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Dielectric characterization of hen eggs during storage

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Abstract

Changes of the dielectric parameters of fresh hen eggs during storage were investigated for the development of a method to assess the main quality indices. Dielectric constant (ε') and loss factor (ε'') were measured in the frequency range 20–1800 MHz using an open-ended coaxial probe on thick albumen and yolk of eggs after 1, 2, 4, 8 and 15 days of storage at room temperature. Tests were also carried out at the 9th day of storage to compare dielectric properties of thick and thin albumen. Statistical analysis was performed at 20, 100, 400, 900 and 1800 MHz. Thick and thin albumen showed slight differences in dielectric properties but significant only for ε'' at 900 MHz. Dielectric constant and loss factor of yolk generally increased with the storage time. The maximum increment from the 1st to the 15th days of storage (22%,) was registered for ε'' at 20 MHz. Methods based on the comparison between the experimental ε'' and calculated values by means of conductivity and frequency can potentially be used in predicting eggs' age.

Keywords: Eggs; Albumen; Yolk; Dielectric constant; Loss factor; Storage; Quality changes

1. Introduction

Dielectric properties can be used in characterizing physical properties of biomaterials. Electric conduction, dipoles, electronic, ionic and Maxwell–Wagner mechanisms directly influence materials' dielectric properties and are closely dependent on electromagnetic wave frequency (Komarov, Wang, & Tang, 2005). Knowledge of the dielectric properties is useful in studying and developing heating processes or grading techniques based on electromagnetic energy.

Investigations to characterize dielectric properties of foods have been carried out over the past fifty years; several comprehensive reviews have been written on this subject (Nelson, 1973; Tang, Feng, & Lau, 2001; Tang, 2005; Venkatesh & Raghavan, 2004). Dielectric properties as related to temperature and frequencies have been reported for different agricultural commodities, including grains and seeds (Nelson, 1965), fruits and vegetables (Feng, Tang, & Cavalieri, 2002; Guan, Cheng, Wang, & Tang, 2004; Ikediala, Tang, Drake, & Neven, 2000; Nelson, 1983; Nelson, Forbus, & Lawrence, 1994; Tran, Stuchly, & Kraszewski, 1984; Wang, Wig, Tang, & Hallberg, 2003; Wang et al., 2003), juice and wine (Garcia, Torres, De Blas, De Francisco, & Illanes, 2004), baked foods and flours (Kim, Morgan, Okos, & Stroshine, 1998; Seras, Courtois, Quinquenet, & Ollivon, 1987; Zuercher, Hoppie, Lade, Srinivasan, & Misra, 1990), dairy products (Green, 1997; Herve, Tang, Luedecke, & Feng, 1998; Kundra, Raghavan, Akyel, Bosisio, & van de Voort, 1992; Sone, Taneya, & Handa, 1970), fish and meat (Bengtsson & Risman, 1971; Lyng, Zhang, & Brunton, 2005), egg white solutions and thermal denatured egg albumen gels (Lu, Fujii, & Kanai, 1998). Studies were also reported that assessed possibility of non-destructively predicting some physical characteristics of agri-food products such as fruits and eggs (Nelson, Forbus, & Lawrence, 1995; Ragni, Gradari, Berardinelli, Giunchi, & Guarnieri, 2006). No similar studies have been reported that relate dielectric properties of egg ingredients to storage quality.

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Nomenclature

ε'	dielectric constant	$\Delta\varepsilon''$	difference between $\varepsilon''$and ε''_σ
ε''	loss factor	f	electromagnetic wave frequency (Hz)
ε''_d	loss due to dipole polarization	H_U	Haugh unit
ε''_σ	loss due to ionic conduction	H_C	thick albumen height (mm)
ε_o	free space permittivity (8.854×10^{-12} F/m)	M	egg mass (g)
σ	electrical conductivity (S/m)		

Several methods can be used for measuring dielectric properties of agri-food products: open ended coaxial probe, transmission line, resonant cavity, free space, parallel plate capacitor. These techniques usually require a network or impedance analyzer or a LCR meter; the choice of a method depends upon the sample physical state (liquid or solid), shape (thickness, flatness) and desired range of frequency and accuracy of measurements (Agilent technologies, 2005; Içier & Baysal, 2004; Komarov et al., 2005; Nelson, 1999). Dielectric properties of agri-food materials are influenced by several factors including moisture content, density, composition and structure, water activity, temperature and frequency of the applied wave (Engelder & Buffler, 1991; Feng et al., 2002; Guan et al., 2004; Kent, 1977; Nelson, 1991, 1992; Nelson & Bartley, 2002; Ohlsson, Bengtsson, & Risman, 1974; Sipahioglu & Barringer, 2003). The open ended coaxial probe method is by far the most popular when studying the influence of frequency and temperature on dielectric characteristics of those materials because of it's ability to scan through a wide range of frequencies with desired accuracy.

Generally, the contents of a hen egg can be divided into two primary constituents; albumen (egg white) and yolk. The albumen is further divided into thin albumen which is the watery part of the egg white located farthest from the yolk, and thick albumen which is the highly viscous part of the egg white adjacent to the yolk. Water is the major constituent of the egg albumen (84–89%, on a wet basis) depending upon the type (thin or thick) of albumen that is stratified (Romanoff & Romanoff, 1949). Proteins are the main content of total solids (about 10% w/w of the albumen) while carbohydrates and inorganic elements contents are about 0.9% and 0.6%, respectively. Ovoalbumine represents more than 50% w/w of the albumen proteins. Ovomucine is the component responsible for the gelatinous structure of the albumen, it represents 3.5% of the total egg protein content. Similar percentage is reached by lysozime, an enzyme that has a lytic action on bacterial cell walls (Johnson, 1966; Li-Chan & Nakay, 1989). Content of water in the yolk is about 50%, lipids (31.8–35.5%), proteins (17.7–16.6%), and carbohydrate contents can reach 1% (Stadelman & Cotterill, 1995).

During storage, eggs undergo major physiological changes such as albumen liquefaction, increase in pH, flattening of yolk and increase in the air cell. The gelatinous structure of thick albumen deteriorates and the albumen becomes thinner (Li-Chan & Nakay, 1989). Changes of the complex lysozime-ovomucine are one of the causes responsible for the albumen liquefaction. The interaction of this complex, in electrostatic nature, is correlated with the viscosity of the albumen and depends on sample pH (Cotterill & Winter, 1955; Robinson & Monsey, 1972). Flattening of yolk depends mainly on water increase due to the osmotic migration from the albumen through the vitelline membrane (Funk, 1948). Air cell increase is caused mainly by loss of water and CO_2 through the shell and it is also influenced by the modifications that occur in the albumen and yolk (Stadelman & Cotterill, 1995).

Eggs represent an interesting yet little explored biomaterial from the point of view of their dielectric characterization. The objectives of this study were to: (1) study the physiological changes of fresh eggs over time at room temperature and (2) investigate the electrical changes of egg constituents (albumen and yolk) via dielectric properties and conductivity during storage periods. Studying the changes of the dielectric parameters of egg constituents during storage represents a first step in the development of a method to assess the main quality indices of eggs that are deeply influenced by storage time. It is believed that a non-destructive technique based on dielectric properties would be advantageous for farmers and egg processors, particularly for countries such as the European community, where the refrigeration is not allowed and eggs are delivered and stored at room temperature (EEC, 1989, 2003).

2. Materials and methods

Dielectric measurements were conducted on albumen and yolk from eggs laid by Plimouth Barred Rock breed hens aged 13 months. The eggs were obtained from a local farm within 12 h they were laid and measurements were conducted after 1, 2, 4, 8 and 15 days of storage at room temperature (22 °C). Those storage test times were chosen considering that the quality parameters associated with the decay of the eggs often show a logarithmic trend (Berardinelli, Giunchi, Guarnieri, & Ragni, 2005; Lucisano, Hidalgo, Comelli, & Rossi, 1996; Rossi, Hidalgo, & Pompei, 1995). For each test run, a randomized sample of 35 eggs was used. After egg cracking, thick albumen of 5 eggs was sucked up with a pipette, collected in a beaker and gently mixed to obtain seven sub-samples of about 50 ml each. Similar sample preparation was carried out for the yolk. Moreover, dielectric measurements were conducted on thick and thin albumen on the 9th day of storage with five samples each consisted of five eggs. It has been established that thin albumen progressively replaces thick albumen during storage; i.e. the thin to thick albumen ratio was expected to be at least 0.5 on the 9th day of storage at room temperature (Rossi, Pompei, & Hidalgo, 1995). This day was chosen because it represents an intermediate storage time that ensures a sufficient quantity of thin albumen for the dielectric measurements.

Eggs used for the above tests were 56 ± 3 g in mass, 42 ± 1 mm in diameter and 57 ± 2 mm in height. The main characteristics of eggs during storage were: thick albumen height, Haugh unit, diameter, height and index of yolk, air cell height, electrical conductivity, pH, and moisture of albumen and yolk. The thick albumen height was calculated by averaging three measurements at about 10 mm from the yolk using a tripod digital caliper. The Haugh unit was determined by means of the following formula (Haugh, 1937):

$$H_{\mathrm{U}} = 100 \log \left[H_{\mathrm{C}} - 5.67 \left(\frac{30\, M^{0.37} - 100}{100} \right) + 1.9 \right] \tag{1}$$

where H_{C} is the mean thick albumen height (mm) and M is the egg mass (g).

The yolk index was obtained by dividing the height by the diameter of the yolk (Funk, 1948).

The air cell height was measured from the base of the shell in three equidistant points on the circumference where the membrane was attached to the shell, and at the middle point of the same membrane. The mean values of these four points were considered.

Electric conductivity was measured for albumen and yolks in each of the seven sub-samples at room temperature using a conductivity meter (CON-500, Cole–Parmer Instrument Co., Vernon Hills, IL, USA), equipped with the platinum/epoxy conductivity probe with built-in temperature sensor. Measurements of pH were carried out in the above mentioned sub-samples using an AP5 pH meter (Fisher Scientific, Pittsburgh, PA, USA). A calibration procedure using appropriate standard solutions was performed for either conductivity or pH meter for each set of measurements. Moisture content of thick, thin albumen and yolk was measured according to the AOAC Official Method No. 925.30 (1990).

Measurements of the dielectric constant (ε') and loss factor (ε'') were carried out for albumen and yolk in the range 20–1800 MHz using a Hewlett-Packard 85070B open ended coaxial probe connected to an Agilent 4291B Impedance Analyzer (Agilent technologies, Inc., Palo Alto, CA). Data of ε' and ε'' were statistically analyzed at 20, 100, 400, 900 and 1800 MHz. Significant differences between means were assessed using ANOVA, or Mann–Whitney Test if significant differences emerged between variance means in the Levene Test or the sample was not represented by a normal distribution. Differences are considered significant if p-level<0.05.

A new method that could potentially be used in determining the age of eggs was also investigated in this study. The method was based on the calculation of the frequency where the dielectric mechanism shifts from ionic conduction-dominated mechanism to dipole polarization and reorientation mechanism. This was accomplished by recognizing that, according to the dielectric theory, the ε'' of a material (at the macro-scale) is a combination of two main mechanisms, namely, dipolar polarization and ionic conduction, and the relationship can be expressed by (Guan et al., 2004):

$$\varepsilon'' = \varepsilon''_{\mathrm{d}} + \varepsilon''_{\sigma} \tag{2}$$

where $\varepsilon''_{\mathrm{d}}$ is the loss due to dipole polarization and ε''_{σ} is the loss due to ionic conduction.

The loss contribution due to ionic conduction ε''_{σ} can further be expressed as (Metaxas & Meredith, 1993).

$$\varepsilon''_{\sigma} = \frac{\sigma}{2\pi f \varepsilon_0} \tag{3}$$

where σ is the electrical conductivity (S/m), ε_{o} is the free space permittivity (8.854×10^{-12} F/m) and f is the electromagnetic wave frequency (Hz). The absolute values of the difference ($|\Delta\varepsilon''|$) between the measured ε'' and ε''_{σ} were calculated; and the frequency at which an inflection point appeared was correlated with age of the egg contents (albumen and yolk).

3. Results and discussion

3.1. Egg characterization

Values of the main parameters characterizing eggs during storage are shown in Tables 1–3. Thick albumen height, Haugh unit, and yolk index significantly decreased with storage at room temperature, while air cell height significant increased with storage time. Electric conductivity of thick albumen increased by about 4% over 15 days of storage, but the difference became statistically significant only at the end of the 15th day of storage. Electric conductivity of yolk increased by about 15% over the first 8-days storage period. The pH of yolk also increased slightly showing a positive correlation to conductivity ($R^2 = 0.621$, Table 7). Table 2 summarizes the main parameters of eggs used to evaluate differences between thick and thin albumen. Thick albumen had lower electric conductivity than that of thin albumen, while pH values for the two egg constituents were not different.

Table 3 summarizes measured moisture contents of thick, thin albumen and yolk over the 15 days of storage. Changes in moisture of thick albumen were slight (1.6%) and seemed to occur the first day. After the 2nd day and until the 15th day the moisture remained constant. The

Table 1
Main characteristics of the eggs over 15 days storage at room temperature

Day	Albumen				Yolk					Air cell height (mm)
	Thick albumen height (mm)	Haugh unit	Conductivity (S/m)	pH	Diameter (mm)	Height (mm)	Yolk index	Conductivity (S/m)	pH	
1	6.6^{a} (1.0)	81^{a} (2)	0.782^{a} (0.016)	8.96^{a} (0.07)	42^{a} (1)	19^{a} (1)	0.44^{a} (0.08)	0.252^{a} (0.012)	5.95^{a} (0.09)	3.6^{a} (0.4)
2	6.1^{b} (0.6)	79^{a} (4)	0.766^{a} (0.011)	9.09^{b} (0.06)	42^{a} (1)	18^{b} (1)	0.43^{a} (0.02)	0.257^{a} (0.016)	6.15^{b} (0.07)	4.3^{b} (0.3)
4	5.6^{c} (0.9)	74^{b} (8)	0.772^{a} (0.005)	9.31^{c} (0.04)	43^{b} (1)	18^{b} (1)	0.42^{a} (0.02)	0.273^{b} (0.012)	6.14^{b} (0.06)	4.8^{c} (0.5)
8	4.5^{d} (0.5)	65^{c} (5)	0.774^{a} (0.010)	9.45^{d} (0.09)	43^{b} (2)	17^{c} (1)	0.38^{b} (0.03)	0.290^{c} (0.008)	6.24^{cd} (0.05)	6.3^{d} (0.4)
15	3.5^{e} (0.5)	55^{d} (7)	0.802^{b} (0.018)	9.55^{e} (0.08)	45^{c} (1)	15^{d} (1)	0.35^{c} (0.01)	0.284^{bc} (0.003)	6.19^{bd} (0.07)	8.2^{e} (0.6)

Differences between means with the same exponent letter within a column are not significant at probability $p < 0.05$. Values in parentheses are the standard deviation.

Table 2
Main characteristics of eggs used in the test to compare dielectric properties of thin and thick albumen (on the 9th day of storage at room temperature)

Thick albumen height (mm)	4.2 (0.8)	
Haugh unit	61 (7)	
Yolk diameter (mm)	44 (1)	
Yolk height (mm)	17 (1)	
Yolk index	0.39 (0.02)	
Air cell height (mm)	6.4 (0.7)	
Kind of albumen	**Thick**	**Thin**
Conductivity (S/m)	0.793^{a} (0.01)	0.830^{b} (0.01)
pH	9.55^{a} (0.05)	9.56^{a} (0.03)

Differences between means with the same exponent letter within a line are not significant at probability $p < 0.05$. Values in parentheses are the standard deviation.

Table 3
Moisture content of albumen and yolk over 15 days storage at room temperature

Day of storage	Thick albumen	Thin albumen	Yolk
1	88.8^{a} (0.6)	89.8^{a} (1.9)	48.0^{a} (0.9)
2	87.8^{b} (0.5)	88.3^{b} (0.9)	48.1^{a} (1.0)
4	87.9^{b} (0.5)	87.8^{b} (0.5)	48.6^{a} (0.6)
8	87.7^{b} (0.6)	87.6^{bc} (0.4)	49.6^{b} (0.7)
15	87.4^{b} (0.8)	87.4^{c} (0.2)	50.5^{c} (0.9)

Differences between means with the same exponent letter within a column are not significant at probability $p < 0.05$. Values in parentheses are the standard deviation.

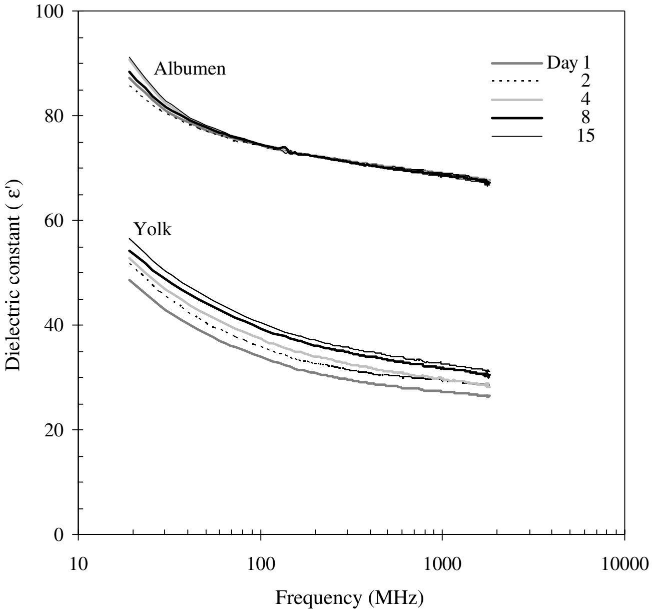

Fig. 1. Dielectric constant (ε′) of thick albumen and yolk stored for 15 days at room temperature in the frequency range 20–1800 MHz.

moisture content of the thin albumen also decreased the first day after which it stayed relatively stable. The moisture of yolk remained constant until the 4th day and then continuously increased during the subsequent days of storage (+5.2%). Similar levels of moisture increase were reported by Fromm (1966) and Heath (1975). The increase of moisture in yolk was likely due to the well-known mechanism of migration of water from albumen (Funk, 1948).

3.2. Dielectric properties of albumen

Dielectric constant (ε′) and loss factor (ε″) of albumen during storage for the entire examined frequency spectrum are shown in Figs. 1 and 2, respectively. Table 4 lists the mean values for the 20, 100, 400, 900 and 1800 MHz frequencies.

A close examination of the data shows that the trend of ε′ appears to converge in the 20–100 MHz range. Significant differences ($p < 0.05$) were found between the mean ε′ at 20 MHz for different days of storage. This behavior of ε′ supports the interpretation that suspensions in an aqueous medium contributes significantly at lower frequencies to the total polarization, and consequently to the total dielectric permittivity of the mixture. Its contribution diminishes at higher frequencies. For eggs, the suspended particles are biomolecules and electrolytes that have a much lower relaxation frequencies than that of water. At 20 MHz, the ε′ showed a relatively low, yet significant linear correlation with the storage time ($R^2 = 0.418$) that increased slightly for a logarithmic regression ($R^2 = 0.436$, considering the variability of the values for each day of storage or $R^2 = 0.500$, Fig. 3). The highest difference was observed to be between the 2nd and the 15th day of storage (7%). Although higher level of signal noise was

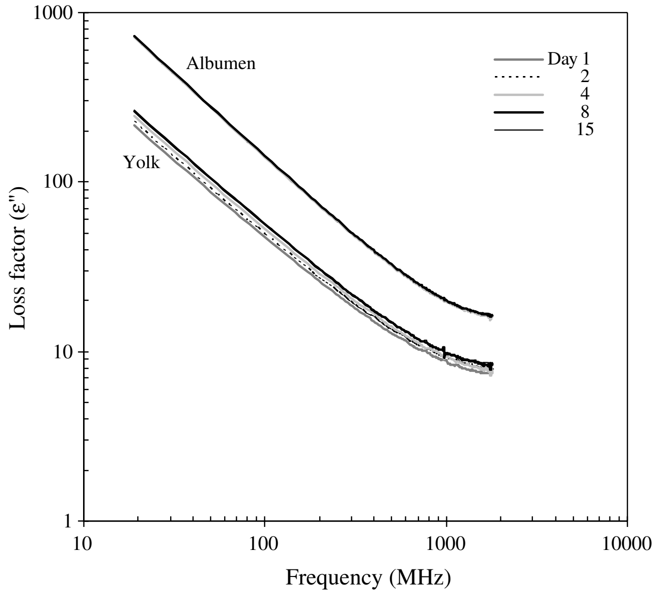

Fig. 2. Dielectric loss factor (ε'') of thick albumen and yolk stored for 15 days at room temperature in the frequency range 20–1800 MHz.

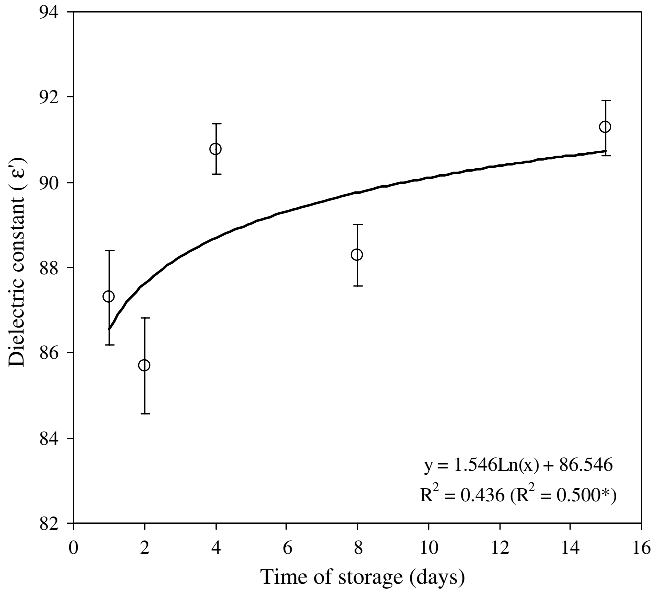

Fig. 3. Dielectric constant (ε') of egg thick albumen during storage at 20 MHz. Error bars are the standard deviation. *Calculated by considering the mean values of ε' for each day of storage.

detected at higher frequencies, a significant difference in ε' was found between the 8th and 15th days at 1800 MHz.

A linear change on the log–log plot was observed for ε'' as influenced by frequency between 100 and 1000 MHz. But the influence of storage time on ε'' was minimal (~2%). Values of dielectric loss experimentally obtained (ε'') and calculated (ε''_σ) with Eq. (3) are shown in Fig. 4. Absolute differences ($|\Delta\varepsilon''|$) between ε'' and ε''_σ values are shown in Fig. 5. It is evident from Fig. 4 that ε'' is very close to the calculated ε''_σ at low frequencies where ionic conduction is the dominant mechanism, but ε'' gradually becomes larger than the calculated ε''_σ at high frequencies where dipole polarization mechanism dominates. The shift between the above two mentioned mechanisms is further exemplified and depicted in Fig. 5, where the specific frequency (referred in this paper as the critical frequency) at which dielectric mechanisms' shift take place is clearly identified. For albumen a quasi-linear negative correlation exists between the critical frequency and time of storage until the 8th day ($R^2 = 0.982$) but the critical frequency drastically increases for the last day of storage.

Comparison between thick and thin albumen readings from the 9th day of storage showed in general that the values of ε' are very close for the two types of albumen (Fig. 6). But as the frequency increased, the ε' of thin albumen became slightly but increasingly lower than ε' of the thick albumen, showing a behavior similar to the one experienced for the thick albumen at different times of storage. Nonetheless, no significant differences were found for the ε' for all the considered frequencies (Table 5); only at 900 MHz ε'' demonstrated a minor (5%) increase for the thick albumen with respect to the thin albumen.

3.3. Dielectric properties of yolk

Values of ε' and ε'' for yolk during storage for the entire selected range of the frequency spectrum are shown in Figs. 1 and 2, respectively. Table 6 provides the mean

Table 4
Dielectric properties of the thick albumen of 15 days stored eggs at room temperature in the frequency range 20–1800 MHz

Day	Dielectric constant					Loss factor				
	Frequency (MHz)									
	20	100	400	900	1800	20	100	400	900	1800
1	87.3^{a} (1.1)	74.4^{a} (1.0)	70.8^{a} (1.1)	69.1^{a} (1.1)	67.7^{ab} (1.1)	722.0^{ab} (20.0)	140.9^{ab} (3.8)	38.9^{ab} (1.0)	21.0^{ab} (0.5)	16.3^{abc} (0.3)
2	85.7^{b} (1.1)	74.2^{a} (1.0)	70.5^{a} (1.0)	69.1^{a} (1.0)	67.1^{ab} (1.0)	709.0^{a} (15.2)	138.5^{a} (2.9)	38.8^{a} (0.8)	20.7^{a} (0.4)	16.0^{a} (0.2)
4	90.8^{c} (0.6)	74.5^{a} (0.3)	70.6^{a} (0.3)	68.8^{a} (0.4)	67.2^{ab} (0.4)	720.2^{ab} (14.9)	140.7^{ab} (2.8)	39.0^{ab} (0.6)	21.1^{b} (0.2)	16.2^{b} (0.1)
8	88.3^{d} (0.7)	74.5^{a} (0.5)	70.6^{a} (0.5)	68.8^{a} (0.4)	67.2^{a} (0.4)	729.1^{b} (13.9)	142.8^{b} (2.6)	39.6^{ab} (0.6)	21.4^{b} (0.2)	16.3^{bc} (0.1)
15	91.3^{c} (0.7)	74.6^{a} (0.4)	70.4^{a} (0.4)	68.6^{a} (0.4)	66.7^{b} (0.5)	728.0^{b} (15.9)	142.8^{b} (3.0)	39.7^{b} (0.8)	21.4^{b} (0.3)	16.5^{c} (0.1)

Differences between the means with the same exponent letter within a column are not significant at probability $p < 0.05$. Values in parentheses are the standard deviation.

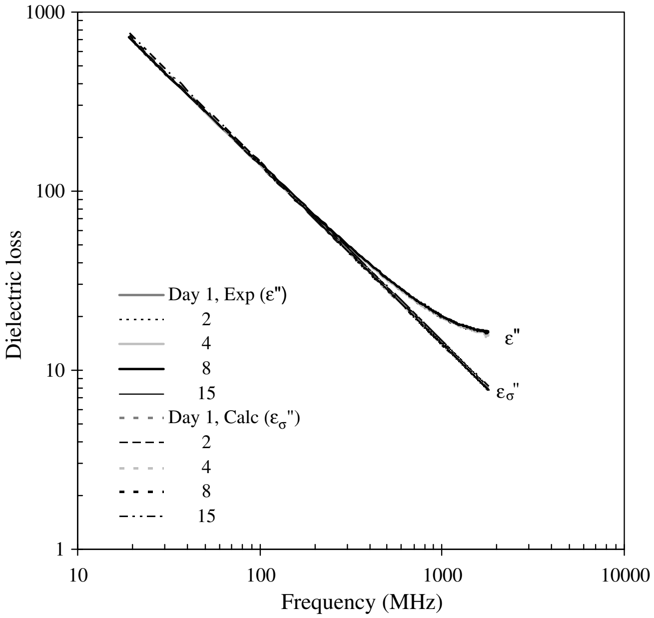

Fig. 4. Experimental (ε'') and calculated (ε''_σ) values of thick albumen dielectric loss of 15 days stored eggs at room temperature in the 20–1800 MHz frequency range.

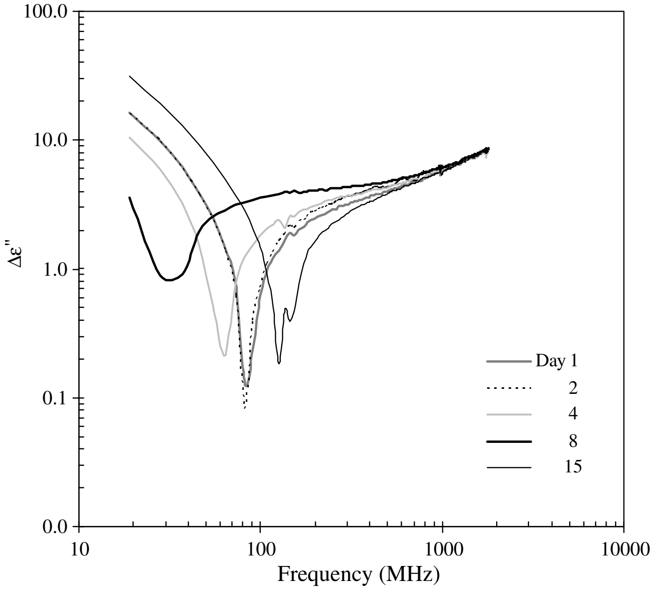

Fig. 5. Numerical absolute differences ($\Delta\varepsilon''$) between experimental (ε'') and calculated (ε''_σ) thick albumen dielectric loss of 15 days stored eggs at room temperature in the frequency range 20–1800 MHz.

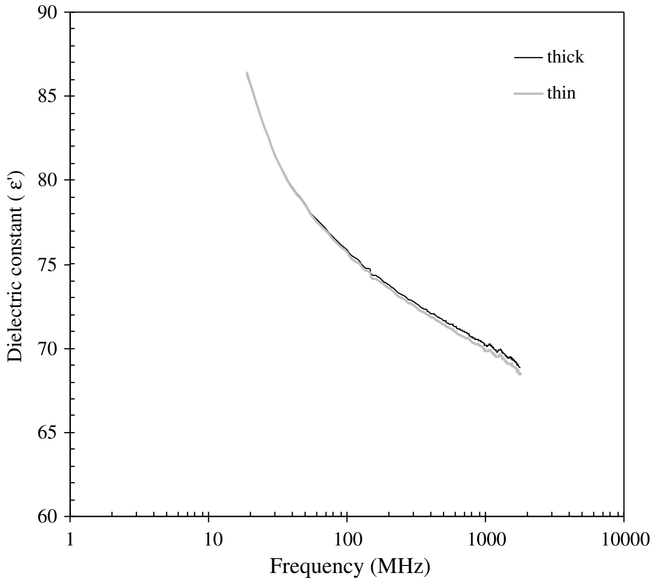

Fig. 6. Dielectric constant (ε') of eggs thick and thin albumen of 9 days stored eggs at room temperature in the frequency range 20–1800 MHz.

values of ε' and ε'' for frequencies of 20, 100, 400, 900 and 1800 MHz.

In general, both the dielectric constant and loss factor of yolk are consistently smaller than that of albumen. A close examination of Fig. 2 shows that the loss factor of albumen is about five times that of the yolk. These differences can be attributed largely to higher electric conductivity (0.77–0.80 S/m) and moisture content (87–89%) in albumen than that of yolk (0.25–0.28 S/m). For all considered frequencies, dielectric properties were positively correlated with storage time (Table 6). Coefficients of determination (R^2) were from 0.295 (ε'' at 900 MHz) to 0.457 (ε' at 400 MHz) for linear correlation and from 0.371 (ε'' at 900 MHz) to 0.555 (ε'' at 20 MHz) for logarithmic correlation. It is useful to emphasize that these coefficients took into account the dispersion of the data at the same days of storage. Using the mean values in Table 6, the coefficient of determination for ε'' at 20 MHz reaches 0.956 for logarithmic correlation (0.963 at 100 MHz) and 0.995 for a second order curve at 100 MHz ($R^2 = 0.538$, considering the data dispersion, Fig. 7). Over the tested storage period, ε' increased by 20% at 400 MHz and ε'' by 22% at 20 MHz.

Table 5
Dielectric properties of thick and thin albumen of 9 days stored eggs at room temperature in the frequency range 20–1800 MHz

Kind of albumen	Dielectric constant					Loss factor				
	Frequency (MHz)									
	20	100	400	900	1800	20	100	400	900	1800
Thick	86.3^{a} (0.6)	75.9^{a} (0.4)	72.1^{a} (0.5)	70.4^{a} (0.5)	68.8^{a} (0.5)	765.1^{a} (22.7)	149.2^{a} (4.3)	41.3^{a} (1.1)	22.2^{a} (0.5)	17.1^{a} (0.3)
Thin	86.4^{a} (0.5)	75.7^{a} (0.4)	71.9^{a} (0.5)	69.9^{a} (0.6)	68.5^{a} (0.6)	778.0^{a} (19.5)	151.6^{a} (3.6)	41.9^{a} (0.9)	21.1^{b} (0.4)	17.2^{a} (0.2)

Differences between the means with the same exponent letter within a column are not significant at probability $p < 0.05$. Values in parentheses are the standard deviations.

Table 6
Dielectric properties of the yolk stored for 15 days at room temperature in the frequency range 20–1800 MHz

Day	Dielectric constant					Loss factor				
	Frequency (MHz)									
	20	100	400	900	1800	20	100	400	900	1800
1	48.7^{a} (4.6)	33.9^{a} (3.4)	28.9^{a} (2.8)	27.5^{a} (2.4)	26.4^{a} (2.1)	216.9^{a} (22.3)	47.4^{a} (4.8)	15.2^{a} (1.5)	9.1^{a} (0.9)	7.4^{a} (0.7)
2	51.7^{ab} (2.3)	35.8^{ab} (1.6)	30.7^{ab} (1.3)	29.4^{a} (1.2)	28.5^{bc} (1.1)	226.3^{a} (15.0)	49.4^{a} (3.0)	15.9^{ab} (0.8)	9.6^{a} (0.4)	7.8^{a} (0.2)
4	52.9^{bc} (3.1)	37.4^{bc} (2.4)	31.9^{bc} (2.2)	29.8^{ab} (2.3)	28.2^{ab} (2.3)	246.3^{b} (15.6)	53.2^{b} (3.5)	16.7^{bc} (1.2)	9.9^{ab} (0.7)	7.8^{ab} (0.6)
8	54.3^{bc} (4.2)	39.4^{cd} (3.1)	33.9^{cd} (2.7)	32.0^{bc} (2.6)	30.5^{cd} (2.5)	263.7^{b} (24.1)	56.6^{bc} (5.1)	17.5^{c} (1.6)	10.3^{b} (0.9)	8.4^{bc} (0.7)
15	56.5^{c} (1.3)	40.4^{d} (1.0)	34.8^{d} (0.9)	32.8^{c} (1.0)	31.1^{d} (0.9)	265.1^{b} (6.4)	57.0^{c} (1.3)	17.7^{c} (0.4)	10.4^{b} (0.4)	8.6^{c} (0.3)

Differences between the means with the same exponent letter within a column are not significant at probability $p < 0.05$. Values in parentheses are the standard deviations.

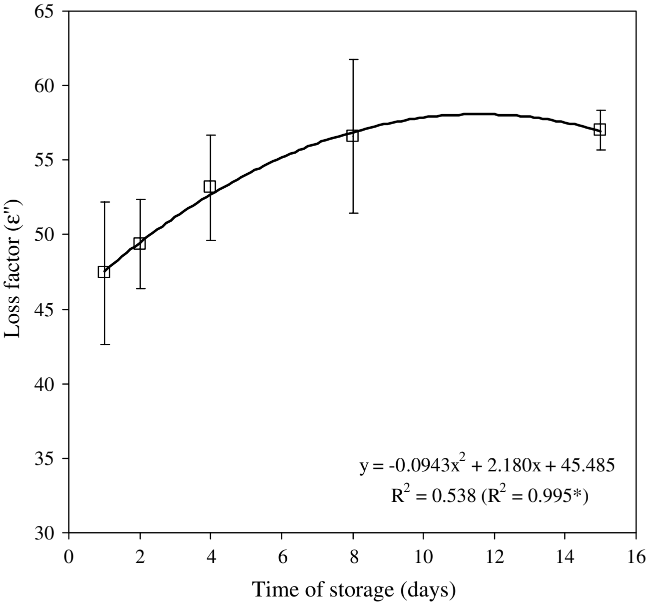

Fig. 7. Loss factor (ε'') of egg yolk during storage at 100 MHz. Error bars are the standard deviation. *Calculated by considering the mean values of ε'' for each day of storage.

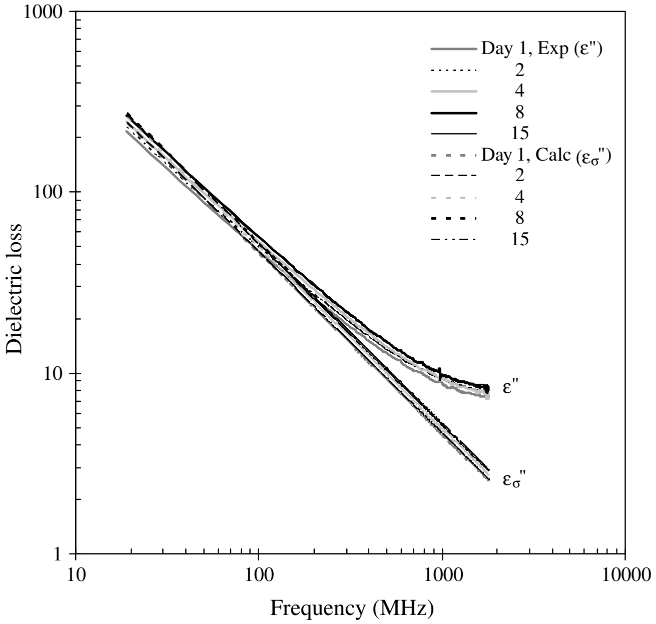

Fig. 8. Experimental (ε'') and calculated (ε''_{σ}) values of yolk in eggs stored for 15 days at room temperature in the frequency range 20–1800 MHz.

The value of ε'' was correlated with electric conductivity at 100 MHz with a strong linear correlation ($R^2 = 0.971$). Values of experimentally obtained (ε'') and calculated (ε''_{σ}) using the Eq. (3) are shown in Fig. 8. Absolute differences $|\Delta\varepsilon''|$ between ε'' and ε''_{σ} are shown in Fig. 9. Again, examination of Fig. 8 indicates where the mechanism of dielectric dispersion changes from ionic conduction-dominated to rotational-dominated behavior. Contrary to the results obtained for albumen, the 'critical' frequency analysis for the yolk appears to provide a more practical and efficient tool to utilize for the effects of storage time. This is strongly supported by a logarithmic correlation between the observed critical frequency and days of storage which results with an R^2 value of 0.999 as depicted in Fig. 10.

Complex biochemical changes in yolk during storage contributed to the trend observed in Fig. 10. As was mentioned previously, the increase in ε' and ε'' can also be attributed to electrolyte content increase due to lipids and proteins physiological change and to the migration of ions and water through the vitelline membrane during storage (Funk, 1948). Increase in pH yolk during storage is due to loss of carbon dioxide through the albumen, and through the pores in the shell (Rossi, Pompei, & Hidalgo, 1995). Both the increase of moisture content in yolk (from 48.0% to 50.5%, Table 3) and the loss of dissolved carbon dioxide (as reflected in increased pH) may have been responsible for the dipole mechanisms to become more predominant and shifted the critical frequency to the low spectrum.

Second order correlations between moisture and dielectric parameters (average values) were characterized by R^2 values of 0.965 (ε' at 20 MHz) and 0.949 (ε' at 1800 MHz) and from 0.934 (ε'' at 20 MHz) to 0.968 (ε'' at 1800 MHz).

A summary of all the above mentioned correlations (both for yolk and albumen) is given in Table 7.

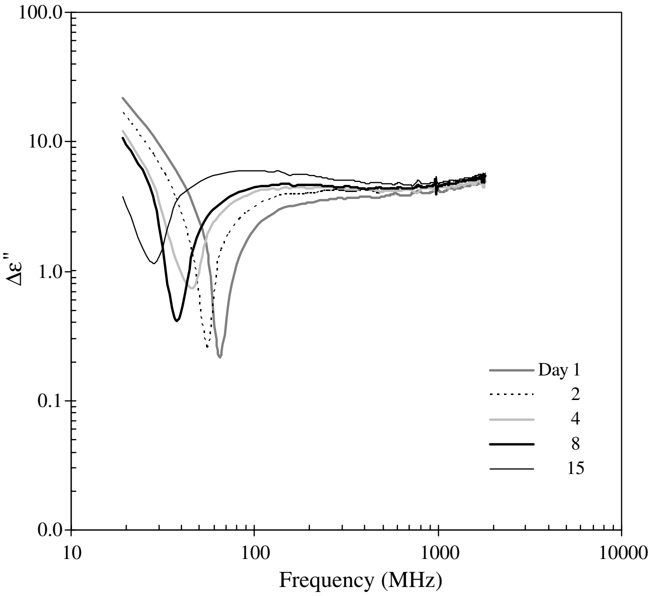

Fig. 9. Numerical absolute differences ($\Delta\varepsilon''$) between experimental (ε'') and calculated (ε''_{σ}) dielectric loss of yolk in eggs stored for 15 days at room temperature.

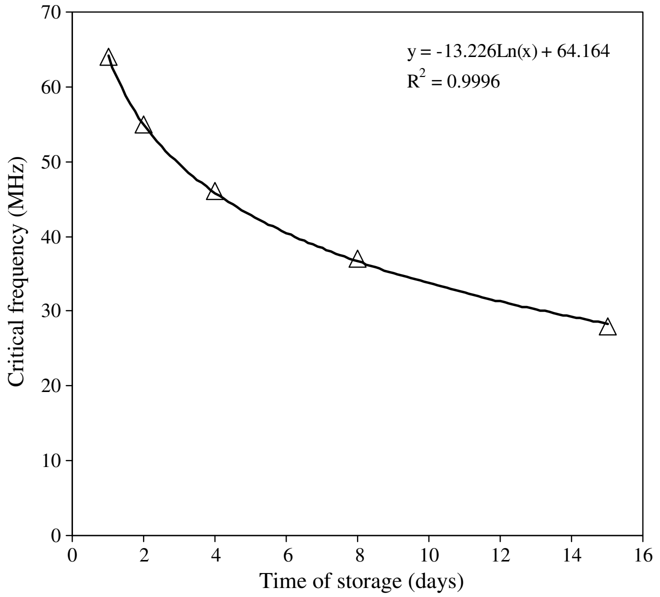

Fig. 10. Critical frequency for yolk in hen eggs during storage at room temperature.

Table 7
Summary of the correlations mentioned in the text

Variable		Function	R^2	Condition
Dependent (y)	Independent (x)			
ε' of albumen	Storage time (days)	$y = 0.287x + 86.946$	0.418	at 20 MHz
		$y = 1.581\text{Ln}(x) + 86.475$	0.436	at 20 MHz
		$y = 1.581\text{Ln}(x) + 86.475$	0.500	at 20 MHz[a]
Critical frequency for the albumen (MHz)	Storage time (days)	$y = -6.8087x + 91.783$	0.982	until day 8[a]
ε'' of yolk	Storage time (days)	$y = 0.086x + 9.341$	0.295	at 900 MHz
		$y = 0.509\text{Ln}(x) + 9.156$	0.371	at 900 MHz
		$y = 19.730\text{Ln}(x) + 216.575$	0.555	at 20 MHz
		$y = 19.730\text{Ln}(x) + 216.575$	0.956	at 20 MHz[a]
		$y = -0.0943x^2 + 2.180x + 45.485$	0.538	at 100 MHz
		$y = -0.0943x^2 + 2.180x + 45.485$	0.995	at 100 MHz[a]
		$y = 3.895\text{Ln}(x) + 47.371$	0.963	at 100 MHz[a]
	Conductivity (S/m)	$y = 254.749x - 16.368$	0.971	at 100 MHz[a]
ε' of yolk	Storage time (days)	$y = 0.389x + 29.704$	0.457	at 400 MHz
Critical frequency for the yolk (MHz)	Storage time (days)	$y = -13.226\text{Ln}(x) + 64.164$	0.999	–[a]
Yolk moisture (%)	ε' of yolk	$y = 0.04663x^2 - 4.562x + 159.516$	0.965	at 20 MHz[a]
		$y = 0.1329x^2 - 7.143x + 144.093$	0.949	at 1800 MHz[a]
	ε'' of yolk	$y = 0.00127x^2 - 0.5709x + 111.980$	0.934	at 20 MHz[a]
		$y = 1.6035x^2 - 23.652x + 135.249$	0.968	at 1800 MHz[a]
Yolk pH	Conductivity (S/m)	$y = 5.5482x + 4.6131$	0.621	–

[a] Calculated by considering the mean values.

4. Conclusions

Analysis of the dielectric dispersion in the range 20–1800 MHz on albumen and yolk of hen eggs stored at room temperature for 15 days led to the following conclusions:

(1) dielectric properties of yolk constituents have been shown to be correlated with aging of eggs. It opens up new perspectives in egg quality characterization by means of dielectric spectroscopy;

(2) the ε' of thick albumen at 20 MHz was influenced significantly by storage time, and the logarithmic correlation between this parameter and the time, although significant, was moderately low ($R^2 = 0.436$). A linear correlation ($R^2 = 0.982$) emerged between the 'critical' frequency, where dielectric mechanism shifts from ionic conduction to dipole polarization, and time of storage until the 8th day;

(3) ε' of yolk demonstrated a continuous increase with storage time (up to 20% at 400 MHz). The loss factor of yolk was also characterized by a general and

significant increase (up to 22%, at 20 MHz) in the considered storage period. Significant correlations emerged between dielectric parameters of yolk and time of storage. A logarithmic correlation was observed between 'critical' frequency and days of storage ($R^2 = 0.999$);

(4) the method based upon the comparison between the experimental ε'' and calculated values by means of conductivity and frequency can potentially be used to determine the age of eggs or monitor freshness of eggs supply.

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References

Agilent technologies. (2005). *Basics of measuring the dielectric properties of materials*. Application note. Palo Alto, CA. p. 31.

AOAC, Association of Official Analytical Chemists. (1990). Official method No. 925.30. Gaithersburg, MD 20877-2417, USA.

Bengtsson, N. E., & Risman, P. O. (1971). Dielectric properties of food at 3 GHz as determined by a cavity perturbation technique II. Measurements on food materials. *Journal of Microwave Power, 6*(2), 107–123.

Berardinelli, A., Giunchi, A., Guarnieri, A., & Ragni, L. (2005). Effetti della durata di conservazione sugli indici qualitativi delle uova da consumo [Effects of the storage duration on the quality indices of shell eggs]. *Rivista di Ingegneria Agraria, 26*(2), 49–55.

Cotterill, O. J., & Winter, A. R. (1955). Egg white lysozime 3. The effect of pH on the lysozime–ovomucine interaction. *Poultry Science, 34*, 679–686.

EEC, Commission Regulation No. 2295/2003. (2003). *Official Journal of the European Union*, L 340/16.

EEC, Council Directive 89/437/EEC. (1989). *Official Journal of the European Union*, L 212.

Engelder, D. S., & Buffler, C. R. (1991). Measuring dielectric properties of food products at microwave frequencies. *Microwave World, 12*(2), 6–15.

Feng, H., Tang, J., & Cavalieri, R. P. (2002). Dielectric properties of dehydrated apples as affected by moisture and temperature. *Transactions of the ASAE, 45*(1), 129–135.

Fromm, D. (1966). The influence of ambient pH on moisture content and yolk index of the hen's yolk. *Poultry Science, 45*, 374–379.

Funk, E. M. (1948). The relation of the yolk index determined in natural position to the yolk index, as determined after separating the yolk from the albumen. *Poultry Science, 27*, 367.

Garcia, A., Torres, J. L., De Blas, M., De Francisco, A., & Illanes, R. (2004). Dielectric characteristics of grape juice and wine. *Biosystems Engineering, 88*(3), 343–349.

Green, A. D. (1997). Measurements of the dielectric properties of cheddar cheese. *Journal of Microwave Power and Electromagnetic Energy, 32*(1), 16–27.

Guan, D., Cheng, M., Wang, Y., & Tang, J. (2004). Dielectric properties of mashed potatoes relevant to microwave and radio-frequency pasteurilization and sterilization processes. *Journal of Food Science, 69*(1), 30–37.

Ikediala, J. N., Tang, J., Drake, S. R., & Neven, L. G. (2000). Dielectric properties of apples and codling moths in relation to microwave and RF heating. *Transactions of the ASAE, 43*(5), 1175–1184.

Haugh, R. R. (1937). A new method for determining the quality of an egg. *U.S. Egg Poultry, 39*, 27–49.

Heath, J. L. (1975). Investigation of changes in yolk moisture. *Poultry Science, 54*, 2007–2014.

Herve, A. G., Tang, J., Luedecke, L., & Feng, H. (1998). Dielectric properties of cottage cheese and cottage cheese surface treatment by microwaves. *Journal of Food Engineering, 37*(4), 389–410.

Içier, F., & Baysal, T. (2004). Dielectric Properties of food materials – 2: Measurement techniques. *Critical Review in Food and Nutrition, 44*, 473–478.

Johnson, L. N. (1966). *The structure and function of lysozyme* (Vol. 54). Oxford: Science Progress, pp.367–385.

Kent, M. (1977). Complex permittivity of fish meal: a general discussion of temperature, density and moisture dependence. *Journal of Microwave Power, 12*(4), 341–345.

Kim, Y. R., Morgan, M. T., Okos, M. R., & Stroshine, R. L. (1998). Measurement and prediction of dielectric properties of biscuit dough at 27 MHz. *Journal of Microwave Power and Electromagnetic Energy, 33*(3), 184–194.

Komarov, V., Wang, S., & Tang, J. (2005). In Kai Chang (Ed.), *Encyclopedia of RF and microwave engineering* (pp. 3693–3711). John Wiley and Sons, Inc.

Kundra, T. G., Raghavan, S. V., Akyel, C., Bosisio, R., & van de Voort, F. R. (1992). Electromagnetic properties of milk and its constituents at 2.45 MHz. *Journal of Microwave Power, 12*(4), 199–204.

Li-Chan, E., & Nakay, S. (1989). Biochemical basis for the properties of egg white. *Poultry Biology, 2*(1), 21–50.

Lu, Y., Fujii, M., & Kanai, H. (1998). Dielectric analysis of hen egg white with denaturation and in cool storage. *International Journal of Food Science and Technology, 33*, 393–399.

Lucisano, M., Hidalgo, A., Comelli, E. M., & Rossi, M. (1996). Evolution of chemical and physical albumen characteristics during the storage of shell eggs. *Journal of Agricultural and Food Chemistry, 44*(5), 1235–1240.

Lyng, J. G., Zhang, L., & Brunton, N. P. (2005). A survey of the dielectric properties of meats and ingredients used in meat product manufacture. *Meat Science, 69*, 589–602.

Metaxas, A. C., & Meredith, R. J. (1993). *Industrial microwave heating*. London, UK: Peter Peregrinus Ltd.

Nelson, S. O. (1965). Dielectric properties of grain and seed in the 1 to 50-mc range. *Transactions of the ASAE, 8*(1), 38–48.

Nelson, S. O. (1973). Electrical properties of agricultural products – a critical review. *Transactions of the ASAE, 16*(2), 384–400.

Nelson, S. O. (1983). Dielectric properties of some fresh fruits and vegetable at frequencies of 2.45 to 22 GHz. *Transactions of the ASAE, 26*(2), 613–616.

Nelson, S. O. (1991). Dielectric properties of agricultural product measurements and applications. *IEEE Transactions on Electrical Insulation, 26*(5), 845–869.

Nelson, S. O. (1992, chap. 6). Estimation of permittivities of solid from measurements on pulverized or granular materials, dielectric properties of heterogeneous materials. In A. Priou (Ed.), *Progress in electromagnetics research* (Vol. 6) (Kong J. A. ch. Ed.). New York: Elsevier Science Publishing Co.

Nelson, S. O. (1999). Dielectric properties measurement techniques and applications. *Transactions of the ASAE, 42*(2), 523–529.

Nelson, S. O., & Bartley, P. G. Jr., (2002). Frequency and temperature dependence of the dielectric properties of food materials. *Transactions of the ASAE, 45*(4), 1223–1227.

Nelson, S. O., Forbus, W. R., Jr., & Lawrence, K. C. (1994). Microwave permittivities of fresh fruits and vegetables from 0.2 to 20 GHz. *Transactions of the ASAE, 37*(1), 183–189.

Nelson, S. O., Forbus, W. R., Jr., & Lawrence, K. C. (1995). Assessment of microwave permittivity for sensing peach maturity. *Transactions of the ASAE, 38*(2), 579–585.

Ohlsson, T., Bengtsson, N. E., & Risman, P. O. (1974). The frequency and temperature dependence of dielectric food data as determined by a cavity perturbation technique. *Journal of Microwave Power, 9*(2), 129–145.

Ragni, L., Gradari, P., Berardinelli, A., Giunchi, A., & Guarnieri, A. (2006). Predicting quality parameter of shell eggs using a simple technique based on the dielectric properties. *Biosystems Engineering, 94*(2), 255–262.

Rossi, M., Hidalgo, A., & Pompei, C. (1995). Classical methods and new proposals for rapid evaluation of shell egg freshness. In *Proceedings of the sixth European symposium on the quality of eggs and egg products*, 25–29 September 1995. Zaragoza, Spain, pp. 23–31.

Rossi, M., Pompei, C., & Hidalgo, A. (1995). Freshness criteria based on physical and chemical modifications occurring in eggs during aging. *Italian Journal of Food Science, 7*(2), 147–156.

Robinson, D. S., & Monsey, J. B. (1972). Changes in the composition of ovomucin during liquefaction of thick egg white. *Journal of the Science of Food and Agriculture, 23*, 29–38.

Romanoff, A. L., & Romanoff, A. (1949). *The avian egg*. New York: John Wiley and Sons Co.

Seras, M., Courtois, B., Quinquenet, S., & Ollivon, M. (1987). Measurement of the complex permittivity of dielectrics during microwave heating: Study of flours and starches. In B. McKenna & M. Roques (Eds.), *Physical properties of foods. 2. COST 90bis final seminar on flours and starches, final seminar proceedings* (pp. 217–223). London: Elsevier Applied Science.

Sipahioglu, O., & Barringer, S. A. (2003). Dielectric properties of vegetables and fruits as a function of temperature, ash, and moisture content. *Journal of Food Science, 68*(1), 234–239.

Sone, T., Taneya, S., & Handa, M. (1970). Dielectric properties of butter and their application for measuring moisture content during continuous processing. In *18th international dairy congress*. IE: 221 (Food Science and Technology Abstract 12P1690 2(12)).

Stadelman, W. J., & Cotterill, O. J. (Eds.). (1995). *Egg science and technology* (4th ed.). New York: Food Product Press.

Tang, J. (2005). Dielectric properties of food. In H. Schubert & M. Regier (Eds.), *Microwave processing of food* (pp. 22–40). London: CRC Press, Woodhead Publishing Limited.

Tang, J., Feng, H., & Lau, Ming. (2001). Microwave heating in food processing. In X. Young, J. Tang, C. Zhang, & W. Xin (Eds.), *Advances in agricultural engineering*. New York: World Scientific Publisher.

Tran, V. N., Stuchly, S. S., & Kraszewski, A. W. (1984). Dielectric properties of selected vegetables and fruits, 0.1–10 GHz. *Journal of Microwave Power, 19*(4), 251–258.

Venkatesh, M. S., & Raghavan, G. S. V. (2004). An overview of microwave processing and dielectric properties of agri-food materials. *Biosystems Engineering, 88*(1), 1–18.

Zuercher, J., Hoppie, L., Lade, R., Srinivasan, S., & Misra, D. (1990). Measurement of the complex permittivity of bread dough by an open-ended coaxial line method at ultra-high frequencies. *Journal of Microwave Power Electromagnetic Energy, 25*(3), 161–167.

Wang, Y., Wig, T., Tang, J., & Hallberg, L. M. (2003). Dielectric properties of food relevant to RF and microwave pasteurization and sterilization. *Journal of Food Engineering, 57*, 257–268.

Wang, S., Tang, J., Johnson, J. A., Mitcham, E., Hansen, J. D., Hallman, G., et al. (2003). Dielectric properties of fruits, nuts, and insect pests as related to radio frequency and microwave treatments. *Biosystems Engineering, 85*(2), 201–212.