2	A novel strategy for improving radio frequency heating uniformity of dry food
3	products using computational modelling
4	Zhi Huang <sup>1</sup> , Francesco Marra <sup>2</sup> , Shaojin Wang <sup>1,3*</sup>
5	
6	<sup>1</sup> College of Mechanical and Electronic Engineering, Northwest A&F University, Yangling,
7	Shaanxi 712100, China
8	<sup>2</sup> Dipartimento di Ingegneria Industriale, Università degli studi di Salerno, Fisciano, SA, Italy
9	<sup>3</sup> Department of Biological Systems Engineering, Washington State University, 213 L.J. Smith
10	Hall, Pullman, WA 99164-6120, USA
11	
12	
13	
14	
15	
16	*Corresponding author: Shaojin Wang, Ph.D, Professor. Tel.: +86-29 87092391; Fax: +86-29
17	87091737; Email address: shaojinwang@nwsuaf.edu.cn
18	

For submission to Innovative Food Science and Emerging Technologies

## 19 Abstract

This study attempted to quantify effects of dielectric properties (DPs) of a surrounding 20 container and treated food products on heating uniformity in a 6 kW, 27.12 MHz parallel plate 21 22 radio frequency (RF) system. A computer simulation model was established with finite element-based commercial software, COMSOL Multiphysics<sup>®</sup>, and experiments with 1.5 kg 23 soybean flour packed in a rectangular polystyrene container were performed to validate the 24 developed model. Surface temperature distributions of soybean flour in three different 25 horizontal layers were obtained with an infrared camera, and temperature-time histories at two 26 27 representative locations inside the container were monitored with two fiber optical sensors. The uniformity index (UI) was used as criterion to evaluate RF heating uniformity within food 28 29 products. Results showed that the temperature uniformity of food samples was clearly 30 influenced by DPs and density of the surrounding container. UI was the lowest when the surrounding container dielectric constant was in a comparable range of the sample's, with the 31 loss factor values of surrounding container lied between 0.01-0.1% of the sample's. The 32 33 optimum RF heating uniformity of food products could be achieved with a smaller density value of the surrounding container. The correlations of DPs and density between surrounding 34 35 container and food products derived from the validated simulation model could provide valuable information and strategy to improve the RF heating uniformity of low moisture 36 foods for insect or microbial control. Thus, the established strategy can further be used for 37 38 developing effective industrial-scale RF treatment protocols after optimization of this process by the food industry. 39

40 Keywords: Computer simulation; Dielectric properties; Heating uniformity; Radio frequency;
41 Soybean flour.

## 42 **1. Introduction**

Low moisture foods, such as wheat flour, corn meal, glutinous rice flour, nuts, spices, and 43 milk powders, are normally considered as shelf stable foods and can be stored for a long time 44 45 due to preventing bacterial growth in its low moisture environment. Soybean flour is a popular food due to its high nutritional value and functional characteristics, which contain 46 flavonoids, fiber and bioactive peptides (Hassan, 2013). Pathogens and insect pests, however, 47 48 are found to survive in a low moisture environment more easily for several months (Finn, 49 Hinton, McClure, Amézquita, Martins, & Fanning, 2013; Johnson, Wang, & Tang, 2010; 50 Mohapatra, Kar, & Giri, 2015). The qualitative and quantitative losses can be reached as high as 30% due to insect damages (FAOSTAT, 2013), which also promote mold growth, toxin 51 52 production, and product degradation in low moisture foods (Jiao, Johnson, Tang, & Wang, 53 2012; Vijay, Bhuvaneswari, & Gajendran, 2015). Several cases of soybean flour contamination with Plodia interpunctella greatly affect the quality and taste properties of the 54 end product made by the flour (Singh, Satya, & Naik, 2013; Taylor, Fields, & Sutherland, 55 56 2007). Radio frequency (RF) heat treatment has been considered as a novel heating technology for controlling insect and microbial populations in several dry products, such as 57 almond (Gao, Tang, Villa-Rojas, Wang, & Wang, 2011), date (Ben-Lalli, Bohuon, Collignan, 58 & Méot, 2013), lentil (Jiao et al., 2012; Wang, Yue, Chen, & Tang, 2008), peanut butter 59 60 cracker (Ha, Kim, Ryu, & Kang, 2013), raisin (Alfaifi et al., 2014), spice (Kim, Sagong, Choi, 61 Ryu, & Kang, 2012), walnut (Mitcham et al., 2004; Wang et al., 2008), and wheat (Shrestha, & Baik, 2013). 62

Although the most important characteristic of RF treatments is fast and volumetric heating generated by dipole rotation and ionic conduction, edge over-heating is still a major problem for foods heated in rectangular containers (Farag, Marra, Lyng, Morgan, & Cronin, 2010; Tiwari, Wang, Tang, & Birla, 2011a,b; Alfaifi et al., 2014). Edges and corners always 67 absorb more electromagnetic energy compared to other regions due to different dielectric properties (DPs) between food products and the surrounding media (usually air), resulting in 68 an uneven electric field distribution (Birla, Wang, Tang, & Hallman, 2004). The non-uniform 69 70 heating in RF treated products may cause either survivals of pathogens/insects or degraded quality (Birla et al., 2004; Jiao et al., 2012; Geveke, Kozempel, Scullen, & Brunkhorst, 2002; 71 72 Kim et al., 2012; Kirmaci & Singh, 2012). A number of methods have been reported for overcoming non-uniform RF heating, such as combining with an external heating or cooling 73 74 (Birla et al., 2004; Hou, Ling, & Wang, 2014; Liu, Wang, Mao, Tang, & Tiwari, 2013; Wang, 75 Tiwari, Jiao, Johnson, & Tang, 2010), enclosing in another medium (Ikediala, Hansen, Tang, Drake, & Wang, 2002; Jiao, Tang, & Wang, 2014a; Luechapattanaporn et al., 2005), mixing 76 77 or rotating food (Birla et al., 2004; Chen, Wang, Li, & Wang, 2015a), modifying electrode 78 shapes (Tiwari et al., 2011a; Alfaifi et al., 2014), and sample movement (Chen, Huang, Wang, 79 Li, & Wang, 2015b; Wang, Monzon, Johnson, Mitcham, & Tang, 2007). The trial and error procedures are time consuming, costly, and often provide limited information, which cannot 80 81 easily identify the mechanism behind non-uniform RF heating. Finite element modelling may serve as valuable tools to acquire deep insights on the heating uniformity of products and 82 83 offer opportunity to clearly understand RF interactions with food components without the necessity of extensive experiments. 84

Modeling RF processes is a multi-physics problem that involves the solution of coupled electromagnetic and heat transfer equations. Several simulation models have been developed to improve the RF heating uniformity for different food materials, such as apple (Birla et al., 2004), fish (Llave, Liu, Fukuoka, & Sakai, 2015), meat batters (Marra, Lyng, Romano, & McKenna, 2007), peanut butter (Jiao, Shi, Tang, Li, & Wang, 2015b), raisins (Alfaifi, et al., 2014), shell eggs (Lau, 2015), soybeans (Huang, Zhu, Yan, & Wang, 2015c), wheat flour (Tiwari et al., 2011a,b), and wheat kernels (Jiao, Deng, Zhong, Wang, & Zhao, 2015a). The

92 simulated uniformity index (UI) has been used as criteria to evaluate the temperature uniformity in RF treated products (Alfaifi et al., 2014; Huang et al., 2015c; Jiao et al., 2015a; 93 Tiwari et al., 2011a; Wang, Yue, Tang, & Chen, 2005). Simulated results show that the RF 94 95 heating uniformity could be improved by immersing the model fruit in water, suggesting that the non-uniform heating is mainly caused by the difference between DPs of food and its 96 97 surrounding medium (Birla et al., 2004; Huang, Zhang, Marra, & Wang, 2016; Jiao, Tang, Wang, & Koral, 2014b). When the dielectric constant of surrounding material is in a 98 comparable range with the sample, the best heating uniformity would be achieved (Tiwari et 99 100 al., 2011a; Huang et al., 2015c). The dielectric constant determines the electric field distribution when the loss factor is far smaller than the dielectric constant (Jiao et al., 2014a,b; 101 102 Metaxas, 1996). Surrounding the container with a more or less thick material, characterized 103 by dielectric constant similar to that of processed samples, provides better RF heating 104 uniformity (Huang et al., 2015c). Following this approach, Jiao et al. (2015b) have shown that 105 the temperature uniformity in peanut butter has been improved by minimizing the difference 106 of dielectric constant between food sample and surrounding material. However, there was no available mathematical modelling in literature to identify the specific relationship of DPs 107 between surrounding material and treated products for RF heating uniformity improvement. 108 109 Therefore, it is desirable to conduct a numerical analysis to systematically study the RF heating characteristics and design treatment protocols to improve RF heating uniformity in 110 111 low moisture foods.

The objectives of the current study were to: (1) develop a computer simulation model to predict the electric field intensity and temperature distribution in three different layers of soybean flour in a rectangular shaped container, (2) conduct experiments with soybeans flour in a 6 kW, 27.12 MHz RF system to verify the simulation results, (3) apply the validated model to evaluate the heating uniformity of soybean flour influenced by DPs and density of the surrounding container and treated products, and (4) establish the DPs and density correlations between surrounding container and treated food sample when the best heating uniformity was obtained.

# 120 **2. Materials and methods**

## 121 2.1. Raw material preparation

122 Soybean flour (Glycine max.) was purchased from a local market in Yangling, Shaanxi, China. A total of 20 kg of soybean flour were kept in polyethylene bags and stored at the 123 constant temperature (20 °C) in a thermostatic and humidity controlled chamber (BSC-150, 124 125 Shanghai BoXun Industrial & Commerce Co., Ltd., Shanghai, China). They were taken out from the chamber and kept at ambient room temperature ( $20 \pm 1$  °C) for 4 h prior to RF 126 processing. The initial moisture content of tested soybean flour was  $7.93 \pm 0.08$  % on wet 127 128 basis (w.b.). The chemical compositions of the flour were measured in the College of Food Science and Engineering, Northwest A&F University, Yangling, China. The measurements 129 were made in three replicates and the standard methods used for each measurement are 130 131 summarized in Table 1.

Compositio	on	Method
Ash (g/100 g)	4.62	AOAC 920.181
Moisture (g/100 g w.b.)	7.93	AOAC 934.01
Fat (g/100 g)	18.26	AOAC 992.06
Protein (g/100 g)	32.64	AOAC 955.04
Carbohydrate (g/100 g)	30.61	CFR 101.9
Calories (Kcal/100 g)	418	CFR 101.9
Dietary fibre (g/100 g)	5.94	AOAC 994.13

132	Table 1. Composition	s and measurement	methods for soybean	flour.
-----	----------------------	-------------------	---------------------	--------

133

## 134 2.2. Container material and dimensions

The polystyrene container was chosen based on the closest dielectric constant to that of soybean flour with a small dielectric loss factor (Table 2). With the main advantage of lower density, cheap, stable, high heat resistance and portable characteristics, polystyrene was widely used in food processing and packaging industry. The inner dimension of the rectangular polystyrene container was  $30 \times 22 \times 6$  cm<sup>3</sup>, the thickness for the bottom and side walls was fixed at 2 cm (Huang et al., 2016).

141

#### 142 2.3. Temperature measurements

143 About 1.5 kg soybean flour samples filled the rectangular container were horizontally divided into three layers (A: z = 6 cm; B: z = 4 cm; C: z = 2 cm) parallel to the container 144 145 bottom by two thin polypropylene films (mesh opening of 0.2 mm) with each filled of 0.5 kg 146 to represent the temperature distribution inside the sample (Fig. 1). An infrared camera (DM63-S, DaLi Science and Technology Co., LTD, Zhejiang, China) with an accuracy of ± 147 2 °C was used for mapping surface temperatures of soybean flour in three different layers 148 149 after RF treatment. The thermal digital infrared camera was first calibrated against a thin thermocouple thermometer (HH-25TC, Type-T, OMEGA Engineering Inc., Stamford, 150 Connecticut, USA) with an accuracy of  $\pm$  0.5 °C and 0.9 s response time. For internal 151 temperature monitoring during RF heating, two optical fiber sensors (1.8 mm diameter) 152 attached to a temperature measurement system (FTS-P104, Xi'an HeQi Opo-Electronic 153 154 Technology Co., LTD, Shaanxi, China) were inserted at the center (B<sub>1</sub>) and near the corner (B<sub>2</sub>) at middle layer (z = 4 cm) of food sample. The optical fiber sensors were calibrated by 155 using ice-water mixture and boiling water before experiment and temperatures were recorded 156 at 1 s intervals. A plastic foam board  $(29 \times 21 \times 2 \text{ cm}^3)$  made from polystyrene was used to 157 cover on the top of the container. The sensors were inserted at the center  $(B_1)$  and corner  $(B_2)$ 158 of the board using a drill to ensure precise hole positioning (Fig. 1). These positions were held 159

by fixing the sensor cables to a stationary position through the feeding inlet of the RFmachine.



### 162

Fig. 1. Position of two fiber optical temperature sensors in the rectangular polystyrene container split into three layers (A-top, B-middle, C-bottom layer) for temperature profile measurements (all measures in cm).

- 166 2.4. Computer modeling of RF heating
- 167 2.4.1. Physical model

A 6 kW, 27.12 MHz free running oscillator pilot scale RF system (COMBI 6-S, 168 Strayfield International Limited, Wokingham, UK) was used in this study (inner dimension 169  $129 \times 109 \times 52$  cm<sup>3</sup>). This system consisted of a chamber with electrically insulated walls and 170 two parallel rectangular electrodes, a generator, power amplifier, matching unit, and a RF 171 applicator. The RF power was set automatically by controlling the voltage to the power 172 amplifier from the generator, which was fed in the middle of upper electrode back side and 173 proportional to the electrode gap by changing the top electrode position with the aid of 174 adjustable screws (Fig. 2). A 1-cm-thick polystyrene sheet with the area of  $83 \times 40$  cm<sup>2</sup> (same 175 as the top electrode) was placed above the bottom electrode to prevent direct contact between 176 the electrode and the container. The polystyrene container with a bottom thickness of 2 cm 177 178 was placed in the center of the polystyrene sheet for achieving better RF heating uniformity

(Huang et al., 2015c; Tiwari et al., 2011a; Jiao et al., 2015a). To ensure repeatability of the
experimental results and limit the influence of non-uniform electromagnetic fields on sample,
the location of the sample was fixed on the center and middle between the top and bottom
electrodes during all experiments (Llave et al., 2015).





184

Fig. 2. Schematic diagram of the 6 kW, 27.12 MHz RF system.

# 185 2.4.2. Governing equations

186 The electric field distribution within the load (food sample and the container) and at any 187 point inside the electrodes were given by the solution of the Gauss law derived from a 188 quasi-static approximation of Maxwell's equations (Marra et al., 2007):

189 
$$\nabla(\varepsilon \cdot \vec{E}) = 0$$
 (1)

190 where  $\varepsilon$  is permittivity of the load and  $\vec{E}$  is the electric field vector. Simulation of RF heating 191 as a quasi-static electric field between the electrodes was preferred since the wavelength (11 192 m) in the 27.12 MHz RF system is often much larger than the analyzed domain sizes. 193 The RF power conversion to thermal energy  $(Q, W m^{-3})$  within the food sample under an 194 electric field intensity  $(|\vec{E}|, V m^{-1})$  are described as follows (Choi & Konrad, 1991):

195 
$$Q = 2\pi f \varepsilon_0 \varepsilon' \left| \vec{E} \right|^2$$
(2)

where *f* is the frequency (Hz),  $\varepsilon_0$  is the permittivity of electromagnetic wave in free space (8.86 × 10<sup>-12</sup> F m<sup>-1</sup>),  $\varepsilon''$  is the relative dielectric loss factor of the sample load, and  $|\vec{E}|$  is the modulus of the *E* (*x*, *y*, *z*) field.

Evaluating the absorbed RF power density at any point inside the sample requires the value of  $\vec{E}$ , which is a function of the sample geometry and DPs as well as the electrode configuration (Jiao et al., 2014b; Marshall & Metaxas, 1998):

202 
$$\left|\vec{E}\right| = \frac{V}{\sqrt{\left(\varepsilon'd_{air} + d_{mat}\right)^2 + \left(\varepsilon''d_{air}\right)^2}}$$
(3)

where  $d_{air}$  is the air gap between top electrode and food sample (m),  $d_{mat}$  is the height of the food material (m), and *V* is the voltage between the two electrodes (V).

The heat equation was solved within the food material and the container, considering the internal conduction and the heat generation due to RF energy, while external convection at the sample surface was taken into account in the boundary conditions. Therefore, the temperature distribution was simulated based on a 3D heat transfer equation, including solution of Fourier heat transfer equation plus a generation term, coupled with the quasi-static electro-magnetic field equations (Uyar et al., 2015):

211 
$$\rho C_P \frac{\partial T}{\partial t} = \vec{\nabla} \cdot k \vec{\nabla} T + Q$$
(4)

where  $\rho$  is density (kg m<sup>-3</sup>),  $C_p$  is specific heat (J kg<sup>-1</sup> K<sup>-1</sup>), k is thermal conductivity (W m<sup>-1</sup>) K<sup>-1</sup>), T is the temperature (K), and t is the time (s). While the solution of heat transfer is 214 needed just within the sample, the Gauss law must be evaluated for the space between the two

electrodes, which includes the sample and the air around it (Marra et al., 2007).

216 2.4.3. Geometric model

217 The 3D geometric model was developed using COMSOL (V4.3a COMSOL Multiphysics, COMSOL, Co., LTD., Shanghai, China) based on the actual structure and size of the 6 kW, 218 27.12 MHz RF system (Fig. 3). The complete geometric model included RF cavity ( $129 \times 109$ 219  $\times$  52 cm<sup>3</sup>), top (83  $\times$  40 cm<sup>2</sup>) and bottom electrode (99  $\times$  59 cm<sup>3</sup>), polystyrene sheet (83  $\times$  40 220  $\times$  1 cm<sup>3</sup>), soybean flour sample (30  $\times$  22  $\times$  6 cm<sup>3</sup>), and polystyrene container (34  $\times$  26  $\times$  8 cm<sup>3</sup>) 221 222 as shown in Fig. 3. Soybean flour samples were placed in the center and middle between the top and bottom electrodes with an air gap of 3 cm. The whole domain in this study was 223 224 divided into three sub-domains, the food sample (SD1), container (SD2), and the surrounding 225 air (SD3). In SD1 and SD2, both heat transfer and Gauss law equations were solved, whereas in SD3 only dielectric phenomenon was considered. An unstructured mesh consisting of 226 227 Lagrange quadratic elements was created over the entire domain of RF cavity. The extremely 228 fine mesh structure was preferred based on the convergence tests even though it led to longer computational times, which consisting of 122,868 domain elements (tetrahedrons), 10,574 229 boundary elements (triangles) and 879 edge elements (linear). The top electrode was drawn as 230 an embedded element (work plane) in 3D to avoid a large number of mesh elements for a thin 231 electrode plate. The X, Y, and Z directions were modeled using a mesh spacing of 5 mm, 232 233 which provided satisfactory spatial resolution for the considered domain and the solution was found to be independent to the grid size with further refinement. 234





Fig. 3. 3D model approach of RF heating system (mesh of 5 mm).

237 2.4.4. Thermo-physical and dielectric properties

The moisture content of soybean flour was determined by drying triplicate 3-5 g flour 238 samples in aluminum moisture dishes in an oven (DZX-6020B, Shanghai Nanrong Co. Ltd., 239 240 Shanghai, China) at 130 °C for 1 h until a substantially constant weight was obtained (AOAC, 2002). The density of soybean flour was calculated from the measured mass and volume at 241 242 room temperature over three replicates. The specific heat and thermal conductivity of soybean 243 flour were measured using a differential scanning calorimeter (DSC, Q2000, TA Instruments, 244 New Castle, PA, USA). An empty sealed aluminium pan was used as a reference and 10 mg 245 soybean flour samples were sealed in another aluminium pan (20 µL). The procedure included 246 cooling the sample from room temperature to 0 °C, equilibrating for 6 min, and then heated in the DSC at a rate of 10 °C/min over a temperature range of 20-80 °C. Each measurement was 247 repeated three times and details of the measurement system and procedure can be found 248

elsewhere (Jiao et al., 2014b). These data were subjected to linear regression analysis for using these properties in simulation model over the treatment temperatures range from 20 to 60 °C (suitable for postharvest pest control). Dielectric properties of the soybean flour sample had a non-linear relationship with temperature and were reported at frequencies of 27 MHz (Guo, Wang, Tiwari, Johnson, & Tang, 2010). Dielectric and thermo-physical properties of the electrode and ambient air at room temperature (20 °C) were adapted from COMSOL material library (Table 2).

256	Table 2. Electrical	and thermo-physical	properties of	materials used i	in computer simulation
-----	---------------------	---------------------	---------------	------------------	------------------------

Property	Electrode <sup>a</sup>	Air <sup>a</sup>	Polystyrene <sup>b</sup>	Soybean flour
Density ( $\rho$ , kg m <sup>-3</sup> )	2700	1.2	25	380
Specific heat $(C_p, J \text{ kg}^{-1} \text{ K}^{-1})$	900	1000	1300	5.8· <i>T</i> +1614
Thermal conductivity $(k, W m^{-1} K^{-1})$	160	0.026	0.036	0.0007· <i>T</i> +0.083
Dielectric constant ( $\varepsilon'$ )	1	1	2.6	3.96 <sup>c</sup>
Loss factor ( $\varepsilon''$ )	0	0	0.0003	0.38 <sup>c</sup>

257

Source: T = temperature (°C)

<sup>&</sup>lt;sup>a</sup> COMSOL material library, V4.2a (2012).

259	<sup>b</sup> Huang et al. (2016)
239	fluang et al. (2010)

<sup>c</sup> Guo et al. (2010).

261 2.4.5. Initial and boundary conditions

The Gauss law needs boundary conditions for the electrodes and the RF cavity walls to determine the electric field distribution inside the system.

264 – Top electrode was set as the electromagnetic source since it introduced high frequency

265 electromagnetic energy from the generator to the heating cavity, which maintained at a certain

266 potential *V* and constant during the processing time.

267 – All the metal shielding parts except for the top electrode were maintained at the ground 268 condition ( $V_0 = 0$ ).

269 – The metallic RF cavity walls and the plastic container were assumed to be electrically 270 insulated  $(\nabla \cdot \vec{E} = 0)$ , and for the left and right sides of the system, the surrounding air was 271 considered.

For the heat transfer equation to determine the temperature distribution inside the system: — The initial temperature of the two electrodes, surrounding air, polystyrene sheet, and polystyrene container was set at room temperature (20 °C), and the initial temperature of the soybean flour was assumed at a uniform temperature ( $T_0 = 20$  °C).

276 — The inlet and outlet of the simulated system were adjusted to meet the condition  $T = T_a$ , 277 where  $T_a$  is the ambient temperature, the top and bottom sides (the external sides of the upper 278 and lower electrodes) were thermally insulated ( $\nabla T = 0$ ).

279 - The load was considered to be subjected to heat exchange with surrounding air by
 280 convection expressed as:

281 
$$-k\frac{\partial T}{\partial n} = h(T - T_a)$$
(5)

Heat exchange was approximated by assuming a convective heat transfer coefficient ( $h = 20 \text{ W m}^{-2} \text{ K}^{-1}$ , which is a typical value used for natural convection in air in the used configuration),  $T_a$  is the air temperature inside the RF cavity (20 °C) and  $\stackrel{1}{n}$  is the normal vector.

286 2.3.6. Constant electric potential

Even though the voltage varies all over the surface of the top electrode, it was assumed to be uniform since the electrode dimensions  $(0.83 \times 0.4 \text{ m}^2)$  were much lower than 30% of the RF wave length ( $\approx 11 \text{ m}$ ). Thus, the calculation with constant electric potential applied at the top electrode was performed by the following equation with the measured anode current ( $I_a$ ) (Zhu, Huang, & Wang, 2014):

292 
$$V = 11242 I_a + 2029.9$$
 (6)

On the basis of the measured anode current (0.18 A), the top electric voltage applied was set at 4050 V for subsequent simulations. Based on our preliminary results, the voltage *V* varies up to 11% between standby and full-load conditions while Marshall and Metaxas (1998) reported that the difference was only 7%. Llave et al. (2015) also reported the similar heating problems associated with the free-running oscillator systems.

298 2.3.7. Model assumptions

In the current simulation study, the following assumptions were considered to simulate temperature distribution of soybean flour subjected to RF heating:

301 – DPs and TPs (thermo-physical properties) of soybean flour were assumed to be
 302 homogeneous and isotropic, the density and dielectric properties were assumed temperature
 303 independence.

The mass and momentum transfers of water were ignored due to a short RF heating
 time (5 min) and the influence on moisture content was unnoticeable (< 2%) based on</li>
 preliminary study.

307 2.3.8. Solution methodology

The Joule heating module of a finite element based software COMSOL was used to solve this multi-physics problem that involves the solution of electromagnetic equation coupled with heat transfer equation. The important stages of the computer simulation included defining transport equations or domain and sub-domains of interest, describing dielectric and thermo-physical properties, setting up boundary conditions, meshing of the domains, and choosing the final numerical solver. All computer simulations were performed on a Dell workstation with an Intel® Core<sup>TM</sup> i5-2400, 3.10-GHz processor and 8 GB RAM running a 315 Windows 8.1 64-bit operating system. The maximum and minimum mesh element sizes were 0.0578 m and 0.000578 m, respectively. Time step was determined by the max iteration per 316 317 time step. If the computations were convergent before the max iteration, the time step could 318 be short and would start next iteration automatically. Based on extensive testing, the maximum iteration per time step was  $10^4$ , and initial and maximum time steps were set as 319 320 0.001 and 1 s, respectively. The direct linear system solver UMFPACK was used for all calculations with a relative tolerance and an absolute tolerance of 0.01 and 0.001. 321 Simultaneous solution of the coupled transient equations took nearly 5400 s (1.5 h) for 322 323 simulating 300 s of RF heating process at 1-s intervals.

324 2.4. RF experiments

In experiments, soybean flour samples with density of 380 kg m<sup>-3</sup> were put into the 325 rectangular container and positioned coaxially in the center and middle between the top and 326 bottom electrodes of the RF system. The lethal temperature range for the complete mortality 327 of many insects including Indianmeal moth at all life stages was within 44-52°C with 328 329 different holding time (Johnson, Wang, & Tang, 2003). To meet the required insect mortality and acceptable product quality, the soybean flour samples were subjected to RF heating for 5 330 min with electrode gap of 12 cm according to our previously published RF power 331 recommendations (Huang et al., 2015c). Sample temperature-time histories at two 332 representative locations in middle layer of soybean flour were measured using a six-channel 333 334 fiber optical temperature measurement system with an accuracy of  $\pm 1$  °C. Probes connected with two channels were inserted into the center and corner of soybean flour through predrilled 335 holes while the remaining channels were used to monitor the air temperature inside the RF 336 337 cavity (Fig. 1). Temperatures were sampled every second and recorded at 1-s intervals over 5 min. After 5 min RF heating, the container was immediately taken out from the RF cavity, 338 and the surface temperature of soybean flour from top to bottom layer was measured by an 339

340 infrared camera. All three thermal imaging recordings were completed in 20 s. The image analysis system (V1.0, DaLi Science and Technology Co., LTD, Zhejiang, China) was used to 341 collect and analyze the surface temperature data of soybean flour, and 97,856 individual 342 343 temperature data were collected in each layer. The whole experiments were replicated five times and the experimental setup for sample temperature measurements during RF heating are 344 shown in Fig. 4. The measured temperature profiles and the isothermal curves of soybean 345 346 flour were used to validate the developed simulation model. Average and standard deviation values in the temperature of the RF heated samples were also compared over three layers. 347





Fig. 4. Experimental setup for sample temperature measurements in RF heating.

Means and standard deviations were calculated with Sigma-Plot 12.0 (Systat Software, Inc., San Jose, CA, USA) statistical software. Sigma-Plot was used to perform t-test, one way analysis of variance (ANOVA) and Least Significant Difference test (LSD) at a 95% confidence level (P < 0.05) to identify difference among samples. To validate the simulation results, the root mean square error (RMSE) was calculated for the comparison of temperature differences between the simulated and experimental profiles over *N* time steps.

<sup>350 2.5.</sup> Statistical analysis

357 
$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^{N} \left[ \left( T_{exp}(i) - T_{sim}(i) \right) / T_{exp}(i) \right]^2}$$
(7)

358 where  $T_{exp}$  and  $T_{sim}$  are the experimental and simulated temperature (°C), respectively, *i* is the 359 data node number, and *N* is the total number of points.

360 2.6. Heating uniformity evaluation

361 Cold spot locations were determined by finding the lowest temperature from thermal images and simulated temperature profiles, which were located at the center part of each layer 362 (Huang, Chen, & Wang, 2015a). In this study, all simulations were conducted based on the 363 top surface center (cold spot) to reach the target disinfestation temperature (50 °C) after 5 min 364 RF heating. The relative sensitivity with respect to model inputs was evaluated using the 365 temperature uniformity index (UI) inside the samples under the same electrode gap of 12 cm. 366 It would reflect the degree to which temperature in the volume deviated from the target 367 temperature (Jiao et al., 2015b): 368

369 
$$UI = \frac{\int_{V_{vol}} \sqrt{(T - T_t)^2} dV_{vol}}{(T_t - T_{initial}) V_{vol}}$$
(8)

where  $T_t$  and  $T_{initial}$  are the target and initial temperatures (°C) inside the dielectric material over the volume ( $V_{vol}$ , m<sup>3</sup>). A smaller index corresponds to a better heating uniformity.

373 2.7. Simulation sequence

# 2.7.1. Simulation with varying DPs of the sample and container

The RF heating uniformity was significantly influenced by DPs of the treated material. After the model has been validated by comparing its results with the experimental ones, to study the effect of container dielectric constant on UI of the sample, simulations were run by changing dielectric constant of the container from 0.1 to 19, with various sample dielectric constants ranging between 2 and 13. Electrode gap in each simulation was set at 12 cm with heating time of 5 min. The top electrode voltage was determined by running multiple simulations with different input voltage values until the lowest temperature at the top surface center (cold spot) to reach 50 °C (Jiao et al., 2015b). Another set of simulations were performed with container loss factor varied between 0.00003 and 0.3 and sample loss factor increased from 0.1 to 5 to study the effect of container loss factor on UI. Regression equations could be established for the DPs of the sample and container based on the minimum UI values.

387 2.7.2. Simulation with varying density of sample and container

388 Since the specific heat and thermal conductivity had a relatively slight effect on RF heating uniformity based on previous studies (Huang, Zhu, & Wang, 2015b; Tiwari et al., 389 390 2011b), a series of simulations were run by changing container density progressively between 20 and 1000 kg m<sup>-3</sup>, while the density of the processed food was fixed as 300, 500, 800, and 391 1000 kg m<sup>-3</sup>, respectively. Trends of the UI of soybean flour in each simulation were 392 determined and the container density values corresponding to the minimum UI could be 393 394 calculated using Matlab 2010 (Mathworks Inc., Natick, MA). Therefore, the density correlations between the surrounding container and treated products could be developed. 395

396 **3. Results and discussions** 

397 3.1. Simulated electric field distribution for soybean flour

Figs. 5a-c show the general trends of electric field distributions between two parallel plate electrodes without dielectric material, and with dielectric material placed on the bottom and middle between two electrodes. The electric field between two parallel plate electrodes was in parallel lines uniformly spaced throughout the region between two electrodes, and perpendicular to their surfaces with no dielectric material placed in it (Fig. 5a). When the dielectric material was placed at the center of the bottom electrode with an air gap over it, the presence of the surrounding air causes an intensification of the heating near the top of the 405 electrode space at the edges of the material (Fig. 5b). As observed in Fig. 5c, the electric field 406 was distorted in the presence of the dielectric material placed in the middle of two parallel 407 plate electrodes. Samples placed in the middle of RF electrodes showed higher electric field 408 intensity at their central section as electric field deflected by both (top and bottom) edges with 409 increased net electric field concentration at the central parts of the sample (Tiwari et al., 410 2011a). This demonstrated the significant effect of surrounding material (usually air) on 411 obtaining uniform electric field distributions inside the product.



Fig. 5. The electric field strength between two parallel plate electrodes with (a) no dielectric sample, (b) dielectric sample placed on the center of bottom electrode, (c) and dielectric sample placed in the center and middle between top and bottom electrodes with a fixed electrode gap of 12 cm and initial temperature of 20 °C.

419 Figs. 6a-c show the simulated spatial E-field distributions of soybean flour in the middle layer of whole sample (z = 4 cm), top horizontal layer (z = 6 cm) and vertical (x = 11 cm) 420 cross section after 5 min RF heating at a fixed electrode gap of 12 cm. In the whole 3-D 421 422 model of the rectangular shaped sample, high electric field concentrations occurred at the corners and edges due to the refraction and reflection of the electrical field at these parts (Fig. 423 424 6b). Horizontally, electric field lines all bending at the interfaces between soybean flour and 425 the contacted container side walls (Fig. 6a). As a result, net electric field increased at four sides and edges of the treated sample. In the vertical center cross section (x = 11 cm), the 426 427 electric field distribution was more uniform in the top and bottom than other sections but concentrated at the side walls and center parts of the sample (Fig. 6c). The merging of two or 428 429 more electric field lines at the edges and corners resulted in a higher volumetric power density, 430 hence, overheating occurred in these areas more than on the flat surfaces (Fu, 2004). The 431 electric intensification was greater for samples either having a higher or lower dielectric constant than for those surrounding materials (Huang et al., 2015c; Tiwari et al., 2011a). 432 433 Therefore, it is necessary to devise means to eliminate the electromagnetic field concentration phenomenon within the treated products. 434



Fig. 6. Simulated electric field direction (arrow), electric field intensity (color surface, V m-1), and electric potential (streamline) of soybean flour  $(30 \times 22 \times 6 \text{ cm}^3)$  in (a) middle layer of

435

438 whole sample (z = 4 cm), (b) top horizontal layer (z = 6 cm), and (c) vertical (x = 11 cm) 439 center cross sections after 5 min RF heating with an electrode gap of 12 cm and initial 440 temperature of 20 °C.

441

455

442 3.2. Simulated temperature profiles for soybean flour

443 The results presented in Fig. 6 showed that corners, edges, and middle parts had higher 444 electric field concentration when the sample was placed in the center and middle between the top and bottom electrodes. Figs. 7a-c corroborate the result as the maximum temperature 445 446 values appeared at the corners and edges of all layers. In three horizontal layers, the temperature values were higher in the middle layer (51-74°C), whereas they were lower in the 447 448 top and bottom layers (50-69°C and 52-67°C). The highest temperature values occurred at the 449 center parts of soybean flour over the volume, placed in the middle (3 cm above the bottom 450 electrode) between two RF electrodes (Fig. 3). The non-uniform temperature distribution 451 could be attributed to the electric field behaviour, which was deflected at the sample corners 452 and edges, resulting in higher temperature values at these parts (Fig. 7). These findings are 453 corroborated by overheating at edges and center parts of the fish (Llave et al., 2015), raisins 454 (Alfaifi et al., 2014), and wheat kernels (Jiao et al., 2015a) subjected to RF treatment.



456 Fig. 7. Simulated temperature distributions (°C) at (a) top (z = 2 cm), (b) middle (z = 4 cm), 457 and (c) bottom (z = 6 cm) layers of soybean flour ( $30 \times 22 \times 6$  cm<sup>3</sup>) placed in the center and

458 middle between two electrodes after 5 min RF heating at an electrode gap of 12 cm and initial
459 temperature of 20 °C.

460 3.3. Computer model validation

461 The simulated spatial temperature profiles of soybean flour over three layers were compared with the experimental results obtained using a thermal imaging camera (Fig. 8). 462 Both results demonstrated that the experimental temperature distribution showed good 463 464 agreements with the simulated data in center parts of the top, middle, and bottom layers. For corners and edges of the sample, the maximum temperature differences in top, middle, and 465 466 bottom layers were about 4, 5, and 3 °C, respectively. The temperature differences between experiment and simulation might be caused by the simplification of the RF units or ignored 467 468 moisture and heat loss in the possible evaporation in the soybean flour samples. Fig. 9 shows 469 the measured and simulated time-temperature profiles of soybean flour at center and corner 470 positions (as demonstrated in Fig. 1). Both sets of data exhibited similar trends, with the 471 heating rate of 5.2 °C/min for corner and 4.8 °C/min for center throughout the duration of the 472 heating process. Acceptable RMSE values were obtained for corner (0.014 °C) and center (0.017 °C), respectively. Table 3 compares simulated and experimental average and standard 473 deviation temperatures of all three layers after 5 min RF heating. Five replicates of 474 experimental average temperatures were slightly lower than simulated ones, which were 475 476 attributed to the heat loss when samples were transferred from the RF cavity to the camera. 477 The simulated standard deviations were comparatively higher than those determined by experiments. This was probably caused by more data points for corners with fined meshes in 478 simulation, while these were equally distributed in experiment. It was corroborated that the 479 480 simulated temperature profile in all three layers could capture the actual temperature field variation, and the model accuracy could be guaranteed. 481

# Experimental

## Simulated



Fig. 8. Comparison of simulated and experimental temperature distributions in top (A), middle (B), and bottom layers (C) of soybean flour placed in a polystyrene container  $(30 \times 22$  $\times 6 \text{ cm}^3)$  in the center and middle between the top and bottom electrodes after 5 min RF heating at a fixed electrode gap of 12 cm and initial temperature 20 °C.

487 Table 3. Comparison between simulated and experimental temperatures (Ave ± SD, °C) in three different horizontal layers of soybean flour placed in the center and middle between two 488 electrodes after 5 min RF heating with an electrode gap of 12 cm and initial temperature of 489 20 °C. 490

	T and a	Simulated (°C)	Experimental (°C)
	Layer —	Ave $\pm$ SD	Ave $\pm$ SD
	Upper	$57.9 \pm 4.4$	$56.8\pm3.9$
	Middle	$59.6\pm5.4$	$58.8\pm4.1$
	Bottom	$56.8\pm3.8$	$56.1 \pm 3.2$
491			
	$\begin{array}{c} 60 \\ 50 \\ 50 \\ - \\ - \\ - \\ - \\ - \\ - \\ - \\ - \\ - \\ $	l central ntral	$\begin{array}{c} 70 \\ 60 \\ 0 \\ 0 \\ 50 \\ 40 \\ 30 \\ 20 \end{array}$
492	0 1 2	3 4 5 6 Time (min)	0 1 2 3 4 5 Time (min)

Fig. 9. Experimental and simulated temperature-time histories of soybean flour at the center (a) 493 494 and corner (b) positions of middle layer (z = 4 cm), placed in a polystyrene container in the center and middle between the top and bottom electrodes during 5 min heating at a fixed 495 electrode gap of 12 cm (bars indicate standard deviation of five runs). 496

497

3.4. Effect of DPs of sample and container on sample temperature distribution 498

499 Fig. 10 shows the electric field intensities in polystyrene container and soybean flour samples with container dielectric constant of 2.6 and 3.96, respectively. Simulated results 500

501 illustrated how electric field patterns were distorted within the rectangular shaped sample, and 502 severe edge heating usually happened at four corners and the bottom sections of sample-wall interfaces. When the dielectric constant differences between surrounding material and treated 503 504 sample was greater, deflection and distortion of the electric field increased more frequently at the contact surface of two different materials (Fig. 10a). The electric field distortion was 505 506 reduced when the container dielectric constant changed to 3.96 (equal to the treated sample). The maximum electric field intensity decreased from 3.32 to  $2.98 \times 10^4$  V m<sup>-1</sup> over the whole 507 treated sample and surrounding container (Fig. 10b). The top electrode voltage used in 508 509 computer simulation was 4200 V determined by running multiple simulations with different inputted voltages until the target temperature (50 °C) was obtained at the top surface center 510 511 (cold spot) of soybean flour (Jiao et al., 2015b). The electric field distortion was reduced with the maximum electric field difference of middle layer reduced from 0.92 to  $0.51 \times 10^4$  V m<sup>-1</sup> 512 after changing dielectric constant of the surrounding container (Fig. 11). The standard 513 514 deviation of temperature and electric field at three horizontal layers of soybean flour was also 515 reduced as shown in Table 4. Simulated UI of soybean flour decreased from 0.112 to 0.083 516 over the volume, demonstrating a rather uniform temperature obtained after 5 min RF heating. 517 Therefore, different electric field patterns between surrounding container and food products could be minimized by appropriately matching DPs of the container with that of treated 518 material. 519



Fig. 10. Simulated electric field direction (arrow), electric field intensity (color surface, V  $m^{-1}$ ), and electric potential (streamline) of soybean flour and polystyrene container with container DPs of (a) 2.6-*j*\*0.0003 and (b) 3.96-*j*\*0.0003 after 5 min RF heating under the fixed sample DPs of 3.96-*j*\*0.38.



Fig. 11. Simulated electric field distribution (V m<sup>-1</sup>) of soybean flour in the middle layer with container DPs of (a) 2.6-j\*0.0003 and (b) 3.96-j\*0.0003 after 5 min RF heating under the fixed sample DPs of 3.96-j\*0.38.

529

Table 4. Simulated temperature (Ave  $\pm$  SD, °C) and electric field intensity (Ave  $\pm$  SD, V m<sup>-1</sup>) of soybean flour in three different layers with container DPs of 2.6-0.0003 $\cdot j$  and 3.96-0.0003 $\cdot j$ under the fixed electrode gap of 12 cm.

Container	2.6-0	).0003 <i>·j</i>	3.96-0.0003 <i>· j</i>			
DPs		-				
Layer	Temperature (°C)	Electric field (V m <sup>-1</sup> )	Temperature (°C)	Electric field (V m <sup>-1</sup> )		
Unner	57 9 + <i>4 4</i>	18886 2 + 1488 4	593+31	19329 6 + 923 3		
Opper	57.7 ± 4.4	$10000.2 \pm 1400.4$	$57.5 \pm 5.1$	$17527.0 \pm 725.5$		
Middle	$59.6\pm5.4$	$19063.3 \pm 1957.5$	$61.8\pm4.3$	$20125.4 \pm 1226.5$		
Bottom	$56.8\pm3.8$	$17151.9 \pm 1019.7$	58.1 ± 2.1	$18255.1 \pm 864.4$		

533

534 3.5. Effect of DPs of sample and container on sample UI

535 Figs. 12a-c summarize the results of a series of simulations with surrounding container dielectric constant ranging from 0.1 to 19, and sample dielectric constants increasing 536 progressively at regular intervals between 2 to 13, based on the previous measured DPs of 537 538 soybean flour (Guo et al., 2010). Simulated results demonstrated that increase in container dielectric constant caused initial reduction and then the increase of UI with the fixed sample 539 dielectric constant values. UI was the lowest when the surrounding container dielectric 540 constant was smaller than the sample's one if dielectric constant of heated sample was < 7. 541 542 But for the dielectric constant of heated sample  $\geq 7$ , UI started to get the minimal value when 543 the surrounding container dielectric constant was greater than the sample's one. It was interesting to notice that UIs had their minimum values whenever the dielectric constants of 544 545 surrounding container were in a comparable range with the sample's one. This was contrary to 546 a general concept that increasing dielectric constant of the surrounding container for equal to 547 the sample's one might obtain the best heating uniformity of the heated products. Logistic regression analysis was performed to determine the minimum values of UI by solving a set of 548 549 quadratic equations based on the simulated trends of UI as illustrated in Fig. 12. Therefore, 550 dielectric constants of the surrounding container could be determined by solving the roots of 551 the regression equations. Table 5 shows the calculated dielectric constants of the surrounding container when the UI achieved the lowest values with various sample dielectric constants. 552 553 The simulated voltages used in the computer simulation were in a range of 2860-9300 V in 554 order to make the lowest temperature reach 50 °C. It is clear that trends of simulation voltage change depended on the dielectric constant of sample and the surrounding container. Since the 555 voltage level along the upper electrode is proportional to the square of dielectric constant of 556 557 treated sample, it also increased with the increase in dielectric constant (Birla, Wang, & Tang, 2008). 558

Fig. 13 shows the general trends of dielectric constant correlations between surrounding container and treated soybean flour for ensuring good RF heating uniformity. It is clear that dielectric constant of surrounding container increased almost linearly with increasing dielectric constant of heated sample. The dielectric constant of surrounding container as a function of sample's one may be predicted according to the following regression equation with a high coefficient of determination ( $R^2 = 0.99$ ):

565 
$$\mathcal{E}'_{con}$$

$$\varepsilon_{container}' = 1.419\varepsilon_{sample}' - 2.543 \tag{9}$$

566 The predicted dielectric constant value of surrounding material was 8.8 with the DPs of wheat flour fixed as 8-10\*i based on the developed regression equation. There was little 567 difference with the literature findings that UI was the lowest when the surrounding material 568 dielectric constant was between 8 and 11 (Tiwari et al., 2011a). The calculated dielectric 569 constant value of surrounding sheets was 3.17 with DPs of peanut butter of 4.03-0.004\**i*. This 570 was in good agreement with previous simulation results that the RF heating uniformity was 571 improved by using the PEI (DPs of 3.15-0.0025\**i*) assisting method (Jiao et al., 2014a, 572 2015b). Obviously, results demonstrated that the developed correlation equation is reliable for 573 choosing the most suitable surrounding material to improve the RF heating uniformity in 574 575 various food products. For example, the estimated dielectric constant value of surrounding container was 2.6 with dry soybeans DPs of 3.6-0.26\*j. This was in accordance with our 576 preliminary studies that UI was the lowest when the surrounding material dielectric constant 577 was between 2.5 and 3.5 (Huang et al., 2015c, 2016). Optimum RF heating uniformity in 578 579 other dry products (such as almond, lentil, wheat, walnut, pistachio nut, raisin, and rice) could 580 be achieved with a particular dielectric constant values of the surrounding material.









Fig. 12. Simulated UI of soybean flour with varying sample dielectric constants between (a)
2-5, (b) 6-9, and (c) 10-13 under various container dielectric constants after 5 min RF heating
with an electrode gap of 12 cm.

583

Table 5. The calculated dielectric constants of the surrounding container ( $\varepsilon'_{container}$ ) when the UI obtained the minimum values and the corresponding inputted voltages (V) with varying sample dielectric constants ( $\varepsilon'_{sample}$ ) in each simulation after 5 min RF heating.

$\mathcal{E}_{sample}^{\prime}$	2	3	4	5	6	7
$\mathcal{E}'_{container}$	0.94	1.68	3.13	4.28	5.93	7.07
Simulation voltage (V)	2860	3540	4000	4490	5000	5600
$arepsilon_{sample}'$	8	9	10	11	12	13

$\mathcal{E}_{container}^{\prime}$	8.59	10.03	11.56	13.03	14.36	16.61
Simulation voltage (V)	6280	6820	7430	8060	8630	9300





591

Fig. 13. Dielectric constant correlations between surrounding container and the treated food
products when the simulated UI obtained the minimum values after 5 min heating time in a 12
cm electrode gap.

595 3.6. Effect of loss factor of sample and container on sample UI

Table 6 compares the simulated UIs of soybean flour with different loss factor values of surrounding container and heated sample. UI showed a sharp decrease first and then a constant as a function of container loss factor. The UI decreased from 5.052 to 0.244 when loss factor of the surrounding container decreased from 0.3 to 0.00003 with the fixed sample loss factor of 0.1. It is also observed that when loss factor of sample increased gradually to 5, 601 UI decreased from 0.472 to 0.321. UI was the lowest compared to other loss factor values 602 when loss factors of the surrounding container were given between 0.00003 and 0.0003, 0.01-0.1% of the sample's one. Loss factor of sample describes the amount of electric energy 603 604 converted to heat and the inputted voltages were lower when the sample loss factor decreased (Table 6). It is clear that the heating uniformity of food products could be improved when loss 605 606 factors are small both for the surrounding container and treated samples (Huang et al, 2015b). The UI may not be further reduced due to the small enough loss factor values of the 607 608 polystyrene container (0.0003) used in this study. Therefore, the dielectric constant of 609 surrounding container is the dominating factor to influence the heating uniformity of treated products. This behaviour has been observed for dry soybeans (Huang et al., 2015c, 2016), 610 611 peanut butter (Jiao et al., 2014a, 2015b), shell eggs (Lau, 2015), and wheat flour (Tiwari et al., 612 2011a) subjected to RF treatment.

613

Table 6. Simulated UI of soybean flour with varying loss factor values of sample ( $\varepsilon_{sample}''$ ) and surrounding container ( $\varepsilon_{container}''$ ) under the corresponding inputted voltages (V) after 5 min RF heating with an electrode gap of 12 cm.

$arepsilon_{sample}''$		0.1				1				
$\mathcal{E}''_{container}$	0.3	0.03	0.003	0.0003	0.00003	0.3	0.03	0.003	0.0003	0.00003
UI	5.052	0.584	0.278	0.247	0.244	0.616	0.286	0.245	0.242	0.241
Simulation voltage (V)	9150	8000	8000	8000	8000	2580	2570	2560	2560	2560
${\cal E}''_{sample}$			3					5		
$\mathcal{E}''_{container}$	0.3	0.03	0.003	0.0003	0.00003	0.3	0.03	0.003	0.0003	0.00003
UI	0.619	0.494	0.482	0.481	0.481	0.472	0.336	0.322	0.321	0.321

Simulation										
	1570	1570	1570	1570	1570	1670	1670	1670	1670	1670
voltage (V)										

617

618 3.7. Effect of density of sample and container on sample UI

Fig. 14 indicates that the smaller densities of surrounding container resulted in better RF 619 620 heating uniformities within the samples. UI decreased slightly with increasing densities of the 621 surrounding container but then increased dramatically under each fixed sample density values. Low densities of the surrounding container should provide a better heating uniformity due to 622 the lower density values of heated material resulted in higher RF heating rate (Huang et al., 623 2015b). Table 7 shows the calculated density values of surrounding container in each fixed 624 625 sample density when the minimum values of UI were obtained. The inputted voltage increased with increasing sample density due to the positive correlation between sample 626 density and top electrode voltage (Birla et al., 2008). The general trend of density correlations 627 628 between the surrounding container and treated soybean flour was shown in Fig. 15 for ensuring good RF heating uniformity. Therefore, a linear relationship for densities of 629 surrounding container and treated products was observed with high coefficient of 630 determination ( $R^2=0.98$ ). 631

$$\rho_{container} = 0.358 \rho_{sample} - 68.10$$

633 (10)

It can be inferred from the above results that uniform RF heating of the sample could be achieved when the surrounding container density was far small than the sample's one. The regression equation developed in this study may be further used in other dry products and other applications for heating uniformity improvement. Similar results have been also obtained by Huang et al., (2016) that the temperature uniformity was greatly improved by placing soybean samples in the polystyrene container (with low density) other than the 640 polypropylene container. This suggests that the developed relationship could possibly be applied to other low moisture foods for overcoming edge heating effect and maintaining good 641 product quality. Therefore, in practical applications for an industrial-scale RF system, which 642 643 was equipped with an auxiliary hot air system and conveyor belt, choosing an optimum container material could be effectively minimize the effect of electric field bending and 644 distortion within the corners and edges of food products during the RF treatment. The 645 developed correlation equations are a very effective tool for choosing the most suitable 646 surrounding material to be used in an industrial application. 647





Fig. 14. Simulated UI of soybean flour with four different sample density values (300, 500, 800, and 1000 kg m<sup>-3</sup>) and various container densities after 5 min RF heating with an electrode gap of 12 cm.

Table 7. The calculated density values of the surrounding container ( $\rho_{container}$ ) when the UI obtained the minimum values and the corresponding inputted voltages (V) with varying sample densities ( $\rho_{sample}$ ) in each simulation after 5 min RF heating.

Sample density ( $\rho_{sample}$ , kg m <sup>-3</sup> )	300	500	800	1000
Container density ( $\rho_{container}$ , kg m <sup>-3</sup> )	50	100	210	300
Simulation voltage (V)	3560	4620	5780	6460

655



656

Fig. 15. Density correlations between the surrounding container and treated food products
when the simulated UI obtained the minimum values after 5 min heating time in a 12 cm
electrode gap.

660 4. Conclusions

661 A comprehensive coupled electromagnetic and heat transfer model was developed considering quasi-static electric fields in a 6 kW, 27.12 MHz RF heating system. Simulated 662 temperature distribution in three horizontal layers of soybean flour were found in good 663 agreement with experimental temperature profiles, except for some corners with maximum 664 difference of 5 °C. The validated model was further used to study the effects of DPs and 665 density of sample and surrounding container on sample UI. Simulated results illustrated that 666 667 the RF heating uniformity could be improved when the dielectric constant and density of surrounding container and sample were in accordance with the established relationships. The 668 669 smaller loss factor values for both surrounding container and heated products provided better temperature uniformities. The obtained regression equations may be useful to obtain a better 670 heating uniformity in disinfestations of dry products and for designing an RF treatment 671 672 protocols.

## 673 Acknowledgements

This research was conducted in the College of Mechanical and Electronic Engineering, Northwest A&F University, and supported by research grants from General Program of National Natural Science Foundation of China (31371853) and Program of Introducing International Advanced Agricultural Science and Technologies (948 Program) of Ministry of Agriculture of China (2014-Z21). The authors thank Qian Hao, Hongxue Zhou, Rui Li, Xiaoxi Kou, and Lixia Hou for their helps in conducting experiments.

### 680 **References**

- AOAC. (2002). Official methods of analysis. Gaithersburg, MD, USA: Association of Official Analytical
   Chemists.
- Alfaifi, B., Tang, J., Jiao, Y., Wang, S., Rasco, B., Jiao, S., & Sablani, S. (2014). Radio frequency disinfestation
   treatments for dried fruit: Model development and validation. *Journal of Food Engineering, 120*,
   268-276.
- Ben-Lalli, A., Bohuon, P., Collignan, A., & Méot, J. M. (2013). Modeling heat transfer for disinfestation and control of insects (larvae and eggs) in date fruits. *Journal of Food Engineering*, *116*(2), 505-514.
- Birla, S., Wang, S., & Tang, J. (2008). Computer simulation of radio frequency heating of model fruit immersed in water. *Journal of Food Engineering*, 84(2), 270-280.
- Birla, S., Wang, S., Tang, J., & Hallman, G. (2004). Improving heating uniformity of fresh fruit in radio
   frequency treatments for pest control. *Postharvest Biology and Technology*, 33(2), 205-217.

- 692 Chen, L., Wang, K., Li, W., & Wang, S. (2015a). A strategy to simulate radio frequency heating under mixing
   693 conditions. *Computers and Electronics in Agriculture*, 118, 100-110.
- 694 Chen, L., Huang, Z., Wang, K., Li, W., & Wang, S. (2015b). Simulation and validation of radio frequency 695 heating with conveyor movement. *Journal of Electromagnetic Waves and Applications*, in proof.
- Choi, C., & Konrad, A. (1991). Finite element modeling of the RF heating process. *IEEE Trans. Magnetics*, 27(5), 4227-4230.
- 698 COMSOL material library, COMSOL Multiphysics, V4.3a, (2012). Burlington, MA, USA.
- FAOSTAT. (2013) Food and Agriculture Organization of the United Nations. (Available at:http://faostat.fao.org/
   site/312/default.aspx).
- Finn, S., Hinton, J. C., McClure, P., Amézquita, A., Martins, M., & Fanning, S. (2013). Phenotypic
   characterization of salmonella isolated from food production environments associated with low-water
   activity foods. *Journal of Food Protection*, 76(9), 1488-1499.
- Farag, K. W., Marra, F., Lyng, J. G., Morgan, D. J., & Cronin, D. A. (2010). Temperature changes and power
   consumption during radio frequency tempering of beef lean/fat formulations. *Food and Bioprocess Technology*, 3(5), 732-740.
- Fu, Y. C. (2004). Fundamentals and industrial applications of microwave and radio frequency in food processing.
   *Food Processing: Principles and Applications*, Blackwell, Iowa, USA, pp. 79-100.
- Gao, M., Tang, J., Villa-Rojas, R., Wang, Y., & Wang, S. (2011). Pasteurization process development for controlling Salmonella in in-shell almonds using radio frequency energy. *Journal of Food Engineering*, 104(2), 299-306.
- Geveke, D. J., Kozempel, M., Scullen, O. J., & Brunkhorst, C. (2002). Radio frequency energy effects on microorganisms in foods. *Innovative Food Science and Emerging Technologies*, *3*, 133-138.
- Guo, W., Wang, S., Tiwari, G., Johnson, J. A., & Tang, J. (2010). Temperature and moisture dependent
   dielectric properties of legume flour associated with dielectric heating. *LWT-Food Science and Technology*, 43(2), 193-201.
- Ha, J. W., Kim, S. Y., Ryu, S. R., & Kang, D. H. (2013). Inactivation of Salmonella enterica serovar
  Typhimurium and Escherichia coli O157: H7 in peanut butter cracker sandwiches by radio-frequency *Food Microbiology*, *34*(1), 145-150.
- Hassan, S. M. (2013). Soybean, Nutrition and Health-Chapter 20. Intech open access publisher. Rijeka, Croatia,
   European Union, pp. 453-473.
- Hou, L., Ling, B., & Wang, S. (2014). Development of thermal treatment protocol for disinfesting chestnuts
   using radio frequency energy. *Postharvest Biology and Technology*, 98, 65-71.
- Huang, Z., Chen, L., & Wang, S. (2015a). Computer simulation of radio frequency selective heating of insects in soybeans. *International Journal of Heat and Mass Transfer*, 90, 406-417.
- Huang, Z., Zhu, H., & Wang, S. (2015b). Finite element modelling and analysis of radio frequency heating rate
   in mung beans. *Transactions of the ASABE*, 58(1), 149-160.
- Huang, Z., Zhu, H., Yan, R., & Wang, S. (2015c). Simulation and prediction of radio frequency heating in dry soybeans. *Biosystems Engineering*, 129, 34-47.
- Huang, Z., Zhang, B., Marra F., & Wang, S. (2016). Computational modelling of the impact of polystyrene
   containers on radio frequency heating uniformity improvement for dried soybeans. *Innovative Food Science and Emerging Technologies*, accepted.
- Ikediala, J., Hansen, J., Tang, J., Drake, S., & Wang, S. (2002). Development of a saline water immersion
   technique with RF energy as a postharvest treatment against codling moth in cherries. *Postharvest Biology and Technology*, 24(2), 209-221.
- Jiao, S., Deng, Y., Zhong, Y., Wang, D., & Zhao, Y. (2015a). Investigation of radio frequency heating
   uniformity of wheat kernels by using the developed computer simulation model. *Food Research International*, 71, 41-49.
- Jiao, S., Johnson, J., Tang, J., & Wang, S. (2012). Industrial-scale radio frequency treatments for insect control in lentils. *Journal of Stored Products Research*, 48, 143-148.
- Jiao, Y., Shi, H., Tang, J., Li, F., & Wang, S. (2015b). Improvement of radio frequency (RF) heating uniformity
   on low moisture foods with Polyetherimide (PEI) blocks. *Food Research International*, 74, 106-114.
- Jiao, Y., Tang, J., & Wang, S. (2014a). A new strategy to improve heating uniformity of low moisture foods in radio frequency treatment for pathogen control. *Journal of Food Engineering*, 141, 128-138.
- Jiao, Y., Tang, J., Wang, S., & Koral, T. (2014b). Influence of dielectric properties on the heating rate in free-running oscillator radio frequency systems. *Journal of Food Engineering*, *120*, 197-203.
- Johnson, J., Wang, S., & Tang, J. (2003). Thermal death kinetics of fifth-instar Plodia interpunctella
   (Lepidoptera: Pyralidae). *Journal of Economic Entomology*, 96(2), 519-524.
- Johnson, J., Wang, S., & Tang, J. (2010). Radio frequency treatments for insect disinfestation of dried legumes.
   In Proc.10th Intl. Working Conf. Stored Product Protection (pp. 688-694), Berlin, Germany: Julius Kühn Institut.

- Kim, S. Y., Sagong, H. G., Choi, S. H., Ryu, S., & Kang, D. H. (2012). Radio-frequency heating to inactivate
   *Salmonella Typhimurium* and *Escherichia coli O157: H7* on black and red pepper spice. *International Journal of Food Microbiology*, *153*(1), 171-175.
- Kirmaci, B., & Singh, R. K. (2012). Quality of chicken breast meat cooked in a pilot-scale radio frequency oven.
   *Innovative Food Science and Emerging Technologies, 14*, 77-84.
- Lau, S. K. (2015). Simulation and validation of radio frequency heating of shell eggs. *Dissertations & Theses in Food Science and Technology*, 61, 1-133.
- Liu, Y., Wang, S., Mao, Z., Tang, J., & Tiwari, G. (2013). Heating patterns of white bread loaf in combined
   radio frequency and hot air treatment. *Journal of Food Engineering*, *116*(2), 472-477.
- Llave, Y., Liu, S., Fukuoka, M., & Sakai, N. (2015). Computer simulation of radiofrequency defrosting of frozen
   foods. *Journal of Food Engineering*, 152, 32-42.
- Luechapattanaporn, K., Wang, Y., Wang, J., Tang, J., Hallberg, L. M., & Dunne, C. P. (2005). Sterilization of
   scrambled eggs in military polymeric trays by radio frequency energy. *Journal of Food Science*, 70(4),
   288-294.
- Marra, F., Lyng, J., Romano, V., & McKenna, B. (2007). Radio-frequency heating of foodstuff: Solution and validation of a mathematical model. *Journal of Food Engineering*, 79(3), 998-1006.
- Marshall, M., & Metaxas, A. (1998). Modeling of the radio frequency electric field strength developed during
   the RF assisted heat pump drying of particulates. *Journal of Microwave Power and Electromagnetic Energy*, 33(3), 167-177.
- 771 Metaxas, A. (1996). Foundations of electroheat: a unified approach. John Wiley & Sons, New York.
- Mitcham, E., Veltman, R., Feng, X., De Castro, E., Johnson, J., Simpson, T., Biasi, W., Wang, S., & Tang, J.
   (2004). Application of radio frequency treatments to control insects in in-shell walnuts. *Postharvest Biology and Technology*, *33*(1), 93-100.
- Mohapatra, D., Kar, A., & Giri, S. K. (2015). Insect pest management in stored pulses: an overview. *Food and Bioprocess Technology*, 8(2), 239-265.
- Shrestha, B., & Baik, O. D. (2013). Radio frequency selective heating of stored-grain insects at 27.12 MHz: a
   feasibility study. *Biosystems Engineering*, 114(3), 195-204.
- Singh, P., Satya, S., & Naik, S. (2013). Grain Storage insect-pest infestation-issues related to food quality and safety. *Internet Journal of Food Safety*, 15, 64-73.
- Taylor, W. G., Fields, P. G., & Sutherland, D. H. (2007). Fractionation of lentil seeds (lens culinaris medik.) for
   insecticidal and flavonol tetraglycoside components. *Journal of Agricultural and Food Chemistry*,
   55(14), 5491-5498.
- Tiwari, G., Wang, S., Tang, J., & Birla, S. (2011a). Analysis of radio frequency (RF) power distribution in dry food materials. *Journal of Food Engineering*, 104(4), 548-556.
- Tiwari, G., Wang, S., Tang, J., & Birla, S. (2011b). Computer simulation model development and validation for radio frequency (RF) heating of dry food materials. *Journal of Food Engineering*, 105(1), 48-55.
- Uyar, R., Bedane, T. F., Erdogdu, F., Palazoglu, T. K., Farag, K. W., & Marra, F. (2015). Radio-frequency thawing of food products–A computational study. *Journal of Food Engineering*, *146*, 163-171.
- Vijay, S., Bhuvaneswari, K., & Gajendran, G. (2015). Assessment of grain damage and weight loss caused by
   *Sitophilus oryzae (L.)* feeding on split pulses. *Agricultural Science Digest*, 35(2), 111-115.
- Wang, S., Monzon, M., Johnson, J., Mitcham, E., & Tang, J. (2007). Industrial-scale radio frequency treatments for insect control in walnuts: I: Heating uniformity and energy efficiency. *Postharvest Biology and Technology*, 45(2), 240-246.
- Wang, S., Tiwari, G., Jiao, S., Johnson, J., & Tang, J. (2010). Developing postharvest disinfestation treatments
   for legumes using radio frequency energy. *Biosystems Engineering*, 105(3), 341-349.
- Wang, S., Yue, J., Chen, B., & Tang, J. (2008). Treatment design of radio frequency heating based on insect control and product quality. *Postharvest Biology and Technology*, 49(3), 417-423.
- Wang, S., Yue, J., Tang, J., & Chen, B. (2005). Mathematical modelling of heating uniformity for in-shell
  walnuts subjected to radio frequency treatments with intermittent stirrings. *Postharvest Biology and Technology*, 35(1), 97-107.
- Zhu, H., Huang, Z., & Wang, S. (2014). Experimental and simulated top electrode voltage in free-running
   oscillator radio frequency systems. *Journal of Electromagnetic Waves and Applications*, 28(5),
   606-617.
- 805