oleifera* seed kernels were influenced by frequency, temperature and moisture content. Both dielectric constant and dielectric loss factor decreased with frequency increasing but increased with the increase of moisture content and temperature. The penetration depth of electromagnetic wave and the maximum RF heating rates could be estimated by the dielectric property parameters. The dielectric properties of *C. oleifera* seed kernels are useful for developing the RF or MW treatment protocols in the future.

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