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Radio frequency treatments to control codling moth in in-shell walnuts

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Abstract

'Diamond' Walnuts (*Juglans regia* L.) in the shell were treated with radio frequency (RF) energy in a 27 MHz pilot-scale system to determine the treatment effect on third- and fourth-instar codling moth, *Cydia pomonella* (L.) (Lepidoptera: Tortricidae), mortality and walnut quality. After 2 and 3 min of RF treatments, infested in-shell walnuts were heated to 43 and 53°C. The corresponding insect mortality reached 78.6 and 100%. The fatty acid (FA) concentration of treated walnuts was not affected by RF treatments. The FA values were $<0.1\%$ after accelerated storage times up to 30 days at 35°C, simulating storage at 4°C for up to 3 years. The effect of RF treatments on walnut oil peroxide values (PV) was not significant. The PV value of walnuts was less than 1.0 meq/kg (the upper limit for good quality walnuts), after 20 days storage at 35°C that simulated 2 year storage at 4°C. The PV was about 1.2 meg/kg after 30 days storage at 35°C. RF treatments can, therefore, potentially provide an effective and rapid quarantine security protocol against codling moth larvae in walnuts as an alternative to methyl bromide fumigation.

Keywords: Radio frequency; Quarantine and phytosanitary treatment; Codling moth; Walnuts; Quality

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1. Introduction

Codling moth, *Cydia pomonella* (L.) (Lepidoptera: Tortricidae), is one of the main insects infesting walnuts (*Juglans regia* L.) in the field. This pest reduces the quality of walnuts by feeding, webbing, and by promoting entry of other walnut storage pests. Codling moth is also targeted by quarantine agencies in many foreign countries (Anon., 1982). Methyl bromide fumigation of in-shell walnuts is used to control codling moth and other pests to meet commercial phy-

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tosanitation requirements (Hartsell et al., 1991). However, the use of methyl bromide will be sharply restricted or eliminated by 2005 (Johnson et al., 1998), because of its high ozone depletion potential (Anon., 1995). Furthermore, an alternative fumigant phosphine used for phytosanitation purposes requires long periods of fumigation time (> 10 h) (Yokoyama et al., 1993) and may be subjected to future environmental restrictions (USEPA, 1998). Therefore, there is an urgent need to develop a practical alternative to control insect pests in walnuts while having minimum impact on walnut quality.

Conventional hot air or hot water disinfestation methods are suggested as alternatives to chemical fumigation. Much research on different fruit types and insect species has been conducted using hot water or air treatments alone or in combination with cold or controlled-atmosphere storage conditions (Tang et al., 2000). Hot water treatment is more efficient than hot air heating. Poor heat conduction within fruits during forced hot air or water heating results in long treatment times (Hansen, 1992) which are often detrimental to the quality of treated commodities.

A potential alternative disinfestation treatment involves the use of radio frequency (RF) energy. RF energy interacts with dielectric materials, such as fruits and nuts, to generate heat as a result of converting electromagnetic energy into thermal energy. The US Federal Communications Commission (FCC) has allocated 13.56 ± 0.067, 27.12 ± 0.160, and 40.68 ± 0.020 MHz radio frequencies for industrial, scientific, and medical (ISM) heating applications. The two main advantages of RF heating for walnuts compared with conventional heating methods are (1) fast and potentially more uniform heating that can lead to the development of continuous treatment processes, and (2) ability to treat walnuts sealed in plastic containers to avoid post-treatment contamination. In addition, RF treatments leave no chemical residue on commodities and have no known impact on the environment. For treating in-shell walnuts, RF energy interacts directly with walnut kernels through the shells and can significantly reduce the amount of time required for walnuts to reach the temperature lethal to insects as compared with conventional heating methods. Earlier research on RF treatments to control insects in cereal grain and pecans has been reviewed by Hallman and Sharp (1994) and Nelson (1996). Appropriately designed RF treatments can kill insects in these cited host materials. Dielectric property measurements suggest possible preferential heating for wheat weevil in wheat in the RF range of 10–100 MHz. But the accepted use of inexpensive chemical fumigation hindered the research using RF treatments (Nelson, 1996). Furthermore, most of earlier insect control studies were conducted without prior knowledge of the thermal death kinetics of target insects. The research most related to this study was reported by Nelson and Payne (1982), in which they applied a 40 MHz RF heat treatment to inactivate pecan weevil, *Curculio caryae* (Horn) (Coleoptera: Curculionidae), both in in-shell and broken pecans. A complete kill of insects was achieved at 53°C in pecan pieces of 2.6% moisture content and at 80°C in pecan pieces of 6.1% moisture content. However, the impact of the treatments on pecan oil quality was not reported.

Walnuts contain approximately 60% lipids and 54% unsaturated fatty acids (Mate and Krochta, 1997). Oxidation of unsaturated fatty acids is the primary reason for quality degradation. Oxidative rancidity results from interactions between radicals and the hydrogen atoms adjacent to unsaturated double bonds. The lipid eventually breaks down to small molecular compounds, such as aldehydes, ketones, alcohol and acids. Some of these compounds have very low flavor threshold concentrations and will result in undesirable off-flavors. Oxidative rancidity is enhanced by the presence of oxygen, increased temperatures and storage times. Peroxides are a product of the first step of oxidation of unsaturated fatty acids. Therefore, the peroxide value (PV) of pressed walnut oil is often selected as an indicator of walnut quality. A PV greater than 1.0 meq/kg is associated with the onset of oxidative rancidity (Forbus et al., 1980; Fourie and Basson, 1989).

Hydrolytic rancidity, on the other hand, results from enzymatic hydrolysis of triacylglyerols and the release of fatty acids (FA). The FA may contribute a soapy flavor and an off-flavor to walnuts. Determination of the PV and FA values will assess the potential effect of RF treatments on walnut quality during short- and long-term storage.

The objective of this study was to develop a postharvest treatment using RF energy to control codling moth larvae in walnuts in the shell based on the available thermal death kinetics of codling moth. A pilot-scale 27 MHz RF system was used to study process parameters leading to a complete kill of third- and fourth-instar codling moth. The effects of selected thermal treatments and storage conditions on the PV and FA values of the treated walnuts were also examined.

2. Materials and methods

2.1. RF heating theory and experimental system

Dielectric materials including most agricultural products store a part of the electric energy and convert a part into heat. The heat generated per unit volume (P in W/m^3) in a dielectric material when exposed to RF energy can be expressed as (Nelson, 1996)

$$P = 5.56 \times 10^{-11} f E^2 \varepsilon'' \quad (1)$$

where f is the frequency (Hz), E is the electric field intensity (V/m), and ε'' is the dielectric loss factor. The power coupled into a sample is nearly constant when the electric field intensity and dielectric loss factor do not vary at a fixed frequency. Therefore, at a constant operating condition, the temperature increase in the sample due to absorbed electromagnetic energy is only a function of the heating time. The temperature increase can be estimated by assuming that the electric field is uniform and dielectric properties are relatively constant. The temperature increase (ΔT in °C) of the sample during RF heating can be expressed as (Halverson et al., 1996)

$$\Delta T = \frac{kP}{C_p m} \Delta t \quad (2)$$

where C_p is the specific heat of the sample (J/kg °C); k is the energy coupling coefficient; m is the sample mass (kg); P is the input power (W); and Δt is the RF heating time (s). According to Eq. (2), the temperature of the sample will increase linearly with the heating time at constant operating conditions.

A 6 kW, 27 MHz pilot-scale RF system (COMBI 6-S, Strayfield International Limited, Workingham, UK) was used to heat walnuts (Fig. 1). The RF system consisted of a transformer, rectifier, oscillator, an inductance-capacitance pair commonly referred to as the ‘tank circuit’, and the work circuit. The transformer raised the voltage to about 9 kV and the rectifier changed the alternating current to direct current. Direct current was then converted by the oscillator into RF energy at 27 MHz. This frequency was determined by the values of the inductance and capacitor in the tank circuit. The parallel plate electrodes, with sample in-between, acted as the capacitor in the work circuit. The gap of the electrode plates can be changed to adjust RF power coupled to the sample between the two plates. Twenty in-shell walnuts were placed in a closed plastic box (19 ×

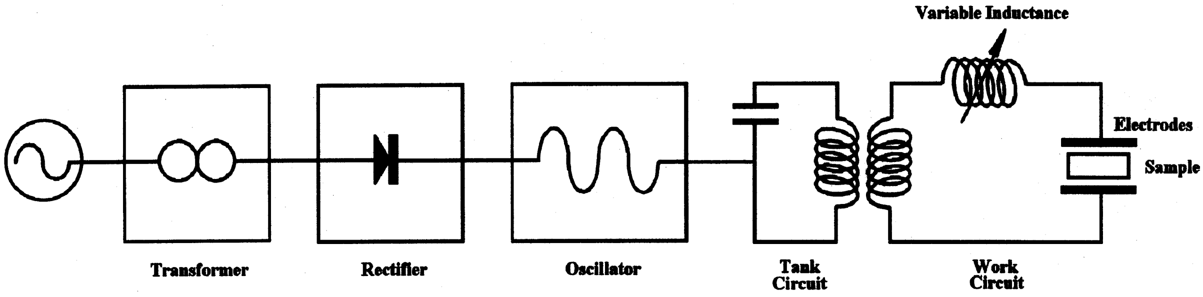

Fig. 1. Schematic view of the radio frequency heating system.

16 × 6 cm) made of polyvinyl chloride (PVC). Air in the gap between the electrodes, and the plastic box did not heat during the RF treatment. The gap of the electrodes was adjusted to 6.12 cm to provide 0.4 kW of power.

When RF waves are directed at walnut kernels infested with codling moth larvae, the absorption of RF energy depends on their dielectric loss factors. The difference between the dielectric properties of insects and the host material is important when considering the possibilities of preferential differential heating or insects. The dielectric properties of codling moth larvae and walnuts were determined by the coaxial probe technique (Engelder and Buffer, 1991) using a HP system (Model: 85070B, Hewlett Packard Corp. Santa Clara, CA). The dielectric probe system was calibrated with a standard air-short-triple deionized water calibration procedure. Typical error of the system was 5%. A detailed description of dielectric property measurement for codling moth larvae and fruits is provided in Ikediala et al. (2000a).

2.2. Determination of RF treatment time

The temperature in the walnuts increases linearly with RF heating time if the coupled power remains constant Eq. (2). The experimental conditions for the disinfestation studies were walnut mass $m = 0.3$ kg and measured RF power input $P = 0.4$ kW. The coupling factor k was selected as 0.2 based on preliminary experiments. The small k value was the result of the small quantity of walnuts used in a large RF system. The RF system thus provided a specific RF power level (kP/m in Eq. (2)) of 0.27 kW/kg. With the specific heat for walnuts of $C_p = 1.4$ kJ/kg per °C, the final walnut temperature after given RF treatment times was estimated using Eq. (2). When heating walnuts from a room temperature of 20°C, the average walnut temperatures calculated by Eq. (2) were 31, 43 and 54°C for 1, 2 and 3 min heating periods, respectively.

Ikediala et al. (2000b) studied the effect of temperature and time combinations on mortality of fifth-instar codling moth using conduction heating blocks. The thermal-death-time (TDT) curve of codling moth larvae suggests a complete kill of 200 insects when heated to 48°C and held for 20 min, 50°C for 5 min or 52°C for 2 min. Combining the information and estimated walnut temperatures due to RF heating and the TDT curve for codling moth larvae, 1, 2 and 3 min were selected so that the insects in the walnuts would reach a range of mortality levels after 5 min of holding time at the final temperature. We expected complete mortality of codling moths after 3 min RF heating and (to reach 54°C) 5 min holding.

2.3. RF treatments

Walnuts were infested with third- and fourth-instar codling moth by drilling a hole through the shell at a ratio of about one larva per walnut at USDA-ARS, Yakima Agricultural Research Laboratory in Wapato, WA, USA. Twelve plastic boxes each containing 20 infested walnuts were shipped by Federal Express to Washington State University for RF treatments. One box of infested in-shell walnuts was used as the control and one box was used for each treatment run. Two preliminary test runs were carried out to verify walnut kernel temperatures for 2- and 3-min periods of the selected RF treatment time. Immediately after RF treatments, a type T thermocouple (0.8 mm diameter and 0.8 s response time) was used to measure the kernel temperatures of 20 walnuts, by inserting the sensor through pre-drilled holes in the shell. A fiber optic temperature sensor (Photonetics Inc., MetriCor Div., Wakefield, MA, USA) was inserted into the center of a walnut kernel to monitor the temperature profile during RF heating and natural cooling periods.

The three selected RF treatments were repeated two to five times. For each test, walnuts at the initial room temperature (20°C) were treated in the RF pilot-scale system to reach the desired walnut kernel temperature. The treated walnuts were then removed from the cavity and held at ambient for 5 min. The walnut kernel temperature decreased < 5°C during the holding period. The walnuts were stored for a day at 4°C to minimize the probable effect of the elevated temperatures on walnut quality, then removed and kept at room conditions for 1 day to minimize the effect of cold stupor on the insects before examination. Insect mortality was analyzed based on the total number (dead or alive) of recovered insects in the infested walnuts. Dead larvae were defined as larvae that exhibited no movement. Moribund larvae which showed very little or sluggish movement when prodded with a blunt instrument were monitored for 5 days at room temperature in immature apples and a 16-h light:8-h dark lighting regime.

2.4. Walnut quality analyses

At elevated temperatures, lipid oxidation reaction can be very rapid. Therefore, walnut quality after the 3 min RF treatment, the most severe treatment used in this study, was analyzed. Twenty freshly harvested walnuts were used as the control, and 120 walnuts were treated in six replicates by 3 min RF heating as tested earlier for the insect mortality. Accelerated shelf life tests of in-shell walnuts were performed in an incubator at 35°C and 30% relative humidity (RH) for 10, 20, and 30 days. These conditions simulated approximately 1, 2 and 3 year storage periods at 4°C, respectively, based on a Q_{10} value of 3.4 at 35°C, which is defined as the increase of shelf-life, as a ratio, when storage temperature is reduced by 10°C (Taoukis et al., 1997). To distinguish the effect of RF treatments from the effect of storage, untreated walnuts were stored at 35°C and 30% RH for up to 30 days as controls.

After pre-determined storage periods at 35°C, oil was pressed from RF-treated walnuts at room temperature using a CARVER Laboratory Press (Fred S. Carver Inc., Summit, NJ, USA). The peroxide value was determined by the official method (Cd 8–53) of the American Oil Chemists Society (AOCS, 1998a). After titration of the walnut oil in acetic acid/choroform solutions and a blank with 0.01 N sodium thiosulfate solution, the peroxide value was calculated by the following equation

$$\mathrm{PV} = \frac{(S - B) \times N_1 \times 1000}{W} \tag{3}$$

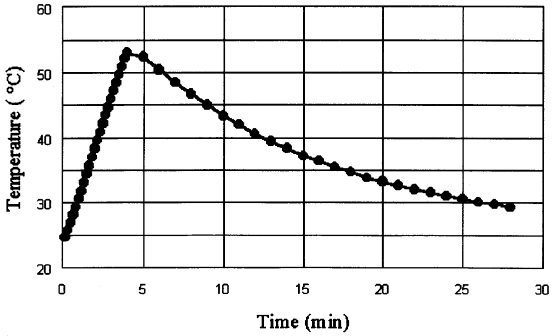

Fig. 2. A typical heating and cooling curve for in-shell walnut kernels subjected to radio frequency energy.

where PV is the peroxide value (meq/kg), B is titration of the blank (ml) which is the mean of triplicate titrations, S is titration of the walnut oil (ml), N_1 is the normality of sodium thiosulfate solution, and W is the weight of the walnut oil (g).

Fatty acid value was determined by the AOCS standard method Ca 5a–40 (AOCS, 1998b). Based on the titration of the walnut oil with 0.1 N NaOH solution, fatty acid value was calculated by the following equation

$$\mathrm{FA} = \frac{V \times N_2 \times 28.2}{W} \tag{4}$$

where FA is defined as fatty acid (% oleic acid), V is titration of the walnut oil (ml), and N_2 is normality of NaOH solution, accurately standardized by the procedure described in AOCS specification H 12–52 (AOCS, 1998c). Assays for both PV and FA values were conducted in duplicate. Analysis of variance (ANOVA) was performed using Minitab statistical software to determine the effects of RF treatments and storage conditions on walnut quality.

3. Results and discussion

3.1. Walnut temperature

A typical temperature-time profile for an in-shell walnut kernel during RF treatment is shown in Fig. 2. Walnut kernel temperature increased

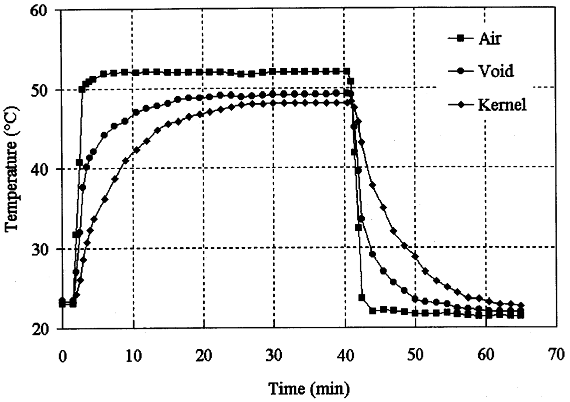

Fig. 3. Typical heating and cooling curves for in-shell walnut void and kernel when subjected to forced hot air treatment (air temperature, 53°C; air velocity, 1 m/s).

linearly with process time as predicted by Eq. (2). The heating rate, as indicated by the slope of the temperature profile during the heating period, can be adjusted by RF power input into the walnuts to control process time while achieving the same final temperature. Thus, RF treatments provide a major advantage over conventional heating methods. In hot air treatments, the heating rate decreases as the product temperature approaches the medium temperature, resulting in prolonged heating (Soderstrom et al., 1996). Typical temperature-time curves for walnut kernels and in-shell air void during a hot-air treatment (53°C at 1 m/s air velocity) are shown in Fig. 3. The rise of kernel temperature lagged significantly behind that of in-shell void temperature which in turn increased very slowly with treatment time once it reached within about 10°C of the hot air temperature. This was the result of significant thermal resistance in the porous walnut shell and the in-shell void that hindered the transfer of thermal energy from the hot air outside of the walnut shell. RF energy, on the other hand, was applied directly to walnut kernels in shell and generated heat in the insects and walnut kernels. The treatment based on RF energy can, therefore, significantly reduce heating time allowing for development of a continuous process.

In the pilot-scale RF system, the walnut kernel temperatures increased to an average of 53°C in about 3 min. Following RF heating, the walnut temperature decreased by less than 5°C during exposure to still air at 20°C for 5 min. This cooling rate can be increased by cold air circulation to reduce the potential heat effects on walnut quality. Walnut kernel temperatures were measured from 20 samples for each 2 and 3 min RF treatments. In the 2 min RF treatment, the mean value and standard deviation of the kernel temperature were 42.6 and 2.5°C. With the 3 min RF treatment, the mean value and standard deviation of the kernel temperature reached 53.3 and 3.6°C. These two temperatures agreed with the calculated temperatures of 43 and 54°C using Eq. (2). The large standard deviation of the measured temperatures was probably due to a non-uniform RF electromagnetic field in the walnut shell and different walnut properties.

3.2. Codling moth mortality

Evaluation of infested and control walnuts indicated no mortality due to shipment of the infested samples for treatment (Table 1). Insect mortality reached 47.5, 78.6 and 100% following 1, 2 and 3 min RF heating treatments, respectively. In general, increasing treatment time under given operating conditions resulted in increased walnut kernel temperatures, and thus increasing larvae

Table 1
Mortality of third- and fourth-instar codling moth larvae in walnuts after radio frequency (27 MHz) treatments

Treatments	# Total recovered	# Alive	# Dead	Repeat number (*n*)	Mortality ± S.D. (%)
Control	17	17	0	2	0
1 min heating	61	32	29	2	47.5 ± 11
2 min heating	84	18	66	5	78.6 ± 6
3 min heating	70	0	70	4	100 ± 0

Table 2
Quality characteristics (means ± S.D. over duplication) of in-shell walnuts treated by radio frequency energy (3 min)

Storage time at 35°C	Peroxide value (meq/kg)		Fatty acid (%)	
Day	Control	RF treated	Control	RF treated
0	0.26 ± 0.04	0.28 ± 0.04	0.08 ± 0.01	0.08 ± 0.01
10	0.49 ± 0.05	0.51 ± 0.05	0.08 ± 0.01	0.09 ± 0.01
20	0.93 ± 0.05	0.98 ± 0.05	0.10 ± 0.01	0.08 ± 0.01
30	1.04 ± 0.05	1.17 ± 0.05	0.11 ± 0.01	0.10 ± 0.01

mortality. A similar effect was observed in an earlier study with codling moth larvae in cherries following 915 MHz microwave treatments (Ikediala et al., 1999). The present study also showed that 1 and 2 min treatment periods were inadequate to control codling moth larvae in walnuts. These results were expected based on the thermal-death-time curve obtained in our earlier study (Ikediala et al., 2000b).

3.3. Walnut quality

The effect of RF treatments on the PV and FA values in pressed walnut oil are summarized in Table 2. The RF treatments did not significantly affect the FA values of RF-treated walnuts during accelerated storage at 35°C for up to 30 days. The mean PV value in the walnuts increased slightly from 0.26 to 0.28 meq/kg due to the 3 min RF treatment, but this difference was not statistically significant ($P > 0.05$). There was no difference between the PV of RF-treated walnuts and the control after storage at 35°C for up to 20 days. The PV of treated walnuts were slightly larger than the control after storage at 35°C for 30 days, which simulates 3-year storage at 4°C. However, walnuts are rarely stored that long. The RF treatments used in our study, therefore, appeared to have no effect on walnut quality. This is likely due to the short times that walnuts were exposed to an elevated temperature. Johnson et al. (1992) also observed that walnuts treated by hot air at 55°C for 1 h showed no increase in rancidity after 9 months of storage at 20°C. The reaction rate can be introduced based on the Arrhenius relationship

$$k = k_{\mathrm{ref}} e^{-E_{\mathrm{A}}/R(1/T - 1/T_{Ref})} \qquad (5)$$

where k is reaction rate constant, E_{A} is activation energy (J/mol), R is universal gas constant (8.314 J/Kmole), T is absolute temperature (K), subscript 'ref' stands for reference. The activation energy E_{A} for lipid oxidation in general is about 100 kJ/mole (Taoukis et al., 1997), and the reaction rate of lipid oxidation at 53°C is about 3.2 times larger than at 43°C. That is, 5 min at 53°C is equivalent to about 16 min at 43°C. A commercial hot air drying process in bins normally lasts more than 10 h at about 43°C (Thompson et al., 1998). Therefore, the RF treatments would cause less lipid oxidation than commercial drying operations.

The PV of the oils pressed from both the RF-treated and control walnuts increased steadily with storage time at 35°C. According to Diamond Walnut Growers, Inc. of California, established standards of the walnut industry for acceptable walnuts contain less than 0.6% FA and PV of less than 1.0 meq/kg oil (Lindsay, private communication, 1999). Bossell (1989) also suggested that freshly refined fats should have PVs of less than 1 meq/kg. Walnuts treated with RF for 3 min in this study had FA and PV values within the limits for quality walnuts even after 20 days storage at 35°C. The PV, however, increased to 1.17 meg/kg after 30 days storage at 35°C. These results showed that 3 min RF heating at 27 MHz was effective for killing all the codling moth larvae in in-shell walnuts with no adverse effect on walnut quality.

3.4. Discussion

The experimental results showed that 3 min of RF heating resulted in 100% mortality of codling moth larvae (Table 1). However, the measured walnut kernel temperatures during each test were not uniform and a minimum final temperature of 49°C was observed. Based on the earlier determined thermal-death-time curve for 100% codling moth mortality, it is expected that a treatment temperature of 49°C with a 5 min holding period is not adequate to completely kill the insects. The unexpected complete kill with 3 min RF treatment can be explained by preferential heating of codling moth larvae infesting the walnuts. Based on RF heating theory and dissipated power calculations in Eq. (1), the absorption of RF energy is proportional to the dielectric loss factor of the materials. The dielectric loss factor was clearly different between codling moth larvae and walnut kernels, especially in the RF range (Fig. 4). The dielectric loss factor of the insects was much higher than that of walnuts, similar to that found by Nelson (1991) for weevils and wheat. Codling moth larvae might absorb more energy than walnuts when subjected to the same electromagnetic field. According to the temperature increase in the material from Eq. (2), a higher temperature in codling moth larvae than in walnut kernels is expected. The much larger dielectric loss factor of codling moths may account for the complete kill of the pests with a 3 min RF heating under our test conditions.

The final temperature of the insects after RF treatments and the post-treatment holding time at or close to the end temperature are the two most important factors that determine insect mortality. The 3 min RF treatment selected in this study to give 100% codling moth mortality in in-shell walnuts is specific to the power level (0.27 kW/kg) selected for our tests, and the mass of the walnuts used for each test. The end temperature can be achieved by different combinations of RF power applied into the walnuts (kP) per unit mass of the walnuts (*m*) and RF treatment time (Δt), as indicated in Eq. (2). This allows for flexibility in future design of RF treatments to satisfy the needs of the walnut industry and allows for a shorter treatment than the current 3 min treatment.

Moist materials in general have high dielectric loss factors and, therefore, RF energy can be preferentially coupled into the products. As a result, RF energy is very effective in drying moist products, and is commercially used to finish-dry bakery products. In-shell walnuts are often washed and air-dried in bins before moving to markets. RF treatments may achieve the tasks of

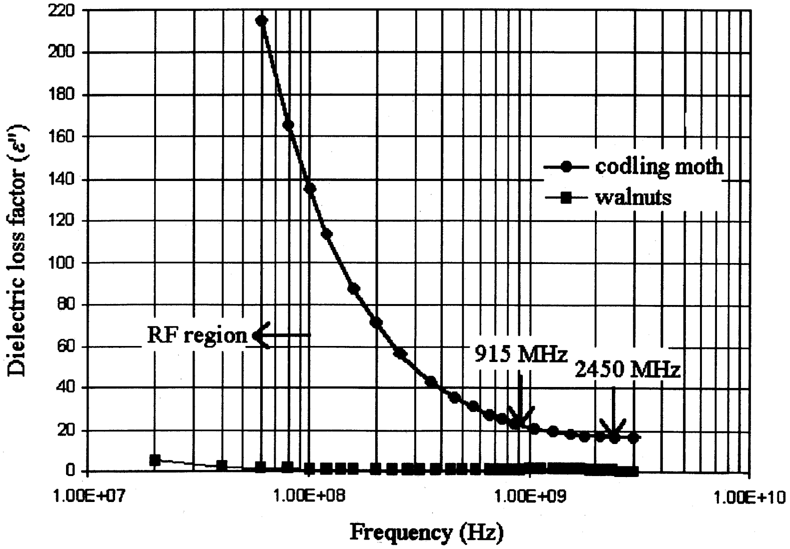

Fig. 4. Dielectric loss factor (ε″) of codling moth larvae and walnuts as a function of frequency.

both controlling insect pests and drying washed walnuts to reduce operation time and cost.

4. Conclusions

The final walnut kernel temperature is a key factor both for insect mortality and walnut quality. The mean kernel temperatures after 2 and 3 min RF treatments in a pilot-scale system were 43 and 53°C. Codling moth larvae mortality reached 47.5, 78.6 and 100% after 1, 2 and 3 min heating periods, respectively.

The effect of RF treatments on PV and FA values of walnuts was not significant, indicating that in-shell walnuts were tolerant of a short exposure to the selected treatment temperatures less than or equal to 53°C.

The results of this study were obtained from experiments using a pilot-scale RF system with relatively small infested in-shell walnuts (~ 300 g) in small boxes (19 × 16 × 6 cm). For industrial applications in which large volumes of walnuts need to be treated, it is necessary to design a continuous treatment system with conveyor belts moving multi-layers of walnuts through RF applicators. Studies of scale-up conditions to provide uniform RF energy to walnuts over relatively wide conveyor belts are necessary.

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