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### High-temperature-short-time thermal quarantine methods

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#### Abstract

In this paper, kinetic models are discussed with respect to their uses in describing the intrinsic thermal mortality of insect pests. A unique heating block system was used to obtain kinetic information for the thermal mortality of codling moth larvae. The kinetic data demonstrated the possibility to develop high-temperature-short-time thermal treatments to control codling moth and reduce thermal impact on product quality. Equations are presented to evaluate cumulative effects of any time–temperature history on the thermal mortality of target insect pests and on the quality of host materials. Computer simulation results demonstrated that the cumulative thermal effects on product quality during the heating period in conventional hot air or hot water treatments are much more important than the cooling period. Radio frequency (RF) heating or microwave heating is suggested as an alternative to reduce adverse thermal impact to treated commodities during the heating period. A case study is presented to demonstrate the effect of RF heating in a high-temperature-short-time thermal treatment to control codling moth larvae in in-shell walnuts. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Quarantine; Heat treatment; Radio frequency; Kinetics; Cumulative heat effect; Codling moth; Nuts

#### 1. Introduction

Methyl bromide (MeBr) fumigation is an effective quarantine treatment against codling moth, *Cydia pomonella* (*L*.) (Lepidoptera: Tortricidae), and a commercial phytosanitation treatment to control other insect pests (Guance et al., 1981; Anon., 1983; Yokoyama et al., 1990; Moffitt et al., 1992). However, MeBr is identified by the

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U.S. Environmental Protection Agency (EPA), under the Federal Clean Air Act (Anon., 1990) and by the Montreal Protocol (Anon., 1995), as having high ozone depletion potential. The EPA has mandated the removal of MeBr from the chemical register and the phase out of its production and import into the United States by December 31, 2005. Similar legislation has passed in other industrialised countries. Due to an uncertain future for chemical fumigation and public concern over residues in treated products, large efforts have been made in developing non-chemical methods, particularly thermal treatments.

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It is a complex task to develop an effective thermal treatment that provides required guarantine security yet has minimum adverse effect on product quality. Systematic research to develop such a treatment protocol requires an understanding of the effects of heat on insect mortality and product quality. In this paper, we examine kinetic models with respect to their use in describing the intrinsic thermal mortality of insect pests. We use kinetic data for codling moth larvae to demonstrate the possibility of developing high-temperature-short-time thermal treatments to control codling moth and reduce thermal impact on product quality. We discuss the inherent limitations of conventional hot air or hot water treatments based on their cumulative thermal effects on insect mortality and product quality. Then we present the results of a case study in which radio frequency energy was used in a high-temperatureshort-time thermal treatment to control codling moth in in-shell walnuts.

## 2. Strategy for developing thermal quarantine treatments

Developing effective thermal treatments to control insects in fruits or nuts is a complicated task that involves entomology, engineering and fruit physiology. An important key to the development of successful thermal quarantine treatments is to balance complete kill of insects with minimised thermal impact on product quality. The effectiveness for developing thermal treatment protocols, therefore, depends upon three important components: (1) knowledge of thermal susceptibility of targeted insects; (2) engineering principles that govern thermal energy delivery methods; and (3) understanding of thermal effects on product quality. It is essential to have a clear knowledge of the intrinsic thermal susceptibility of insects independent of thermal energy delivery methods. Knowledge of the minimum required thermal energy to kill insects over a relatively large range of temperatures should provide flexibility for the design of thermal processes. Equally important is an understanding of fundamental engineering principles that govern various means to transfer energy to and within host commodities. Information for the thermal resistance of targeted insects needs to be combined with fundamental heat transfer theories and product quality data to develop effective thermal treatment protocols that deliver required energy to control target insects while minimising adverse impact on the quality of the treated commodities.

### 3. Thermal susceptibility of codling moth

Different methods have been used to study and analyse thermal resistance of insects. Probit analysis is a commonly used method. This analysis is based on the assumption that the frequency of individual deaths in an insect population under constant temperatures follows standard normal distribution over time. Although probit analysis is effective to confirm quarantine treatments (e.g. probit 9 criteria or 99.9968% mortality), it may not be suited for developing thermal methods. The analysis does not give kinetic data for insect mortality and does not provide sufficient information from which an optimal temperature—time combination can be chosen to reduce food quality loss.

### 3.1. Thermal kinetic models

The food industry has long been working on preservation technologies to kill pathogenic and spoilage bacteria and to minimise adverse thermal impact on food quality. The success of those developments relied heavily on the theory of thermobacteriology which provides an understanding of how bacteria respond to heat. Most important concepts in this theory are the *D* and *z* values determined from thermal death time (TDT) lethality curves for thermally inactivating pathogens (Stumbo, 1973). Those concepts are based on the observation that mortality of micro-organisms at a given temperature follows a logarithmic constant rate:

$$\log N = \log N_0 - \frac{t}{D} \tag{1}$$

where  $N_0$  and N are the initial and surviving number of bacteria after heat treatment, respec-

tively, and t is the treatment time (min). The susceptibility of bacteria to heat at a specific temperature is characterised by the value of D, the time (min) required to obtain one log reduction (tenfold reduction) in the bacterial population. The sensitivity of thermal susceptibility of bacteria to temperature change is characterised by a z value (the degree of temperature increase to result in one log reduction in D value). That is:

$$z = \frac{T_2 - T_1}{\log D_{T_1} - \log D_{T_2}} \tag{2}$$

where  $D_T$  represents value of D measured at temperature T, and  $T_1$  and  $T_2$  are two different temperatures.

Alternatively, changes in bacteria population N can be expressed as a first order kinetic model:

$$\frac{\mathrm{d}N}{\mathrm{d}t} = -kN\tag{3}$$

where k is the rate constant (min<sup>-1</sup>). Value of k changes with temperature, following an Arrhenius relationship:

$$k = k_{\text{ref}} e^{(-E_a/R)((1/T) - (1/T_{\text{ref}}))}$$
(4)

where T is absolute temperature (K),  $k_{\text{ref}}$  is the reaction rate constant at the reference temperature  $T_{\text{ref}}$ ,  $E_{\text{a}}$  is the activation energy (J/mol), and R is the universal gas constant (8.314 J/mol K).

The D and z values from the TDT curves are related to first order kinetic parameters k and  $E_{\rm a}$ . These relationships are:

$$D = \frac{2.303}{k} \tag{5}$$

and

$$E_{\rm a} = \frac{2.303RT_{\rm min}T_{\rm max}}{z} \tag{6}$$

where  $T_{\rm min}$  and  $T_{\rm max}$  are the minimum and maximum temperature (K) of a test range, respectively. That is, the D and z values in the theory of thermobacteriology can be calculated from the value of rate constant and activation energy in the classic reaction kinetic theory, or vice versa.

Once the D and z values, or k and  $E_a$  values, for a target bacterium are determined, the cumu-

lative temperature—time effect of a thermal treatment with a known temperature-time history on reduction of the bacteria can then be predicted with good accuracy. This method has been the basis for calculating the thermal processing times for commercial food thermal processes (Stumbo, 1973). The theory and concepts evolved from the TDT curves and the classic thermal kinetic model for describing thermal mortality of bacteria may be readily used to study thermal mortality of insects. Thomas and Mangan (1997) critically reviewed several models for Mexican fruit flies. They recommended the use of thermal kinetic models for estimating quarantine treatment levels to develop new treatment methods, and recommended use of the traditional probit analysis for validation tests. They demonstrated that the thermal kinetic model reliably describes death kinetics of Mexican fruit flies subjected to constant temperatures. Jang (1986, 1991) also discussed the advantages of kinetic models for determining the heat treatment kinetics of fruit flies. In general, knowledge of the fundamental kinetics for the thermal death of insects will allow the prediction of lethal times over a range of temperatures.

A significant advantage of the thermal kinetic model or the use of D and z values over the probit analysis is the ability to predict the efficacy of a thermal process based on time-temperature history in host materials (see Section 3.2 and Section 4.1). At low temperatures, a lag or shoulder, however, often preceded the logarithmic order reduction in tests with micro-organisms. The shoulder section decreases with increasing temperature (King et al., 1979). Similar deviations from logarithmic order were reported for insects (Jang, 1986, 1991). The deviations, however, become less important at high temperatures ( $\geq$  45°C) and long periods of exposure time (Jang, 1991).

# 3.2. Calculation of cumulative time-temperature effects based on kinetic information

In thermal treatments, temperature within fruit changes with time. The time-temperature history in fruit depends on methods of heating and cool-

ing. When developing optimised thermal treatments, it is important to be able to compare the effectiveness of different treatments. This can be done by calculating the cumulative thermal effect for a given treatment, based on kinetic data for the thermal mortality of target insects and for product quality losses.

Given a time-temperature history of T(t), the cumulative thermal mortality of this treatment can be calculated to an equivalent length of time,  $M_{\rm ref}$  minutes, at a reference temperature  $T_{\rm ref}$  by using the following relationship:

$$M_{\rm ref} = \int_0^t 10^{-(T(t) - T_{\rm ref})/z} \, \mathrm{d}t$$
 (7)

where T(t) is the time-temperature history obtained by real-time measurements, z value is from TDT studies of thermal mortality of target insect pests, and t is time. Detailed description of this relationship can be found in Stumbo (1973) for thermal mortality of micro-organisms. The above integration can be carried out analytically if the mathematical expression for T(t) is available, or numerically (e.g. using a spreadsheet) when the defined only by experimental T(t)is measurements.

Similar calculations can be made to evaluate the impact of a thermal treatment to quality, if kinetic information for quality losses due to thermal treatments is available:

$$Q_{\text{ref}} = \int_{0}^{t} 10^{-(T(t) - T_{\text{ref}})/z} dt$$
 (8)

where the value for z is for quality. z value varies between 17 and 45°C for overall sensory quality, texture and color changes in foolds (Holdsworth, 1997). But it is normally taken as 33°C for calculations in canning operations (Rao and Lund, 1986; Taoukis et al., 1997). No z value has been reported for quality changes of fresh fruits and nuts during thermal treatments against insect pests. But we used z = 33°C in this paper to demonstrate its use in evaluating the influence of heat treatments on quality.

The equivalent minutes at a reference temperature  $T_{\rm ref}$  for insect mortality,  $M_{\rm ref}$ , and for fruit quality,  $Q_{\rm ref}$ , can be used in combination with standard thermal-death-time (TDT) curves to

compare the effectiveness of a particular treatment. Information on TDT curve and z value for 5th instar codling moth is given in Section 3.5. Since our research activities have been focused on control of codling moth in fresh fruits and nuts, our following discussions on insect thermal mortality will be mostly limited to codling moth.

### 3.3. Thermal mortality data for codling moth

Several researchers have reported thermal mortality of codling moth instars (Yokoyama et al., 1991; Neven, 1994; Neven and Rehfield, 1995; Neven and Mitcham, 1996; Ikediala et al., 2000a). Yokoyama et al. (1991) reported that the 5th instar is the most heat tolerant developmental stage of the codling moth. A summary of reported mortality for 5th instar codling moth is shown in Table 1.

As shown in Table 1, different experimental methods were used to assess the thermal mortality of codling moth. There seems to be a lack of a consistent method to obtain experimental data that truly reflect the intrinsic insect susceptibility when subjected to heat. The reported thermal mortality information for codling moth was often confounded with heat transfer phenomena. Furthermore, information is lacking on codling moth heat resistance for short time (< 6 min) heat treatments. Furthermore heating rate is believed to have a significant effect on insect mortality rates (Neven, 1998a,b).

# 3.4. Limitation of reported methods for insect thermal mortality

Several experimental methods are reported to characterise the effect of thermal treatment on insect pests. Direct hot water bath immersion is a commonly used technique (Hayes et al., 1984; Jang, 1986; Yokoyama et al., 1991; Thomas and Mangan, 1997). For example, Thomas and Mangan (1997) and Jang (1986) placed fruit fly larvae in mesh bags (150 or 30 ml) and immersed the bags in a water bath at selected temperatures. This method results in almost instantaneous wetheating of the insects, which may not be similar to hot air treatments, or even water dip treatment of

actual commodities where air pockets may exist around the insect. Direct contact of insects with water at elevated temperature may lead to drowning of the insects (Hansen and Sharp, 1998, 2000). Other researchers used an indirect immersion method. For example, Neven (1994) and Yokoyama et al. (1991) placed codling moth larvae in glass vials (7.5 and 15 ml, respectively) before heating in water baths. With these methods, the actual heating rates of insects decrease with time due to the decreasing temperature difference between the heating medium and the insects.

Another common method to determine thermal mortality characteristics of insects is to heat infested commodities to a selected temperature and hold the commodities for pre-determined times. Information from those experiments is only applicable to the tested products and the specific test conditions. It is, thus, difficult to extract true information for the susceptibility of the insects to heat, because the experimental data are confounded with the effects of heat transfer, which in turn is dependent on the heating methods, thermal properties and size of the products, as well as position of insects in the host material. Furthermore, a large amount of the reported data is in the form of LT<sub>95</sub> (min) or LT<sub>99</sub> (min), which demonstrates relative thermal susceptibility of insects, but cannot be used directly in combination with fundamental heat transfer theory to develop thermal treatment protocols.

# 3.5. Thermal block system and thermal kinetic data for codling moth larvae

An experimental heating block system was developed at Washington State University (WSU) to

| Table 1 |           |    |         |      |               |
|---------|-----------|----|---------|------|---------------|
| Thermal | tolerance | of | codling | moth | fifth-instars |

| Temperature (°C) | Medium and heating methods                                 | 100% mortality<br>(min) | Source                    |
|------------------|--|-------------------------|---------------------------|
| 45               | 15 ml vials (constant temperature water bath) <sup>c</sup> | >55                     | Yokoyama et al. (1991)    |
|                  | 0.48 L jars (walk-in chamber) <sup>d</sup>                 | 1440 <sup>a</sup>       | Soderstrom et al. (1996)  |
|                  | Infested cherries <sup>e</sup>                             | 60                      | Neven and Rehfield (1995) |
|                  | Infested cherries (in test chamber)f                       | 124 <sup>b</sup>        | Neven and Mitcham (1996)  |
| 46               | 15 ml vials (water bath) <sup>c</sup>                      | > 55                    | Yokoyama et al. (1991)    |
|                  | In artificial diet and thinning apples <sup>g</sup>        | 480                     | Neven and Rehfield (1995) |
|                  | 7.5 ml vials (temperature ramp water bath) <sup>h</sup>    | 97                      | Neven et al. (1996)       |
| 47               | 15 ml vials (constant temperature water bath) <sup>c</sup> | 25                      | Yokoyama et al. (1991)    |
| 48               | 15 ml vials (constant temperature water bath) <sup>c</sup> | 10                      | Yokoyama et al. (1991)    |
|                  | 7.5 ml vials (temperature ramp water bath) <sup>h</sup>    | 177                     | Neven et al. (1996)       |
|                  | 7.5 ml vials (temperatue ramp water bath) <sup>i</sup>     | 73                      | Neven et al. (1996)       |
| 49               | 15 ml vials (constant temperature water bath) <sup>c</sup> | 7.5                     | Yokoyama et al. (1991)    |
| 50               | 15 ml vials (constant temperature water bath) <sup>c</sup> | 4                       | Yokoyama et al. (1991)    |
| 51               | 15 ml vials (constant temperature water bath) <sup>c</sup> | 3                       | Yokoyama et al. (1991)    |

<sup>&</sup>lt;sup>a</sup> Time to achieve a probit 9.

<sup>&</sup>lt;sup>b</sup> Time to achieve LT<sub>99</sub>.

<sup>&</sup>lt;sup>c</sup> Heating rate not reported, 44–112 5th instars each treatment.

d Heating rate not reported, 100-125 diapausing larvae each treatment.

<sup>&</sup>lt;sup>e</sup> Quoted by Neven and Rehfield (1995) as unpublished data for 5th instar at heating rate, 4.4°C/min, number of insects and heating method not reported.

f Heating rate of the air in test chambers, 1.66°C/min, 200 5th instars each treatment.

<sup>&</sup>lt;sup>g</sup> Heating rate of moist hot air in a test chamber, 4°C/h, 28 days of cold storage at 0-2°C, 240 larvae each treatment.

<sup>&</sup>lt;sup>h</sup> The temperature ramp rate was set to simulate heating of apples in moist air in a test chamber, heating rates not reported, 500 5th instars each treatment.

<sup>&</sup>lt;sup>i</sup> The temperature ramp rate was set to simulate heating of apples in vapour forced air in a test chamber, heating rates not reported, 500 5th instars each treatment.

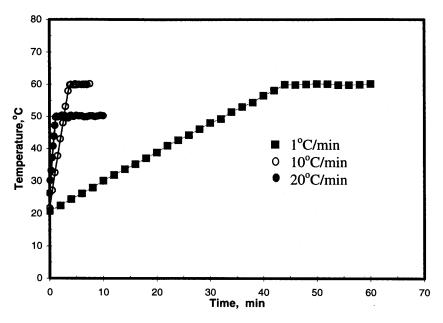


Fig. 1. Temperature of the WSU heating block at different heating rates (Ikediala et al., 2000a).

study thermal susceptibility characteristics of insects. The heating system consists of two metal blocks with heating pads capable of producing from 0.2 to 28°C/min heating rates. An aluminium plate with four square openings is sandwiched between the two metal blocks to form compartments, each capable of holding several insects. The thickness of the sandwiched plates can be adjusted for different insect size to ensure direct contact between insects and metal blocks during heating tests. With a solid state relay and a data acquisition system, the block temperatures can be controlled to increase to the desired set point within  $\pm 0.3$ °C at selected heating rates (Fig. 1). The data acquisition software displays the real time data to enable monitoring and confirmation of treatment heating rate, set point temperature, and holding time. An engineering analysis of the heating block system shows that for fifth instar codling moth (12-20 mm long and 2.0-2.5 mm wide), the temperature differential (lag) between the block and the centre of insect body was less than 0.2°C and 0.6°C at the 5°C/ min and 20°C/min heating rates, respectively, 30 seconds after reaching the selected holding temperature. Detailed information on this system is reported elsewhere (Ikediala et al., 2000a).

With the WSU test system, the effect of heating rate on thermal lethality and the thermal death time (TDT) kinetics of insects can be determined, since heating rate is believed to have a significant effect on insect metabolism and on the physiological adjustment to the heat treatment (Evans, 1986; Neven, 1998a,b). Neven (1998a) reported that at slow heating rates, insect larvae must be exposed to the final temperature for a longer period of time to achieve 95% mortality. Knowledge of the effect of a fast heating rate is needed when developing RF and microwave treatments.

The WSU system was used to subject fifth instar codling moth to various combinations of temperature—time treatments to study the thermal mortality characteristics (Fig. 2). The dashed line (TDT curve) in Fig. 2 defines the boundary for 100% mortality in the upper right region for 200 insects. This line presents a semi-logarithmic relationship between minimum time and temperature combinations to achieve more than 2.3 log reduction (>2.3 D reduction) of fifth instar codling moth larvae. A probit 9 process is equivalent to a 4.5 D reduction [log(1 – 0.999968)  $\approx$  4.5]. That is, doubling the treatment time on the TDT curve in Fig. 2 for each temperature (2 × 2.3D = 4.6D)

would lead to a better than probit 9 efficacy. For example, based on Fig. 2, we expect that holding fifth instar codling moth larvae at 52°C for 4 min or 54°C for 1.2 min would lead to more than probit 9 efficacy.

The z value for the fifth-instar codling moth was determined from the graph in Fig. 2 to be 4°C, and the activation energy ( $E_a$ ) was estimated to be 499 kJ/mole based on Eq. (6). This activation energy is slightly greater than that for thermal inactivation of pathogenic microbial spores ( $E_a = 210-350$  kJ/mole; Lund, 1977), and much greater than for softening of texture or lipid oxidation (e.g.,  $E_a = 100$  kJ/mole or z = 33°C; Rao and Lund, 1986; Taoukis et al., 1997). Therefore, codling moth larvae are much more sensitive to increase in treatment temperature than is fruit quality.

Similar to an insect thermal death time curve, a quality curve may also be plotted that defines a region in which the quality of the commodity will not suffer when it is treated below different combinations of temperature and time. Because the

activation energy for quality changes is generally smaller than that of insect mortality (e.g.  $E_{\rm a}=100$  kJ/mole for texture softening, Rao and Lund, 1986; Taoukis et al., 1997), the slope of this quality curve should be smaller than that of the insect mortality curve (Fig. 3). The exact location and the slope of this curve depends on the commodity. The overlap between the lower region of the quality curve and upper region of the insect mortality curve defines the potential for developing thermal quarantine treatments. This region highlights the need to develop high-temperature-short-time (HTST) processes to provide desired lethality to insects yet avoid adverse impact on product quality.

#### 4. High-temperature-short-time treatments

The concept of a high-temperature-short-time (HTST) process is used extensively in the food industry to minimise thermal degradation of processed foods. For example, HTST processes are

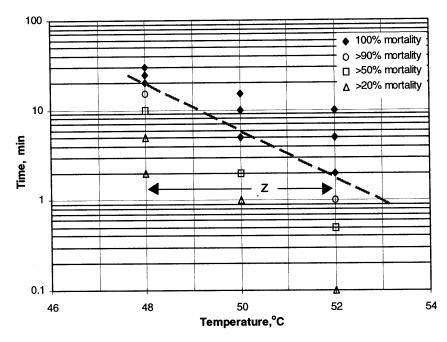


Fig. 2. Thermal-death-time (TDT) curve for 5th instar codling moth larvae at  $20^{\circ}$ C/min heating rate. Each point represents 200 larvae. The dashed line defines the boundary for 100% mortality in the upper right region (from Ikediala et al., 2000a). z value indicates the temperature increase corresponding to a log reduction in time.

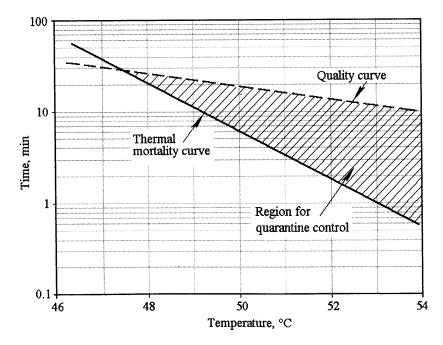


Fig. 3. Hypothetical commodity quality curve and insect mortality curve that define possible region for developing high-temperature-short-time quarantine thermal treatments.

used in commercial pasteurisation and sterilisation to kill pathogenic micro-organisms in foods and result in better quality retention (Lund, 1977; Ohlsson, 1980). A similar concept can be used for the development of thermal treatments to control insect pests. The potential for successful development of an effective HTST thermal treatment to control insects and to minimise detrimental thermal effects on the host products relies on the differences in the temperature-dependent kinetics for insect mortality and quality degradation. The activation energy for softening of plant tissues and lipid oxidation is about 100 kJ/mole (or z = 33°C, Rao and Lund, 1986; Taoukis et al., 1997). Based on Eq. (4) and the value of  $E_a$ , it was estimated that every 10°C increase in treatment temperature would lead to a 3-fold increase in the rate of quality loss. For mortality of codling moth, on the other hand, the activation energy is estimated to be 499 kJ/mole. Using this value of  $E_{\rm a}$  in Eq. (4), we expect that a temperature increase from 44 to 54°C will increase the thermal lethal effect 300-fold. That is, 1/300 of the time is needed at 54°C as compared to at 44°C to achieve

the same lethality. Thus, a thermal treatment at  $54^{\circ}$ C equivalent in insect thermal mortality as  $44^{\circ}$ C would reduce quality loss by a factor of 100 (= 300/3). For codling moth, an effective HTST treatment may be developed at temperatures between 50 and  $54^{\circ}$ C.

Based on experiments conducted at 50 and 51°C, Yokoyama et al. (1991) also concluded that high temperatures and short treatment times might be suitable for control of codling moth in stone fruit and nuts. The potential for HTST processes will depend on how thermal energy is delivered to the insects in the host materials.

# 4.1. Conventional thermal treatments for HTST processes

Thermal treatment methods using hot water, vapor or hot air have been investigated extensively as alternatives to MeBr fumigation (Sharp et al., 1991; Yokoyama et al., 1991; Moffitt et al., 1992; Neven, 1994; Neven and Rehfield, 1995). Varying degrees of efficacy have been reported using different thermal treatments alone or in

combination with cold or controlled atmosphere storage conditions (Toba and Moffitt, 1991; Sharp, 1993; Neven and Mitcham, 1996; Neven et al., 1996; Soderstrom et al., 1996). Larvae usually bore into fruits or nuts in shells to feed on the centre flesh, seeds or kernels. Thus, the commodity centre must be heated to desired temperatures during the thermal treatment. However, a common difficulty with conventional heating methods is the slow rates of heat transfer that often led to long treatment times (Hansen, 1992). Prolonged heating may be detrimental to the quality of treated products, and may not be practical in industrial applications.

The conventional heating consists of convective heat transfer from the heating medium to the fruit surface and conductive heat transfer from the surface to the core. A clear understanding of those heat transfer processes is necessary in order to appreciate the limitations of conventional heat methods as HTST processes.

For conventional heating, air or water is the source of thermal energy. Thermal energy is transferred from the heating medium to the fruit surface  $(r=r_0)$  by convection as described by the following equation:

$$-K\frac{\partial T}{\partial r}\Big|_{r=r_0} = h[T(r_0,t) - T_e]$$
Heat flow from fluid

(Convection to heat fruit surface)

(9)

where K is thermal conductivity of fruit (W/m°C), h is the surface heat transfer coefficient (W/m²°C), r is the radial coordinate (m),  $r_0$  is the fruit radius, T is the fruit temperature (°C), and  $T_e$  is the fluid temperature (°C). According to the above relationship, heat flow from the medium to the fruit surface is proportional to the surface heat transfer coefficient, h, and temperature difference between the medium and fruit surface. The value of h may vary from 6 W/m²°C in still air to 30 W/m²°C in moving air, to 1200–6000 W/m²°C in circulating water (Earle, 1983). Therefore, increasing air speed or using circulating water can increase thermal energy transfer at the fruit surface.

Once the thermal energy is transferred to the fruit surface, this energy is further moved into the fruit interior by conduction. For spherical fruits such as cherries, apples, and oranges, heating rate

at various locations within the fruit is a function of radial position (r) and treatment time (t). This function is governed by a general heat transfer equation in spheres:

$$\rho C_{\rm p} \frac{\partial T}{\partial t} = K \left( \frac{\partial^2 T}{\partial^2 r} + \frac{2}{r} \frac{\partial T}{\partial r} \right) + \underbrace{Q}_{\text{Heat ing rate}}$$
Heat conduction within fruit (10)

where  $C_{\rm p}$  is fruit specific heat capacity (J/kg°C), Q is additional heat generation within the fruits (W/m³), and  $\rho$  is fruit density (kg/m³). Q is zero when fruits are subjected only to conventional heat treatments and when heat generation due to respiration is neglible. Heat conduction within fruits, as represented by the first term on the right hand side of Eq. (10), is small due to a relatively small thermal conductivity value for fruits ( $K \approx 0.5 \text{ W/m°C}$ , as compared to 50--400 W/m°C for metals). As a result, the heating rate at the core of a fruit can be very small, especially for large fruits such as apples.

To demonstrate the influence of heating medium and fruit size on rate of heating in the core of fruits, computer simulation results are presented in Fig. 4 and Fig. 5 for apples and cherries, respectively. The simulation program was validated with analytical solutions and experimental results. The physical properties used for the simulation are from the literature (Incropera

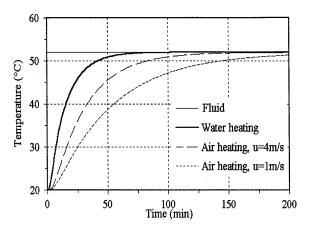


Fig. 4. Simulated core temperatures of an apple (d=8 cm) diameter) when heated by water or air (u in the legend represents air speed).

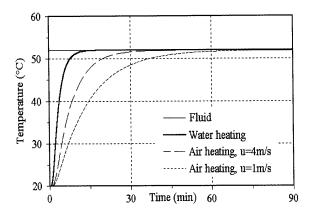


Fig. 5. Simulated core temperatures of a cherry (d=3 cm) diameter) when heated by water or air (u in legend represents air speed).

and DeWitt, 1996) and listed in Table 2. A representative fruit size was chosen for cherries and apples.

When heated in circulating air at 1 m/s and 52°C, the core temperatures of an apple of 8 cm diameter increased very slowly to approach the medium temperature. For example, it took about 143 min for the core temperature to increase from 20 to 50°C (Fig. 4). By increasing circulating air speed to 4 m/s, the time for the same core temperature rise was reduced to about 83 min. When heated in a circulating water bath at 52°C, the

Table 2
Thermal properties used in computer simulation for cherries and apples

| Parameters  | Apple | Cherry |
|---|-------|--------|
| Diameter d (cm)                                       | 8     | 3      |
| Density $\rho$ (kg/m <sup>3</sup> )                   | 840   | 1010   |
| Specific heat capacity $C_p$ (J/kg°C)                 | 3600  | 3300   |
| Thermal conductivity $K(W/m^{\circ}C)$                | 0.513 | 0.527  |
| Surface heat transfer coefficient $h$ $(W/m^{2o}C)^a$ |       |        |
| Air heating at air speed 1 m/s                        | 17    | 25     |
| Air heating at air speed 4 m/s                        | 39    | 57     |
| Water heating at water speed 1 m/s                    | 1911  | 2830   |

<sup>&</sup>lt;sup>a</sup> Estimated from the correlation for spheres in Dincer (1997).

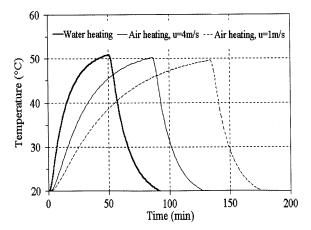


Fig. 6. Simulated core temperatures of an apple (d = 8 cm) by different heating methods to deliver the same lethal dose at the core (equivalent to 52°C for 4 min) followed by hydraulic cooling (water at 18°C and 1 m/s).

rate of fruit core temperature increase was significantly improved. Yet, it still took about 42 min for the core temperature to rise to 50°C. As the heating proceeds, the difference between the core temperature and the medium temperature becomes smaller and so does the heating rate at the fruit cores (Fig. 4).

For smaller fruits such as cherries, the heating is much faster. For the case demonstrated in Fig. 5, it took about 36 min for the core temperature of a cherry of 3 cm diameter to reach 50°C when heated in air at 52°C moving at 1 m/s. This heating time was reduced to 18 min when the air speed was increased to 4 m/s. When using circulating water, the time required to raise core temperature from 20 to 50°C was reduced to 4 min.

With the heating curves shown in Fig. 4 and Fig. 5, Eq. (7) was numerically integrated to determine appropriate treatment times to deliver the desired thermal mortality. Fig. 6 and Fig. 7 show the appropriate heating times to deliver mortality equivalent to a full exposure of 4 min at  $52^{\circ}$ C ( $M_{52^{\circ}$ C} = 4 min, a probit 9 process based on Fig. 2) at the centre of apples and cherries, respectively. The curves in Fig. 6 and Fig. 7 illustrate that water heating takes much less time than hot air heating to deliver the same thermal mortality to insect at the centre of the fruits. Fruit size also has a major influence on treatment times. Fig. 6

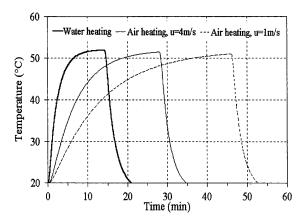


Fig. 7. Simulated core temperatures of the cherry (d = 3 cm) by different heating methods to deliver the same lethal dose at the core (equivalent to 52°C for 4 min) followed by hydraulic cooling (water at 18°C and 1 m/s).

and Fig. 7 also show temperature—time histories for cooling using circulating water at 18°C and 1 m/s.

Although the treatments represented in Fig. 6 and Fig. 7 have the same effect on insect mortality in fruits, these processes have vastly different impacts on fruit quality. The cumulative effect of the heating and cooling on the quality index  $Q_{52^{\circ}C}$  was estimated by numerically integrating Eq. (8) using time–temperature history data and a z value of 33°C. Since it is expected that the fruit surface will be heated most severely during heating, and the fruit core will be more affected during cooling, values of  $Q_{52^{\circ}C}$  (min) only for those locations are reported in Table 3. Based on the  $Q_{52^{\circ}C}$  values, it is clear that a water-heating treatment should have the least impact on fruit quality among the three heating methods. But the

water treatment may still severely damage fruit at the surface. For apples, the effect of water heating on the fruit surface is equivalent to directly exposing the tissues to 52°C for 43.9 min. The cooling contributes 11.1 min to the total  $Q_{52^{\circ}C}$  value. For cherries,  $Q_{52^{\circ}C}$  is 11.1 min for the heating and 1.9 min for cooling when using the hot water heating method. These results indicate that the heating period had a major impact on fruit quality.

The use of circulating water as a heating medium represents a practical best-case scenario in terms of delivering thermal energy to the fruit surface with conventional heating methods. But once the energy is delivered to the surface, little can be done with the heat conduction within the fruits. Therefore in those processes, by the time the treatment delivers sufficient thermal energy to the fruit centre to kill infesting insects, the fruit surfaces are exposed to high temperatures for an extended period, which might cause severe and visible thermal damage.

## 4.2. Thermal treatments based on electromagnetic energy

Radio frequency (RF) and microwave energies interact with the commodity's interior to quickly raise the centre temperature. Electromagnetic energies are, therefore, more compatible with HTST processing than conventional heating methods. The RF or microwave treatments can be designed as a continuous flow process to allow processing of large quantities of products in a short period of time, a critical requirement for an alternative treatment useful to control insects in fruits or nuts destined for domestic and international markets.

Table 3 Impact of treatment on fruit quality [ $Q_{52^{\circ}C}$  values (min)]. The Q values were calculated for the fruit surface during heating and for the fruit centre during cooling based on a hypothetical z value of 33°C. All treatments deliver equal lethal dose (equivalent to 52°C for 4 min) to 5th instar codling moths at the center of fruits

| Fruits | Heating methods |             |             | Subsequent cooling (using water at 18°C) that followed heating methods |             |             |
|--------|-----------------|-------------|-------------|--|-------------|-------------|
|        | Water           | Air (4 m/s) | Air (1 m/s) | Water  | Air (4 m/s) | Air (1 m/s) |
| Apple  | 43.9            | 54.4        | 71.5        | 11.1   | 10.9        | 10.6        |
| Cherry | 12.0            | 17.8        | 25.4        | 1.9  | 1.8         | 1.8         |

RF and microwave energies (13–2450 MHz) are used successfully in the food processing industry. Treatments using electromagnetic energies at RF and microwave frequencies leave no chemical residues on products and have minimal impact on the environment.

Researchers have explored the feasibility of using electromagnetic energies to disinfest insect pests. Two studies carried out in Japan demonstrated that 100% of tobacco moth larvae, Ephestia elutella (Hubner) (Lepidoptera: Pyralidae), and cigarette beetles. Lasioderma serricorne (F.) (Coleoptera: Anobiidae), were killed when heated by 2450 MHz microwaves to 55°C for 30 s (Hirose et al., 1970a,b). A recent study demonstrated the possibility of using 2450 MHz microwaves to destroy woodworms by heating the larvae to 52-53°C for less than 3 min (Andreuccetti et al., 1994). Hallman and Sharp (1994) summarized research on the application of RF and microwave treatments to kill selected pests in many postharvest food crops and recommended exploration of synergistic effects of radio frequency and conventional heating methods. Nelson (1996) summarized more than five decades of research on the susceptibility of various stored grain insect species to RF and microwave treatments and reiterated that RF frequencies between 10 and 100 MHz should preferentially heat insects in grains.

Early research efforts using RF and microwave energy for insect control were, however, generally hindered by comparatively inexpensive chemical fumigation. Nelson (1972) appropriately stated "the potential for application of electromagnetic energy to insect control problems hinged on economical factors and continued acceptability of current practical methods." The cost of RF and microwave equipment has since dropped to a level comparable to conventional heating equipment, and the need for alternatives to chemical fumigation is becoming inevitable.

Radio frequency and microwave treatments involve the application of electromagnetic energy at 10–30000 MHz (Orfeuil, 1987). Because of the congested bands of RF and microwaves already being used for communication purposes, the U.S. Federal Communication Commissions allocated five frequencies for industrial, scientific, and medi-

cal applications, 13.56, 27.12 and 40.68 MHz in the RF range and 915 and 2450 MHz in the microwave range.

A unique feature of RF or microwave treatments is that RF and microwave energies are directly coupled to a dielectric material, such as most agricultural commodities, to generate heat Q (in Eq. (10)) within the material. This can significantly increase the heating rates and reduce heating time. The magnitude of the heat generation is proportional to the loss factor,  $\varepsilon''$ , at a given frequency and electric field:

$$Q = 5.563 \times 10^{-11} f \varepsilon'' E^2 \tag{11}$$

where f is frequency (Hz), and E is the electric field intensity (V/m).

For a fast heating process in which heat conduction is relatively small, Eq. (10) reduces to:

$$\Delta T = \frac{Q}{\rho C_{\rm p}} \Delta t \tag{12}$$

That is, the temperature increases linearly with treatment time. Adjusting RF or microwave power can control the rate of the increase. In appropriately designed RF or microwave systems, the fruit core can be linearly heated to a desired temperature in a short time. For example, using Eq. (8), it is estimated that ramping from 20 to 52°C in 3 min using a RF process contributes only 1.2 min to  $Q_{\rm 52^{\circ}C}$  value. The shortened heating time will significantly reduce the total thermal impact on fruit quality. This is a major advantage of RF or microwave processes over conventional heating methods. The major challenges in developing effective RF or microwave processes are to provide uniform heating among fruits and to develop means to monitor and control the end product temperature.

# 4.2.1. Dielectric properties of insects, fruits and nuts

The dielectric properties data for the targeted insects and host fruits or nuts provide general guidance for selecting optimal ranges of frequency and the design of the thickness of the treated bed for uniform RF and microwave treatments. The dielectric properties of codling moth and selected host materials were measured at WSU using a

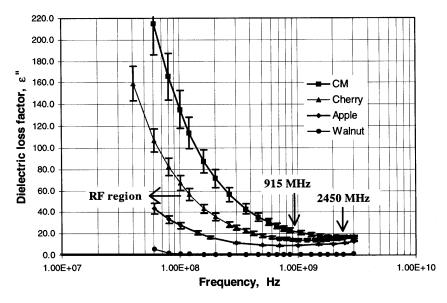


Fig. 8. Dielectric loss factors of codling moth larva (CM) cherry, apple and walnut at RF and microwave frequencies.

coaxial probe technique (HP Dielectric Probe Kit, Model 85070B, Hewlett Packard Corp., Santa Clara, CA). Detailed information on dielectric properties of codling moth larvae and some host material is presented elsewhere (Ikediala et al., 2000b).

The results of  $\varepsilon''$  for codling moth larvae and selected host materials such as cherry, apple and walnuts are summarized in Fig. 8. The dielectric loss factor  $\varepsilon''$  of codling moth larvae is greater than that of the tested host materials (Fig. 8), particularly in the RF frequency range. According to Eq. (11), a larger  $\varepsilon''$  value results in more generation of thermal energy at a given frequency, f, and electric field intensity, E. Therefore, codling moth may absorb much more energy than the host materials, especially walnuts, leading to possible preferential heating of the larvae while maintaining product quality.

#### 4.2.2. Penetration depth

The penetration depth (dp, m) of RF and microwaves in a lossy material increases with decreasing frequency as described by (Von Hippel, 1954):

$$dp = \frac{c}{2\pi f \sqrt{2\varepsilon' \left[\sqrt{1 + \left(\frac{\varepsilon''}{\varepsilon'}\right)^2 - 1}\right]}}$$
(13)

where c is the speed of light in free space  $(3 \times 10^8)$ m/s) and  $\varepsilon'$  is the relative dielectric constant of the treated material. For example, the penetration depth of microwaves in apples and potatoes at room temperature increases from 1.2 cm and 0.9 cm to 4.3 cm and 2.2 cm when the frequency decreases from 2450 to 915 MHz (Tang, 2000). Limited penetration depth leads to non-uniform heating in large objects, and may in part explain the disappointing results reported by Seo et al. (1970) and Hayes et al. (1984) in which 2450 MHz microwaves were used to treat mangoes and papaya. Microwaves at 2450 MHz, however, may provide much deeper penetration in nuts and dried fruits due to low moisture content and, thus, may provide more uniform heating.

In addition to 2450 MHz microwaves, longer wavelength microwave (915 MHz) and RF energies at 27.12 MHz can be used to take advantage of deeper penetration depths and potential selective heating of insects in the host material.

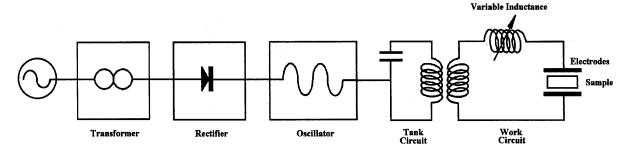


Fig. 9. Schematic view of a radio frequency heating system.

### 5. Thermal treatments based on RF energy

A HTST thermal treatment was developed that used RF energy to control codling moth in inshell walnuts. A 6 kW, 27 MHz pilot-scale RF system (COMBI 6-S, Strayfield International Limited, Wokingham, UK) (Fig. 9) was used for the tests. 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. The oscillator converted direct current into RF energy at 27 MHz. 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.

RF and microwave treatments have particular advantages over conventional hot air heating in treating walnuts in shells. The shell and the air spaces in the shell act as layers of insulation and slow down heat transfer with conventional heating methods, while electromagnetic energy directly interacts with the kernel inside the shell to generate heat. Fig. 10 shows typical temperature-time profiles for in-shell walnut kernel during hot air treatment (53°C at 1 m/s air velocity) and the RF treatment. With hot air treatment, the heating rates decreased as the product temperature approached the medium temperature, resulting in prolonged heating time of more than 40 min (Fig. 10). The kernel temperature lagged significantly behind that of the hot air temperature, increasing very slowly with treatment time once it reached within about 10°C of the air temperature. The small thermal conductivity of the porous walnut shell and the in-shell void hindered the transfer of thermal energy from the hot air outside of the walnut shell.

With RF treatment, walnut kernel temperature increased linearly with process time (Fig. 10), as predicted by Eq. (12). Increasing RF power input can further increase the heating rate. We used a 27.12 MHz pilot RF system to treat in-shell walnuts infested with 3rd instar codling moth larvae, using a RF specific power of 0.27 kW/kg walnuts. After 2 and 3 min treatments, the mean kernel temperature reached 43 and 53°C, respectively (Table 4). According to Fig. 2, 5 min holding at 53°C should lead to more than probit 9 efficacy.

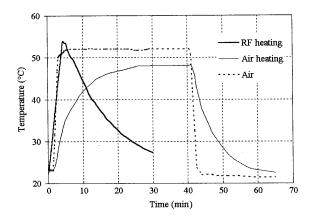


Fig. 10. Typical heating and cooling curves for in-shell walnut kernels when subjected to forced hot air (air temperature, 53°C; air velocity, 1 m/s) and radio frequency (RF) treatments.

Table 4 Mortality results of codling moth larvae in walnuts (20 walnuts per run and five replicates)

| Mortality of 3rd and<br>4th-instars in walnuts (%) |
|--|
| 0  |
| 78.6   |
| 100  |
|  |

One hundred in-shell walnuts, infested at one larva per walnut, were heated by RF energy in the pilot-system to 43 or 53°C, and held in air for 5 min to impart desired lethality to the codling moth before air cooling at 4°C. The results of the mortality tests are shown in Table 4. The 2 and 3 min RF treatments resulted in 78.6 and 100% mortality, respectively (Table 4).

The standard deviations in the final temperatures of the walnut kernels suggested that some walnuts were not heated to a level that would cause 100% mortality after 5 min holding time (e.g.  $\sim 48^{\circ}$ C after 3 min RF treatment). However, the actual kill at these temperatures far exceeded the mortality rate estimated from the TDT curves (Fig. 2). We speculate that this might have been the result of possible preferential heating in codling moth larvae due to their much larger dielectric loss factors at the RF frequency, as compared to that of walnuts (Fig. 10). Nelson and Payne (1982) reported 100% kill in the pecan weevil, Curculio caryae (Horn), with 40 MHz RF treatment of shelled and in-shell pecans when the kernel temperature was raised to 53°C in 15 s. A hot air treatment at 45°C, on the other hand, would require approximately 24 h to achieve a probit 9 efficacy (Soderstrom et al., 1996).

An accelerated shelf-life study (ASLT) was conducted to determine effect of the short time RF treatment (53°C final temperature and 5 min holding time) on walnut quality during long-term storage. Detailed information on the theoretical basis for ASLT can be found in Taoukis et al. (1997). The RF-treated walnuts were held at 35°C for 10 and 20 days, equivalent to 1 and 2 years storage at 4°C. The measured peroxide value and fatty acid values (Table 5) suggest that the RF treatments did not significantly affect walnut quality. It is, therefore, possible to develop a HTST continuous process for walnuts and other similar commodities to satisfy the quarantine security requirements of the industry against codling moth.

#### 6. Need for furture research

Further research is needed to determine the most heat resistant life-stage of target insect pests of economic importance and obtain the intrinsic kinetics information of this life-stage for developing treatment protocols. There is also need for studies to obtain fundamental kinetic information regarding time-temperature effects on quality of commodities. This information would allow us to establish a quality curve for each commodity (e.g. in Fig. 3) and for calculating the z value or activation energy for use in Eq. (8). Because of complicated nature of thermal degradation, there may exist different z value for different temperature ranges. Nevertheless, the kinetic information for insect mortality and commodity quality can be used in Eq. (7) and Eq. (8) in combination with

Table 5 Quality characteristics of in-shell walnuts treated by radio frequency energy (3 min)

| Storage time at 35°C (day) | Peroxide value (m | neq/kg)         | Fatty acid (%)  |                 |
|----------------------------|-------------------|-----------------|-----------------|-----------------|
|                            | Control           | RF treated      | Control         | RF treated      |
| 0                          | $0.26 \pm 0.04$   | $0.28 \pm 0.04$ | $0.08 \pm 0.01$ | $0.08 \pm 0.01$ |
| 10                         | $0.49 \pm 0.05$   | $0.51 \pm 0.05$ | $0.08 \pm 0.01$ | $0.09 \pm 0.01$ |
| 20                         | $0.93 \pm 0.05$   | $0.98 \pm 0.05$ | $0.10 \pm 0.01$ | $0.08 \pm 0.01$ |

temperature-time history of different treatment methods to select optimized process conditions, which will provide adequate thermal mortality of target insects and minimum impact on commodity quality. Validation tests can then be conducted with infested commodities to confirm the efficacy of the selected treatment protocols. An advantage of this approach will be two-fold: (1) the fundamental kinetic information regarding insect mortality and quality should help gain general insights into the physiological nature of the thermal death of the insect and the quality losses of commodities. Although different insects or commodities may have different tolerance to heat, they should share similar underlying mechanisms. Kinetic information on one specific insect or commodity may not be directly applicable to others, but the fundamental trend should be similar; (2) The tests for insect mortality and quality can be conducted independent of the final treatment protocols and the procedures can be standardised to allow laboratories in different nations or regions to collaborate on similar projects. Such a systematic approach should reduce redundant experimentation.

#### 7. Summary

Fundamental kinetic studies for insect pests can be combined very effectively with time-temperature history data to evaluate the efficacy of a thermal treatment and assess its impact on fruit quality. This approach can be used to develop an optimized treatment that delivers required mortality to insect, yet has minimum impact on fruit quality. Our kinetic data for codling moth suggest the possibilities of developing a high-temperatureshort time treatment as an optimized treatment protocol. But this protocol may not be possible using conventional hot water or hot air heating methods for heat-sensitive fruits or nuts, due to slow heat conduction within those commodities. Our results suggest that radio frequency heating may be effective in high-temperature-short time control of codling moths in in-shell walnuts.

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