



## Treatment design of radio frequency heating based on insect control and product quality

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### ABSTRACT

Non-uniform heating caused by different size, geometry, and properties of agricultural commodities is a major challenge that needs to be addressed in developing industrial-scale radio frequency (RF) treatments for postharvest phytosanitary and quarantine applications to dry grains or nuts. A mathematical model based on normal distributions of product temperatures and taking into account insect thermal mortality and product quality was developed to predict treatment temperature–time ranges as a function of number of mixing. To demonstrate its applications, this model considered two boundary conditions: complete mortality of fifth-instar navel orangeworm with temperature–time relationships (46, 48, 50, 52, and 54 °C for 140, 50, 15, 6, and 1 min, respectively), and maximum temperature tolerance relationship for walnut quality (53, 69, and 75 °C for 240, 5, and 1 min, respectively). This model was validated by experimental data of the uniformity index and applied to walnut, soybean, lentil, and wheat, each with its own unique RF heating characteristics. The results showed that the operation ranges (temperature–time combinations) for effective control of pests without causing adverse quality changes expanded with increasing mixing number and the improved heating uniformity. This study suggested more flexibility in developing RF treatments of pest control for small-size crops with better heating uniformity, as compared to large-size crops.

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### 1. Introduction

Agriculture commodities, such as wheat, soybean, lentil, and walnut, are important contributors to exported goods from the United States. Agricultural commodities are, however, natural carriers of exotic insect pests. These pests can cause major local economic losses (Mumford, 2002). To reduce the risk of introducing pests, importing countries or regions impose quarantine or phytosanitary requirements for commodities that host targeted pests. Methyl bromide is an important quarantine and phytosanitary treatment method, but it is a highly toxic gas and listed as an ozone-depleting chemical under the Montreal Protocol of 1992 (UNEP, 1992). Recent regulatory actions have limited its use to quarantine treatments, requiring industries to apply for yearly Critical Use Exemptions for phytosanitary applications through a complicated procedure, thus creating an urgent need to find environmentally friendly and effective alternatives.

Thermal treatments can be non-chemical alternatives for postharvest insect control in agricultural commodities. Radio frequency (RF) energy, proposed as a novel form of delivering thermal energy to commodities, has been widely used as an advanced thermal technology in food processing over conventional hot air and water heating (Tang et al., 2000). Research on applications of RF heating in disinfesting agricultural products has been hindered by the low price of methyl bromide fumigation practices and a lack of heating uniformity in the RF-treated products (Frings, 1952; Hallman and Sharp, 1994; Nelson, 1996). Recent studies on thermal death kinetics for insects and on dielectric properties of the insects and the related agricultural materials (Wang et al., 2002a,b, 2003, 2005; Johnson et al., 2003, 2004) has made it possible for the development of laboratory and industrial scale RF pest control treatments for walnuts with acceptable product quality (Wang et al., 2001, 2002c, 2006, 2007a,b; Birla et al., 2004; Mitcham et al., 2004).

Since dried materials are more heat tolerant than fresh produce, RF treatments hold greater potential in disinfesting wheat, soybean, lentil, and walnut. Heating non-uniformity is a major problem in RF treatments, which would result in either insect survival or product damage (Birla et al., 2004). Mixing can reduce commodity tem-

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perature variations in RF treatments as it eliminates the position effect in a container caused by non-uniform electrical field in RF systems (Wang et al., 2005). A mathematical model based on normal temperature distribution of RF-treated walnuts has been used to determine the intermittent mixing number to meet the required insect mortality. The concept of heating uniformity index was used in this model to quantify the severity of non-uniformity problem (Wang et al., 2007a,b). However, the selection of the product mean temperature as a key operation parameter in that model was based only on insect mortality, with no consideration for product quality. It is desirable to develop a model that takes into account both insect thermal mortality and product quality. It is also important to extend our research from walnuts into similar low-moisture and small particle crops, such as soybeans, lentils, and wheat.

The objectives of this study were to determine RF heating characteristics of walnut, soybean, lentil, and wheat, to develop a mathematical model that takes into account insect mortality and product quality in providing time–temperature parameters for the four crops, to validate the model with experimental data of the uniformity index, and to analyse the influence of mixing numbers and required security levels on time–temperature regimes for the four crops.

## 2. Materials and methods

### 2.1. Temperature distributions after RF treatments

Based on a previous study (Wang et al., 2005), walnut product temperatures after RF treatments follow a normal temperature distribution. To further confirm if the normal distribution was suitable for the simulation model development, four different commodities, namely wheat, soybean, lentil, and walnut, were heated in a 12 kW, 27 MHz pilot-scale RF system (Strayfield Fastran with E-200, Strayfield International Limited, Wokingham, UK). These samples were purchased from local grocery stores and stored at room temperature before tests. Samples of 11.3 kg in weight were held in a plastic container (43.2 cm × 34.3 cm × 12.7 cm) and heated between the two parallel plate electrodes (Fig. 1). The gap between the electrodes was fixed at 165 cm to have a suitable heating rate with a RF power of 6–7 kW. After a predetermined heating time, the container was immediately taken out of the RF system for temperature measurement. Two different sets of experiments were conducted, including RF heating without mixing for 0, 1, 2, and 3 min to determine the rise in mean and standard deviation of sample temperatures, and RF heating with two intermittent mixings to determine the sample temperature distribution. For mixing tests, the RF-treated samples were placed in a large container (55 cm × 40 cm × 14 cm) and mixed manually for 20 s. After the mixing was complete, the samples were placed back into the RF cavity for further heating under the same conditions.

The sample surface temperature in the container was measured by an infrared camera (Thermal CAM™ SC-3000, N. Billerica, MA)

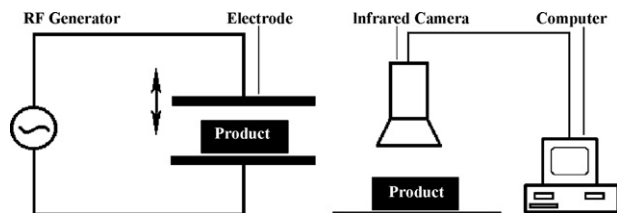


Fig. 1. Schematic view of the 12 kW, 27.12 MHz radio frequency (RF) unit showing the plate electrodes and the surface temperature measurement using a thermal imaging camera.

with an accuracy of  $\pm 2^\circ\text{C}$  before and after RF treatments (Fig. 1). After calibrations, each thermal image showing the temperature distributions could rapidly determine the relatively accurate heating uniformity. Each measurement took about 3 s. In each of the thermal images, 45,056 individual temperature data were collected over the container and used for statistical analyses. The probability density frequency (PDF) versus the sample temperatures was plotted to determine the temperature distribution. The left and right tail probabilities between the experimental temperature distribution after RF heating and the associated normal distribution were compared.

### 2.2. Temperature limits for insect control and product quality

To determine the optimal mixing number in the RF treatments, appropriate temperature limits should be selected to meet the requirements for both the insect control and crop quality. Different insect species and crop varieties have different tolerances to heat. Thermotolerance of insects can be defined by thermal-death-time (TDT) curves previously determined for four economically important insect species at different life stages. The minimum exposure time required to completely kill test insects at a lethal temperature followed a semi-logarithmic relationship (Wang et al., 2002a,b; Johnson et al., 2003, 2004):

$$\log_{10}(\tau) = c_1 - c_2L \quad (1)$$

where  $\tau$  is the exposure time (min),  $c_1$  and  $c_2$  are positive constants that depend upon insect species, life stage, and pretreatment condition, and  $L$  is the lethal temperature ( $^\circ\text{C}$ ). Likewise, if the exposure time is set at  $\tau$ , the minimum lethal temperature is given by:

$$L = \frac{c_1 - \log_{10}(\tau)}{c_2} \quad (2)$$

To maintain the crop quality and shelf-life, the maximum temperature and time quality curve should be determined. The quality curve is more difficult to define than the insect mortality curve, because each quality attribute may react differently to different temperature ranges. If any attribute becomes unacceptable to the consumer, the treated crop loses its market value. Based on some discrete experimental data for major crop quality attributes, a negative logarithmic relationship similar to that for insect mortality can be used (Lau et al., 2000):

$$\log_{10}(\tau) = c_3 - c_4D \quad (3)$$

where  $c_3$  and  $c_4$  are positive constants depending upon the crop types, and  $D$  is the maximum temperature before crop quality deteriorates ( $^\circ\text{C}$ ). If the same exposure time as for insect control is set at  $\tau$ , the maximum temperature for the crop to maintain a good quality is given by:

$$D = \frac{c_3 - \log_{10}(\tau)}{c_4} \quad (4)$$

Generally,  $D$  is larger than  $L$  for most thermally disinfesting crops, especially for low-moisture crops such as walnut, lentil, soybean, and wheat because the insects were more susceptible to the increased temperature than crops (Wang et al., 2002a). For the same exposure time, the large gap between  $D$  and  $L$  makes it easy to achieve the complete insect control and maintain good crop quality.

### 2.3. Model development

The assumptions similar to a previous study (Wang et al., 2005) were used in the current model development, namely RF treatments consisted of  $n+1$  number of RF units with  $n$  number of mixings in between with negligible heat loss during crop mixing,

and the mixing of crops only randomly changed their positions in the container without reducing their temperature. During the short exposure time ( $\leq 5$  min), the mean and standard deviation values of product temperatures remained the same.

It is observed from previous research that the standard deviation ( $\sigma$ , °C) for the temperature of RF-treated walnuts increases linearly with the increase of the mean temperature ( $\mu$ , °C) (Wang et al., 2005). The ratio of the increase in standard deviation to the unit increase in the mean temperature in a specific RF unit is defined as the uniformity index ( $\lambda = \Delta\sigma/\Delta\mu$ ). This parameter reflects the degree of heating uniformity for a fixed RF configuration and a given load.

Assuming that the initial product temperature is  $\mu_0$  (°C), and that the initial standard deviation is negligible, as the mean temperature increases from  $\mu_0$  to  $\mu_T$  (°C) with  $n$  mixing times during the heating process in  $n+1$  RF units, the final standard deviation (°C) increases from 0 to  $\sigma_T$  according to:

$$\sigma_T = \lambda_n(\mu_T - \mu_0) = \frac{\lambda(\mu_T - \mu_0)}{\sqrt{n+1}} \quad (5)$$

where  $\lambda$  and  $\lambda_n$  are the uniformity indexes before and after  $n$  intermittent mixings. To ensure the insect mortality and maintain desired crop quality, the following probability ( $P$ ) requirements on the crop temperature  $X$  should be met:

$$\begin{aligned} P(X \leq L) &\leq p_1 \\ P(X \geq D) &\leq p_2 \end{aligned} \quad (6)$$

where  $p_1$  and  $p_2$  are sufficiently small probabilities to ensure that the temperature variation is within two standard deviations of the mean. When  $X$  is normally distributed, Eq. (6) can be rewritten as:

$$z_{p_1} \geq \frac{L - \mu_T}{\sigma_T} = \frac{\sqrt{n+1}(L - \mu_T)}{\lambda(\mu_T - \mu_0)} \quad (7)$$

and

$$z_{(1-p_2)} \leq \frac{D - \mu_T}{\sigma_T} = \frac{\sqrt{n+1}(D - \mu_T)}{\lambda(\mu_T - \mu_0)} \quad (8)$$

where  $z_{p_1}$  and  $z_{(1-p_2)}$  are the normal scores corresponding to the probabilities  $p_1$  and  $1-p_2$ . Clearly,  $z_{p_1}$  is negative but  $z_{(1-p_2)}$  is positive under normal distribution. The normal scores as a function of the desired probability can be found from a statistical handbook (Neter et al., 1996) or calculated by the function of NORMSINV in Excel. Combining Eqs. (7) and (8), we obtained the following mean temperature range:

$$\mu_{T_{\min}} \leq \mu_T \leq \mu_{T_{\max}} \quad (9)$$

where the minimum final mean temperature is:

$$\mu_{T_{\min}} = \frac{\lambda\mu_0 z_{p_1} + \sqrt{n+1}(c_1 - \log \tau)/c_2}{\lambda z_{p_1} + \sqrt{n+1}} \quad (10)$$

and the maximum final mean temperature is:

$$\mu_{T_{\max}} = \frac{\lambda\mu_0 z_{1-p_2} + \sqrt{n+1}(c_3 - \log \tau)/c_4}{\lambda z_{1-p_2} + \sqrt{n+1}} \quad (11)$$

Clearly, the final mean temperature range is a function of the holding time  $\tau$  and number of mixings  $n$ . As  $\tau$  and  $n$  increase, the temperature range reduces to zero when  $\mu_{T_{\min}} = \mu_{T_{\max}}$ . The regimes corresponding to  $\mu_{T_{\min}} > \mu_{T_{\max}}$  are unsuitable for pest control. The crossing point ( $\tau_c, \mu_{T_c}$ ) at the time–temperature plot is determined when  $\mu_{T_{\min}} = \mu_{T_{\max}} = \mu_{T_c}$  as follows:

$$\mu_{T_c} = \frac{c_1 - c_3 + \lambda\mu_0(c_2 z_{p_1} - c_4 z_{1-p_2})/\sqrt{n+1}}{c_2 - c_4 + \lambda(c_2 z_{p_1} - c_4 z_{1-p_2})/\sqrt{n+1}} \quad (12)$$

$$\log \tau_c = c_1 + \frac{\lambda c_2 z_{p_1} \mu_0}{\sqrt{n+1}} - c_2 \left( \frac{\lambda z_{p_1}}{\sqrt{n+1}} + 1 \right) \mu_{T_c} \quad (13)$$

Based on the initial temperature distributions and the uniformity index, the treatment temperature and time range can be determined as a function of the number of mixings ( $n$ ) for given  $L, D, p_1$ , and  $p_2$  to satisfy the desired insect control and quality attributes.

## 2.4. Model validation

According to Eq. (5), the uniformity index after  $n$  intermittent mixings can be estimated from that without mixings by:

$$\lambda_n = \frac{\lambda}{\sqrt{n+1}} \quad (14)$$

After experimentally determining the heating uniformity index for the four crops without and with two intermittent mixings in the RF system, Eq. (14) was used to validate the above mathematical model. The experimental uniformity index after RF treatments for 3 min with two intermittent mixings ( $n=2$ ) was compared with that predicted by the mathematical model (Eq. (14)). The validated model can be used for further applications.

## 2.5. Model applications

### 2.5.1. Typical values of $D$ and $L$

Different crops have different temperature–time limits for targeted insect control and product quality. The insect heat tolerance depends on the thermal death kinetics of the targeted species, life stages and populations. The main quality attributes depend on the consumer acceptance and product marketability. Fig. 2 shows the typical temperature–time combinations for the required insect thermal mortality and walnut quality. The thermal death time curve of the most heat resistant insect in walnuts, fifth-instar navel orangeworm, was expressed as a negative logarithmic relationship with the exposure time (Wang et al., 2002b), with a  $c_1$  value of 14.19 and  $c_2$  value of 0.26 in Eq. (1). Walnut kernels contain approximately 60% lipids and 54% unsaturated fatty acids (Wang et al., 2001). Oxidation of unsaturated fatty acids renders walnuts one of the most heat sensitive low-moisture commodities. If the quality curve of walnuts could be used in the modeling, the required final average temperature for given mixing numbers would provide a

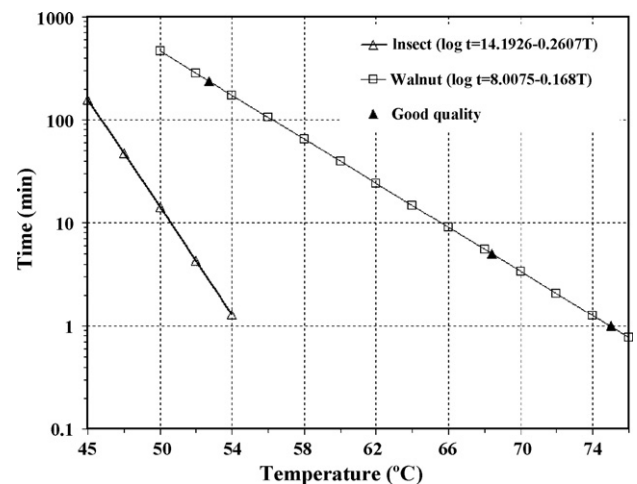


Fig. 2. Typical insect thermal-death-time curves and typical product quality curves based on the experimental temperature limits of walnuts from the literatures (Wang et al., 2002b, 2007b; Mitcham et al., 2004).



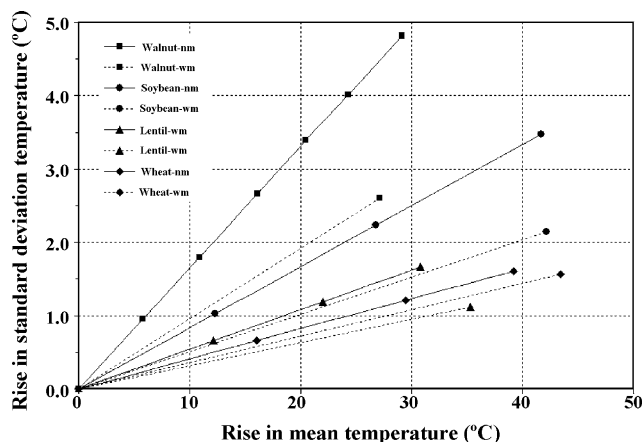


Fig. 4. Relationship between the rise in standard deviation and the rise in mean of wheat, lentil, soybean, and walnut temperatures with (wm) and without (nm) mixing (the slope defined as a uniformity index  $\lambda$ ).

### 3.2. Heating uniformity and model validation

Fig. 4 shows the experimental relationship between the rise in standard deviation and the rise in mean temperature of RF-treated crops without and with two mixings. The heating uniformity index obtained from the slopes indicated that different crop responded differently to the same RF unit heating. The heating uniformity index was in decreasing order for walnut, soybean, lentil, and wheat, which most likely depends on the crop size. After two intermittent mixings, the heating uniformity indexes for all four crops were clearly reduced (Table 2). In our previous studies, heating uniformity was further improved by moving containers on a conveyor belt in an industrial-scale in addition to mixing. The uniformity index for walnuts was reduced from 0.167 for in the batch type RF system to 0.062 for a continuous treatment with added hot air for surface heating (Wang et al., 2007a).

Table 2

Comparisons of the heating uniformity index of the four crops between experiment and simulation with two mixings after RF heating for 3 min

Crops	Experiment		Simulation (with two mixings)
	Without mixings	With two mixings	
Wheat	0.041	0.024	0.024
Lentil	0.054	0.031	0.031
Soybean	0.080	0.046	0.046
Walnut	0.165	0.096	0.095

It was observed that the heating uniformity index without mixing for soybean, lentil, and wheat was still smaller than that for walnuts with two mixing, suggesting that the RF treatments could be more easily applied into soybean, lentil, and wheat to achieve the same desired insect control and product quality.

The model validation based on the reduced standard deviation after mixing was shown in Table 2. The predicted heating uniformity index with two mixings using Eq. (14) was in good agreement with the experimental value for all four crops. Therefore, the established mathematical model can be used to determine the optimal mixing number needed in RF-treated products.

### 3.3. Suitable treatment temperature–time range of RF treatments

Fig. 5 shows time–temperature limitations of the four crops as a function of mixing number at the high security level. When the mixing numbers were 0 and 1, no solution was obtained for walnuts because  $\mu_{T_{min}}$  (blue) was higher than  $\mu_{T_{max}}$  (red). That is, with no or one-time mixing, it is impossible to control pests without causing significant quality losses. But for soybean, lentil, and wheat, the operational treatment temperature–time ranges were large even without mixing and the ranges increased with the mixing number. The differences in the operational treatment temperature–time range among the four crops were mainly caused

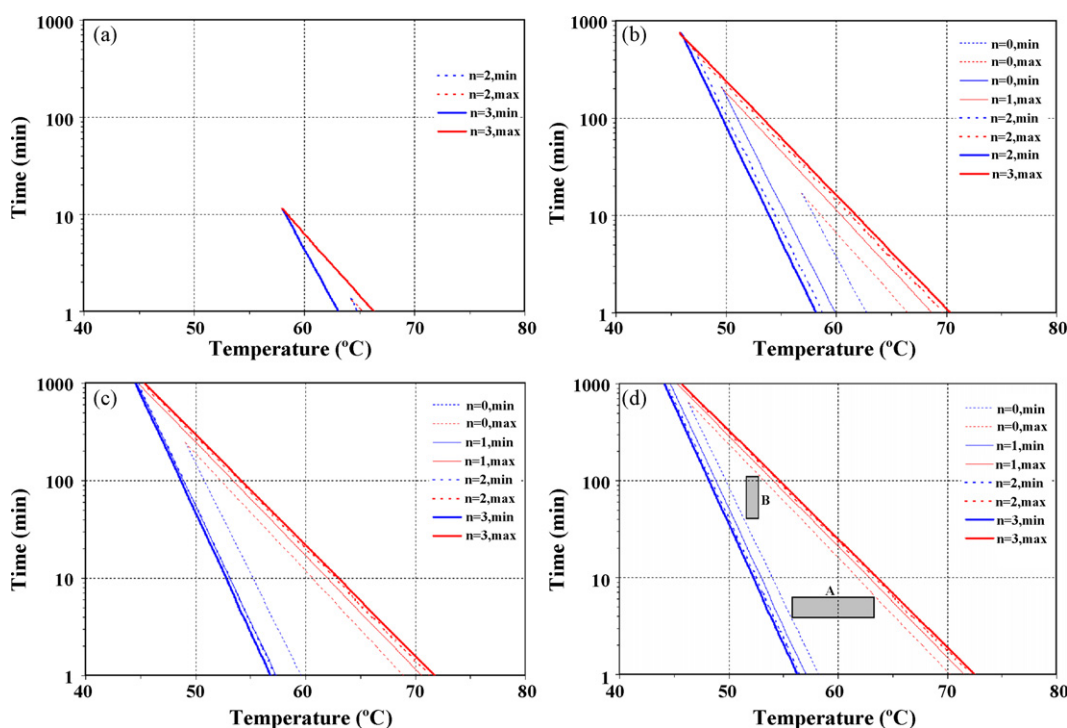


Fig. 5. Time–temperature high (max) and low (min) limitations of walnut (a), soybean (b), lentil (c), and wheat (d) as a function of mixing number ( $n$ ) at desired security levels of  $p_1 = 0.000032$  for insect control and  $p_2 = 0.01$  for product quality.

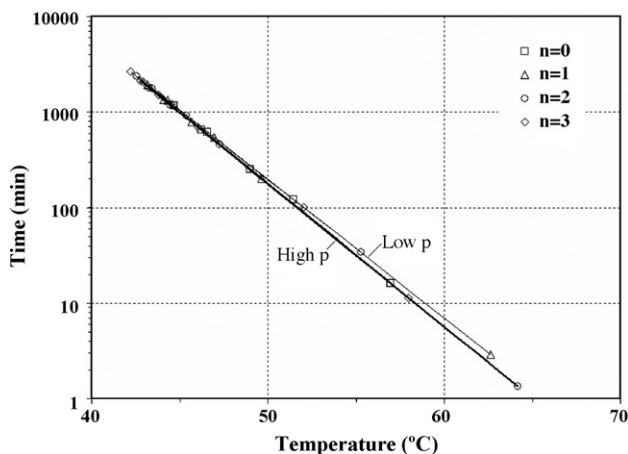
**Table 3**

Suitable final temperature ranges (°C) after 5-min holding as a function of mixing number for four crops to achieve the high ( $p_1 = 0.000032$  for insect control and  $p_2 = 0.01$  for product quality) and low ( $p_1 = 0.001$  and  $p_2 = 0.05$ ) security levels

Security levels	Mixing No.	Walnut	Soybean	Lentil	Wheat
High	0	–	59.4–61.0	56.5–63.1	55.3–64.3
	1	–	56.8–62.9	54.4–64.6	54.2–65.4
	2	–	55.8–63.8	54.3–65.2	53.7–66.0
	3	59.7–60.8	55.2–64.4	54.0–65.6	53.4–66.3
Low	0	–	57.8–62.9	55.6–64.6	54.6–65.4
	1	–	55.8–64.4	54.4–65.6	53.7–66.3
	2	59.2–62.0	55.0–65.1	53.9–66.1	53.3–66.7
	3	58.0–62.8	54.5–65.5	53.6–66.4	53.1–66.9

by their heating uniformity indexes listed in Table 2. The operational treatment temperature–time range increased slightly when the mixing number increased from 2 to 3, especially for the crops with good heating uniformity. The operational temperature–time range expanded with increasing mixing numbers. The operational treatment temperature–time range defined in this study quantitatively confirmed the research concept and strategy proposed by Tang et al. (2000). The possible operating range could be low temperature and long time (zone B in Fig. 5d) or high temperature and short time (zone A in Fig. 5d). Two major advantages of operating conditions in zone A zone B are high throughputs due to short holding times and accommodation for large temperature variations. This was further confirmed in our development of the high temperature and short-time RF processing reported in Tang et al. (2000).

Table 3 lists the calculated treatment temperature ranges for a 5-min short exposure time as a function of mixing number for four crops to achieve the high and low security levels. The temperature range increased with the mixing number for each crop and with improved heating uniformity from walnut ( $\lambda = 0.165$ ) to wheat ( $\lambda = 0.041$ ). With the reduced security level, the temperature range slightly increased for each crop with the same mixing number. The temperature range was very limited for walnuts heated in the batch mode of RF systems under the given stationary conditions. However, this temperature range for walnuts could be extended to those between soybean ( $\lambda = 0.080$ ) and lentil ( $\lambda = 0.054$ ) because the heating uniformity index of walnuts in an industrial-scale RF system with continuous process and hot air surface heating was reduced to 0.062 (Wang et al., 2007a). The heating uniformity was, therefore, the key issue to develop successful postharvest quarantine treatments using RF energy.



**Fig. 6.** Time–temperature relationship of crossing points of four crops as a function of mixing number ( $n$ ) at high ( $\log t = 0.1494T + 9.7199$ ) and low ( $\log t = 0.1448T + 9.5355$ ) security levels.

Fig. 6 shows the time–temperature relationship for the crossing points of four crops as a function of mixing number at high- and low-security levels. There was a clear linear correlation between the time and temperature of the crossing points. For each crop, the treatment temperature decreased and exposure time increased with the increasing mixing number. For each mixing number, the temperature decreased and time increased with decreasing size of crops or decreasing uniformity index value. The slope decreased slightly from the high security level to the low one, suggesting that small percentage of extra temperature and time was needed to increase the efficacy of RF treatments for these four crops from 99.9% mortality to 99.9968% (Probit 9).

#### 4. Conclusions

Heating uniformity in RF systems can be improved by intermittent mixings. The established mathematical model based on normal distribution of the product temperatures was used to predict the suitable operational time–temperature range for different mixing numbers to meet both the insect control and product quality. The suitable temperature–time range increased with decreasing size of crops or decreasing uniformity index value, increasing mixing number, and reducing security level. The heating uniformity was an important parameter to increase the suitable treatment temperature–time range in RF-treated products. For industrial RF applications, it is important to improve the heating uniformity by designing a good RF cavity with the best possible RF field distribution, and by using moving conveyor belt and applying additional hot air heating. It is interesting to note that no mixing is needed to completely control the insects and maintain the acceptable product quality in small-size crops such as lentil and wheat. RF treatment is, thus, holding potential as an environment-friendly alternative to chemical fumigation for control of insects in low-moisture crops.

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