PASTEURIZATION OF DEHYDRATED FOOD POWDERS WITH RADIO FREQUENCY HEATING

by

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ABSTRACT

Low-moisture foods with water activity ($a_w \leq 0.6$) associated *Salmonella* outbreaks have become a major food safety concern, which brought attention the food industry, government and scientific community to develop technology for its remediation. Current conventional decontamination methods for low -moisture foods including dehydrated vegetable powders and spices have not been widely accepted due to the public health concern or quality degradation. Radio frequency (RF) heating as an alternative novel technology provides a great chance for pasteurization of dehydrated vegetable powders and spices with reduced come-up time, which allows to design a faster heating process to inactivate *Salmonella* with minimized quality degradation in food products.However, the major challenge for developing an effective RF heating system for microbial inactivation is the non-uniform and unpredictable heating pattern. Thus, the focus of this research was on developing a strategy to improve heating uniformity by investigating the influence of multiple factors including dielectric properties (DP),moisture content, electrode gap and packaging material on the heating rates and uniformity in various low-moisture foods and assessing the effectiveness of RF heating pasteurization for *Salmonella* on the food powders were used corn flour, paprika, white pepper and cumin powder, and evaluate *Enterococcus faecium* NRRL B-2354 (*E. faecium*) as a surrogate for RF heating inactivation of *Salmonella*. DP values of various vegetable powders and spices were determined using a precision LCR meter and liquid test fixture at frequency ranging from 1 to 30 MHz.RF heating uniformity and temperature profiles of each sample as treated under a to RF heating system (27.12-MHz, 6-kW) were obtained with an infrared camera and data logger with a fiber optic sensor. Finally, the effectiveness of RF heating on pasteurization of *Salmonella* and its potential surrogate *E. faecium* was evaluated. In this study, RF heating can be used effectively to pasteurize corn flour, paprika and white pepper. Additionally, a combination of RF heating and post-freezing storage at -20°C for 48h helped to reduce survival of microorganisms, and *E. faecium* was found as a potential surrogate of *Salmonella* on the powders were used corn flour, paprika, white pepper and cumin powder to validate RF heating process.

INDEX WORDS: Radio Frequency Heating, Low-Moisture Foods, *Salmonella*, *Enterococcus faecium* NRRL B-2354, Dehydrated Food Powders

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A Dissertation Submitted to the Graduate Faculty of The University of Georgia in Partial Fulfillment of the Requirements for the Degree

DOCTOR OF PHILOSOPHY

ATHENS, GEORGIA

2018

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May 2018

DEDICATION

To my great parents Mrs. Sebahat and Mr. Ahmet, my brothers, Mr. Yakup and Mr. Yasin, for their unconditional love and support

ACKNOWLEDGEMENTS

I would like to gratefully acknowledge my major advisor, Dr. Fanbin Kong for his mentorship, guidance and financial support during my Ph.D. program. It has been such an honor to be your graduate student. I also extend my sincere gratitude to my Co-Advisor Dr. Rakesh K. Singh and other committee members, Dr. Jinru Chen, Dr. Sudhagar Mani, and Dr. Abhinav Mishra, for challenging me and providing me with thoughtful feedback. Without a doubt, I have grown as a researcher and as a person during my doctoral studies because of your influence. I also like to thank Dr. Samir Trabelsi for his help and advice to improve my dissertation research.

I would like to thank to my great lab mates, Duc, Luna, Yiwen, Lingling, Nakia, Jiannan, Damla and Anuj for their splendid friendship and mental support. I also like to thank Dr. Ali Halalipour and Dr. Bahadir Namdar for their endless friendship and support during our challenging times. Last but not least, I would like to express my sincere appreciation to my family for their love, encouragement, and support. They always believed in me. I also gratefully acknowledge the Turkish Ministry of National Education Council for providing the chance for my graduate studies in U.S.A. Finally, I want to thank my family, advisors and friends for their patience, strength and support during my time in graduate school.

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CHAPTER 1

INTRODUCTION

Many foods eaten today are known as low water activity ($a_w \leq 0.6$) foods, which are foods with moisture content less than 20% w.b. (wet basis). These foods include dried vegetables and fruits, spices, chocolate, peanut butter and peanut-containing foods, raw nuts, children's snack foods, hydrolyzed vegetable protein, and powdered infant formula. Even though there is a common misconception that low moisture foods are unable to support the growth of foodborne pathogens (Leistner and Rödel, 1976), a low number of *Salmonella* in low-moisture foods can still survive and impact a large number of people (Gill et al., 1983; Greenwood and Hooper, 1983; Scott et al., 2009)). Recently, a number of outbreaks of salmonellosis have been associated with composition of low-moisture foods and it has become a major safety issue (CDC, 2007; (Finn et al., 2013; Gill et al., 1983). A large number of clinical cases in young children were reported after consuming such snack foods (Lambertini et al., 2016; Rico et al., 2010; Schweiggert et al., 2007; Waje et al., 2008). Spices such as white and red peppers are commonly used seasonings and ingredients, with moisture content at around 5-15% (w.b.) and a_W 0.1-0.2 (Doymaz and Pala, 2002). In 2009, a salami product made with contaminated black and red pepper spice caused a multistate outbreak of *Salmonella* Montevideo infections in USA in which hundreds of people became ill (CDC, 2010;(Rychlik and Barrow, 2005; Tiozzo et al., 2011). In this case, the presence of microorganism in low moisture foods must be prevented before eating or importing. However, to eliminate the presence of *Salmonella* in these foods is more difficult than in high moisture content foods with the current conventional thermal methods or other applications

(Harris, 2008) because thermal resistance of *Salmonella* in dry foods is higher than in a wet environment, and chemical residuals are not allowed (Harris, 2008). Due to long processing time requirements and quality concerns of the conventional pasteurization methods, radio frequency (RF) heating offers a potential for short time heating and high quality, therefore; improving final product quality and reducing treatment cost. This study was designed to develop a RF-based pasteurization technology for dried vegetables and spices with maintained product quality. The outcomes of the project will include determining and optimizing the processing technology and a strategy for efficient pasteurization, the kinetic parameters of thermal resistance of *Salmonella* in dried vegetables and spices, the dielectric properties of selected products, and the quality properties and temperature distribution in dried foods under RF heating pasteurization process. The proposed research will also generate guidelines for selection of proper packaging material with improved stability during RF heating.

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CHAPTER 2

LITERATURE REVIEW

Introduction

Low moisture foods have been considered as microbiologically low risk foods because they are not supporting to grow of pathogenic microorganisms at water activity ($a_w \leq 0.6$) (Leistner and Rödel, 1976), such foods are not exposed to any further thermal processing before reaching costumers. However, a recent study reported that pathogenic microorganisms like *Salmonella* do not require any growth to survive and cause outbreaks under such insufficient environments (Scott et al., 2009). Recent *Salmonella* outbreaks associated with consumption of low moisture foods have brought great attention to government agencies and researchers to establish regulation guidelines to assure the microbial safety and to address public and industry concerns (USDA, 2009). Due to the large number of *Salmonella* related outbreaks all over the United States, there is an urgent need of a pasteurization technology for bacterial inactivation in low moisture foods. Traditional thermal pasteurization methods are not able to eliminate foodborne pathogens with maintaining food quality in low moisture foods because of longer processing time to achieve the required pasteurization levels due to their poor thermal conductivity properties. Radio frequency (RF) heating as a volumetric heating have shown a great potential to inactivate foodborne pathogens in low moisture foods with shorter processing time while maintaining food quality (Gao et al., 2011). The objective of this review is to provide a basic knowledge about *Salmonella*, *Enterococcus faecium,* RF heating and dielectric properties and strategies for heating uniformity improvement.

Salmonella outbreaks in low moisture foods

Salmonella is one of the most challenging food borne pathogens which is responsible over 1 million illness and approximately 450 deaths in the United States annually (Johnson et al., 2014). The majority of these illness related to foods including animal origin or raw agricultural products occurs as a result of *Salmonella* contamination. Historically, low moisture foods with $a_w \leq 0.6$ have been considered as low risk or safe foods which do not support growth of any microorganism or considered as carrier of any pathogenic bacteria. However, *Salmonella* can survive under insufficient environments for extended storage time (Keller et al., 2013; Podolak et al., 2010; Van Doren et al., 2013), and may cause infection with a population counts as low as ≤ 1 CFU/g of sample in many low moisture foods (Gill et al., 1983; Greenwood and Hooper, 1983; Scott et al., 2009). *Salmonella* contamination in low moisture food may occur during the harvest due to contaminated soils or cross-contamination because of poor sanitation practices, inadequate plants, equipment design and inappropriate maintenance during processing (Carrasco et al., 2012). Furthermore, the presence of water in processing environment is one of the most important threats for low moisture foods which causes *Salmonella* contamination and supports growth and spread of microorganism subsequently in the plant resulting in further product contamination. As a bacterium has attached a dry-food processing environment, it is a challenge to remove immediately

Most of low moisture foods are categorized as ready-to-eat (RTE) foods including chocolate, salami, nut, dry vegetables or fruits and spices which do not undergo any further cooking process prior to consumption. A big majority of low moisture foods including oat cereal (CDC, 1998), peanut butter (CDC, 2007), raw almond (CDC, 2004), various species including red, white and black pepper (CDC, 2010), paprika (Lehmacher et al., 1995) and cumin powder (Moreira et al.,

2009) have been implicated in food borne pathogens, especially, *Salmonella* which leads to many product recall and illnesses. From 2007 to 2012 on the CDC website, 119 recalls linked with different low moisture foods including pet food, powdered infant formula, peanut butter, spices, dry nuts, dry milk, and seeds in United States were reported.

The majority of survival of *Salmonella* in low moisture foods is depended on a_w which plays an important role on the interaction between microbial cell and water which reflects the ability of bacterial growth (Bowman et al., 2015). The minimum a_w for growth of most microorganisms and mycotoxin production by molds was reported as 0.87 and 0.80, respectively (Beuchat et al., 2013). It was examined that reduced a_w results in decrease of microbial cells growth, however, it also causes the increase in heat resistance and survival ability of *Salmonella* in low moisture foods (Beuchat et al., 2013). Therefore, it is a challenge to control *Salmonella* in a dry environment. Other factors such as food matrix, storage temperature or pH also affect the amount of survival in food. For example, Miller et al. (1972) reported the survival population of *S.* Typhimurium during spry dying as inlet and outlet air temperature were 165 and 225°C, and 67 and 93°C, respectively. Additionally, Park et al. (2002) showed the survival of *Salmonella* in spray dried milk and chalk during the 120 days and 6 months, respectively, under storage conditions. Therefore, low moisture foods are not called anymore as safe foods and need to be decontaminated by an effective process. To ensure the safety of low moisture foods, a minimum inactivation in the range of 10^5 to 10^7 CFU/g is required according to strain and host to cause foodborne illness (Hara-Kudo and Takatori, 2011; Teunis et al., 2010).

Enterococcus faecium* as a surrogate of *Salmonella

Food and Drug Administration (FDA) requires food industry to control and validate their processing methods to secure food safety (FDA, 2015). However, using actual foodborne pathogens like *Salmonella* in food process environment resulting further health risk for worker or food products is not allowed, therefore, FDA recommends using non-pathogenic surrogate microorganisms with equal or slightly higher thermal resistance than that of pathogen bacteria as an alternative way to control their process (Jeong et al., 2011). It is important to identify a valid surrogate microorganism for determining efficiency of processing methods on decontamination of foodborne pathogen in food product. *Enterococcus faecium* NRRL B-2354 (*E. faecium)* is known as a Gram-positive and non-pathogenic bacterium (Byappanahalli et al., 2012), which has been commonly used as a potential surrogate for *Salmonella* to validate thermal processing methods in the production of low-moisture foods including almonds (Almond Board of California, 2007), almond kernels (Villa-Rojas et al., 2013), wheat flour (Liu et al., 2018; Tiwari et al., 2011), oat flour (Verma, 2017), chicken meat powder, pet food and savoury seasoning (Rachon et al., 2016). The organism has showed a great potential as a surrogate of *Salmonella* with a thermal resistance higher than *Salmonella* under the same environmental conditions. The use of *E. faecium* as a surrogate for validation of microbial inactivation has been evaluated in different pasteurization techniques including extrusion (Bianchini et al., 2012).

Dielectric properties of food

Material with high electrical resistivity is defined as dielectric or insulator. A good dielectric material needs to be a good insulator or capacitor. Dielectric properties are known as dielectric constant (ε') and dielectric loss factor (ε''). They are also defined as relative permittivity which is fundamental properties of materials identifying the interaction level between materials and electric field or designating their ability for how to reflect, store or transmit electromagnetic energy as placed in an electromagnetic area. The dielectric properties can be stated as:

$$\varepsilon = \varepsilon' - j\varepsilon'' \qquad \text{Eq. (1)}$$

where ε represents the complex permittivity (where $j = \sqrt{-1}$), dielectric constants (ε') is defined the ability of material for storing electromagnetic energy and dielectric loss factor (ε'') is expressed the ability of a material to convert the electromagnetic energy into heat.

The calculation of dielectric properties of materials is based on the capacitance (C_p) and resistance (R_p) of material in the range of applied frequency, which are expressed as (Agilent Technologies, 2000);

$$\varepsilon' = \frac{tC_p}{A\varepsilon_0} \qquad \text{Eq. (2)}$$

$$\varepsilon'' = \frac{t}{2\pi f R_p \varepsilon_0 A} \qquad \text{Eq. (3)}$$

where t is the gap (m) between electrodes of the test fixture, C_p is capacitance (F), R_p is the resistance (Ω), f is the frequency (Hz), ε_0 is the permittivity of vacuum (8.854×10^{-12} F m^{-1}), and A is the electrode area (m^2).

The another main parameter which needs to be determined to apply radio frequency (RF) or microwave (MW) is the penetration depth which can be defined as depth into a sample where the power has dropped to 1/e (e=2.718) of the power value at the surface (Guan et al., 2004). The penetration depth is a function of dielectric properties of food material, and it can be calculated using the following equation (Buffler, 1993; Von Hippel, 1954)

$$d_p = \frac{c}{2\pi f\sqrt{2\varepsilon'\left(\sqrt{1+(\tan\delta)^2}-1\right)}}$$ Eq. (4)

where, c is the speed of light in air (3×10^8 m s^{-1}), f is the frequency (Hz) and $\tan\delta$ is defined as ($\frac{\varepsilon''}{\varepsilon'}$), ε' and ε'' are the measured dielectric constant and dielectric loss factor values of materials.

The dielectric properties are mainly depending on various factors including frequency (MHz), temperature (°C), moisture content (%, wet or dry basis), food composition or density (g cm^{-3}). In the range of RF at room temperature, dielectric properties of many low moisture foods with a_w ≤ 0.6 and moisture content < 20 % (w.b.) are relatively low less than < 20. As the applied frequency less than 100 MHz which is in the range of RF, the prevailing mechanisms of energy loss in material change as ionic conduction, bound water relaxation and Maxwell -Wagner effect (Figure 2.1) (Feng et al., 2002; Harvey and Hoekstra, 1972; Jiao, 2014; Metaxas and Meredith, 1983). The ionic conduction effect is not effective as much as other two dominating mechanisms on dielectric properties due to the lack of free water in low moisture foods. It is known that the mobility of bound water is in between ice and free water, therefore, the relaxation frequency of bound water in sample is lower than that of free water at room temperature as applied frequency is less than 100 MHz (Ryynänen, 1995). In addition, Maxwell-Wagner's effect is known as interfacial loss mechanism, which occurs as a result of substitute aggregation of charged ions at the integrate of heterogeneous dielectrics. That happens as a conducting phase is attached to non-conductive phase, the charged ions accumulate at the integrate and move between two close integrates in an oscillating filed (Meakins, 1961; Metaxas and Meredith, 1983). It could be interpreted as the contribution of both bound water effect and Maxwell-Wagner's effect on dielectric properties is less than that of free water and ionic. It may explain the reason of lower dielectric properties in low moisture foods. Furthermore, Kannan et al. (2013) reported that an increase in temperature of sample enhances mobility of free ions and activity of their polarization, which results in charging more electrons in the sample leading to an increase in the dielectric properties. Moreover, beside the water content of foods, all other food composition facts such as carbohydrates, protein, ash and fat, which are not dissolved in water, also play an important role on the dielectric properties (Calay et al., 1994; Venkatesh and Raghavan, 2004). Recent studies showed that dielectric properties of low moisture foods with high fat content such as butter and butter oil or rice slurry decreased as fat content increased (Ahmed et al., 2007a; Ahmed et al., 2007b).

Current practice in decontamination of low moisture foods

Current sterilization and pasteurization practices, such as conventional thermal processing involving conductive heat transfer to inactivate pathogens, appear inadequate to address the contamination problem in low moisture foods. Compared with intermediate and high moisture foods, *Salmonella* is not easily eliminated from dry foods by thermal treatment. Thermal tolerance of *Salmonella* increases with decreasing water activity, although the relationship is more complicated when it comes to low moisture systems (Mattick et al., 2000). It is known that thermal resistance of *Salmonella* is several orders of magnitude higher in a dry environment than in a wet environment (Archer et al., 1998). Subsequently, commercial steam treatment of packaged low moisture foods requires long time heating leading to quality deterioration with significant loss of color and flavor. The material at the periphery of the container receives much more severe treatment than required to achieve commercial sterility, leading to severely degraded quality at the overheated corners and edges of low moisture foods. Several other decontamination methods have been developed to reduce the microbial loads of low moisture foods, including fumigation with ethylene oxide and irradiation. Ethylene oxide, however, is generally regarded as a carcinogen; irradiation has not found wide consumer acceptance although it is allowed for the decontamination of foods in numerous countries (Schweiggert et al., 2007). Accordingly, there is a need for the development of safe technologies for disinfecting low moisture foods while maintaining the quality.

Development of effective decontamination techniques requires in-depth understanding in the mechanisms of microbial inactivation in low moisture foods. A number of researchers have reported the complexity in the interactions among temperature, water activity, water motility, and food composition in the low moisture foods. The influence of water activity in heat tolerance of microorganism is dependent upon temperature of the foods; Mattick et al. (2000) observed that *Salmonella* at low a_w (0.65) were more heat tolerant at temperatures above 70 °C than when at higher a_w (0.90) but the reverse was true at lower temperatures. Moreover, the effect of a_w in heat tolerance of *Salmonella* is affected by product composition (Finn et al., 2013). (Farakos et al., 2013) investigated microbial inactivation mechanisms in the low moisture foods and found that the thermal resistance of microorganisms in the low water activity foods is not only affected by the a_w but also by water mobility. However, more study remains to be done to understand the survival, growth and inactivation kinetics of *Salmonella* in low moisture foods as affected by multiple factors in order to develop efficient decontamination technology.

Radio frequency heating and its application in low moisture foods

Dielectric heating, that is, microwave and radio frequency (RF) heating, offers the possibility of fast heating in solid and semi-solid foods. An advantage of microwave and RF heating over the conventional thermal processing is the rapid heating by direct interaction between electromagnetic waves and foods that are hermetically sealed in packages (Gao et al., 2010; Lagunas-Solar et al., 2007; Marra et al., 2009; Tiwari et al., 2011). The uniformity of microwave and RF heating need also to be improved to some extent. Thus, microwave and RF pasteurization or sterilization involves short time heating that can potentially protect the product quality (Jiao et al., 2012). The main difference between the RF and microwave is frequency (or wavelength). RF heating involves lower frequencies (13.56, 27.12, and 40.68 MHz) and thus longer wavelengths, and deeper penetration depth compared with those of microwaves at 915 or 2450 MHz. Therefore, the RF heating is particularly useful when applied to institutional-sized packaged food products because of its deep penetration. There have been numerous studies supporting the RF heating as a new thermal treatment method for heating, pasteurization, drying and insect control for relative high-water activity foods such as fruits, meat, juices, milk, nuts (Jiao et al., 2012; Luechapattanaporn et al., 2005; Piyasena et al., 2003; Wang et al., 2012; Wang et al., 2003; Zhu et al., 2012). A recently published paper (Kim et al., 2012) has looked at the potential of RF pasteurization process for black and red pepper spices, however, this study only investigated effects of the RF treatment time and the maximum reduction of S. Typhimurium was 4.29 log CFU/g.

The dielectric properties and a_w of samples, the influence of multiple factors in the heating rates and uniformity of the RF process and strategy to improve heating uniformity involved in the RF heating. And inactivation effect of *Salmonella* has not really been investigated. To our knowledge, no systematic study has been done in the application of the RF process in the pasteurization of dried vegetables and paprika, cumin powder and white pepper. While the RF holds potential for short time heating, thus improving quality attributes including color, texture, flavor and nutritive properties of final products, major challenges for the RF processing are the non-uniform and often unpredictable heating patterns.

The electromagnetic field can be influenced by many factors such as food properties, package geometry, and location of the product inside the RF applicator; it is difficult to predict the heating patterns of the food inside the packages. Since non-uniform heating leads to cold and hot spots within the food, it is critical to find ways to understand and improve uniformity in the heating pattern, including the temperature distributions in order for the RF processes to satisfy food safety requirements for dried vegetables and spices (Tiwari et al., 2011). Moreover, pre-packaged foods subjected to the RF processes may induce adverse alterations on the food packages, which may cause safety problems in the following storage & distribution phase. Thus, it is also critical to study how RF electromagnetic waves interact with different packaging materials, subsequent changes in the physical and chemical properties of in the materials and the influence on food quality and safety.

Improving efficiency of radio frequency heating in microbial inactivation

Compared with high water activity foods, more severe RF conditions are required for substantial microbial reduction in low a_w foods, which could in turn induce changes in quality properties. A combination of RF heating and other technology may reduce the heating severity necessary for the RF heating process, thus improving quality retention. One possibility is to combine post-freezing storage process for inactivating sublethal microorganisms. In higher moisture foods, moist heat is responsible for inactivation of the bacteria through denaturation of proteins in which sufficient water is required to hydrate proteins (Mattick et al., 2000). For low moisture foods, insufficient water is present to denature proteins, so the inactivation of microorganisms is associated with damage to the outer cell membrane through lipid fluidization (Rychlik and

Barrow, 2005). The bacteria with damaged outer cell membrane are called sublethally injured microorganisms because such cells can auto-recover from the outer membrane damage and become active when sufficient moisture is present. In the RF heating process, microorganisms could be subjected to sublethal temperatures due to the non-uniform distribution of temperatures, especially in the low temperature area. Therefore, RF treatments combined with other preservation methods may enhance microbial inactivation in a synergistic manner through the inhibition of repair processes and recovery of sublethally injured microorganisms.

The RF heating has caused damages in the cell membranes, and the sublethal microorganisms were sensitive to freezing. Thus, post-freezing storage treatment is a processing technique that improves the efficacy of microbial inactivation in low moisture foods when used in conjunction with the RF heating. When the RF heating is used in combination with post-freezing storage process, the strength of applied the RF heating could be reduced and will contribute to better retention of quality of low moisture foods. Despite of improved heating uniformity as compared to microwave heating, the non-uniformity and run-way heating are still major challenges to apply RF heating pasteurization system in food industry. The variation in temperature distribution of food during RF heating can lead to overheated hot area causing severe quality deterioration and under-heated cold spot. Several researches have been conducted to improve RF heating uniformity using alternative ways. Wang et al. (2010) reported that as legumes heated by RF oven using combination of forced hot air and shaking container on a conveyor belt resulted in decrease between hot and cold spot temperatures. Combination of hot air heating or preheating with hot water with the RF process may improve temperature uniformity since hot air and hot water can contribute to fast increase in the temperatures in the periphery area and surface (Tiwari et al., 2008). Studies are needed to find optimum conditions for those technologies in combination with the RF technology in order to improve temperature uniformity and reduce heating time. For example, when preheating step is involved, the optimum preheating temperature and time as well as the influence in the following the RF parameters are all important for designing an efficient thermal inactivation system.

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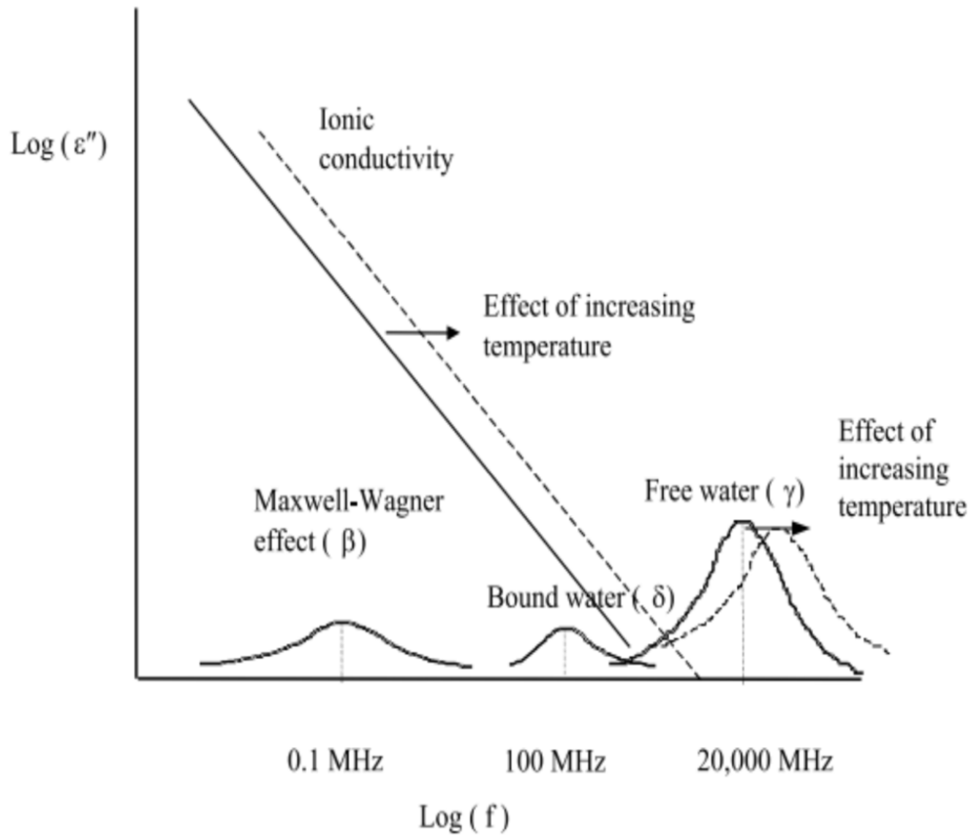

Figure 2.1. Dominating mechanisms on the dielectric loss factor of food materials as influenced by frequency (Feng et al., 2002; Harvey and Hoekstra, 1972; Jiao, 2014; Metaxas and Meredith, 1983)

CHAPTER 3

DIELECTRIC PPOPERTIES OF DRIED VEGETABLE POWDERS AND THEIR TEMPERATURE PROFILE DURING RADIO FREQUENCY HEATING [1]

[1]S. Ozturk, F. Kong, R.K. Singh, S. Trabelsi. Accepted by. *Journal of Food Engineering*. Reprinted here with permission of the publisher.

Abstract

Dielectric properties of selected vegetables powders as influenced by moisture content (MC), compaction density and temperature were determined using a precision LCR meter and liquid test fixture at radio frequency (RF) ranging from 1 to 30 MHz. The RF heating rate of samples was evaluated using a 27.12-MHz, 6-kW RF oven.

The results showed that dielectric properties, namely, the dielectric constant, ε', and loss factor, ε'', of vegetable powders were influenced by MC, compaction density and temperature of the samples and the RF frequency. Both the dielectric constant and loss factor increased with increasing MC and temperature but decreased with increasing frequency. Additionally, dielectric properties of samples increased with compaction density to a peak, then decreased. The relationship between MC, temperature and dielectric properties of broccoli powder at 13.56 and 27.12 MHz can be described by quadratic models with high correlation coefficients ($R^2 > 0.96$). The RF heating rate in samples increased linearly with dielectric loss factor and MC. The information provided in this study is useful to develop an effective RF heating strategy to pasteurize dried vegetable powders.

Keywords: Radio frequency heating; Broccoli powder; Dielectric properties; Heating rate

Introduction

Low-moisture foods, with water activity (a_w) level less than 0.7 (Blessington et al., 2013), are commonly used for various applications in food industry. In the last decade, *Salmonella* contaminated low-moisture foods, including powdered vegetables and infant formula, dry fruit and nuts, spices etc., have become a major safety concern (CDC, 2007, 2010); Sotir et al. (2010) reported an outbreak of *Salmonella* Wandsworth and Typhimurium in vegetables powders such as broccoli powder used for coating of snacks. The high heat resistance of *Salmonella* in low-moisture foods makes pasteurization of dried foods a difficult task. The conventional pasteurization methods such as steam and hot air heating require long processing times due to the slow heating rate, leading to severe quality degradation. RF heating is a volumetric heating with the radiation frequency ranging from 3kHz to 300 MHz, but only 13.56, 27.12 and 40.68 MHz are used for industrial, scientific and medical applications (Wang and Tang, 2001; Sotir et al., 2010). When the dielectric materials are exposed to an alternating electric field at RF and microwave (MW) frequency ranges, they directly convert electrical energy into heat. Therefore, RF heating is more rapid than conventional heating, requiring less process time that can result in improved final product quality and reduced treatment cost (Guo et al., 2011; Nelson, 1996). RF heating systems have been applied in the food industry for various applications such as disinfestation, enzyme inactivation, pasteurization, sterilization, and insect control (Gao et al., 2011; Lagunas-Solar et al., 2007; Manzocco et al., 2008). Because of its fast heating rate and relatively low cost, RF heating can be applied as an alternative pasteurization method for low-moisture foods, including dried broccoli powder, chili powder and onion powder, tapioca flour, and potato starch.

To develop an effective RF pasteurization method for food products, it is important to understand dielectric properties, the major factor characterizing the interaction between the electromagnetic energy and the food. Several studies have been conducted to investigate the dielectric properties of various food powders such as grain seeds, flour, and coffee (Berbert et al., 2001; Lawrence et al., 1990; Nelson and Trabelsi, 2006; Nelson, 1984; Shrestha and Baik, 2015; Trabelsi et al., 1998). The dielectric properties of materials and the RF heating rate are mainly dependent on MC, temperature, and bulk density of sample, and applied frequencies (Piyasena et al., 2003). The influence of frequency, temperature and MC on dielectric properties of chickpea, legume and chestnut flour, and red pepper were reported (Guo et al., 2008; Guo et al., 2010; Guo et al., 2011; Guo and Zhu, 2014; Zhu et al., 2012a). However, there is no information published in the dielectric properties of dried vegetable powder as well as their heating rate during RF treatment. It is also important to understand how compaction densities affect the dielectric properties of food powder, as it can change greatly when subjected to compression pressure, which is frequently encountered during storage and transportation.

The objective of this study was to investigate the dielectric properties of selected vegetables powders, including broccoli, chili and onion powder, tapioca flour, and potato starch, and determine the temperature increase profile during RF heating. Different factors including RF frequency (ranging from 1 to 30 MHz), MC (6.9-14.9%, w.b.), temperature (from 20 to 80°C), and compaction density (0.14 to 0.88 g/ml) were studied for their influence on dielectric constant, loss factor and penetration depth. Mathematical models were developed describing broccoli powder's dielectric properties as function of MC and temperature at selected frequencies. The temperature history profiles of the vegetable powders at 27.12 MHz were determined, and the heating rate of RF was evaluated as a function of MC and dielectric loss factor. The information obtained from this study is expected to help develop guidelines for using

RF technology to pasteurize dried vegetables.

Material and methods

Physical characterization of vegetable powders

Broccoli powder was obtained from Z Natural Food Products (West Palm Beach, FL, 33407). Chili and onion powders, tapioca flour, and potato starch were purchased in a grocery store. The initial MCs of the food powders, determined by drying the samples in a vacuum oven at 105 °C for 16 h (AOAC, 1998), were for onion powder 1.4%, broccoli 3.9%, tapioca flour 5.7%, chili 7.8%, and potato starch 12.6%. The pictures of the four products are shown in Figure 3.1.

To study the effect of moisture, samples of broccoli powder with MC 6.9, 9.1, 12.2, and 14.9% were obtained by spraying distillated water to the powder. The mixtures were stored in plastic containers for two days at 4 °C and shaken twice in a day to obtain a uniform moisture distribution throughout the sample.

Determination of dielectric properties of vegetable powder

The dielectric properties of the vegetable powders were determined by measuring the parallel capacitance (C_p) and resistance (R_p) with an Inductance Capacitance and Resistance (LCR) meter (4285A, Agilent Technologies, Palo Alto, CA) and a dielectric liquid test fixture (16452, Agilent Technologies, Palo Alto, CA). Detailed information about the system was presented in the paper of Izadifar and Baik (2008) and Shrestha and Baik (2013). All parts of the test fixture were washed with distillated water and completely dried before and after each test. To minimize the biased error and random error, the system was calibrated before taking measurement. For each test, vegetable powder (2 g) was placed into the test fixture, which was then tightly closed. The test fixture was placed into a temperature chamber (625G, Thermo Fisher Scientific Inc., Waltham, MA, USA) and connected to the LCR meter by its BNC connecter as described in the

Figure 3.2. The sample was heated in the chamber for 50 min to reach target temperatures between 20 to 80 °C with 10 °C interval. The change in temperature of the sample during heating was recorded using a data logger with fiber optic temperature sensor (Fiso Tech. Inc., Quebec, Canada). The C_p and R_p of the vegetable powders were measured with changing frequency in a range from 1 to 30 MHz at each temperature.

The obtained values of C_p and R_p by the LCR meter were used to calculate the values of dielectric constant ε′ and loss factor ε″ using the following equations (Agilent Technologies, 2000; Halliday, 2001; Von Hippel, 1954):

$$\varepsilon' = \frac{tC_p}{A\varepsilon_0} \quad \text{Eq. (1)}$$

$$\varepsilon'' = \frac{t}{2\pi f R_p \varepsilon_0 A} \quad \text{Eq. (2)}$$

where, t is the gap (m) between electrodes of the test fixture, C_p is parallel capacitance (F), R_p is the resistance (Ω), f is the frequency (Hz), ε_0 is the permittivity of vacuum (8.854×10^{-12} F m^{-1}), and A is the electrode area (m^2).

To determine influence of compaction density on dielectric properties of selected food powders, different sample weight ranging from 0.5 to 3 g were placed into the test fixture, which has a 1.5 mm depth fixed by a spacer. The sample was tapped 10 times to spread uniformly and compressed by the top of the test fixture to squeeze the air out from inside powder. Then the test fixture was tightly closed, and its inlet and outlet were sealed from inside to prevent air and moisture escaping from the fixture. The compaction density (g mL^{-1}) was calculated by the sample weight divided by the volume, which equals to $\pi \cdot (0.01m)^2 . 0.0015m = 4.71\times10^{-7}$ m^3, where 0.01 m is the radius of inside bottom of the fixture, 0.0015 m is the depth.

Determination of power penetration depth (*d_p*)

Guan et al. (2004) defined the power penetration depth of RF as the distance below a material surface where the power has decreased by 1/e (e=2.718) of the power value at the surface. The penetration depth is an important parameter to evaluate heating uniformity and design a RF heating system. It can be calculated using the following equation (Buffler, 1993; Von Hippel, 1954):

$$d_p = \frac{c}{2\pi f\sqrt{2\varepsilon'\left(\sqrt{1+(\tan\delta)^2}-1\right)}} \qquad \text{Eq. (3)}$$

where, c is the speed of light in air (3×10^8 m s^{-1}), f is the frequency (Hz) and tanδ is defined as ($\frac{\varepsilon''}{\varepsilon'}$) $\varepsilon'$and $\varepsilon''$are the measured dielectric constant and dielectric loss factor val- ues of broccoli powder sample, respectively. Using the value of ε' and ε'', the power penetration depth d_p (m) of RF at frequencies of 13.56 and 27.12 MHz in the dry food samples was calculated at temperatures from 20 to 80 ℃ for all moisture levels and for different compaction densities.

Temperature history of vegetable powder during the RF heating

The RF heating rates in the different vegetable powders and broccoli powder with different MCs were evaluated using a 27.12-MHz, 6-kW RF oven (COMBI 6-S, Strayfield International, Wokingham, UK). The samples were placed into a small plastic bottle (inner diameter = 2.9 cm) and placed between the parallel electrodes in the RF heating system. The height of sample in the plastic bottle was 4.8 cm. The gap between two electrode plates in the system was fixed as 9 cm.

The samples were heated individually to reach 80 to 90 °C, and then the RF oven was turned off. The samples were kept inside the oven for a couple of minutes before moved out for inspection. For real-time temperature measurement and heating rate determination, a fiber optic temperature sensor (Fiso Tech. Inc., Quebec, Canada) was placed in center of the plastic bottle to monitor the change in temperature of the samples during the RF heating (Figure 3.3). The temperature was recorded every 5 s using a data logger (UM14, Universal Multichannel Instrument, Fiso Tech. Inc., Canada) connected to the fiber optic temperature sensor.

Statistical analysis

All measurements, including dielectric properties and heating rates, were done in triplicate. The mean value and standard deviation (SD) were reported. The regression equation and correlation coefficients (R^2) between dielectric properties and heating rates and moisture, temperature, and compaction density were derived using SAS 9.4 software and Excel 2010.

Results and Discussion

Dielectric properties of vegetable powders at different frequencies

The dielectric properties of vegetable powders at room temperature (22±3°C) with applied frequency are shown in the Figure 3.4. It shows that vegetable powders have the different values of dielectric constant and dielectric loss factor, and the values decreased as the frequency increased from 1 to 30 MHz. The dielectric constant values follow the order chili powder>potato starch>tapioca flour>Broccoli powder>onion powder. As seen in the Figure 3.4, the highest values of dielectric constant and loss factor were 6.85 and 0.39, respectively, for chili powder. It is widely known that MC is the one of the most important parameters affecting the dielectric properties of foods. The decreasing order in ε′ and ε″ of different food powders generally agrees with their MC, except for chili powder and potato starch, as potato starch has a higher MC (12.9 % w.b) than chili powder (7.8%) but smaller ε′ and ε″ (Figure 3.4). This could be explained by the fact that the dielectric properties are not only affected by MC, also by bulk density and particle size of the product (Nelson 1996). Additionally, the lowest dielectric constant values were 3.11 for broccoli (MC: 3.9%) and 2.93 for onion powder (MC: 1.4%), respectively. The dielectric loss factors follow the similar order as dielectric constant but are very low for all the samples due to the low moisture content. The ε″ for broccoli powder and onion powder was not shown in the figure because the values were too low, and the instrument could not give stable values.

The decrease in dielectric constant and loss factor in food with frequency may be explained by the changes in ionic conduction and mobility of ions in sample. Previous studies indicated that ionic conduction is dominant mechanism for dielectric heating below 300 MHz (Ryynänen, 1995), and ionic conduction decreases with increase in frequency, causing a decrease in value of ε″ (Shrestha and Baik, 2013). Similar trends were reported for dielectric constant and loss factor of different low moisture food samples including chickpea, green pea, lentil and soybean flour (Guo et al., 2011) and wheat flour (Nelson and Trabelsi, 2006), and the dielectric constant and loss factors for chickpea, green pea, lentil and soybean flour ranged from 5 to 10, and 0.1 to 1.6, respectively.

Effect of compaction density on dielectric properties and penetration depth

During packaging and transportation, the packages may be subject to different levels of shaking and compression leading to increase in compaction density. The compaction density (g/ml) dependence the dielectric constant and loss factor of selected vegetable powders at 13.56 and

27.12 MHz at 22±3 °C were shown in the Table 3.1 and Figure 3.5. It can be seen that at a constant frequency, both ε′ and ε″ increased with compaction density for different food powders until the density reached a certain level, then started to decrease. Previous researchers have found that the bulk density impacted dielectric properties, and reported that increase in bulk density often leads to higher dielectric properties and dielectric heating rate in flaxseed and other foods (Kent (1977); Kent and Kress-Rogers (1986); Nelson (1983); Nelson (1984). Nelson and You (1989) also reported that increase in bulk density of sample or bulk density of air-particle mixture resulted in increased dielectric properties and dielectric heating rate, which was related to the decrease in the gap between food particles causing a low energy dissipation in the air spaces (Nelson and Datta., 2001). However, as far as we know, this is the first report in literature that ε′ and ε″ will decrease when compaction density continues to increase to a certain level. Although the reason is not clear, it may be related to the caking of powders at high pressure. Food powders are inclined to cake as they are exposed to external compression/pressure, and the level of stickiness and caking in food powder is based on the humidity, particle size and composition of food (Adhikari and others, 2001). The air removal and cake formation should change the moisture distribution, and lead to decreased capacitance and increased resistance of the sample, and eventually decrease in dielectric properties. Figure 3.5. also shows that change in both dielectric constant and dielectric loss factor was more dramatic for samples with high MC. Further studies will be conducted to investigate the mechanisms responsible for the effect of compaction density. The results indicate that it is important to consider the change in the compaction density in the food powders in order to design a proper RF heating protocol.

The penetration depths (d_p)calculated from obtained dielectric constants and loss factors of vegetables samples using Eq. 3 are listed in the Table (3.1) at interested frequencies and different compact densities. The penetration depths in different samples increased to a certain level and then decreased, corresponding to the change in both dielectric constant and dielectric loss factor with compaction densities. The maximum and minimum penetration depths were 0.67 and 0.49 m for tapioca flour and chili powder, respectively, at 27.12 MHz and 0.58 g/ml compaction density. The penetration depths for broccoli powder and onion powder were not calculated because of failure to obtain stable values of dielectric loss factor for these samples due to their low moisture content.

Effect of moisture, temperature on dielectric properties and penetration depth

Broccoli powder was used to study the influence of moisture and temperature on dielectric properties, and the results of the dielectric constant and loss factor of broccoli powder were plotted against frequency with various MC and temperatures in the Figures 3.6 and 3.7. Also, the values of ε' and ε'' for two frequencies (13.56 and 27.12 MHz), four moisture levels (6.9-14.9% w.b.), and seven temperatures (20 to 80°C) are summarized in Table 2. Influence of moisture and temperature at a constant frequency on the dielectric properties was also shown in Figure 3.8. The compaction density was 0.58 g/ml for all the tests, in which 2 g of the powder was used to fill the sample chamber. Figures 3.6 and 3.7 show that the dielectric properties of broccoli powder decreased with frequency but increased with temperature for all moisture levels. Calay et al. (1994) reported that the change in dielectric properties of food materials with MC is related to the amount of free and bound water contents in food. The amount of free water content in a given sample contributes to dielectric polarization much more than bound water when sample is placed in an alternating electric field. Therefore, dielectric properties of broccoli powder increased with increase in MC at constant frequency and temperature (Figure 3.6). Previous studies have also reported similar trend in dielectric properties for different low moisture food products including legume flours and wheat flour (Guo et al., 2008; Guo et al., 2010; Jiao et al., 2011; Nelson and Trabelsi, 2006).

Furthermore, the effect of frequency on both dielectric constant and loss factor was more significant at the higher MCs than at the lower levels (Table 3.2). When the frequency increased from 1 to 27.12 MHz at 40 °C, dielectric constant decreased from 7.23 to 5.52 for the sample with 9.1% MC, but from 23.24 to 13.1 for the sample with 14.9% MC (Figure 3.6). Similarly, at higher temperatures, the influence of frequency on both the dielectric constant and loss factor was much more pronounced than that at the lower temperatures (Figure 3.7). For example, as the frequency increased from 1 to 27.12 MHz, dielectric constant of broccoli powder with 14.9 % MC decreased from 16.6 to 10.6 at 20°C (Figure 3.7). However, it decreased from 64.6 to 24.7 at 80 °C. The loss factor decreased from 4.1 to 0.82 at 20 °C, and from 56.30 to 6.4 at 80 °C (Figure 3.7). Zhu et al. (2012b) reported that the dielectric constant of chestnut flour 11.6 % w.b, MC increased from 2.1 to 10.7 with at temperature ranging from 20 to 60 °C at 27.12 MHz. (Shrestha and Baik, 2013) reported that increase in temperature enhances mobility of ions in the sample, which charges more electrodes, leading to increase in value of ε'. In addition, a rapid change in dielectric properties was observed at high MC (12.2 and 14.9 %), especially above 40°C (Figure 3.8). When the temperature increased from 20 to 80 °C, for the 12.2 % moisture sample, dielectric constant increased from 7.52 to 18.51 and loss factor increased from 0.38 to 4.23 at 27.12 MHz (Figure 3.8). But for sample with 14.9% w.b., ε' and ε'' increased from 10.8 and 0.81 to 25.42 and 6.73 respectively. It is expected that the same trends should exist for the dielectric properties of other vegetable powders as for the broccoli powder.

The penetration depth calculated from measured values of ε' *and* ε'' of broccoli powder using Eq. 3 are listed in the Table 3.2, for frequencies of 13.56 and 27.12 MHz, temperatures from 20 to 80°C, and all MCs (6.9, 9.1,12.2 and 14.9% w.b.). The penetration depth decreased with increasing temperature, frequency and MC as seen in the Table 3.2. Similar trends were reported in previous studies for low-moisture foods at the same and higher frequency ranges (Guo et al., 2008; Jiao et al., 2011; Wang et al., 2003a). The penetration depth of ground almond shells with MC ranging from 6 to 36 % (w.b.) at 90 °C were reported as 2.7 to 0.15 m at 27.12 MHz (Gao et al., 2012). The high penetration depth in broccoli powder can be explained by its low MC level and low dielectric properties so that the energy absorption level on the surface of the powder is lower as compared to the high moisture food samples at the same temperature and frequency. To obtain an appropriate heating uniformity and disinfection effectivity in food materials with dielectric heating, the thickness of food material should not be thicker than penetration depth of dielectric system (Schiffmann, 1995). Therefore, the penetration depth of RF range as shown in the Table 3.1 and 3.2 could be used as practical guidance for the thickness of the packages to be processed in the RF oven in order to develop an industrial scale RF pasteurization system for the broccoli powder.

The data in the Table 3.2 were analyzed using SAS 9.4 software to obtain regression models to describe the dielectric constant and loss factor and penetration depth as a function of temperature and MC. The regression equations for 13.56 and 27.12 MHz are shown in Table 3.3. Analysis of variance (ANOVA) was carried out to determine if MC or temperature had a significant effect on ε', ε'' and d_p of broccoli powder. Each regression model provided a good fit to the experimental data at the significance level ($p<0.0001$) and with a high coefficient of determination ($0.9683 \leq R^2 \leq 0.998$). In addition, the values of RMSE of models (goodness fit) were also shown in Table 3. Those regression models can be used to predict the dielectric constant and loss factor of broccoli powder with MC between 6.9-14.9%, temperature between 20-80 °C, and heating frequency of 13.56 and 27.12 MHz.

The RF heating rate of vegetable powders

Figure 3.9 shows experimental temperature and time histories of different vegetable powders including broccoli, onion and chili powder, tapioca flour and potato starch with initial MC when heated in the 27.12-MHz, 6-kW RF oven. The gap between parallel electrodes was 9 cm. The temperatures were measured from the center of the container, and the initial sample temperature was around 22 ± 3 °C.

As seen in the Figure 3.9, a linear increase and decrease in the temperature profile of vegetable powders was observed when the RF oven was on and off. The sample temperatures reached 85-100 °C in 30 to 150 s. Similar linear increase in temperature during RF heating was reported in wheat flour and almond kernels (Nelson and Trabelsi, 2006; Wang and Tang, 2001; Wang et al., 2013), tropical fruits, (Wang et al., 2003a), cowpea weevil, black eyed peas and mung beans (Jiao et al., 2011; Jiao et al., 2014; Wang et al., 2013). The heating rates, as calculated by dividing the temperature increase by heating time, are 0.41, 0.56, 0.71, 0.82 and 2.04°C s^{-1} for onion powder, tapioca flour, broccoli powder, potato starch and chili powder, respectively. The increasing order of the heating rate for different samples corresponds well with their dielectric constant as shown in the Table 1.The influence of dielectric properties and MC of the material on RF heating rate has been also reported for various food materials and insects (Nelson and Trabelsi, 2006; Nelson, 1996; Shrestha and Baik, 2013). The heating rate of almond kernel, and temperature difference between kernel and insect during RF heating were reported as 10°C min^{-1} and 5 °C min^{-1}, respectively (Wang et al., 2013). In our experiment, it was also noticed that there was around 10 to 20 °C continuous increase in temperature of samples after RF oven was turned off, which may be related to the non-uniform temperature distribution in the sample. The increase in temperature may be the result of heat transfer by conduction from hot to cold areas. Thus, in practical RF applications, it is important to improve temperature uniformity with suitable technologies (Wang et al., 2013).

To further study the influence of MC on RF heating rate, experimental temperature time profile of the broccoli powder with different MCs were obtained and are shown in Figure 3.10. The heating rates were determined as 0.84, 0.91, 1.48 and 2.12 °C s^{-1}, for moisture of 6.9, 9.1, 12.2 and 14.9% w.b., respectively. The absorbance of RF energy in food depends on free water content; therefore, foods with higher MC have faster heating rates. Metaxas (1996) reported that the electrical conductivity of food materials plays an important role in heating mechanism which changes from dielectric heating to resistive heating when the distance between surface of food material and electrode is minimized to zero. Low-moisture food has lower electrical conductivity and thus less heat generation rate than high-moisture food because of low mobility of charged ions (Gao et al., 2012). The effect of MC on ionic conductance has been reported for various food materials such as fresh cut fruits and vegetables, and insects (Guo et al., 2008; Wang, 2005; Wang et al., 2008b). Nelson (1996) also noticed that there is a linear relationship between dielectric loss factor and energy absorption as material is exposed to RF or MW heating.

The linear relationship between heating rate and MC and dielectric loss factor of broccoli powder are shown below:

$$y = 16.262^{*}\varepsilon'' + 0.5047 \qquad R^2=0.99 \qquad \text{Eq. (10)}$$

$$y = 0.1663^{*}MC - 0.4539 \qquad R^2 = 0.95 \qquad \text{Eq. (11)}$$

where y is the hating rate (°$C^{*}s^{-1}$), MC is the moisture content (% w.b), $6.9 \leq MC \leq 14.9$; *t* is treatment time (s). The equations have good fit with experimental data as indicated by high correlation coefficients (R^2) and can be used for predicting temperature increase in broccoli powder at given MC (6.9 to 14.9 %) when heated at 27.12 MHz.

Conclusions

The dielectric constant and loss factor of the vegetable powders decreased with frequency and compaction density and increased with MC or temperature. The changes in dielectric properties of broccoli samples with moisture and temperature were greater at lower frequencies (<10 MHz) than at higher ones. Both dielectric constant and loss factor increased with compaction density but decreased when compaction density reached a certain point. The penetration depth in broccoli samples decreased with MC, temperature of sample, and frequency of the electric field. The moisture and temperature dependence of the dielectric properties of broccoli powder at 13.56 and 27.12 MHz were described with quadratic equations. The regression equation models provided a good fit to predict dielectric properties of broccoli powder samples at a significance level of 0.0001.

The RF heating rates of vegetable powders, ranging from 0.56 to 2.12 °C s^{-1}, were linearly related to moisture, and dielectric loss factor. The penetration depth indicated that RF heating can be used to treat vegetable powders in large package sizes. The fast RF heating rate as observed in this study provided evidence that RF heating could be an effective method to pasteurize dried vegetables in short time that can potentially improve food quality.

Acknowledgments

This study was funded by the USDA-NIFA AFRI. Grant Number: 2013-67005-21295.

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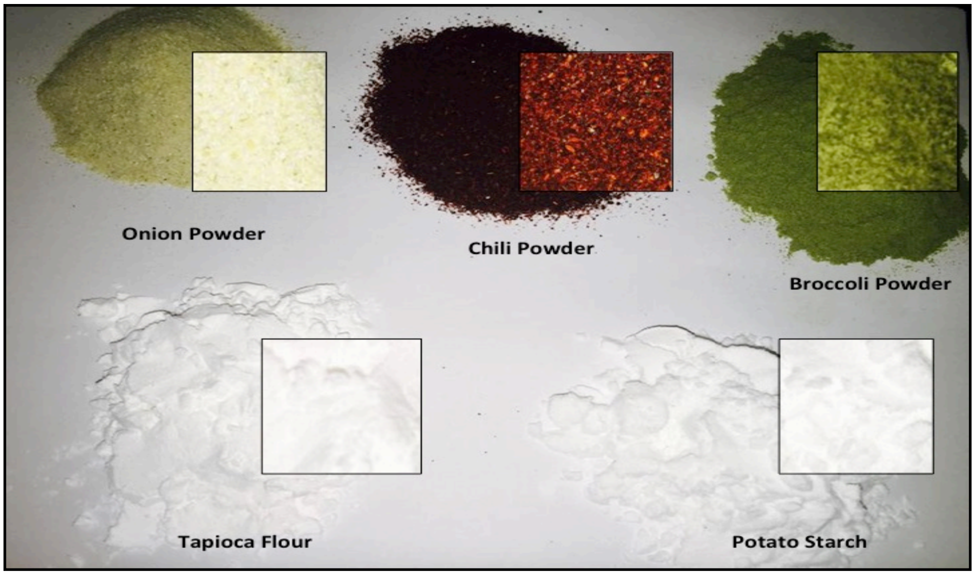


Figure 3.1. Vegetable powder samples use in the study

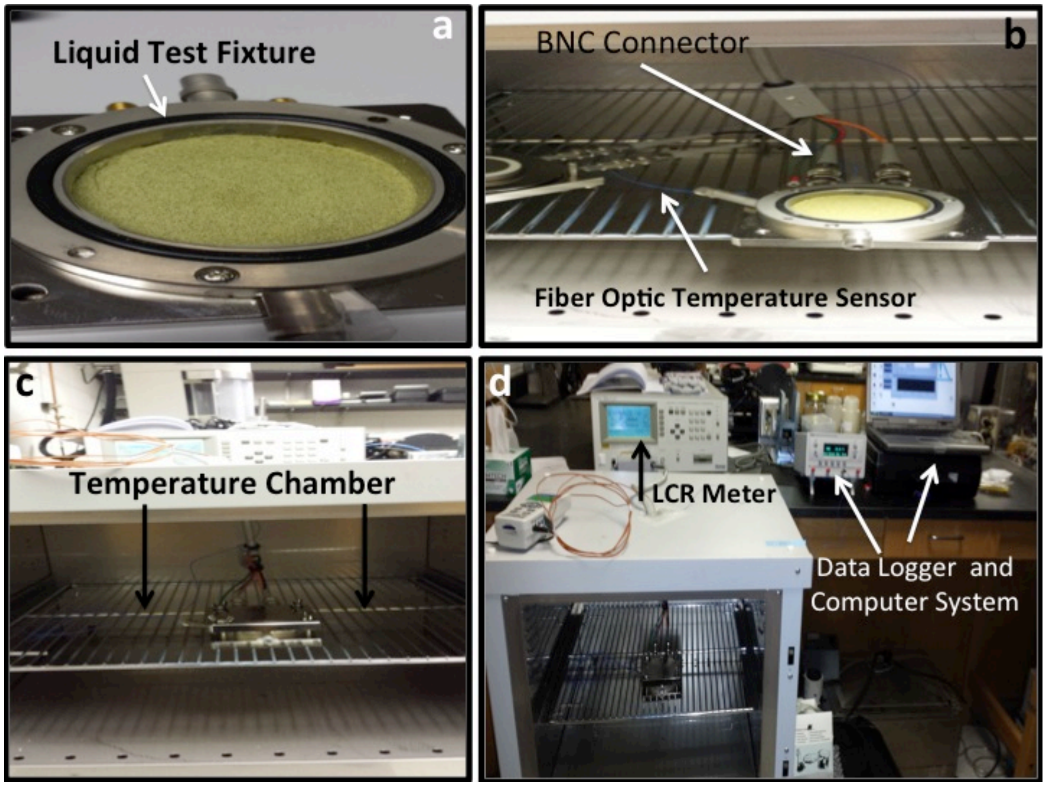


Figure 3.2. Instrumentation setup to measure values of C_p and R_p for calculation of dielectric properties

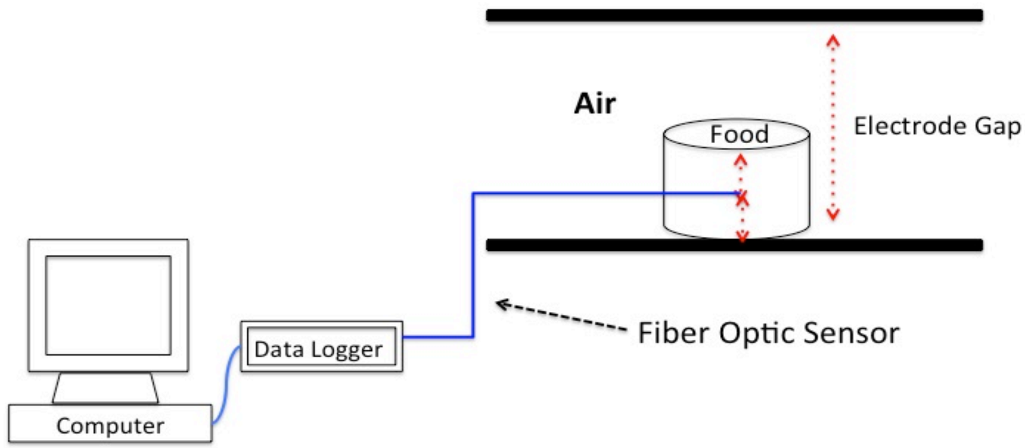


Figure 3.3. Experimental setup for RF heating with a fiber optic temperature measurement system

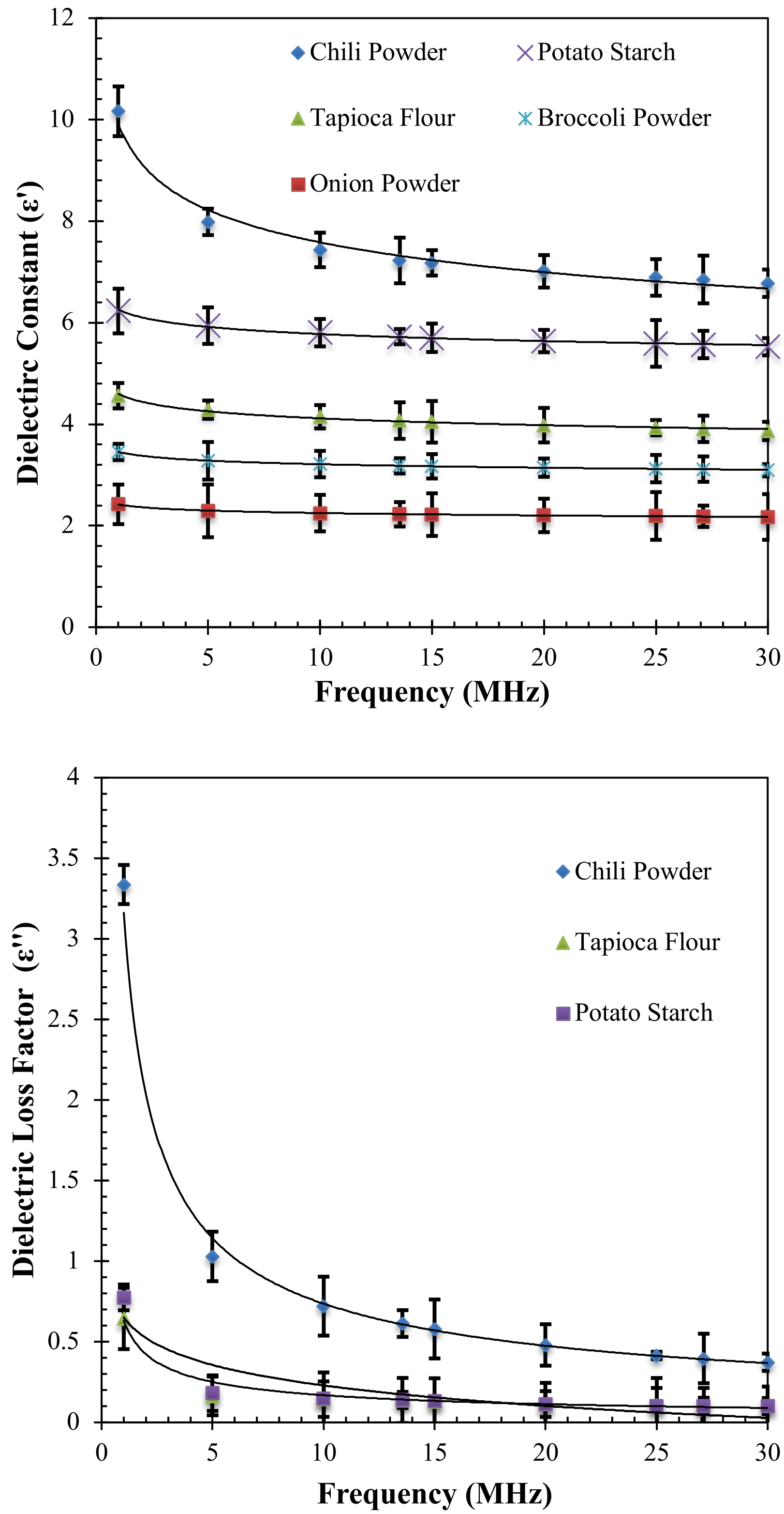


Figure 3.4. The dielectric constant and loss factor of vegetable powders at 27.12 MHz

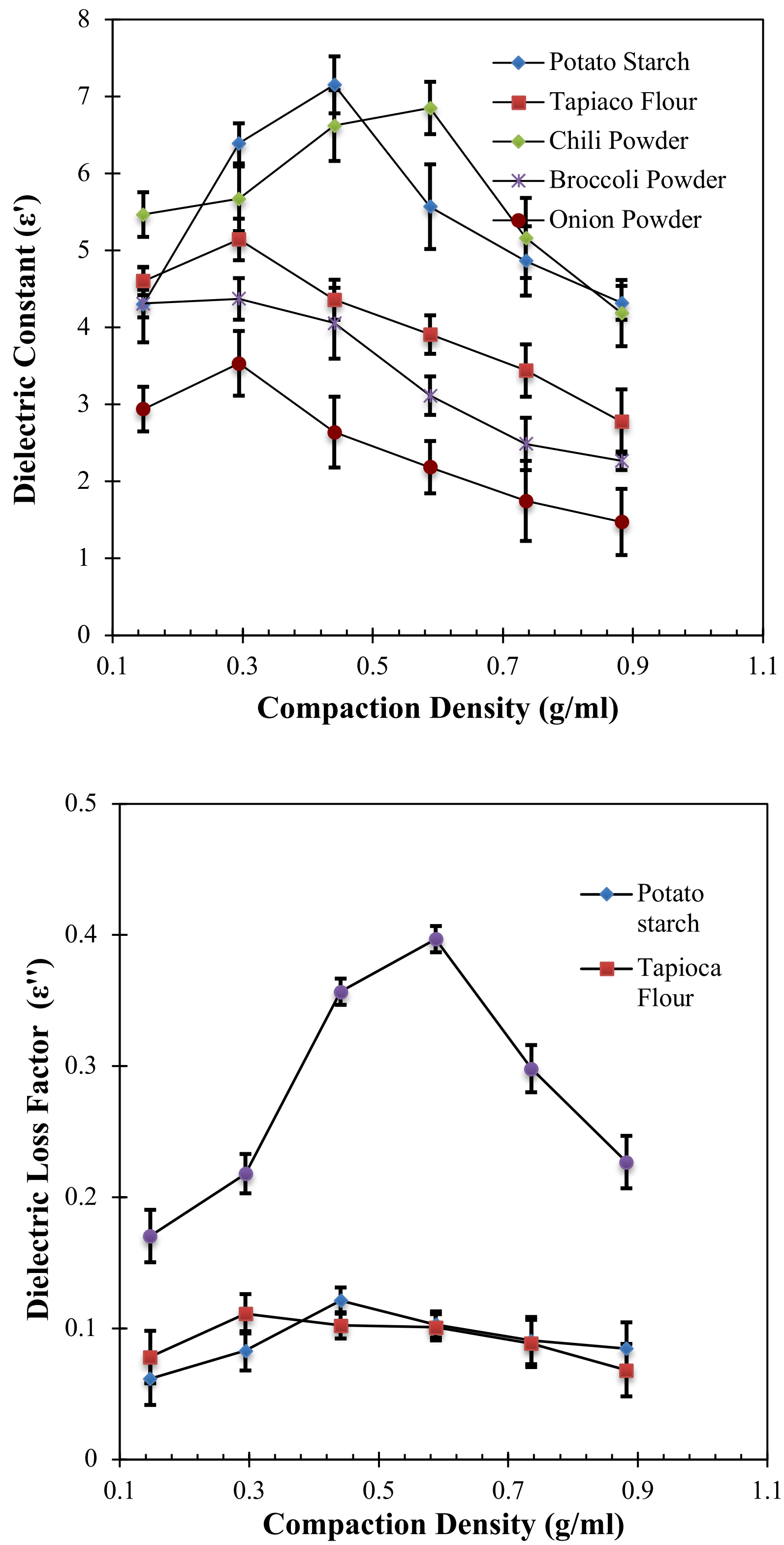


Figure 3.5. Influence of compaction density on dielectric constant and loss factor of vegetable powders at 27.12 MHz

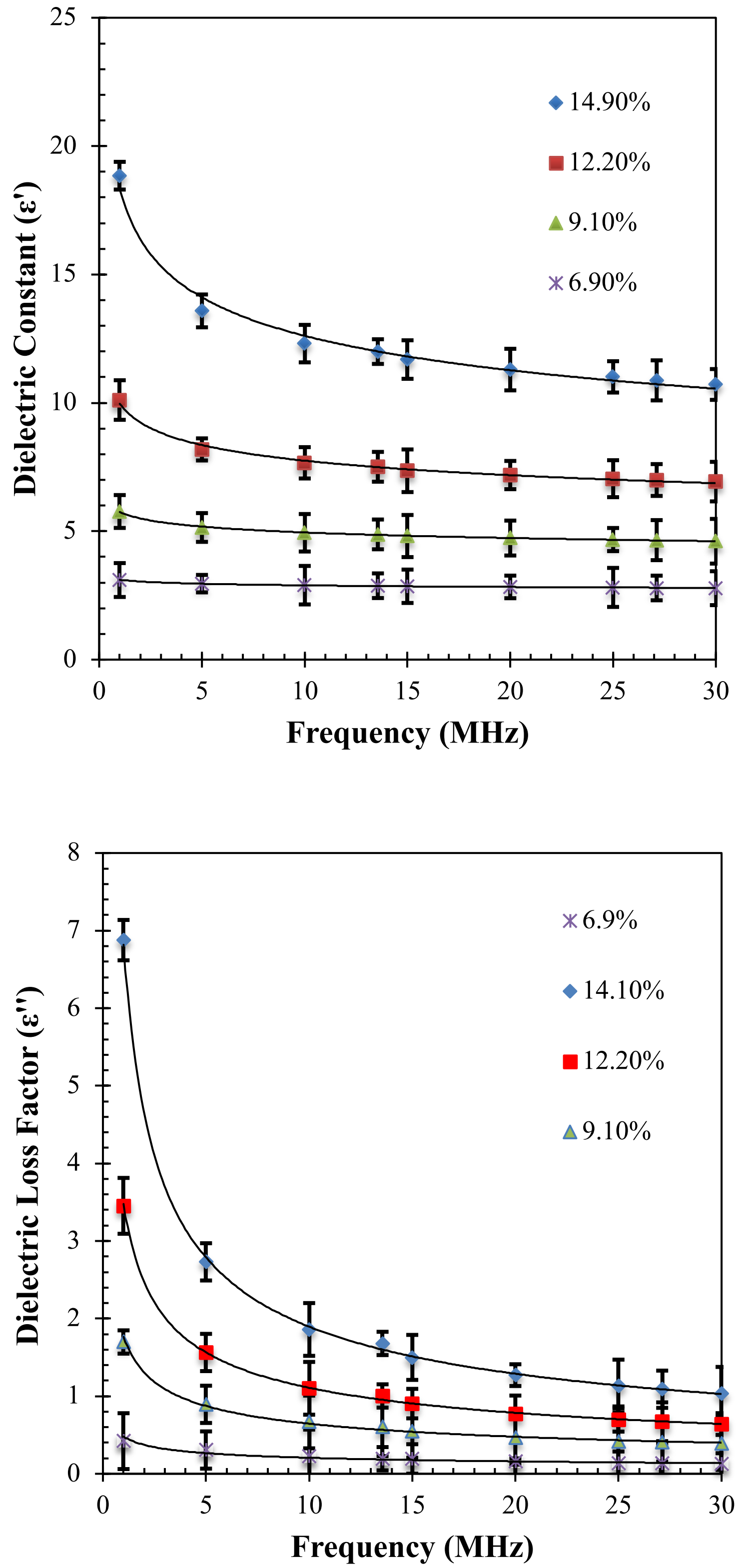
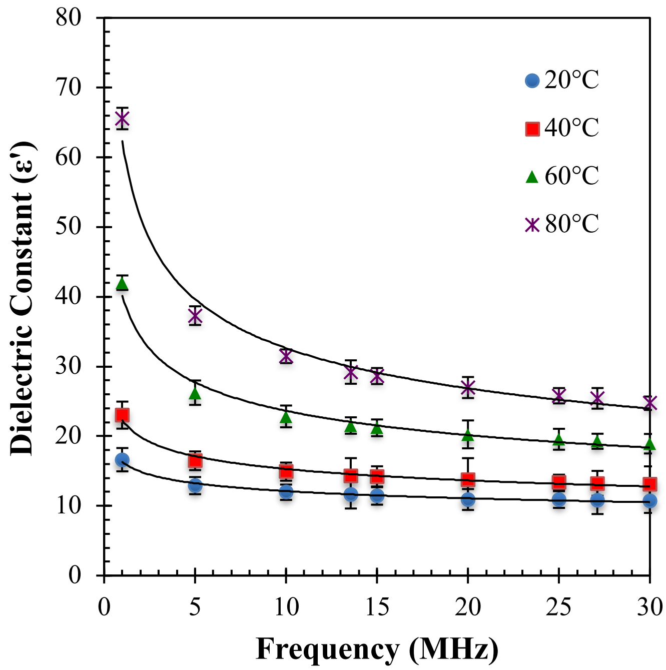


Figure 3.6. Frequency dependent dielectric constant and loss factor with four different MC of the broccoli powder at the frequency range 1 to 30 MHz



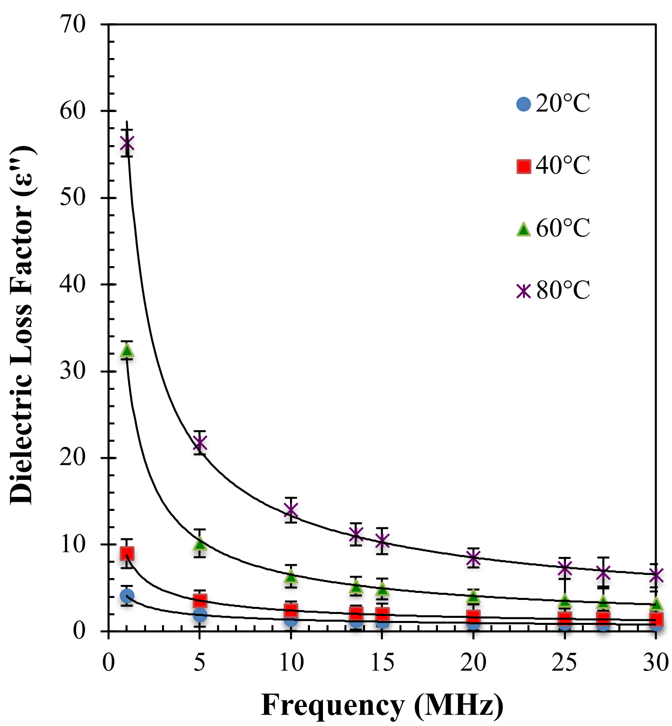


Figure 3.7. Frequency-dependent dielectric constant and loss factor of broccoli powder with MC of 14.9 %, at different temperatures over the frequency range of 1–30 MHz

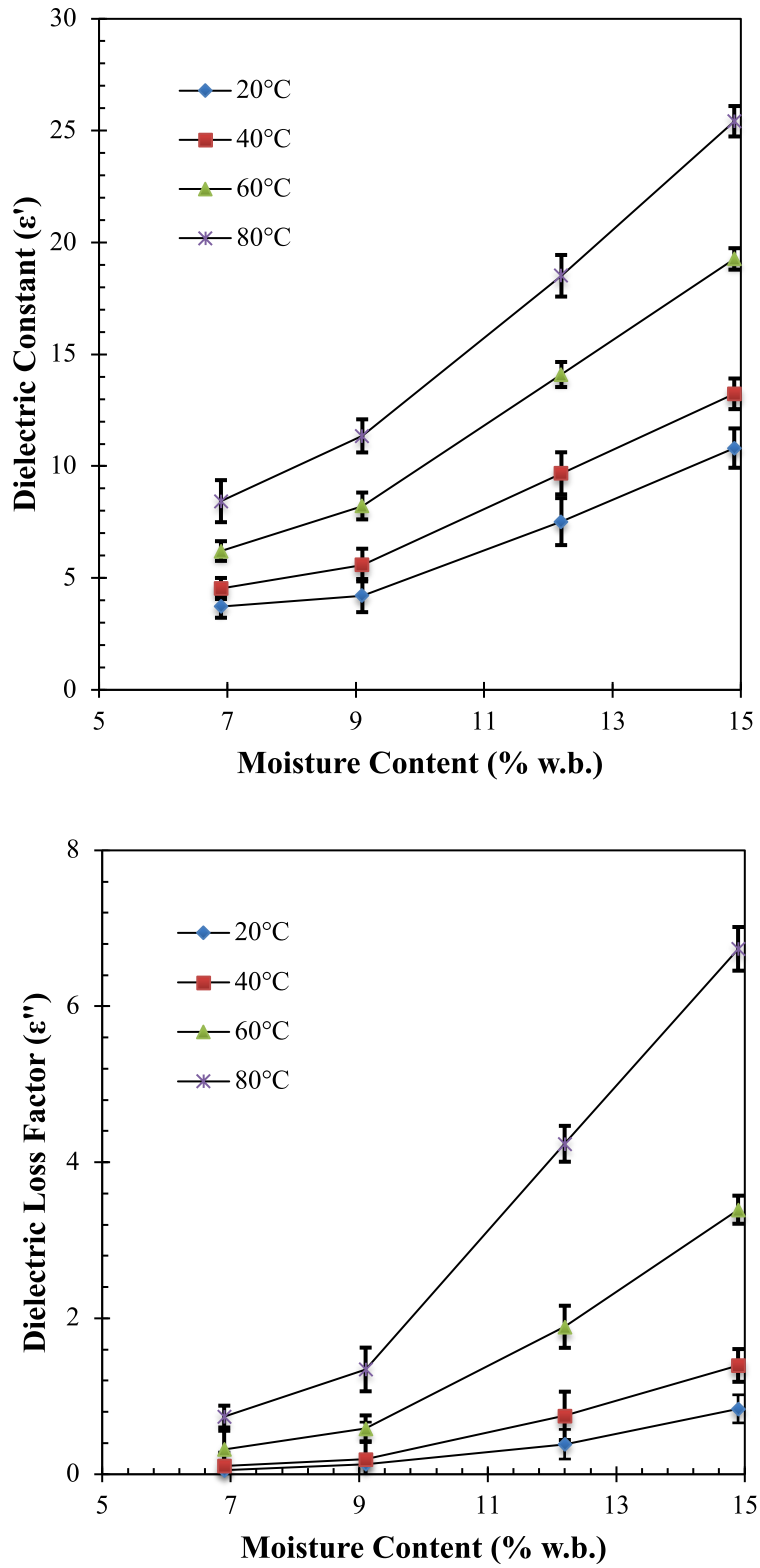


Figure 3.8. Moisture dependent dielectric constant and loss factor of the broccoli at 27.12 MHz and different temperatures

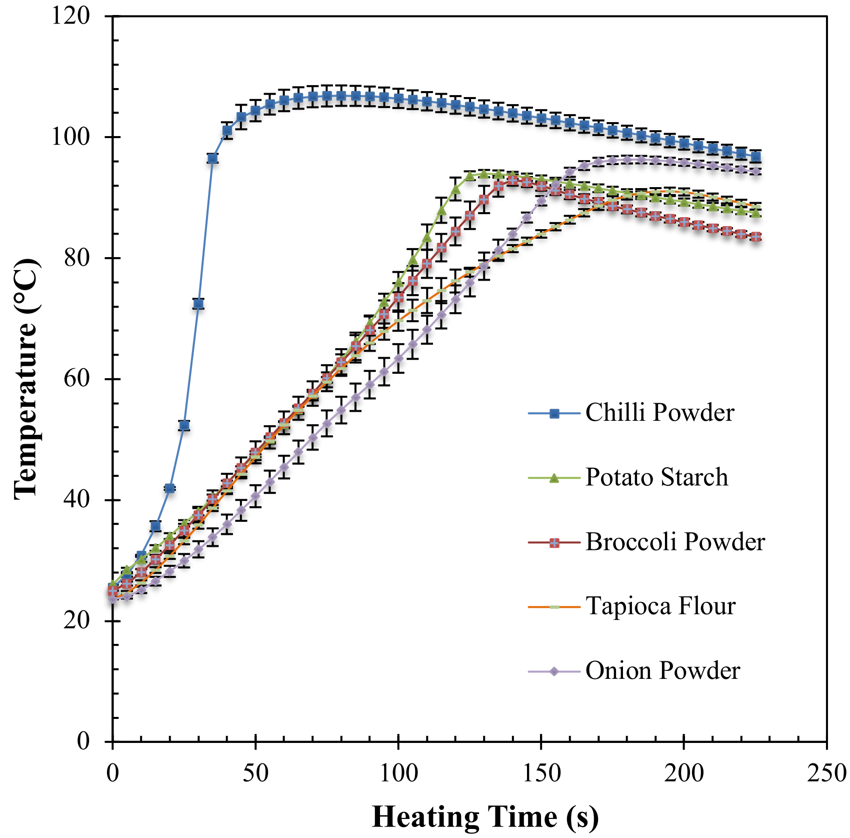


Figure 3.9. Temperature-time profiles of onion powder (1.4%), broccoli powder (3.9%), tapioca flour (5.7%), chili powder (7.8%), and potato starch (12.6%) during the RF heating at 27.12 MHz

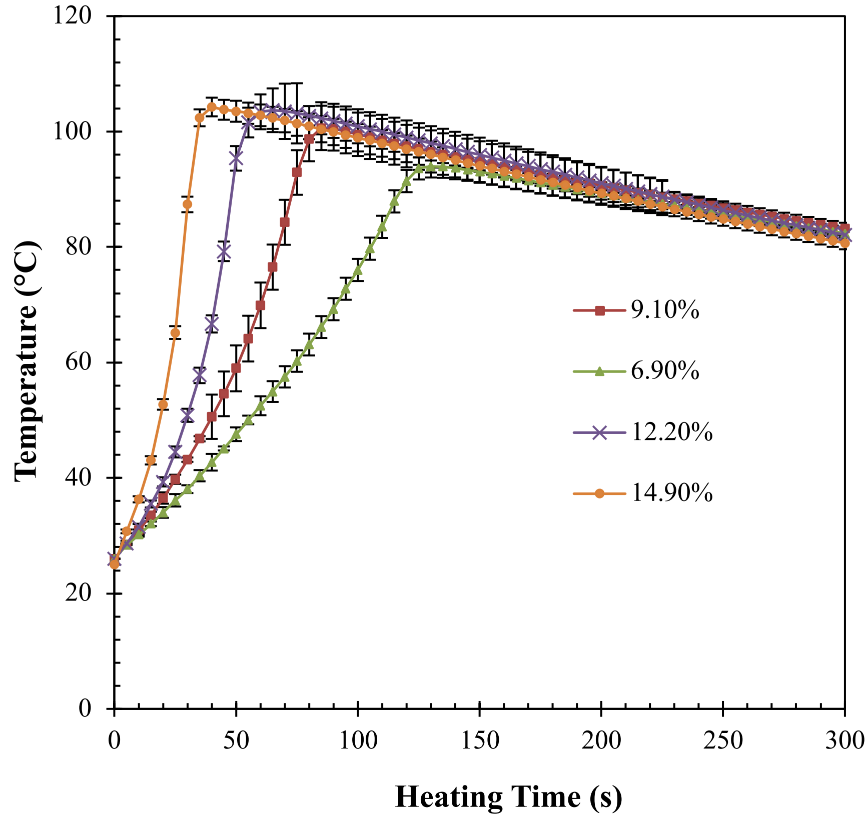


Figure 3.10. Temperature profiles in broccoli powder with different moisture content during the RF heating at 27.12 MHz

Table 3.1. Dielectric properties (mean ± SD of replicates) of the vegetable powders with different compaction densities at 22±3°C. The MC is in the range of 1.4 to 12.6 % (wet basis)

	Compaction Density (g/ml)	Dielectric Constant (ε')		Dielectric Lose Factor (ε'')		Penetration Depth (m)	
		13.56 MHz	**27.12 MHz**	**13.56 MHz**	**27.12 MHz**	**13.56 MHz**	**27.12 MHz**
Broccoli Powder	0.14	4.37±0.1	4.31±0.48	-	-	-	-
	0.29	4.45±0.24	4.37±0.3	-	-	-	-
	0.44	4.13±0.14	4.05±0.46	-	-	-	-
	0.58	3.17±0.41	3.11±0.25	-	-	-	-
	0.73	2.53±0.02	2.48±0.16	-	-	-	-
	0.88	2.31±0.46	2.26±0.34	-	-	-	-
Chili Powder	0.14	5.69±0.31	5.46±0.2	0.31±0.04	0.17±0.11	1.09±0.08	0.55±0.04
	0.29	5.93±0.22	5.67±0.42	0.37±0.07	0.21±0.05	1.06±0.04	0.54±0.02
	0.44	6.97±0.14	6.62±0.25	0.56±0.02	0.35±0.08	0.97±0.19	0.5±0.09
	0.58	7.22±0.42	6.85±0.46	0.61±0.13	0.39±0.07	0.95±0.25	0.49±0.12
	0.73	5.47±0.01	5.16±0.02	0.45±0.11	0.29±0.14	1.11±0.21	0.57±0.19
	0.88	4.42±0.24	4.18±0.16	0.35±0.19	0.22±0.02	1.25±0.25	0.64±0.17
Tapioca Flour	0.14	4.71±0.13	4.6±0.27	0.07±0.09	0.02±0.19	1.21±0.04	0.61±0.18
	0.29	5.32±0.46	5.14±0.54	0.16±0.17	0.11±0.1	1.13±0.08	0.57±0.42
	0.44	4.52±0.25	4.35±0.38	0.14±0.27	0.1±0.13	1.24±0.2	0.63±0.11
	0.58	4.07±0.06	3.91±0.13	0.13±0.03	0.09±0.09	1.31±0.16	0.67±0.17
	0.73	3.57±0.18	3.44±0.17	0.11±0.01	0.08±0.45	1.41±0.17	0.72±0.19
	0.88	2.88±0.11	2.77±0.54	0.09±0.12	0.06±0.01	1.61±0.25	0.82±0.09
Potato Starch	0.14	4.32±0.24	4.29±0.39	0.01±0.13	0.06±0.14	1.27±0.12	0.63±0.05
	0.29	6.54±0.14	6.39±0.46	0.13±0.03	0.08±0.02	1.01±0.04	0.51±0.2
	0.44	7.33±0.42	7.15±0.13	0.16±0.07	0.12±0.05	0.95±0.01	0.48±0.01
	0.58	5.72±0.37	5.56±0.27	0.14±0.27	0.1±0.01	1.08±0.1	0.55±0.4
	0.73	4.98±0.24	4.86±0.41	0.12±0.05	0.09±0.45	1.17±0.04	0.59±0.1
	0.88	4.45±0.19	4.31±0.19	0.11±0.13	0.08±0.19	1.25±0.13	0.63±0.7
Onion Powder	0.14	2.96±0.09	2.93±0.25	-	-	-	-
	0.29	3.58±0.16	3.53±0.34	-	-	-	-
	0.44	2.68±0.4	2.64±0.41	-	-	-	-
	0.58	2.22±0.46	2.18±0.17	-	-	-	-
	0.73	1.77±0.17	1.74±0.48	-	-	-	-
	0.88	1.50±0.27	1.47±0.54	-	-	-	-

Table 3.2. Dielectric properties (mean ± SD of replicates) of the broccoli powder at seven temperatures and three levels of MCs (wet basis)

Moisture Content (%w.b)	Temperature (°C)	Dielectric Constant (ε')		Dielectric Lose Factor (ε'')		Penetration Depth (m)	
		13.56 MHz	27.12 MHz	13.56 MHz	27.12 MHz	13.56 MHz	27.12 MHz
6.9	20	3.84±0.55	3.72±0.71	0.09±0.73	0.05±0.71	31.89±0.61	21.27±0.78
	30	4.14±0.78	4.02±0.54	0.11±0.65	0.07±0.82	27.37±0.73	18.83±0.5
	40	4.66±0.83	4.53±0.63	0.16±0.57	0.11±0.63	21.01±0.91	14.26±0.77
	50	5.46±0.86	5.26±0.34	0.26±0.44	0.18±0.77	14.13±0.4	9.52±0.41
	60	6.58±0.61	6.20±0.87	0.46±0.61	0.31±0.72	8.86±0.59	5.98±0.75
	70	7.86±0.67	7.39±0.61	0.73±0.51	0.49±0.65	5.75±0.55	4.10±0.69
	80	9.11±0.92	8.43±0.75	1.11±0.74	0.74±0.81	4.04±0.33	2.89±0.81
9.1	20	4.66±0.84	4.21±0.48	0.17±0.52	0.12±0.64	12.48±0.72	8.86±0.75
	30	5.12±0.67	4.93±0.75	0.21±0.61	0.14±0.72	10.49±0.63	7.03±0.84
	40	5.78±0.54	5.58±0.56	0.28±0.55	0.19±0.68	7.95±0.72	5.17±0.56
	50	6.95±0.69	6.7±0.63	0.45±0.41	0.33±0.54	5.01±0.77	3.19±0.65
	60	8.75±0.47	8.22±0.34	0.81±0.67	0.54±0.64	3.43±0.53	2.48±0.71
	70	10.66±0.54	9.85±0.46	1.37±0.59	0.91±0.72	2.41±0.68	1.73±0.66
	80	12.48±0.75	11.35±0.62	2.1±0.71	1.34±0.86	1.88±0.78	1.27±0.88
12.2	20	7.82±0.64	7.52±0.52	0.56±0.83	0.38±0.77	17.65±0.94	12.54±0.42
	30	9.09±0.72	8.63±0.61	0.72±0.61	0.52±0.68	14.83±0.77	9.94±0.39
	40	10.31±0.81	9.68±0.73	1.01±0.77	0.74±0.81	11.24±0.72	7.32±0.84
	50	12.68±0.77	11.78±0.51	1.77±0.81	1.34±0.54	7.09±0.65	4.52±0.66
	60	15.41±0.84	14.09±0.42	2.86±0.71	1.89±0.46	4.86±0.78	3.50±0.88
	70	18.41±0.68	16.52±0.76	4.45±0.93	2.92±0.67	3.41±0.96	2.45±0.87
	80	21.27±0.77	18.51±0.82	6.14±0.87	4.23±0.83	2.67±0.87	1.80±0.72
14.9	20	11.60±0.81	10.8±0.68	1.09±0.81	0.81±0.91	10.97±0.6	7.18±0.65
	30	12.72±0.73	11.98±0.86	1.29±0.73	0.94±0.55	9.69±0.55	6.46±0.71
	40	14.27±0.65	13.23±0.27	1.79±0.87	1.26±0.72	7.43±0.88	5.07±0.56
	50	17.47±0.86	15.93±0.58	3.03±0.61	1.91±0.45	4.86±0.65	3.67±0.39
	60	21.52±0.53	19.26±0.72	4.84±0.77	3.03±0.62	3.39±0.73	2.55±0.78
	70	25.45±0.72	22.41±0.81	7.21±0.89	4.53±0.79	2.48±0.57	1.84±0.85
	80	29.19±0.81	25.42±0.73	9.68±0.65	6.73±0.67	2.0±0.81	1.32±0.63

Table 3.3. Regression equations relating dielectric properties, and penetration depth to MC and temperature in broccoli powder at 13.56 and 27.12 MHz

Frequency (MHz)	Regression Equations	Equation Number	R^2	RMSE
13.56	$\varepsilon' = 13.696 - 0.307*T + 2*10^{-3}T^2 - 1.797W + 0.095W^2 + 0.027WT$	(4)	0.995	0.54
	$\varepsilon'' = 11.839 – 0.281*T + 10^{-3}T^2 - 1.585W + 54*10^{-3}W^2 + 18*10^{-3}WT$	(5)	0.968	0.52
	$d_p = 78.49 - 0.63T - 8.027W + 0.2485W^2 + 0.032WT$	(6)	0.827	3.36
27.12	$\varepsilon' = 10.464 - 0.2176T + 0.002T^2 - 1.398W + 0.081W^2 + 0.021WT$	(7)	0.994	0.46
	$\varepsilon'' = 6.458- 0.164T + 0.001T^2 - 0.854W + 0.029W^2 + 0.012WT$	(8)	0.975	0.29
	$d_p = 53.3206 - 0.4234T - 5.472W + 0.17W^2 + 0.0219WT$	(9)	0.825	2.33

W is the moisture content (% w.b), $6.9 \leq W \leq 14.9$; T is temperature (°C), $20 \leq T \leq 80$

CHAPTER 4

DIELECTRIC PROPERTIES, HEATING RATE, AND HEATING UNIFORMITY OF VARIOUS SEASONING SPICES AND THEIR MIXTURES WITH RADIO FREQUENCY HEATING[2]

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[2]S. Ozturk, F. Kong, R.K. Singh, J. D. Kuzy, C. Li, S. Trabelsi. Accepted by. *Journal of Food Engineering*.

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Abstract

Low moisture foods, including seasoning spices, have been associated with a number of multi-state outbreaks of salmonellosis in the past decade. The long-term objective of this study was to develop an effective in-package pasteurization treatment for seasoning mixtures based on radio frequency (RF) heating. Seasoning spices obtained from grocery stores included red, white, and black pepper; cumin; curry powder; and garlic powder with moisture contents ranging from 3.1-12.3% (wet basis). The dielectric properties (DP) of the seasoning mixtures as influenced by frequency, mixing fraction and salt content were determined using a precision LCR meter and liquid test fixture at frequency ranging from 1 to 30 MHz. The RF heating rates of each spice and their mixtures were evaluated using a 27.12-MHz RF system with 105 mm gap between electrodes. To evaluate the effect of mixing on heating uniformity, a sample (50 g) was placed into a polystyrene plastic cylindrical container and heated to 70 °C, and surface images were taken by an infrared camera. The results showed that the relationship among moisture content, temperature and DP of white pepper can be explained by a second-order model at 13.56 and 27.12 MHz. The DP and heating rates of spice mixtures ranged between the highest and lowest values of their respective individual spices. Increase in salt content resulted in a decrease in heating rate and uniformity index. The RF heating rate of samples ranged from 2.97 to 18.61 (°C min^{-1}). The highest heating rate in the samples was in correspondence to the worst heating uniformity and highest average temperature on the sample surface. The best heating uniformity index was obtained for garlic powder as 0.012 at 70 °C. The information obtained from this study is important to develop an effective RF heating strategy for pathogen control in seasoning mixture.

Key Words: Radio frequency heating; Seasoning spices; Mixing; Heating rate; Heating uniformity

Introduction

Foods with low water activity such as vegetable powders and spices have been used as flavoring and seasoning agents. Over the past decade, however, outbreaks were frequently reported in connection with *Salmonella* contamination of spices, including white, red and black pepper (CDC, 2010), paprika (Lehmacher et al., 1995), cumin (Moreira et al., 2009), turmeric and curry powder (Hara-Kudo et al., 2006), and garlic powder (Banerjee and Sarkar, 2003). Although the growth of microorganisms is not supported by foods with low water activity, spices are liable to be contaminated by microorganisms at all stages through production and supply chain including growing, harvesting, processing, packing, handling and transportation (McKee, 1995). Previous studies found that *Salmonella* in foods with low water activity can survive for extended storage time and has high heat tolerance (Hiramatsu et al., 2005; Lehmacher et al., 1995; Ristori et al., 2007). Recent studies have shown that the presence of *Salmonella* in spices can lead to serious foodborne illnesses when added to foods that do not undergo further thermal processing (Rico et al., 2010; Waje et al., 2008).

In general, spices are widely known to contain inhibitory compounds that can inhibit the growth of foodborne pathogens during storage (Arora and Kaur, 1999; Billing and Sherman, 1998; Ceylan and Fung, 2004; Dorman and Deans, 2000; Weerakkody et al., 2011), but it is not clearly known whether or not the antimicrobial effect of spices is sufficient to resist microbial growth. Several post-harvest decontamination methods such as fumigation with ethylene oxide, irradiation, and steam treatment have been used to pasteurize contaminated spices. For example, to pasteurize black pepper, packaged black pepper is traditionally treated by steam for 16 min at 1020 mbar and 100 °C. However, the steam treatment of black pepper caused quality degradation with loss of color and flavor (Waje et al., 2008). Due to its carcinogen effect, the use of ethylene oxide is forbidden by the Europe Union, and irradiated foods have not found acceptance by the customers even if it is allowed to be used for pasteurization of spices (Farkas, 2006; Schweiggert et al., 2007).

Consequently, there is a need to develop a more effective alternative pasteurization technique while maintaining the quality of spices. Radio frequency (RF) heating, also known as dielectric heating, has been applied as a promising technique for various food products (Balakrishnan et al., 2004; Bengtsson and Risman, 1971; Wang et al., 2005c; Zielinska et al., 2013). RF is in the range of 1 to 300 MHz, particularly 13.56, 27.12 and 40.68 MHz, when used for commercial applications, which provides longer wavelength and deeper penetration than those of microwaves at 915 or 2450 MHz (Luechapattanaporn et al., 2005; Marra et al., 2009; Metaxas and Meredith, 1988). As heat is generated volumetrically in foods, RF heating offers significant advantages such as faster heating, better quality, more uniform heat distribution and higher energy efficiency for solid and semi-solid foods with low thermal conductivity as compared to other conventional treatment methods (Casals et al., 2010; Luechapattanaporn et al., 2005; Marra et al., 2009; Pereira and Vicente, 2010). Recent studies also suggested that RF heating has great potential to be used for postharvest disinfection (Jiao et al., 2012; Lagunas-Solar et al., 2007), and pasteurization of dry foods (Gao et al., 2012; Jeong and Kang, 2014; Kim et al., 2012).

Although RF heating is considered a promising pasteurization method, its application is still limited due to challenging in non-uniform heating which is related to dielectric properties of materials and heating rate. The dielectic properties (DP) of food materials are the most important factors affecting RF heating since the DP values play an important role in energy absorption and conversion in food, which directly affect heating rates and uniformity (Tang, 2005). The DP of foods are mainly influenced by moisture content, temperature, frequency, bulk density and salt content (Nelson, 1996; Orsat and Raghavan, 2005; Tang, 2005). Additionally, because different spices are often mixed to achieve a desirable flavor, the DP values of these mixtures differ from the DP of their individual spices. While dielectric properties of various spices have been investigated separately, dielectric properties of spice mixtures have never been measured to our knowledge. Dielectric properties of mixtures of spices at room temperature are important for the efficient use of radio frequency to initiate dielectric heating for thermochemical conversion processes. Thus far, there is a lack of in-depth knowledge regarding the DP, heating rate and heating uniformity of spices and their mixtures. Such knowledge is important for developing an effective RF pasteurization procedure for spices. Mixture equations have been reported to predict the DP of air-particle mixtures (Lal and Parshard, 1973; Nelson and Datta., 2001; Sihvola and Kong, 1988), but have not been used to describe the DP of spice mixtures.

The aim of this study is 1) to determine the DP of selected spices and their mixtures as influenced by frequency (1-30 MHz), moisture content (from 10.2 – 21.7 % w.b.) and temperature (20 to 90 °C), 2) to develop regression models to describe the DP of spices as a function of temperature and moisture content at selected frequencies (13.56 and 27.12 MHz) using white pepper as an example, and predict the DP of spice mixtures using different mixture equations, and 3) to evaluate the heating rate and heating uniformity of spices and their mixtures.

Material and Methods

Sample preparation and characterization

Garlic powder; curry powder; cumin; turmeric; paprika; and black, white and red pepper were purchased from a local grocery store. The moisture contents (w.b.) of all spices were determined as garlic powder 3.1 %, curry powder 8.3 %, cumin 9.2 %, turmeric 9.5 %, paprika 12.3%, black pepper 11.02 %, white pepper 10.2 % and red pepper 10.5 % by drying samples in a vacuum oven at 105°C for 16 h (AOAC, 1998).

To study the effect of moisture on the DP values, the moisture content of white pepper was adjusted by adding distillated water to a sample to reach 13.7, 17.1 and 21.7 % (w.b.). To obtain a uniform moisture distribution in white pepper samples, water-added samples were stored in Ziploc bags for 48 h at 4°C and shaken twice daily.

Spice mixtures were prepared using garlic powder (GP); curry powder (CP); paprika (P); black (B), white (W) and red (R) pepper; and salt. Mixtures were made including paprika, curry powder and black pepper (P-CP-B), white pepper and red pepper (W-R), black pepper and garlic powder (B-GP), black pepper and salt and garlic powder and salt with different proportions on mass basis. The mixtures were stored in Ziploc bags at 4°C for 48 h and manually mixed twice daily to obtain moisture equilibrium and uniform distribution throughout the mixture. The mixtures were placed at room temperature for 12 h for temperature equilibration before measurement.

The bulk density (ρ) of each sample and mixture were determined gravimetrically as;

$$\rho = \frac{m}{v}$$

where m is total mass (g) of the sample and v is its volume (cm^{-3}) (Trabelsi et al., 2001). The particle density of each material in mixture were measured with an air-comparison pycnometer (Nelson, 2001), represented in Table 4.2.

Dielectric properties measurement

The DP of each seasoning spice and their mixtures were determined by measuring capacitance (C_p) and resistance R_p with different frequencies (ranging from 1 to 30 MHz) using the parallel plate method with an Inductance Capacitance and Resistance (LCR) meter (4285A, Agilent

Technologies, Palo Alto, CA) and a dielectric liquid test fixture (16452, Agilent Technologies, Palo Alto, CA). Before taking a measurement, the LCR meter was manually calibrated using a BNC cable to minimize biased error and random error. Detailed information about the system and experimental procedure were described by Ozturk et al. (2016). The liquid test fixture was filled with 2 g sample and then tightly sealed before measurement. To study the effect of temperature on DP values of white pepper, the tightly closed test fixture was placed into a temperature chamber (625G, Thermo Fisher Scientific Inc., Waltham, MA, USA,) to achieve temperatures ranging from 20 to 90 °C with 10 °C intervals. The selected temperature range in this study covers the inactivation temperature for bacteria with high thermal resistance in foods with low water activity. Obtained C_p and R_p values were used to calculate dielectric constant(ε') and loss factor (ε'') for samples using the following equations (Agilent Technologies, 2000; Halliday, 2001; Von Hippel, 1954).

$$\varepsilon' = \frac{tC_p}{A\varepsilon_0} \qquad \text{Eq. (1)}$$

$$\varepsilon'' = \frac{t}{2\pi f R_p \varepsilon_0 A} \qquad \text{Eq. (2)}$$

where t is the gap (m) between electrodes of the test fixture, C_p is capacitance (F), R_p is the resistance (Ω), f is the frequency (Hz), ε_0 is the permittivity of vacuum (8.854 x 10^{-12} F m^{-1}), and A is the electrode area (m^2).

Dielectric mixture equations

To estimate the DP of an air-particle mixture sample, different mixture equations have been proposed by other researchers (Lal and Parshard, 1973; Nelson and Datta., 2001; Sihvola and

Kong, 1988). The following three equations were used to predict both ε′ and ε″ of spice mixture samples in this study:

Complex Reflective Index Mixture Equation (CRIME) $\varepsilon = \left[v_1\varepsilon_1^{1/2} + v_2\varepsilon_2^{1/2}\right]^2$ Eq. (3)

Landau and Lifshitz, Looyenga equation (LLLE) $\varepsilon = \left[v_1\varepsilon_1^{1/3} + v_2\varepsilon_2^{1/3}\right]^3$ Eq. (4)

Lichtenecker Equation (LE) $\log\varepsilon = v_1\log\varepsilon_1 + v_2\log\varepsilon_2$ Eq. (5)

where ε represents the complex permittivity ($\varepsilon = \varepsilon' - \varepsilon'' j$) of the mixture, ε_1 and ε_2 are the complex permittivity of respective components in mixture, and for the two phase (air-particle) mixture $v_1 + v_2 = 1$, and permittivity of air is $1 - j0$, (where $j = \sqrt{-1}$). Complex permittivity of the solid material particles can be calculated, where $v_2 = v_s$. The v_s is volume fraction of representative component in mixture, which can be obtained if the bulk density (ρ_m) of mixture and the density (ρ_s) of the solid particulate material are known, since $v_s = \frac{\rho_m}{\rho_s}$ (Nelson, 2001).

Radio frequency heating rate and uniformity

To investigate the heating rate and heating uniformity of selected spices and their mixtures during the RF heating, polystyrene cylindrical petri dishes with diameter 100 mm and height 15 mm were used to hold the samples. Recent research has indicated that placing a sample in the middle of two parallel electrode plates provides better heating rate with more uniform temperature distribution for different foods during the RF heating (Tiwari et al., 2011). Prior to the RF heating, samples were taken from refrigerator and hold at the room temperature (23 ± 2 °C) for temperature equilibration. Then, the polystyrene petri dishes were filled with 50 g sample and placed into the middle of two parallel electrodes with a gap of 105 mm (Figure 4.1). Samples in petri dishes were treated in a 27.12-MHz, 6-kW RF system (COMBI 6-S, Strayfield International, Wokingham, UK) to reach 70°C. To calculate the heating rate and to determine the heating profile of samples, changes in temperature were recorded at the geometric center of the cylindrical petri dishes using a fiber optic temperature sensor with an accuracy of ± 1 °C (Fiso Tech. Inc., Quebec, Canada) connected to a data logger. When the center temperature reached 70°C, the RF system was turned off and the sample was removed. The top surface image was taken by an infrared camera (FLIR T440, FLIR Systems, Inc., North Billerica, MA, USA) with an accuracy of ± 2 °C. The thermal images were used to determine uniformity index values and average temperature distribution on the surface of each sample using the FLIR Tools Software (FLIR Systems, Inc., North Billerica, MA, USA).

The heating uniformity of each sample was evaluated using the uniformity index (UI), which has been applied to determine the RF heating uniformity for various foods (Hou et al., 2014; Jiao et al., 2012; Pan et al., 2012; Wang, 2005; Wang et al., 2008; Wang et al., 2010; Wang et al., 2005b). The uniformity index (UI) was defined by Wang et al. (2005a) as the proportion of the increase in standard deviation of heated sample temperature to the increase in the mean sample temperatures as heated by RF system. The UI value is calculated by following equation (Wang, 2005):

$$\lambda = \frac{\Delta\sigma}{\Delta\mu} \quad \text{Eq. (6)}$$

where $\Delta\sigma$ is the increase in the standard deviation (SD) of sample temperature (°C), and $\Delta\mu$ is the increase in the average temperature (°C) during the RF heating. The smaller UI values refer to more uniform RF heating in the sample. It is noted that in this study, only surface temperature of the sample was used to calculate the heating uniformity. As the sample holder (Petri dish) is very thin and the sample thickness is about (15 mm), the UI values calculated from the top surface temperature are used as an indicator of the heating uniformity for the entire sample volume.

Data analysis

All measurements, including dielectric properties, heating rate, and heating uniformity were done in triplicate. Mean value and standard deviation (SD) were reported. Excel (Microsoft office, Redmond, WA, USA) was used to process and analyze data. To predict the DP of spice mixtures, the mixture equations were applied by using measured DP of each spice and their mass fractions in mixture. Error bars represent the standard deviation of each replicated measurements.

The dielectric properties of white pepper as affected by moisture content and temperature were analyzed by SAS software 9.3 (SAS Institute Inc., Cary, NC, USA). Regression analysis was applied for experimental results to fit a model for two selected frequencies (13.56 and 27.12 MHz). The fitted second-order model was described by the following equations;

$$\varepsilon'_{13.56\,MHz} \text{ or } \varepsilon''_{13.56\,MHz} = \beta_0 + \beta_1 W + \beta_2 T + \beta_{12} WT + \beta_{11} W^2 + \beta_{22} T^2 \quad \text{Eq. (7)}$$

$$\varepsilon'_{27.12\,MHz} \text{ or } \varepsilon''_{27.12\,MHz} = \beta_0 + \beta_1 W + \beta_2 T + \beta_{12} WT + \beta_{11} W^2 + \beta_{22} T^2 \quad \text{Eq. (8)}$$

where ε' and ε'' are dielectric constant and loss factor at 13.56 and 27.12 MHz, β_0, β_1, β_2, β_{12}, β_{11} and β_{22} are regression coefficients, W is moisture content (%, w.b.) $10.2 \le W \le 21.7$; and T is temperature (°C), $20 \le T \le 90$. The fitted models were verified for their suitability in predicting the dielectric constant and loss factor at selected frequencies (13.56 and 27.12 MHz) as a function of moisture content and temperature based on the coefficient of determination (R^2), and root mean square error (RMSE), which were calculated by;

$$\text{RMSE} = \sqrt{\frac{1}{n}\sum(P-M)^2} \qquad \text{Eq. (9)}$$

where, n is the number of data points, P is the predicted value, and M is the measured value.

Results and Discussion

Dielectric properties of spices and their mixtures as influenced by frequency

The trends of both ε' and ε'' of seasoning spices over the measured frequency at room temperature exhibited similar patterns when graphed. Figure 4.2 shows the effect of frequency on the DP values of each spice sample with initial moisture contents (3.1 to 12.3 %). Both ε' and ε'' decreased as frequency increased from 1 to 30 MHz at room temperature (23 ± 2 °C). The dielectric constant values of samples follow the order: paprika (moisture content: 12.3 %) > white pepper (10.2 %) > black pepper (11.0 %) > red pepper (10.5 %) > turmeric (9.5 %) > curry powder (8.3 %) > cumin (9.2 %) > garlic powder (3.1 %). As seen in Figure 4.2, the highest ε' and ε'' values were 3.45 and 0.15 for paprika at 27.12 MHz, respectively. This order among the DP values agrees with initial moisture content of samples except for red pepper (11.5 %), black pepper (10.4 %) and turmeric (9.5 %). This could be the result of the DP values being not only influenced by moisture content in the samples but also affected by frequency of the electromagnetic waves, temperature, food composition (especially for salt and fat content) (Calay et al., 1994; Sun et al., 1995; Tang, 2005; Venkatesh and Raghavan, 2004), and bulk density and particle size (Nelson, 1996). The lowest ε' values reported were for cumin and garlic powder as 2.1 and 1.7 at 27.12 MHz, respectively. However, the positive ε'' values for cumin and garlic powder cannot be determined by the LCR meter at the RF range (1-30 MHz). Calay et al. (1994) reported that the decreasing effect of frequency on DP values of samples especially at lower moisture content is most probably due to reduced mobility of charged ions and ionic conduction, which causes a decrease in the ε″. Ryynänen (1995) also reported that ionic conduction plays an important role in dielectric heating in the range of RF frequency (1-300 MHz) as the dipole rotation is ineffective. Previous studies reported similar trends for both ε′ and ε″values of various food samples including chickpea, green pea, lentil and soybean flour (Guo et al., 2010); coffee bean (Pan et al., 2012); black and red pepper (Jeong and Kang, 2014); and wheat flour (Nelson and Trabelsi, 2006), peanut kernels (Zhang et al., 2016) as frequency increased. The ε′ values of black (10.1%) and red pepper (12.6%) at 27.12 MHz were reported as 1.72 and 3.17, and the ε″ were 0.02 and 0.03, respectively (Jeong and Kang, 2014).

Moisture content and temperature dependency

Moisture and temperature are widely known as the most important factors affecting the DP of foods. In this study, we used the white pepper as an example to demonstrate the effect of moisture content and temperature on the DP values. The moisture and temperature dependent DP values of white pepper are shown in Figures 4.3 and 4.4, respectively. In general, both ε′ and ε″ of white pepper in the range of frequency (1-30 MHz) increased with increase in moisture content (Figure 4.3). The ε′ of white pepper at room temperature increased from 3.36 to 4.45 with increasing moisture content from 10.2 to 13.7 % (w.b.) at 27.12 MHz. As seen in Figure 4.3, the decrease in both ε′ and ε″ of white pepper with moisture content 21.7 % (w.b.) was much higher than the DP of white pepper with moisture content 10.2 % (w.b.) as frequency increased from 1 to 30 MHz. The DP of white pepper in the range of radio frequency increased slowly as moisture content increased from 10.2 to 13.7 % (w.b.), then increased fast as moisture content increased from 13.7 to 21.7 % (w.b.) at room temperature (23 ± 2 °C) (Figure 4.3). Calay et al. (1994) and Tang (2005) reported that when a sample is subjected to an electromagnetic field in the range of radio frequency or microwave frequency, both free and bound water content in that sample contribute to an increase in the DP values. Therefore, the increase in the DP of white pepper can be explained by an increase in free moisture content for any frequency at room temperature. These results are in agreement with earlier studies for various low moisture foods including broccoli powder (Ozturk et al., 2016), egg white powder (Boreddy and Subbiah, 2016), legume flours, and wheat flour (Guo et al., 2008; Guo et al., 2010; Jiao et al., 2011b; Nelson and Trabelsi, 2006). Figure 4.4 shows temperature dependent measured and predicted DP of white pepper with moisture contents of 10.2, 13.7, 17.1 and 21.7 % (w.b) at 27.12 MHz. The DP of white pepper increased as temperature increased from 20 to 90 °C. The increase in DP values of white pepper agrees with observations made with chickpea samples at higher temperatures (Guo et al., 2008). The ε' for white pepper with 17.1% (w.b) moisture content increased from 5.35 to 19.72 with an increase in temperature from 20 to 80 °C, whereas it increased from 5.35 to 9.53 as temperature increased from 20 to 50 °C at 27.12 MHz. This increase in temperature may be contributed to ionic conductivity and water dipolar activity in sample, which resulted in the increase in the DP of white pepper. Furthermore, as the temperature increased from 20 to 70 °C, the ε'' at 27.12 MHz increased from 0.59 to 6.89 at 17.1% (w.b.). Kannan et al. (2013) reported that an increase in temperature enhances mobility of free ions and activity of their polarization, which results in charging more electrons in the sample leading to an increase in the ε''. These results are in agreement with Zhu et al. (2012), which reported both ε' and ε'' for chestnut flour with 11.6 % moisture content increased from 2.1 to 10.7 with an increase in temperature from 20 to 60°C at 27.12 MHz. Similar observations for both ε' and ε'' with increasing moisture and temperature at given frequency were reported for legume and wheat flours as frequency increased from 10 to 1800 MHz (Guo et al., 2008; Guo et al., 2010; Jiao et al., 2011a; Nelson and Trabelsi, 2006). It is expected that the same trends should exist for the dielectric properties of other seasoning spices and their mixtures as for white pepper.

Predicting moisture content and temperature dependency of dielectric properties

Regression analysis for the DP of white pepper at two selected frequencies was performed with SAS 9.3. Table 4.1 represents the regression constant and coefficients of the second-order models describing the relationship between moisture content (*W*), temperature (*T*) and both ε' and ε'' of white pepper at 13.56 and 27.12 MHz. The second order models, as shown in Eq. 7 and 8 with the coefficients in Table 4.1, could be used to predict the DP of white pepper in the moisture and temperature levels at 13.56 and 27.12 MHz. The predicted values of both ε' and ε'' for white pepper were plotted against temperature in Figure 4.4 for 27.12 MHz. As seen in Figure 4.4, both the predicted and measured values are generally in good agreement. The trend of the residual plot against the predicted loss factor was found to be random in nature for the models at 27.12 MHz in Figure 4.4. Therefore, the second-order models satisfactorily described the variation in the both ε' and ε'' of white pepper as a function of moisture content and temperature in the experimental range at their represented frequencies. The second order model equations for both ε' and ε'' in this study can be used to predict temperature profiles of white pepper subjected to RF heating.

Effect of mixing and salt content on dielectric properties

Both ε' and ε'' of different composite samples were calculated from the measured C_p and R_p data using Eq. 1-2. Table 4.2 shows the DP values of P-CP-B mixtures at selected frequencies (13.56 and 27.12 MHz) at room temperature. Nelson (2001) reported that the dielectric properties of air-solid mixtures are not only dependent on each single complex permittivity of representative sample in mixture, but also bulk density of mixture. For example, as paprika, curry powder and black pepper samples were mixed in mass basis with proportions of P (50%), CP (25%) and B (25%), the bulk density of representative mixture is 0.439 g cm^{-3} even though the bulk density of each sample is 0.437, 0.409, and 0.487 g cm^{-3}, respectively (Table 4.2). As seen in Table 4.3, the DP of mixtures is also affected by bulk density of mixture and volume fraction of each solid particle material. The ε' for P-CP-B mixture was 2.64 at 27.12 MHz, which is less than the ε' of paprika and higher than the ε' of black pepper and curry powder (Figure 4.2). Additionally, mixture equations (Eq. 3, 4 and 5) were used to predict the complex permittivity of the P-CP-B mixture for 13.56 and 27.12 MHz at room temperature, respectively. The measured and predicted dielectric properties of P-CP-B mixtures using mixture equations were represented in Table 4.3. Furthermore, to better evaluate the mixing effect on dielectric properties, the DP of red and white pepper (R-W) mixtures were also analyzed experimentally and calculated by mixture equations at selected frequencies. Table 4.4 shows the experimental and calculated the DP of R-W mixture with various fractions at 13.56 and 27.12 MHz. The DP of R-W mixture decreased as the mass fraction of red pepper increased from 10-90 % (mass basis) in mixture as well as frequency increased from 13.56 to 27.12 MHz (Table 4.3), which may also be because of change in bulk density of R-W mixture. A similar trend was also measured for the DP of B-GP mix (Table 4.5) with an increase in the fraction of garlic powder to black pepper. However, only the $\varepsilon''$of B-GP (90-10%) and B-GP (75-25 %) mixtures were able to be determined by the LCR meter analyzer. This may be explained by the increase in the fraction of garlic powder in the mixture causing low ionic conduction and dipolar activity at low frequency range. As shown in Tables 4.3 and 4.4, the dielectric constant and loss factor values of mixtures calculated from the LE and LLLE equations provides the closest values with the measured DP values. Results are in agreement with Nelson (2001) who used the same mixture equations to compare measured and calculated complex permittivity of coal and limestone powders using volume fraction of each single component in mixture. Although the LE and LLLE equations provide a good match with measured dielectric properties of selected mixtures, the difference between calculated and measured values still needs to be reduced with a better prediction models. Therefore, further study is needed to improve mathematical models to better predict the DP of seasoning spice mixtures.

On the other hand, to observe the influence of seasoning salt content on the DP of seasoning mixture, the black pepper, garlic powder and salt were mixed separately with different mass fractions. Table 4.6 shows the dielectric constant of black pepper-salt, and garlic powder-salt mixtures at 13.56 and 27.12 MHz. The ε' of seasoning salt were obtained as 1.65 and 1.64 at 13.56 and 27.12 MHz, respectively. Although the ε' of black pepper, garlic powder and salt mixtures decreased as the salt fraction increased from 10 to 40 % in the mixture, the ε' of salt was not influenced significantly by frequency. On the other hand, we were not able to measure the positive ε'' of black pepper-salt, garlic powder-salt, and salt in the range of 1-30 MHz, it may be because of reduced mobility of charged ions and ionic conduction in mixtures. The observation for the effect of salt content was not in agreement with pistachio kernel and cheese in which both ε' and ε'' increased as salt content increased at both radio and microwave frequencies (Fagan et al., 2005; Ling et al., 2015), which may be resulted from the different preparation methods as solid salt particles were used in black pepper and garlic powder mixtures while dissolved salts were added to pistachio kernel and cheese.

Radio frequency heating rate

Figure 4.5 shows the increase in temperature of various seasoning spices with initial moisture content subjected to RF heating including P, CP, cumin, turmeric, GP, W, B and R peppers. The samples were stored in polystyrene plastic petri dishes placed in the middle of two parallel electrodes with a gap of 105 mm. Temperature time profiles of each sample were obtained during the RF heating (Figure 4.5). Similar temperature and time profiles were also reported for different foods including wheat flour and almond kernels (Nelson and Trabelsi, 2006; Wang et al., 2001; Wang et al., 2013); tropical fruits (Wang et al., 2003); cowpea weevil, black eyed peas and mung beans (Jiao et al., 2011b; Jiao et al., 2014; Wang et al., 2013); and potato flour, broccoli and chili powders (Ozturk et al., 2016), shell almond (Li et al., 2017) when treated in an RF system. The heating times required to reach the target temperature (70°C) ranged from 1.85 (paprika) to 16 min (garlic powder) for samples (Figure 4.5). It is commonly known that dielectric materials convert electric energy to heat when placed into an electromagnetic field in the range of radio and microwave frequencies. The heat energy P (W/m^3) generation in food when placed in an electric field is determined by using the equation $P = 2\pi f E^2 \varepsilon_0 \varepsilon''$, where f is applied frequency (Hz), E is the electric field strength (V/m) in food, ε_0 is the permittivity of vacuum (8.854×10^{-12} F m^{-1}), and ε'' is the dielectric loss factor of the sample. It means that foods with high dielectric loss factor will enhance conversion of more electromagnetic energy to heat than food with lower dielectric loss factors, which corresponds to faster heat generation during the RF heating. The increase in temperature of a food sample as heated by RF heating system with a fixed electrode gap was described using dielectric properties by Jiao Y. et al., 2014. The heating rates of samples, calculated by dividing increase in temperature by heating time, ranged from 2.97 (garlic powder) to 23.61 °C min^{-1} (paprika). The order of the heating rates for samples mostly represented a good match with their DP values as shown in Figure 4.2. Moreover, the lower the ε″, the longer heating time was required to reach target temperature during RF heating. To further study the influence of mixing on the RF heating rate, experimental temperature time profiles of the mixed paprika (P), curry powder (CP) and black pepper (B) with different proportions were obtained and shown in Figure 4.6. Similarly, an increase in temperature was obtained for all samples. The heating rate of mixtures ranged from 6.12 to 12. 28°C min^{-1} in the following order: P-CP-B (50-25-25%) > P-CP-B (25-25-50%) > P-CP-B (33-33-33%) > P-CP-B (25>50>25%). The heating rates of mixtures were also in agreement with their DP values (Table 4.3). As explained above, the ε″ value of samples plays an important role in heating rate during the RF heating. Additionally, the mixture of black pepper (B) and garlic powder (GP) was also tested for heating rates, and the heating rate increased with an increasing proportion of black pepper in the mixture, which resulted in an increase in DP values of the black pepper and garlic powder mixture (Figure 4.7). For instance, as the fraction of black pepper in mixture increased from 25 to 75 %, the heating rate increased from 2.95 to 5.91 °C min^{-1}. On the other hand, the salt content caused a decrease in heating rate of black pepper (B) - salt and garlic powder (GP) - salt mixtures (Figure 4.8). Results agreed with DP values of both salt mixture samples. As shown in Figure 4.8, the increase in salt content from 20 to 40 % resulted in a decrease from 6.42 to 5.26 °C min^{1} and from 2.47 to 1.03 °C min^{-1} for B-salt and G-salt mix, respectively. These results indicate that the RF heating rates of spices can be modulated through adjusting mixing rates of different spices.

Radio frequency heating uniformity

Figure 4.9 shows the temperature distribution of the upper surfaces for all samples at 105 mm electrode gaps as treated in the RF system. In general, an edge heating was observed in the top surface of samples, and cold spots appeared in the center area. This result was in agreement with previous studies, which reported similar heating patterns for coffee bean (Pan et al., 2012), rice (Zhou et al., 2015), wheat flour (Tiwari et al., 2011), mung beans (Huang et al., 2015), chili powder (Li et al., 2016), potato starch (Zhu et al., 2017), corn grains (Zheng et al., 2017), and corn flour (Ozturk et al., 2017). The average temperature and uniformity index values (UI) of RF heated samples were exhibited in Table 4.6. The UI values ranged from 0.012 to 0.091 and were used to compare RF heating uniformity in spices after heating. As seen in Table 4.7, the highest and lowest UI values were obtained for paprika and garlic powder as 0.091 and 0.012, respectively, which means that the least and most uniform heating were for paprika and garlic powder, respectively. The UI values are also in agreement with the heating rates of paprika and garlic powder; in other words, higher heating rates resulted in worse heating uniformity in samples. Moreover, during the longer heating times, heat conduction also plays an important role in temperature distribution throughout the sample. The central temperature on the top surface after heating was lower than the geometric center of the sample in the container, which could be due to higher electromagnetic energy existing in the middle region between the two electrodes as well as heat loss from the surface to the air. The effect of mixing on heating uniformity is shown in Figure 4.10. The increase in proportion of curry powder in P-CP-B mixture resulted in a decrease in UI value and average surface temperature. As previously stated, the DPof P-CP-B (25-50-25) mixture was lower than those of other P-CP-B mixtures, resulting in a lower heating rate and a better heating uniformity. A similar observation was also obtained for black pepper and garlic powder mix. To further study the effect of salt content, heating uniformity was also determined for GP-salt and B-salt mixtures. The thermal images (Figure 4.11) indicate that the increase in salt content of mixtures resulted in a better RF heating uniformity after heating and a decrease in average surface temperature.

Conclusions

The parallel plate method combined with mixture equations were used to study the dielectric properties of spices and their mixtures. Inverse, linear and quadratic relationships between dielectric properties of white pepper and frequency, moisture content and temperature were observed. The second-order models provided a good fit to predict dielectric properties of white pepper samples at a significance level of 0.0001. The mass fraction and volume fraction of each spice and salt affected the dielectric properties of seasoning mixtures. RF heating rates of spices and their mixtures were linearly related to moisture content and their mass fraction. The LE and LLLE equations provide a better prediction for DP values of spice mixtures than that of the CRIME equations. The heating rates and uniformities were in agreement with measured dielectric properties. The information provided in this study are useful for designing an effective in package RF pasteurization method for spices and their mixtures with improved heating rates and uniformity.

Acknowledgments

This study was funded by the USDA-NIFA AFRI. Grant Number: 2013-67005-21295. Authors also would like to thank the Turkish Ministry of National Education Council for providing the opportunity to Samet Ozturk for his Ph.D. program.

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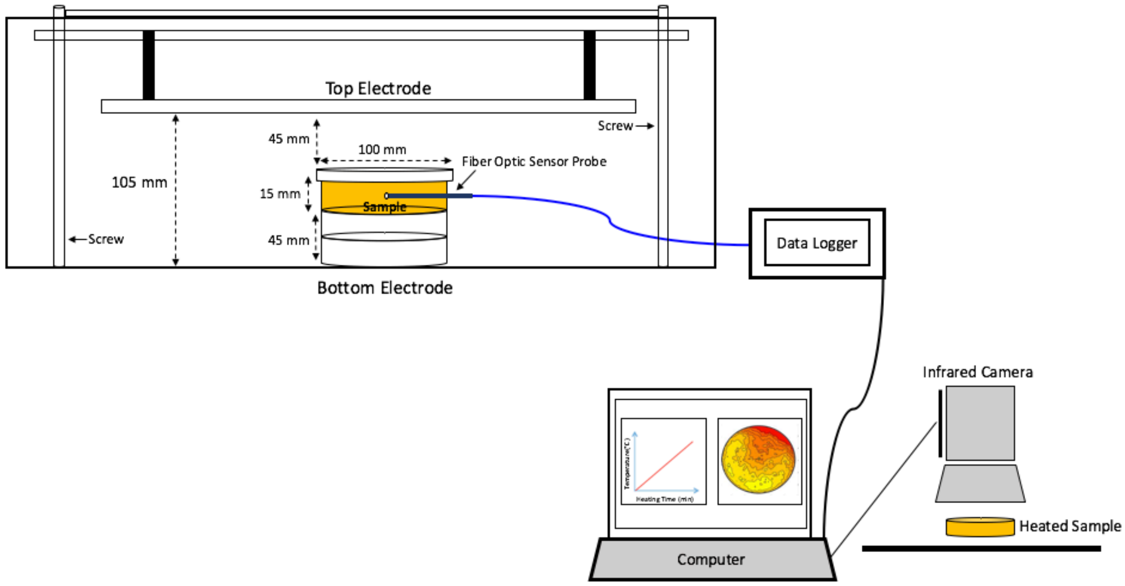
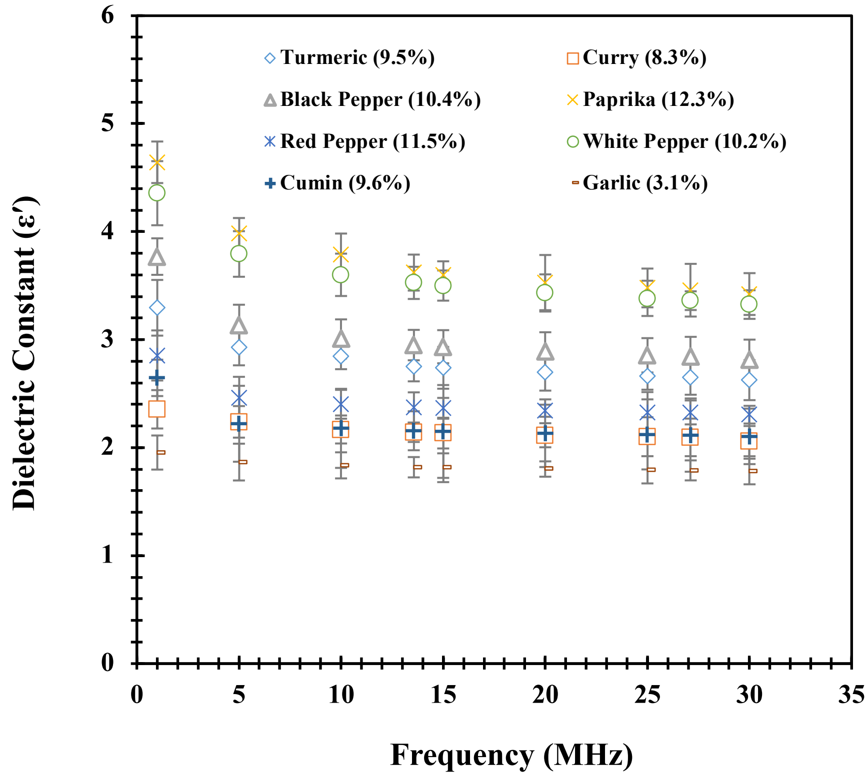


Figure 4.1. Scheme view of sample filled polystyrene petri dish placed in the middle of two parallel electrodes in a 6 kW 27.12 MHz RF system with the fiber optic sensor to monitor the central temperature in the sample, and the infrared camera system



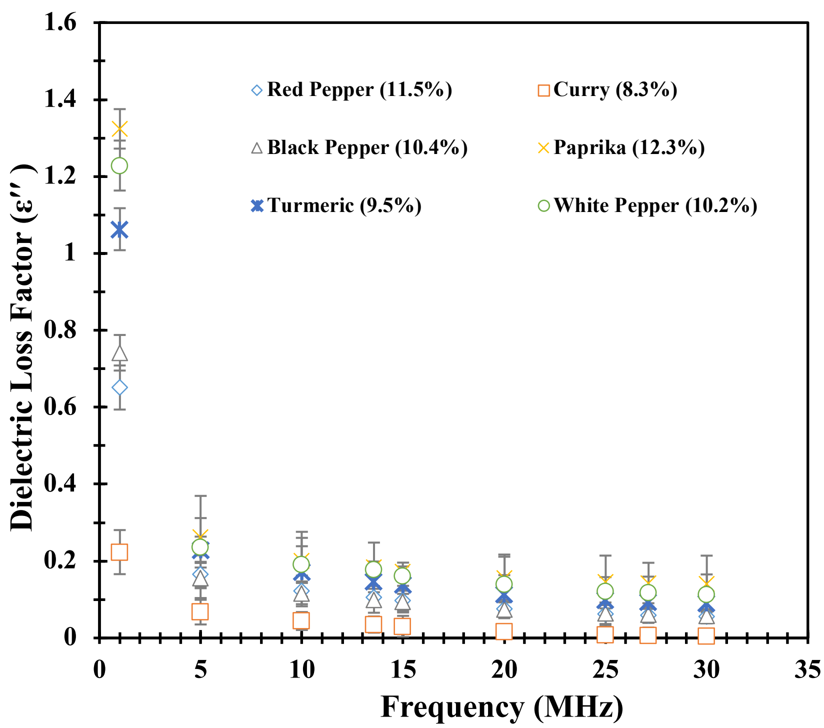


Figure 4.2. Dielectric constant and loss factor of eight different spices in the range of RF frequency (1-30 MHz) at room temperature (23 ± 2 °C)

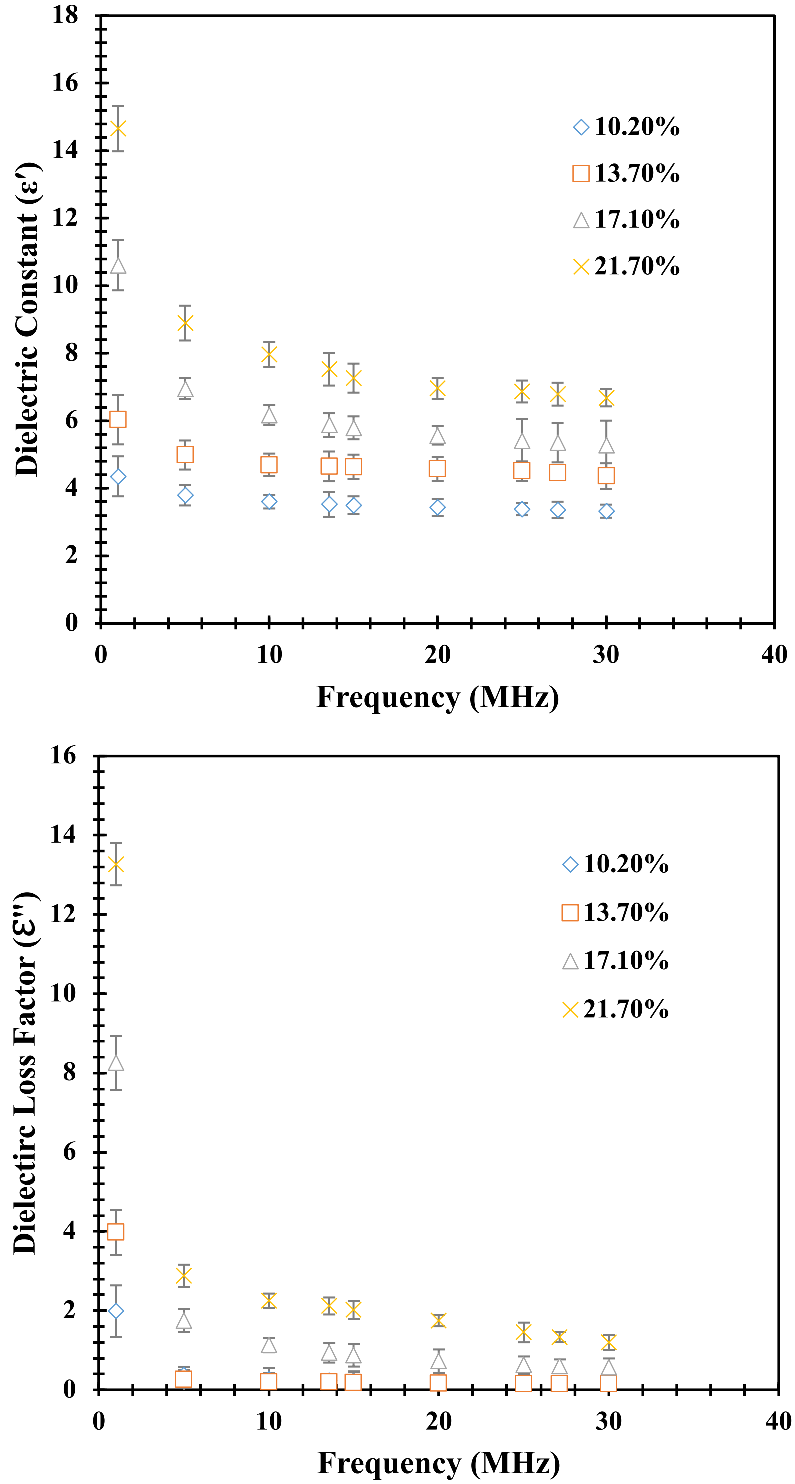


Figure 4.3. Dielectric properties of white pepper as influenced by moisture content (w.b.) in the range of RF frequency (1-30 MHz) at room temperature (23 ± 2 °C)

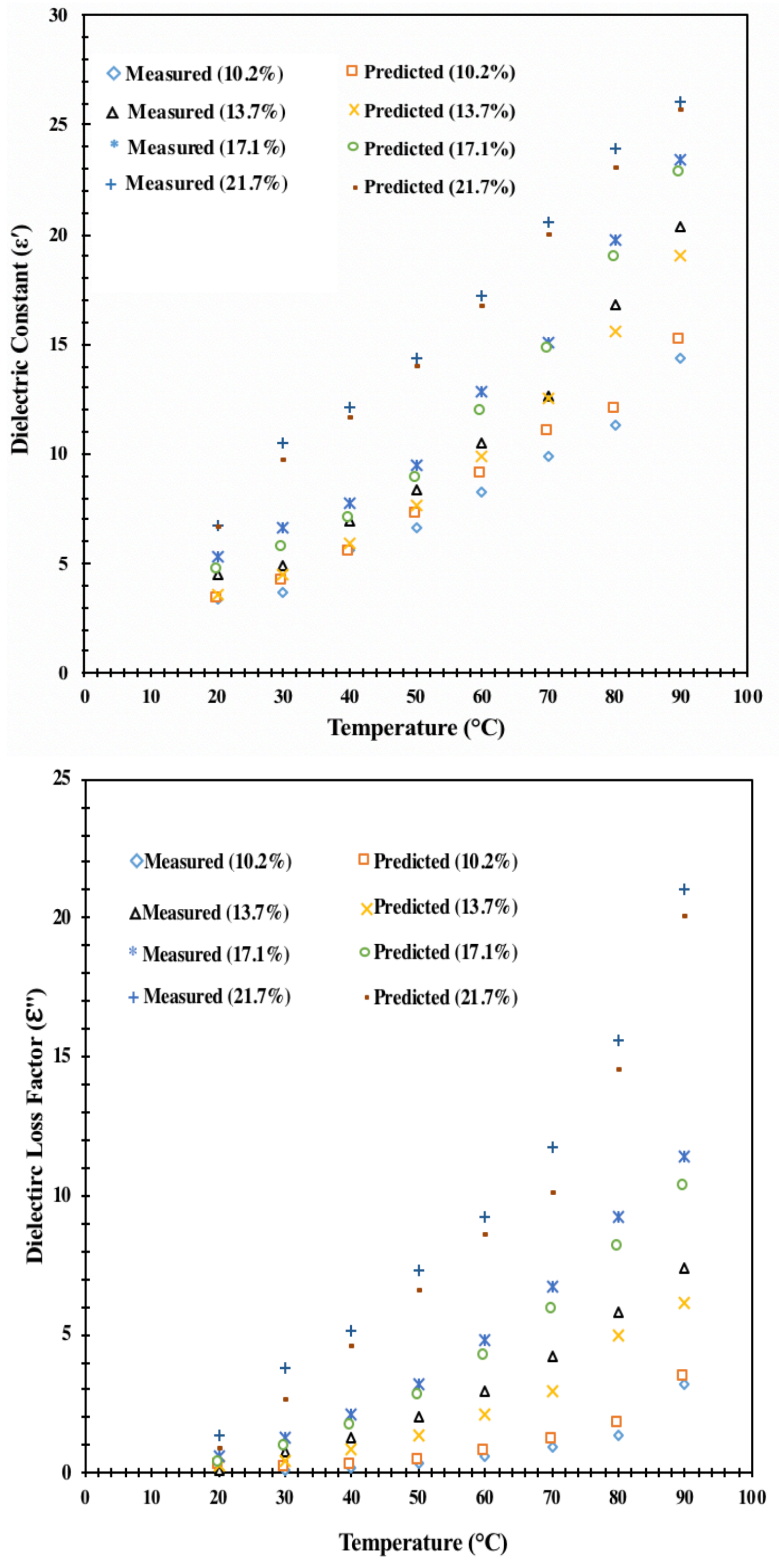


Figure 4.4. The measured and predicted dielectric properties of white pepper as influenced by moisture (10.2, 13.7, 17.1 and 21.7 (w.b.) %) and temperature (from 20 to 90°C) at 27.12 MHz

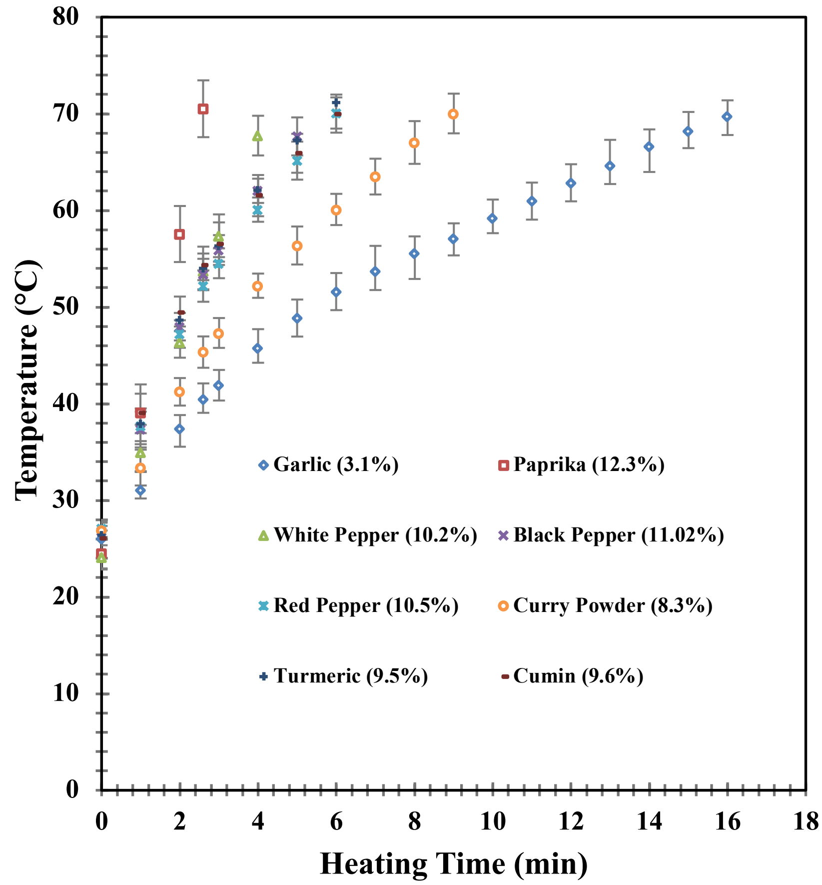


Figure 4.5. Temperature-time profiles of selected spices as heated by the RF system at 105 mm electron gap

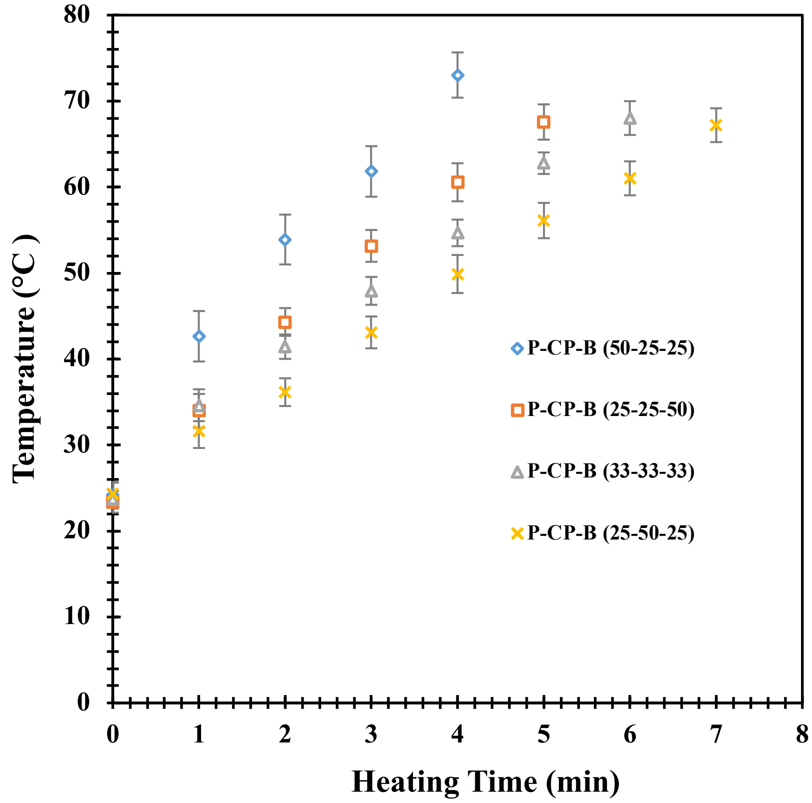


Figure 4.6. Temperature-time profiles of P-CP-B mixture with different mass fractions as heated in the RF system at 105 mm electron gap

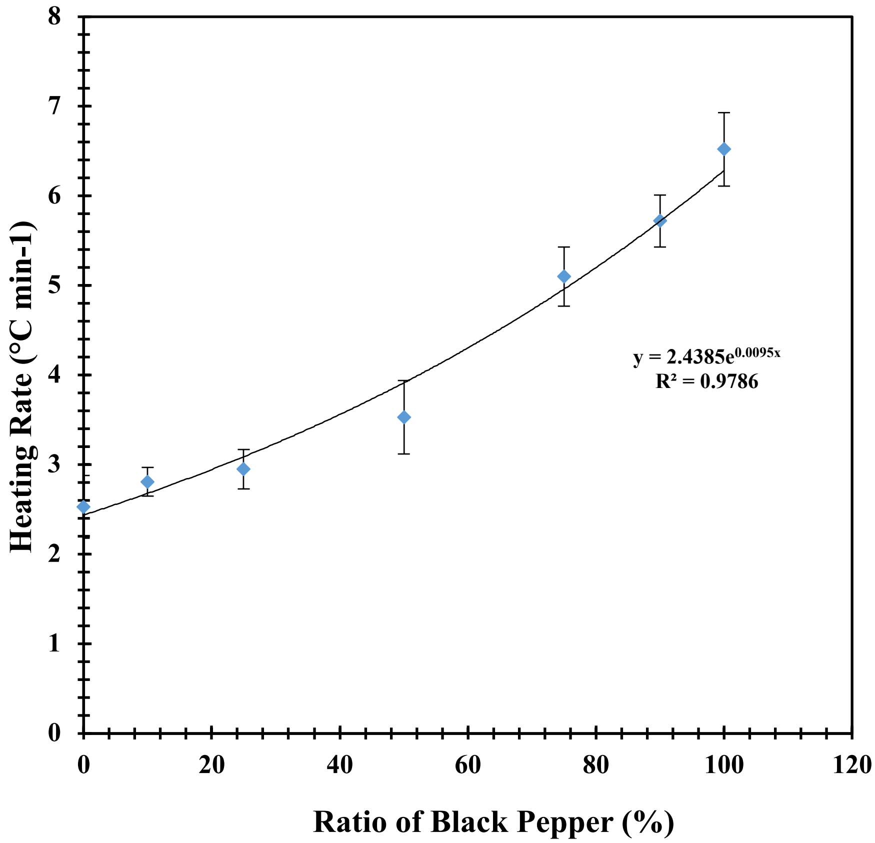


Figure 4.7. Heating rate of black pepper - garlic powder mixture with different mass fractions of black pepper as heated by the RF system at 105 mm electron gap

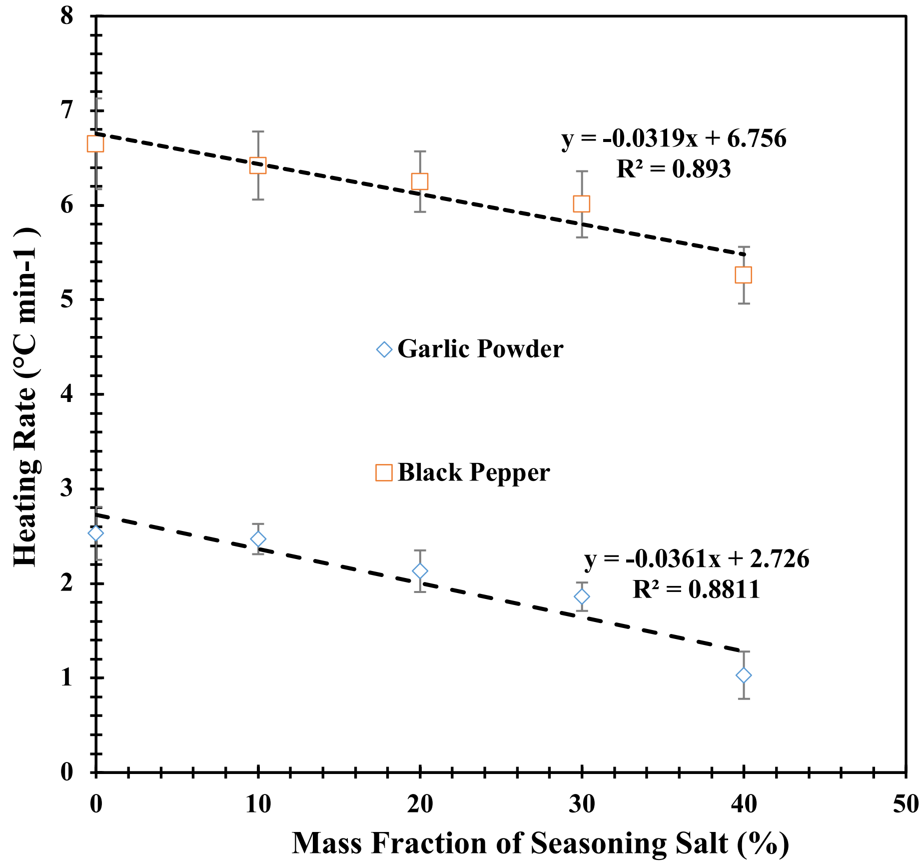


Figure 4.8. Effect of seasoning salt fraction on the heating rate of black pepper, garlic and salt mixture as heated by the RF system

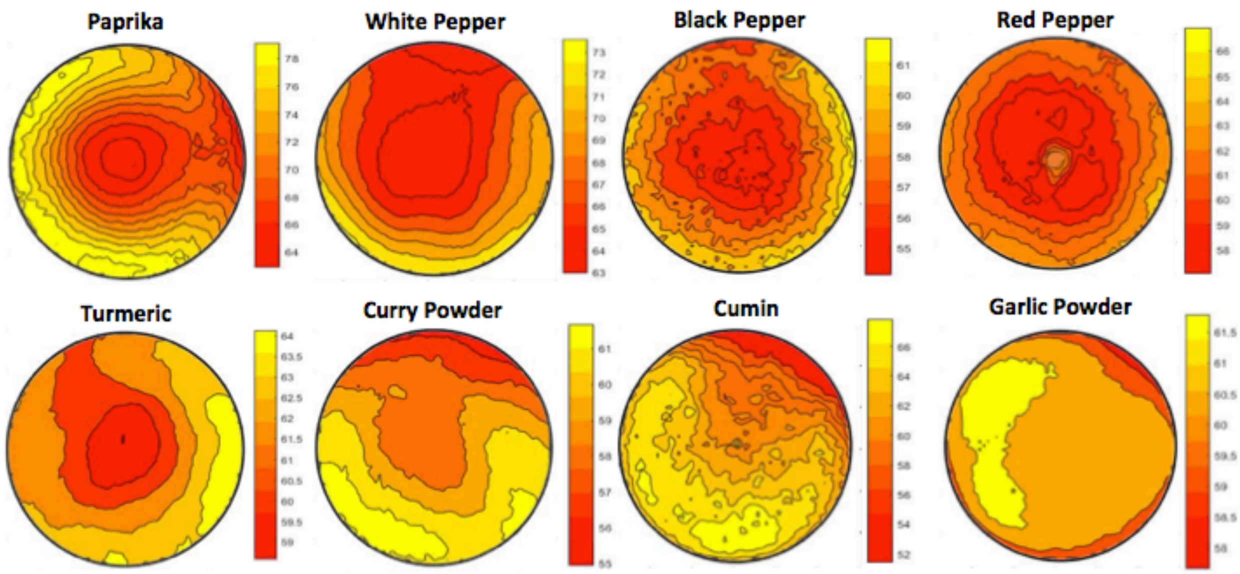


Figure 4.9. Top surface temperature distribution (°C) of spices (50 g) held in a polyester petri dish after heated in the RF system with electrode gap of 105 mm

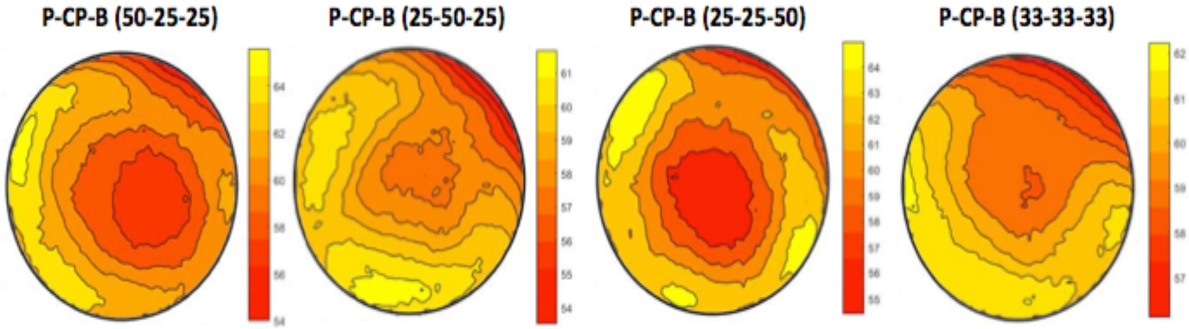


Figure 4.10. Top surface temperature distribution (°C) of paprika (P), curry powder (CP) and black pepper (B) mixtures (50 g) with different mass fractions held in a polyester petri dish after heated in the RF system with electrode gap of 105 mm

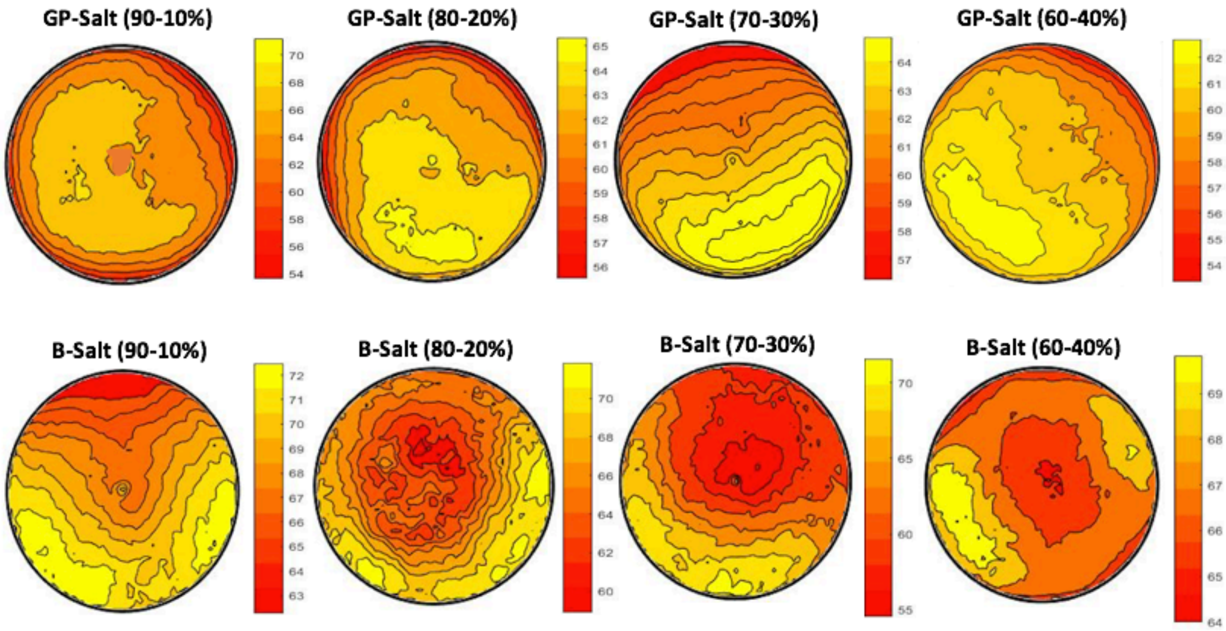


Figure 4.11. Top surface temperature distribution (°C) of black pepper (B), garlic powder (GP) and salt mixtures with different mass fractions held in a polyester petri dish after heated in the RF system with electrode gap of 105 mm

Table 4.1. Regression coefficients of the second-order models to predict the dielectric properties of white pepper at two selected frequencies

	ε'		ε''	
	13.56 MHz	27.12 MHz	13.56 MHz	27.12 MHz
β_0 (Intercept)	2.8529114	1.9670315	12.909306	12.01745
β_1	-0.0055414	-0.02794617	-0.8003574	-1.1320052
β_2	-0.17480004	-0.09898213	-0.5740432	-0.33730058
β_{12}	0.003197467	0.010291806	-0.00489054	0.01927392
β_{11}	0.001847021	0.00664336	0.00267513	0.02520065
β_{22}	0.015351603	0.001515069	0.0316823	0.00158318
R^2	0.96	0.96	0.96	0.98
RMSE	0.27	0.55	0.67	0.54

*RMSE is root mean square error calculated by Eq. (9)

Table 4.2. Bulk and particle density of each solid particulate material in mixtures

	Bulk Density (g cm^{-3})	Particle Density (g cm^{-3})
White Pepper	0.539	1.01
Black Pepper	0.487	1.27
Red Pepper	0.423	1.07
Paprika Powder	0.438	1.21
Curry Powder	0.401	1.15

Table 4.3. Measured and predicted DP values of P-CP-B mixture using mixture equations at 13.56 and 27.12 MHz

Mixture (Mass Bass) (%)	Mixture Bulk Denisty (g/cm^3)	Volume Fraction			MHz		Observed	LE	LLLE	CRIME
		v_P	v_{CP}	v_B						
P-CP-B (25-50-25)					13.56	ε'	2.353	2.484	2.367	3.167
						ε''	0.075	0.068	0.072	0.061
	0.446	0.324	0.387	0.309	27.12	ε'	2.296	2.410	2.321	2.912
						ε''	0.039	0.023	0.034	0.049
P-CP-B (50-25-25)					13.56	ε'	2.782	2.828	2.794	3.512
	0.439	0.312	0.379	0.327		ε''	0.102	0.108	0.106	0.041
					27.12	ε'	2.641	2.693	2.669	3.453
						ε''	0.057	0.059	0.052	0.028
P-CP-B (25-25-50)					13.56	ε'	2.687	2.735	2.718	3.356
	0.442	0.315	0.354	0.307		ε''	0.084	0.089	0.082	0.043
					27.12	ε'	2.615	2.711	2.663	3.291
						ε''	0.050	0.061	0.052	0.037
P-CP-B (33-33-33)					13.56	ε'	2.413	2.579	2.492	3.363
	0.417	0.334	0.362	0.327		ε''	0.081	0.088	0.079	0.041
					27.12	ε'	2.354	2.493	2.327	3.329
						ε''	0.048	0.069	0.041	0.027

* The v_P, v_{CP}, and v_B represent the volume fraction of paprika, curry powder and black pepper in mixture, respectively.

Table 4.4. Measured and predicted DP values of red (R) and white (W) pepper mixture using mixture equations at 13.57 and 27.12 MHz

Mixture (%)	Bulk Density (g/cm^3)	Volume Fraction v_R	Volume Fraction v_W	MHz		Observed	LE	LLLE	CRIME
R-W (90-10)	0.4606	0.431	0.453	13.56	ε'	2.551	2.467	2.579	1.982
					ε''	0.082	0.059	0.069	0.073
				27.12	ε'	2.467	2.413	2.541	1.475
					ε''	0.079	0.051	0.062	0.012
R-W (75-25)	0.439	0.401	0.437	13.56	ε'	2.971	2.898	3.113	2.619
					ε''	0.116	0.121	0.128	0.049
				27.12	ε'	2.858	2.752	2.921	2.214
					ε''	0.045	0.061	0.054	0.012
R-W (60-40)	0.459	0.442	0.408	13.56	ε'	3.301	3.012	3.242	2.771
					ε''	0.155	0.112	0.137	0.072
				27.12	ε'	3.160	2.795	3.172	2.518
					ε''	0.075	0.054	0.082	0.028
R-W (50-50)	0.418	0.391	0.413	13.56	ε'	3.428	3.016	3.459	2.302
					ε''	0.191	0.137	0.108	0.036
				27.12	ε'	3.212	2.796	3.411	2.021
					ε''	0.102	0.084	0.092	0.019
R-W (40-60)	0.542	0.422	0.44	13.56	ε'	3.545	3.208	3.731	1.947
					ε''	0.195	0.145	0.159	0.011
				27.12	ε'	3.276	2.931	3.421	2.171
					ε''	0.114	0.089	0.096	0.002
R-W (25-75)	0.446	0.415	0.429	13.56	ε'	3.706	3.192	3.671	2.934
					ε''	0.203	0.154	0.172	0.074
				27.12	ε'	3.318	3.065	3.591	2.872
					ε''	0.125	0.091	0.101	0.045
R-W (10-90)	0.476	0.443	0.469	13.56	ε'	3.913	3.488	3.924	3.145
					ε''	0.221	0.169	0.231	0.064
				27.12	ε'	3.376	3.239	3.541	3.065
					ε''	0.137	0.109	0.195	0.049

* The v_R and v_W represent the volume fraction of red and white peppers in the mixture, respectively.

Table 4.5. Dielectric constant of black pepper (B) and garlic powder (GP) mixtures with various mass fractions at 13.56 and 27.12 MHz

		Black Pepper Mass Fraction (%)											
		10		25		40		60		75		90	
	MHz	ε'	ε''	ε'	ε''	ε'	ε''	ε'	ε''	ε'	ε''	ε'	ε''
B-GP	13.56	1.87±0.04	-	1.94±0.012	-	1.98±0.021	-	2.09±0.017	-	2.16±0.14	2.16±0.14	0.07±0.14	0.084±0.023
	27.12	1.82±0.03	-	1.89±0.015	-	1.92±0.013	-	2.01±0.022	-	2.11±0.15	2.11±0.15	0.04±0.15	0.047±0.02

Table 4.6. Dielectric constant of black pepper and salt, and garlic powder and salt mixtures with different mass fractions of salt at 13.56 and 27.12 MHz

		Salt Mass Fraction (%)			
		10	20	30	40
	MHZ	ε'	ε'	ε'	ε'
Black Pepper- Salt	13.56	2.56	2.10	1.83	1.67
	27.12	2.44	2.06	1.77	1.61
Garlic Powder-Salt	13.56	1.84	1.47	1.19	0.77
	27.12	1.79	1.41	1.14	0.72

Table 4.7. Average surface temperature and uniformity index of spices as heated by the RF system

	Cumin	White Pepper	Black Pepper	Red Pepper	Paprika	Curry Powder	Garlic Powder	Turmeric
Uniformity Index (λ)	0.057	0.086	0.069	0.051	0.091	0.043	0.012	0.078
Average Temperature (∘C)	62.9 ± 3.2	67.3 ± 1.4	66.9 ± 1.8	64.3 ± 2.3	72.9 ± 4.5	60.9 ± 1.64	58.7 ± 0.7	62.1 ± 1.53

CHAPTER 5

RADIO FREQUENCY HEATING OF CORN FLOUR: HEATING RATE AND UNIFORMITY[3]

[3]S. Ozturk, F. Kong, R.K. Singh, J.D Kuzy, C. Li. Accepted by *Innovative Food Science and Emerging Technologies.*

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Abstract

Non-uniform heating is a major challenge for using radio frequency (RF) heat treatment in pasteurization of low moisture food products. The objective of this study was to evaluate the effect of different electrode gaps, moisture content (MC), bulk density and surrounding materials on RF heating uniformity and rate in corn flour. Additionally, the dielectric and thermal properties of corn flour were determined as affected by MC, temperature (°C), and frequency (MHz). Changes in MC, water activity (a_w) and color in the sample after RF heating were measured to evaluate treatment effect on food quality. A precision LCR meter and a liquid test fixture were used to study DP of the sample at RF frequency ranging from 1 to 30 MHz. The RF heating uniformity and temperature profiles of corn flour as exposed to RF heating were obtained with an infrared camera and data logger using a fiber optic sensor. The DP values increased with increasing MC and temperature but decreased with increasing frequency. The heating rate increased from 3.5 to 6.8 °C min^{-1} with increasing MC (from 10.4 to 16.7%) but decreased from 12.7 to 5.2 °C min^{-1} with increasing electron gap (from 11 to 15 cm). The corner and edge heating were observed at all layers of the samples for all the distances, and the hottest and the most uniform layer were determined as the middle layer at an electrode gap of 15 cm. Glass petri dish provided better uniformity than those of polyester plastic petri dish. Covering by foam led to more uniform RF heating uniformity in corn flour, and better moisture and a_w distribution. This study provided useful information to develop an effective RF process as an alternative of conventional thermal treatments for pasteurization of low-moisture products.

Key words: Radio frequency heating, Corn flour, Heating uniformity, Heating rate

Introduction

Low moisture foods including dried spices, vegetable powders, whole milk powder, corn and wheat flour, are defined as food with water activity (a_w) less than 0.7 or moisture content (MC) of below 20% (Blessington, Theofel, & Harris, 2013). Although the low water activity environment is not suitable to grow microorganisms, low water activity foods are liable to be contaminated by microorganisms during harvesting, processing or transportation. A number of recent outbreaks of salmonellosis were associated with contamination of low moisture foods (CDC, 2007, 2010), and caused severe safety problems leading to foodborne illness such as diarrhea, abdominal pain, mild fever, and chills (Bairdparker, 1990; Rhee, Lee, Dougherty, & Kang, 2003). The pasteurization of *Salmonella* contaminated low moisture foods is difficult because of the high heat resistance of *Salmonella* in low water activity environment. Different decontamination methods have been applied to reduce the microbial hazard of low moisture foods, such as steam, hot air and irradiation. However, conventional methods including steam and hot air require long processing time, which leads to severe quality degradation in low moisture foods, due to their low thermal conductivity. Vancauwenberge, Bothast, and Kwolek (1981) reported the inactivation effect of dry hot air on corn flour contaminated with eight different Salmonella serotypes. Their results showed that corn flour with moisture content 10 and 15% required 5.8 and 2.2 h for 99% log reduction at 49°C, respectively. On the other hand, irradiated foods are not readily accepted by consumers due to health concerns (Farkas, 2006; Schweiggert, Carle, & Schieber, 2007). As a method of volumetric heating, radio frequency (RF) heating offers the possibility to rapidly pasteurize low moisture foods while maintaining the food quality. The RF covers a wide band of frequencies ranging from 1 to 300 MHz, but only 13.56, 27.12 and 40.68 MHz are used for industrial, scientific and medical applications (Wang and

Tang, 2001). Studies were reported recently in using the RF treatments for postharvest disinfection (S. Jiao, Johnson, Tang, & Wang, 2012a; Lagunas-Solar, Pan, Zeng, Truong, & Khir, 2007), and pasteurization of low moisture foods (Gao, Tang, Johnson, & Wang, 2012; Jeong & Kang, 2014; Kim, Sagong, Choi, Ryu, & Kang, 2012). Compared to microwave (MW) heating which involves high frequency (915 or 2450 MHz), RF heating ensures more uniform heating and deeper penetration depth in solid and semi-solid low moisture foods due to the lower frequency range and longer wavelengths (Luechapattanaporn, Wang, Wang, Tang, Hallberg, & Dunne, 2005; Marra, Zhang, & Lyng, 2009). Despite of improved heating uniformity as compared to microwave heating, the non-uniformity and run-way heating are still major challenges to apply RF heating pasteurization system in food industry. The variation in temperature distribution of food during RF heating can lead to overheated hot area causing severe quality deterioration and under-heated cold spot. Several researches have been conducted to improve RF heating uniformity using alternative ways. Wang et al. (2010) reported that as legumes were heated by RF oven using combination of forced hot air and shaking container on a conveyor belt, a decrease between hot and cold spot temperatures was achieved. There are several factors affecting heating uniformity during the RF Heating including electron gap, power, packaging material and geometry, dielectric properties, and physical and chemical properties of heated sample (S. Wang, Monzon, Gazit, Tang, & Mitcham, 2005). Recently, it was reported that surrounding materials, moisture content and bulk density can also impact the RF heating uniformity (Huang, Zhu, & Wang, 2015; Jiao, Tang, & Wang, 2014; Zhang, Zhu, & Wang, 2015). Jiao et al. (2014) studied the effect of surrounding material, which has low dielectric and thermal properties, on the RF heating uniformity in peanut butter and wheat flour. They found that covering the plastic container with PEI material resulted in decrease in the average temperature difference (from 13 to 7°C) on the surface layer. Furthermore, the effect of polyurethane foam sheets on the RF heating uniformity in low moisture foods including walnut and bread have been studied by Liu, Wang, Mao, Tang, and Tiwari (2013) and Wang et al. (2010). However, there is not any reported study on the effect of foam as used surrounding material. Therefore, it is important to experimentally determine the effect of MC and bulk density, and different surrounding material on RF heating uniformity in order to develop optimized RF heating parameters and to minimize unfavorable effect on food quality.

This study aims to investigate the heating behavior of corn flour during RF treatment as affected by different factors during RF heating, and to explore methods to improve heating uniformity by using polystyrene foam and PEI plates as surrounding materials. The obtained information will help to develop an effective RF pasteurization method for packaged low moisture foods.

Material and methods

Physical characterization of corn flour

Commercially processed corn flour was purchased from Georgia Spice Company (Atlanta, GA USA). The initial MC of the corn flour was determined by drying the sample in a vacuum oven at 105 °C for 16 h (AOAC, 1998). To study the effect of moisture on dielectric and thermal properties, heating uniformity and rate, MC of the corn flour was adjusted by spraying distillated water to the samples held in ziplock bags, and shaken manually for 10 min. The water sprayed corn flour samples were stored for two days at 4 °C and shaken twice a day to obtain a uniform moisture distribution throughout the sample.

To determine the effect of bulk density on RF heating uniformity and rate, different bulk densities of corn flour with initial MC (10.2 % w.b.) at room temperature (23 ± 2°C) were obtained by a basic volume method using a polystyrene plastic petri dish (100 x 15 mm). For each density level, the cylindrical petri dish was filled with corn flour and then weighed. The bulk density was calculated as weight of flour/volume (g/cm^3).

Thermal conductivity, diffusivity and specific heat of corn flour with three different MC at room temperature were measured with a KD2 Pro thermal analyzer (Decagon Devices, Inc., USA) using a T1 probe, which is suitable for solid and granule food materials. For each measurement, almost 20 g sample was placed into a plastic container and measurement probe was inserted in the sample to determine thermal properties of corn flour.
All measurements were done in triplicate. The mean value and standard deviation (SD) were reported**.**

Determination of Dielectric properties and penetration depth

The DP of the corn flour were determined using parallel plate method with an Inductance Capacitance and Resistance (LCR) meter (4285A, Agilent Technologies, Palo Alto, CA) with a dielectric liquid test fixture (16452, Agilent Technologies, Palo Alto, CA). The LCR meter was calibrated manually to minimize the biased error and random error using a BNC cable before taking measurement. Dielectric properties of corn flour with various MC (10.4, 13.6 and 16.7 % (w.b.)) at temperature range between 20 to 80°C were measured at 13.56 and 27.12 MHz, as determined by the capacitance (C_p) and resistance (R_p) values obtained from LCR meter. The experimental procedure was described by Ozturk, Kong, Trabelsi, and Singh (2016). The corn flour sample (2 g) was placed into the test fixture. The fixture was then tightly closed and placed into a temperature chamber (625G, Thermo Fisher Scientific Inc., Waltham, MA, USA,) and C_p and R_p values were measured with different frequencies (ranging from 1 to 30 MHz) and temperatures (ranging from 20 to 80°C with 10°C interval). The dielectric constant (ε') and loss factor (ε'') of the corn flour were calculated using the following equations (Agilent Technologies, 2000; Halliday, 2001; Von Hippel, 1954).

$$\varepsilon' = \frac{tC_p}{A\varepsilon_0} \qquad \text{Eq. (1)}$$

$$\varepsilon'' = \frac{t}{2\pi f R_p \varepsilon_0 A} \qquad \text{Eq. (2)}$$

where, t is the gap (m) between electrodes of the test fixture, C_p is parallel capacitance (F), R_p is the resistance (Ω), f is the frequency (Hz), ε_0 is the permittivity of vacuum (8.854×10^{-12} F m^{-1}), and A is the electrode area (m^2).

When exposed to an electromagnetic field, food materials, which have poor electric conductivity, are able to store and release electrical energy as heat energy (Buffler, 1993). The penetration depth of RF and MW can be defined as depth into a sample where the power has dropped to 1/e (e=2.718) of the power value at the surface (Guan, Cheng, Wang, & Tang, 2004). The penetration depth is a function of dielectric properties of food material, and it can be calculated using the following equation (Buffler, 1993; Von Hippel, 1954)

$$d_p = \frac{c}{2\pi f\sqrt{2\varepsilon'\left(\sqrt{1+(\tan\delta)^2}-1\right)}} \qquad \text{Eq. (3)}$$

where, c is the speed of light in air (3×10^8 m s^{-1}), f is the frequency (Hz) and $\tan\delta$ is defined as ($\frac{\varepsilon''}{\varepsilon'}$), ε' and ε'' are the measured dielectric constant and dielectric loss factor values of corn flour with various MC, respectively. Using obtained dielectric properties, d_p (m) values of RF at frequencies of 13.56 and 27.12 MHz in corn flour were calculated at room temperature (23 ±

2°C) and target temperature of RF heating (80 ℃).

Determination of heating uniformity and temperature time profile of corn flour during RF heating

Heating uniformity tests were conducted by placing corn flour in a rectangular polyetherimide (PEI) container (Inner dimension: 7 (H) x 24 (W) x 30 (L) cm^3) (Figures 5.1 and 5.2) and treated in a 27.12-MHz,6-kW RF system (COMBI 6-S, Strayfield International, Wokingham, UK). To develop an effective RF heating protocol for better RF heating uniformity, the optimal electrode gap was determined first. Prior to the RF treatment, the corn flour with MC of 10.4 % (w.b.) were equilibrated at the room temperature (23 ± 2°C). The PEI container was filled with 3.2 kg corn flour, and placed in the middle of two parallel electrodes (Figure 5.1) for RF heating at various electrode gaps including 11, 13 and 15 cm. In the view of previous studies, placing the sample in the middle of parallel electrodes during RF heating can provide better heating uniformity and rate (Tiwari, Wang, Tang, & Birla, 2011). The sample in the PEI container was heated from room temperature (23 ± 2°C) to 80 ± 5°C, which is in the range of lethal temperature required kill most insects and microorganisms at all growth stages (Armstrong, Tang, & Wang, 2009; Johnson, Valero, Wang, & Tang, 2004). To obtain the temperature time history and calculate the heating rate in corn flour during RF heating, the change in temperature of the sample during heating was recorded at the geometric center of the PEI container using a fiber optic temperature sensor with accuracy of ± 1°C (Fiso Tech. Inc., Quebec, Canada) connected to a data logger. When the center temperature reached 80 °C, the sample was taken out immediately in order to take a thermal image. The whole samples in the PEI container were divided into three different interior layers (Figure 5.2) separated by a thin cheese cloth (with mesh opening of 1 mm) to map the horizontal surface temperature using an infrared camera

(FLIR T440, FLIR Systems, Inc., North Billerica, MA, USA) with an accuracy of ± 2 °C. After RF treatment, the top surface and interior layer images were taken by infrared camera to analyze the surface temperature distribution. To determine the effect of the cheese cloth on RF heating uniformity, the interior temperature distribution of each layer at 13 different locations (Figure 5.3) was also measured by portable data loggers (Model RDXL4SD, OMEGA Engineering Inc., Norwalk, CT, USA) using T type thermocouples (Model 91100-20, Cole-Parmer Instrument Company, Vernon Hill, IL, USA) with having an accuracy of ±2°C after RF heating. All the treatments were replicated three times. The average and standard deviation (SD) values of the surface and interior sample temperatures were used to evaluate the RF heating uniformity. The mapped temperature distribution for different layers and locations were used to determine hot and cold spots in corn flour after RF heating.

The RF heating uniformity in corn flour of four different layers with different distances from surface and bottom (Figure 5.2) at the target temperatures was evaluated using the heating uniformity index (UI), which has been used to evaluate RF heating uniformity for many studies (Hou, Ling, & Wang, 2014; S. Jiao, Johnson, Tang, & Wang, 2012b; Pan, Jiao, Gautz, Tu, & Wang, 2012; S. Wang, Luechapattanaporn, & Tang, 2008; S. Wang, Monzon, M., Gazit, Y., Tang, J., Mitchan, E.J., Armstrong, J.W., 2005; S. Wang, Tiwari, Jiao, Johnson, & Tang, 2010). S. Wang, et al. (2005) defined the uniformity index as the ratio of the rise in standard deviation of sample temperatures to the rise in the average sample temperatures during the RF heating. It can be calculated using the following equation (S. Wang, Monzon, M., Gazit, Y., Tang, J., Mitchan, E.J., Armstrong, J.W., 2005):

$$\lambda = \frac{\Delta\sigma}{\Delta\mu} \qquad \text{Eq. (4)}$$

where $\Delta\sigma$ is the rise in the standard deviation (SD) of sample temperature (°C), and $\Delta\mu$ is the rise of the mean temperature (°C) over the RF heating time. The smaller UI values indicate better RF heating uniformity in the sample.

As a comparison, the corn flour filled PEI container was placed into a temperature chamber (625G, Thermo Fisher Scientific Inc., Waltham, MA, USA) with 85°C temperature until the center temperature reached the target temperature (80°C) in the middle of container. The change in temperature of the sample during heating was recorded using a data logger with fiber optic temperature sensor (Fiso Tech. Inc., Quebec, Canada). The time required to reach the target temperature were compared with RF heating. The color of the samples from the two methods was also compared to show the effect of the heating process on food quality.

Effect of surrounding material, moisture content and bulk density on heating uniformity

Based on the recent studies, surrounding materials with similar dielectric properties to treated sample can help improve heating uniformity during the RF heating (Y. Jiao, et al., 2014). In this study, the effect of four surrounding materials, including glass, polystyrene, polyurethane foam and polyetherimide were studied. It was reported that the squared, sphere, and cylinder shape containers commonly used in RF heating systems for different foods demonstrated similar heating uniformity pattern in during RF heating (Alfaifi, et al., 2014; Birla, Wang, & Tang, 2008). For our research, to investigate the influence of surrounding materials on heating uniformity, average temperature and heating rate in corn flour during RF heating, polystyrene and glass cylindrical petri dishes (100 x 15 mm) were used to hold sample. The dielectric properties of surrounding materials, including glass, polystyrene, polyurethane foam and polyetherimide, are shown in Table 5.1. The polystyrene and glass cylindrical petri dishes were filled with 50 g corn flour before placed in the middle of two parallel electrodes at a gap of 15 cm. Samples were heated to reach 70°C, and the change in temperature was recorded using a data logger with a fiber optic sensor, which was placed into the geometric center of cylindrical petri dishes. After RF heating, sample was taken off immediately from the RF oven, and the top surface image was taken by an infrared camera to evaluate the effect of different surrounding material on heating uniformity using the UI values. Furthermore, three different approaches were also tested to determine effect of surrounding material on RF heating uniformity and rate: 1) the polystyrene and glass cylindrical containers were covered all around with polyurethane foam sheet (5 mm thickness), and 2) the petri dish containers was covered by PEI cylindrical blocks with different thickness (diameter * thickness: 100 x 10 or 20 mm) on both top and bottom surface or 3) only one side of the sample container was covered by PEI block to determine the position of PEI blocks on the heating uniformity. In the view of recent studies, the foam box and PEI cylindrical blocks helps to absorb more electrical energy and to enhance the electromagnetic field uniformity around the food sample (Y. Jiao, et al., 2014; S. Wang, et al., 2010; Y. Y. Wang, Zhang, Gao, Tang, & Wang, 2014)

To evaluate the effect of moisture content and bulk density on RF heating uniformity and heating rate, corn flour with various moisture content and bulk density levels (0.42, 0.53 and 0.61g cm^{-3}) at room temperature were heated in the polystyrene plastic petri dish. The thermal images of the top surface and temperature time profiles were used to determine uniformity index values and heating rate in corn flour.

Evaluation of color and moisture distribution in RF treated samples

RF treated corn flour samples prepared with foam covered PEI container were used to evaluate the influence of heating on the quality of final products. Color was selected as the quality parameter to evaluate corn flour quality before and after as the quality parameters to evaluate corn flour quality before and after RF heating. L*, a*, and b* values were obtained from five different locations in the PEI container (Figure 5.2), and analyzed using a Minolta colorimeter (model CR300, Minolta Co., Osaka, Japan). L*, a*, and b* values indicate the lightness of color, whiteness, and yellowness of the sample, respectively. The average of the five locations was used to represent color change in sample after RF heating. In addition, MC and a_w of the corn flour were also used to evaluate the effect of RF heating. To determine moisture distribution in container after RF heating, samples were obtained from selected locations (Figure 5.2) throughout the PEI container, and MC were determined by the vacuum oven drying method. Water activity at different locations was also measured using a water activity meter (Aqualab serious 3TE, Decagon 221Devices Inc., Pullman, WA, USA) at room temperature. All measurements were conducted three times.

Data Processing and Analysis

All measurements, including dielectric properties and heating rates, were done in triplicate. The mean value and standard deviation (SD) were reported. Excel (Microsoft office, Redmond, WA, USA) was used to process and analyze data.

Results and Discussion

Dielectric and thermal properties of corn flour as influenced by moisture content, temperature and frequency

The dielectric properties of corn flour with various moisture levels (10.4, 13.6 and 16.7 % w.b.), and two different temperatures at frequency of 13.56 and 27.12 MHz are listed in Table 5.2. Both ε' and ε'' of corn flour increased with increasing MC and temperature but decreased with increasing applied frequencies. The increase in both ε' and ε'' can be explained with increased amount of free and bound water in corn flour, and the increase in mobility of water molecules as temperature increased. Furthermore, the effect of frequency on decrease on both ε' and ε'' of corn flour can be explained by the changes in ionic conduction in the samples. Ryynänen (1995) reported that dielectric heating below 300 MHz is based on the ionic conduction, which decreases as frequency increases, resulting in decrease in dielectric constant (Shrestha & Baik, 2013). On the other hand, the penetration depth of RF in corn flour decreased with increasing MC and temperature, and frequency (Table 5.2). It means that lower frequencies (1-300 MHz) provide deeper penetration than those of high frequencies (915 or 2450 MHz), which means that high frequency results in much more surface heating than that of RF range. Other researchers also reported decreases in penetration depth as frequency increased for various food powders including wheat flour (Nelson & Trabelsi, 2006), soybean flour (Guo, Wu, Zhu, & Wang, 2011), and broccoli powder (Ozturk, et al., 2016). The penetration depth is an important parameter to evaluate efficiency of RF heating in low moisture foods, and to design an effective RF heating system. The Results indicate that it is important to consider the moisture content of low moisture foods to design a high penetration capacity RF heating system.

Table 5.2 also shows the thermal properties values including conductivity, diffusivity and specific of corn flour with various MC at room temperature. Thermal conductivity of corn flour lies in range of 0.123 - 0.166 (W/ m °C) as MC content increased from 10.4 to 16.7 % (w.b) at room temperature (23 ± 2°C). It is important to know the thermal properties of corn flour since as RF heating generates heat in the food, heat transfer happens as conduction throughout sample and convection between container surface and surrounding air, which affect heat distribution. Bozikova (2003) reported a linear relationship between moisture content and thermal conductivity for corn and wheat flour, and low moisture foods has low thermal conductivity which require a long heating time with conventional thermal treatment.

Determination of RF heating rates in the corn flour at three different electrode gaps

Figure 5.4 shows the temperature change in corn flour with MC 10.4 % (w.b.) and 0.501 a_w at the geometric center of container in the middle of two parallel electrodes during RF heating for three selected electrode gaps of 11, 13 and 15 cm, with heating times required to raise the temperature from 23 ± 2°C to 80°C for each gap being about 4, 9 and 13 min, respectively. A linear increase in temperature of corn flour was obtained for all electron gaps during RF heating (Figure 5.4). The heating rate in corn flour during the RF heating decreased from 12.71 to 5.2 °C min^{-1} as the electron gap increased. The high heating rates correspond to higher throughputs and shorter processing times, which are desired, but it adversely affects the heating uniformity in sample during the RF heating because of rapid and run-away heating. Similar temperature time profiles to corn flour as heated by RF heating were also reported for different low moisture agriculture foods including almond, lentil, chickpea, and green peas (Gao, Tang, Wang, Powers, & Wang, 2010; S. Wang, et al., 2010). To compare the efficiency of RF heating with conventional heating system, sample was also heated by hot air in temperature chamber, and it took almost 210 min with 0.28°C min^{-1} heating rate to reach target temperature because of poor heat conduction in corn flour, which has a low thermal conductivity (0.13 W/m °C) at 10.4 % (w.b.) MC (Table 5.2). Although RF heating was much faster than hot air heating in corn flour, its heating uniformity is a major challenge to achieve safe products with high food quality. Therefore, an electron gap of 15 cm corresponding to a relatively longer heating time (13 min) was selected for RF pasteurization of corn flour, as it has better heating uniformity than those of smaller electron gaps.

Radio Frequency heating uniformity in the corn flour

Figure 5.5 shows the experimental temperature distribution of the corn flour in top and middle layers after RF heating for each electrode distance. The overheating of corner and edge were observed in both top and middle layers of sample for all distances, and cold spots were located at center area in each layer. Similar observations were reported for RF heated coffee bean (Pan, et al., 2012), rice (Zhou, Ling, Zheng, Zhang, & Wang, 2015), and wheat flour (Tiwari, et al., 2011). The more detailed comparison of temperature distribution, and uniformity index values in RF heated corn flour for each layer at different electrode gaps are shown in Table 5.3. The UI was used to compare heating uniformity in corn flour for different distances after RF heating. The average temperature in middle layer was higher than in top layer at each electrode gap (Table 5.3), which could be caused by high electromagnetic fields in the middle of two parallel electrodes, and heat dispersion on top surface to the surrounding air. The most uniform layer, which corresponds the smallest UI value, was observed in the middle layer of each treatment at different electrode gaps. The RF heating uniformity in corn flour was gradually improved as the electrode gap increased from 11 to 15 cm due to reduced over-heating and run-away energy. Heat conduction may have also helped improve the heating uniformity throughout the PEI container due to the slow heating rate at 15 cm and the longer heating time. On the other hand, the thermocouple measured RF heating uniformity index and average temperature of each layer were only slightly different than those from the infrared camera. For example, the uniformity index and average temperature of the layer with 1 cm depth at 15 cm electrode gap were determined as 0.042 ± 0.019 and 68.37 ± 1.65°C, respectively, using type T thermocouples, as compared to 0.044 ± 0.011 and 70.36 ± 1.88 °C , measured by the infrared camera. The results show that the cheese cloth did not affect significantly temperature distribution and heating pattern in the PEI container.

Effect of surrounding material on the RF heating uniformity

Figure 5.6 shows the top surface temperature distribution in polystyrene and glass cylindrical petri dishes with two MC levels, and with and without foam sheet. As seen in the Figure 5.6, glass cylindrical petri dish with filled corn flour provides a better heating uniformity (IU=0.036) than those (UI=0.051) of polystyrene after RF heating. The average temperature in top surface of corn flour (10.4 % (w.b.) with bulk density (0.42 g cm^{-3}) in glass petri dish was higher than that of polystyrene. It may be explained by higher dielectric property values of glass which results in higher energy absorption and dispersion as heat energy to the sample. Furthermore, RF heating rates in glass and polystyrene with filled corn flour were 4.52 °C $min^{-1}$and 3.85 °C min^{-1}, respectively. On the other hand, foam sheet helped improve RF heating uniformity and rate in both glass and polystyrene cylindrical containers (Figure 5.6). The uniformity index values of top surface in glass and polystyrene was decreased by covering foam sheet (5 mm thickness) from 0.036, 0.051 to 0.023, 0.039, respectively. The smaller uniformity index values correspond to a better temperature distribution, and small variations between hot and cold spots after RF heating.

The heating uniformity index, average temperature and heating rate in top surface of corn flour in polystyrene petri dish as sandwiched by a pair of PEI block are shown in Table (5.4). As seen in Table 5.4., the UI decreased as the thickness of PEI blocks increased, which means that PEI blocks improved the heating uniformity. When the thickness of PIE block increased from 1 to 2 cm, the average temperature increased from 62.3 ± 2.3 to 64.3 ± 2.7 °C. That is probably because the thicker PEI block can absorb more electrical energy, which results in increase in heating rate with better heating uniformity as compared to uncovered polystyrene container. Y. Jiao, et al. (2014) also came up a similar effect of PEI block on the heating uniformity of peanut butter. Although the PEI cylindrical blocks provided more uniform RF heating and higher average temperature on top surface than those of uncovered polystyrene container, the hot and cold spots still stay at the same locations (Figure 5.7). Furthermore, other approaches were also tried including covering only one side (bottom or top surface) of polystyrene cylindrical container to determine the best position for PEI blocks to have a better heating uniformity and rate in corn flour. As the PIE block with 1 cm thickness was placed only on bottom side, the UI increased from 0.0435 to 0.078, which means worsen heating uniformity, and the average temperature decreased from 60.9 to 49.9°C on top surface (Figure 5.8). This indicates less energy absorption on the top surface area during RF heating. The average surface temperatures of corn flour (10.4 % (w.b.)) as placing PEI blocks placed at bottom were 60.9 and 62.45 °C for 1 and 2 cm thicknesses, respectively (Figure 5.8). In this case, covering both side of polystyrene cylindrical container by PEI blocks provides better heating uniformity and higher energy absorption, which results in increase in average surface temperature, than those of covering only one side. Result showed that heating uniformity and average temperature on top surface in polystyrene container could be improved by adding PEI blocks on both sides of container. However, average temperature in top surface is still lower than center temperature of corn flour in polystyrene container. Other methods like hot air heating may be used as in combination with RF heating to further improve the heating uniformity in order to determine the effect of foam on heating rate and uniformity. Furthermore, the PEI container was surrounded by the foam sheet (5 mm thickness). The average temperatures of corn flour in different layers increased slightly, and the heating uniformity was improved with reduced standard deviations (Table 5.3). The reason of that might be the enhanced electromagnetic field around the container when foam sheet was used (S. Wang, et al., 2010). Additionally, heating rate in PEI container by covering foam sheet increased from 5.52 to 5.91°C min^{-1}. Thus, PEI container covered with foam sheet (5 mm thickness) as a surrounding material seemed to be an effective strategy to improve RF heating effects.

Effect of moisture content and bulk density on the RF heating uniformity

The effect of MC and bulk density on heating uniformity in top surface of corn flour as heated in polystyrene petri dish are shown in Table 5.5. As MC increased the UI also increased, which results in worse temperature distribution and uniformity. It may be explained with increase of run-away energy due to the increased dielectric properties of corn flour. The heating pattern with edge and corner effects obtained for three moisture samples in the polystyrene cylindrical petri dish were similar to the sample with initial MC heated in PEI container. The average surface temperatures in higher moisture (16.7 % w.b.) corn flour were higher than those of lower moisture samples (10.4 and 13.6 % w.b.) (Table 5.5). The heating rate also increased as MC increased. The relationship among the heating rate, dielectric loss factor and MC of the corn flour is shown in the Figure 5.9. The effect of moisture content on dielectric loss factor of various foods were reported (Guo, Wang, Tiwari, Johnson, & Tang, 2010; S. Jiao, Johnson, Tang, Tiwari, & Wang, 2011; Ozturk, et al., 2016). It is possible to adjust RF heating rate in food by controlling the moisture content (S. Jiao, et al. (2011). Furthermore, the heating uniformity was also gradually increased with increasing bulk density (from 0.42 to 0.61 g cm^3) of the samples in polystyrene petri dish based on reduced UI values (Table 5.5). The hot spot and cold spot after RF heating were located in edges and center of the cylindrical petri dish. Similar results were reported in RF heated wheat flour (Tiwari et al., 2011), lentil (Jiao et al., 2011), coffee bean (Pan et al., 2012), chestnut (Hou et al., 2014) and rice (Zhou et al., 2015). These results suggest that a greater bulk density would help achieve a better temperature distribution which results in a better RF heating uniformity.

Effect of RF heating on color and moisture distribution in corn flour

The change in color, MC and a_w of corn flour along the PEI container with and without foam sheet at five different locations, which cover hot and cold spots, is presented in the Table 5.6, for three electron distances. There were slight changes in color before and after RF treatments. The results showed that RF heating had no significant impact on corn flour color for all treatments, which is also in good agreement with literature for other RF treated low moisture foods including lentil (Jio et al. 2011), coffee bean (Pan et al. 2012), and legumes (Wang et al. 2010). The slight change of color values (L*, a* and b*) after RF heating was probably because of different temperature distributions along the treated sample. Furthermore, moisture migration occurred during the RF heating. The highest MC and a_w of RF heated samples were determined in the center of container, which is the coldest part of sample. As the PEI container was covered with foam sheet, the water migration in corn flour during the RF heating was reduced and moisture distribution throughout the container was more uniform which helps to maintain food quality. These results indicate that a foam layer can be used to cover corn flour container during RF heating treatment that can not only improve heating uniformity but maintain good product quality.

Conclusions

RF heating of corn flour can be obtained with a heating rate of 5.52 °C min^{-1} as compared with hot air heating 12.71°C min^{-1}. The dielectric and thermal properties of corn flour were influenced by MC, temperature and frequency, which are also influencing factor on the RF heating uniformity and rate. The heating uniformity was improved with increase in bulk density but decreased with increase in moisture content. For the sample heated at the central position between the two parallel electrodes, the middle layers were hotter than bottom and top layers, and the hot spot and cold spot were located in edges and center of the sample for all experimental setup, respectively. Adding and increasing the thickness of cylindrical PEI blocks on top and bottom of polystyrene container increased RF heating uniformity, rate and average temperature of corn flour. RF heating uniformity was also improved by addition of surrounding polyurethane foam sheet leading to decreased difference between hot and cold spot temperatures. The results showed that the RF heating offers an effective pasteurization method for corn flour with fasted heating rate and improved product quality as compared to hot air heating. Further research will be conducted to confirm the effectiveness of the RF heating in reduction of salmonella inoculated in the corn flour.

Acknowledgments

This study was funded by the USDA-NIFA AFRI. Grant Number: 2013-67005-21295.

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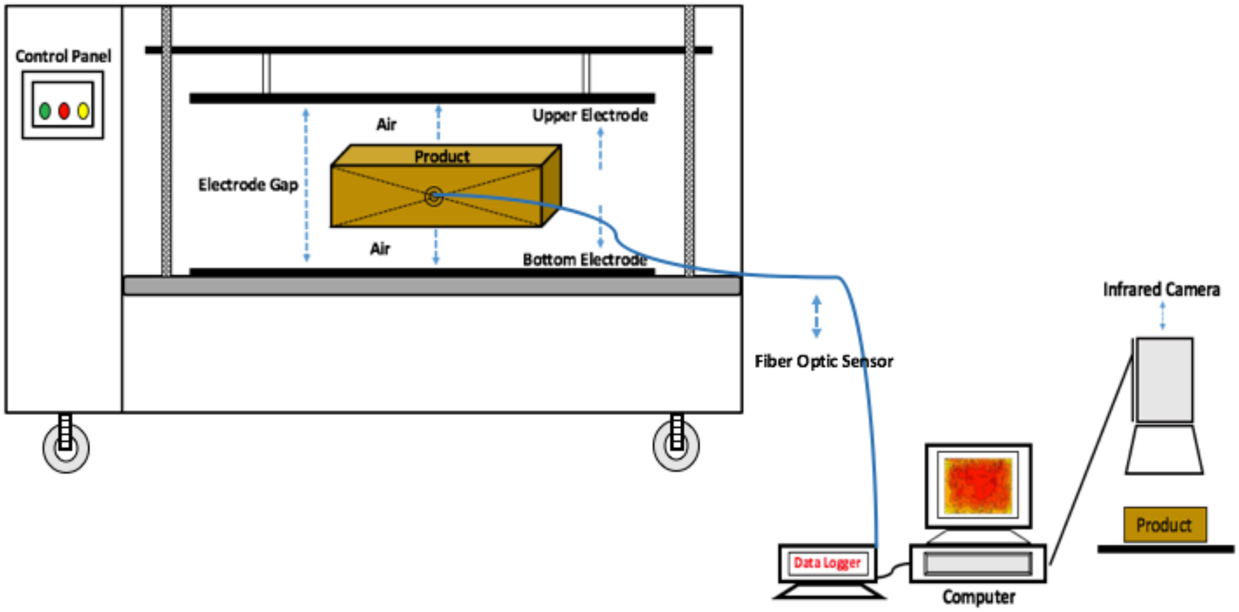

Figure 5.1. Scheme view of corn flour filled PEI rectangular container placed in the middle of two parallel electrodes in a 6 kW 27.12 MHz RF oven with the fiber optic sensor to monitor the central temperature in the sample, and the infrared camera system

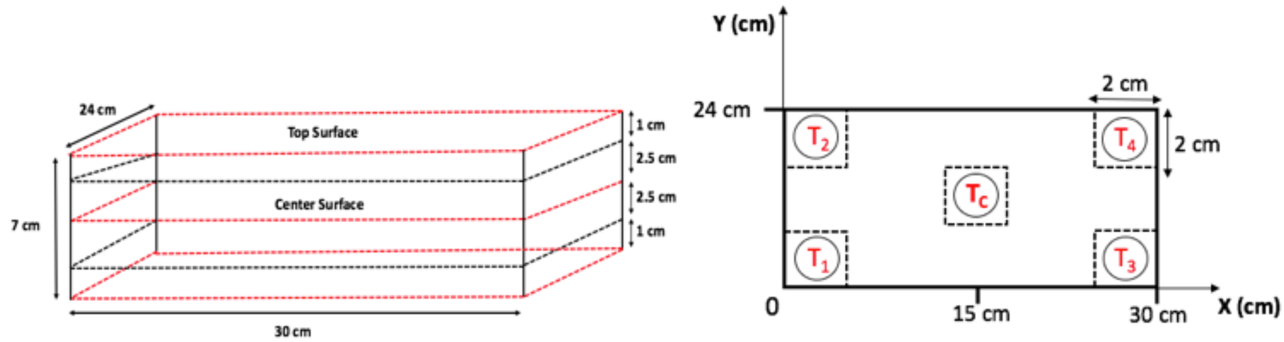

Figure 5.2. A rectangular PEI container with filled corn flour separated into three interior layers for temperature distribution measurement, and top view of 5 different locations for moisture distribution and color measurements

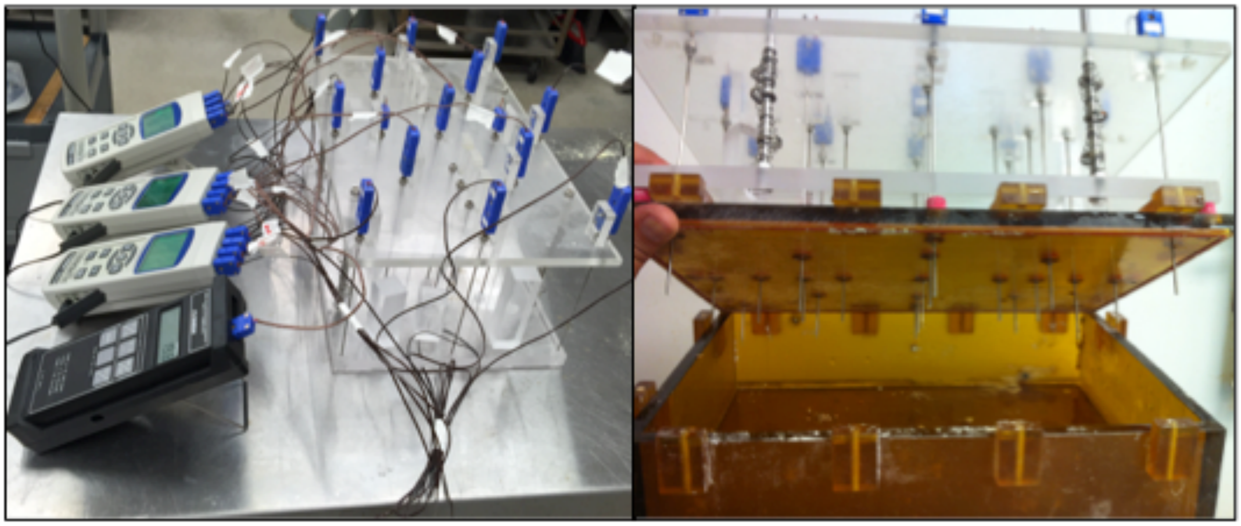

Figure 5.3. Rectangular PEI container and the layout of T type thermocouples. The springs are installed in each thermal couple to allow easy adjustment of insert depth into the sample

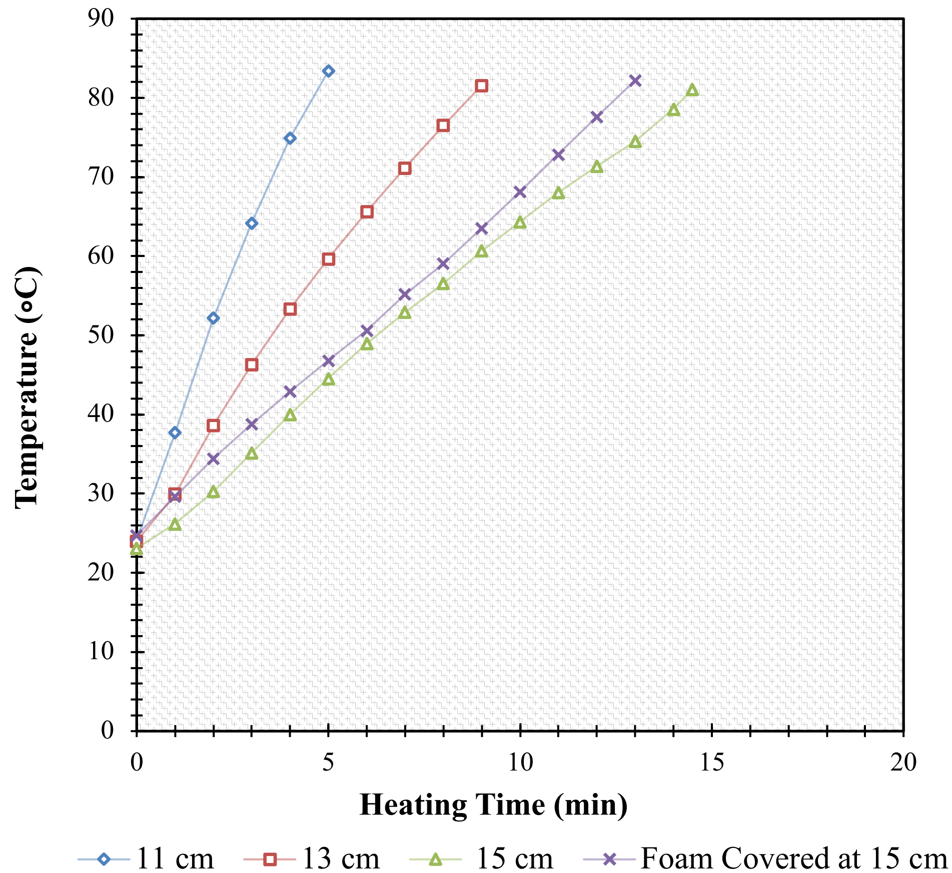

Figure 5.4. A typical temperature–time curve of corn flour subjected to RF heating at three different electrode gaps

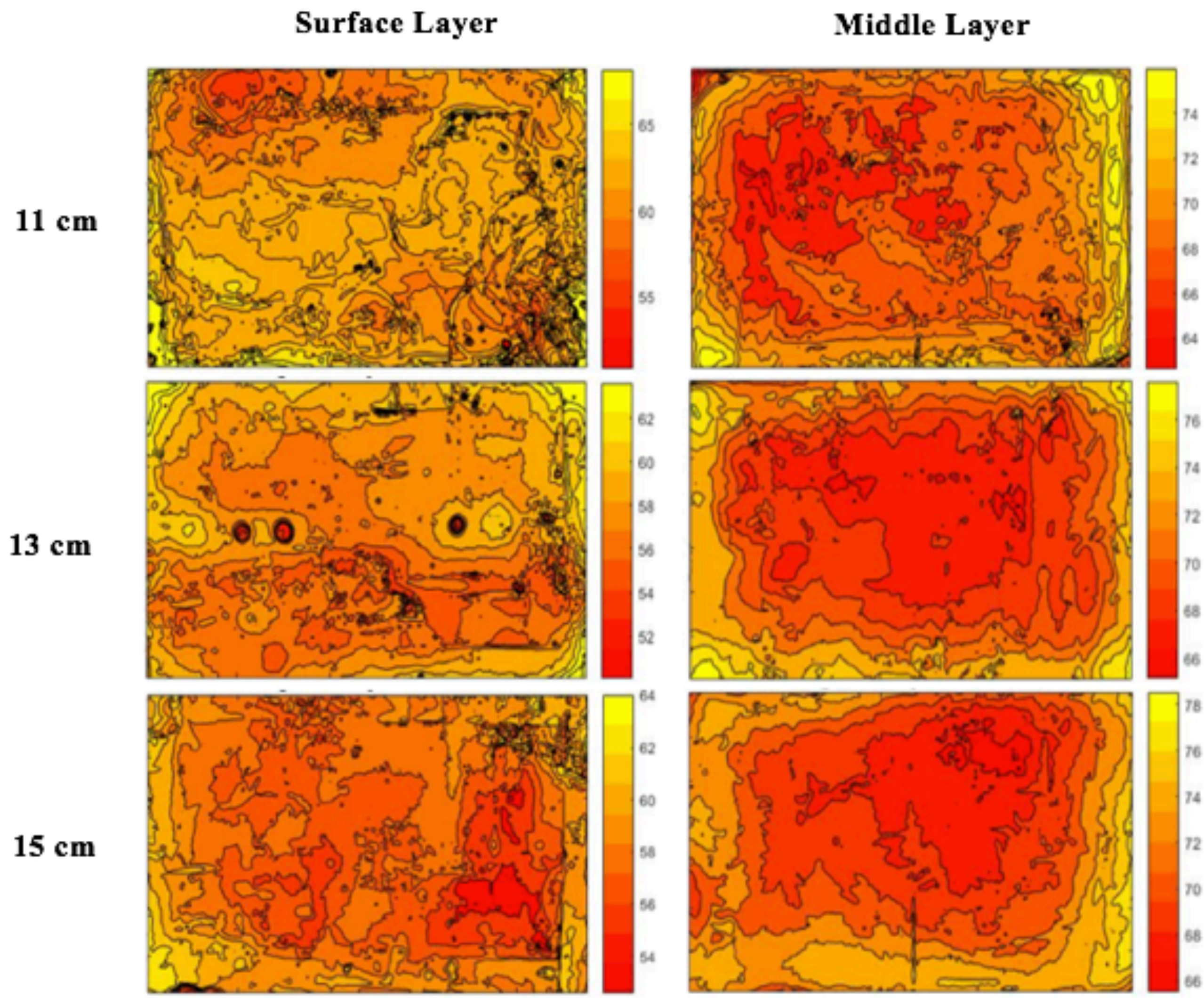

Figure 5.5. Top and middle surface temperature distribution (°C) of corn flour in a PEI container heated in the middle of two parallel electrodes of a RF oven with three electrode gaps (11, 13 and 15 cm)

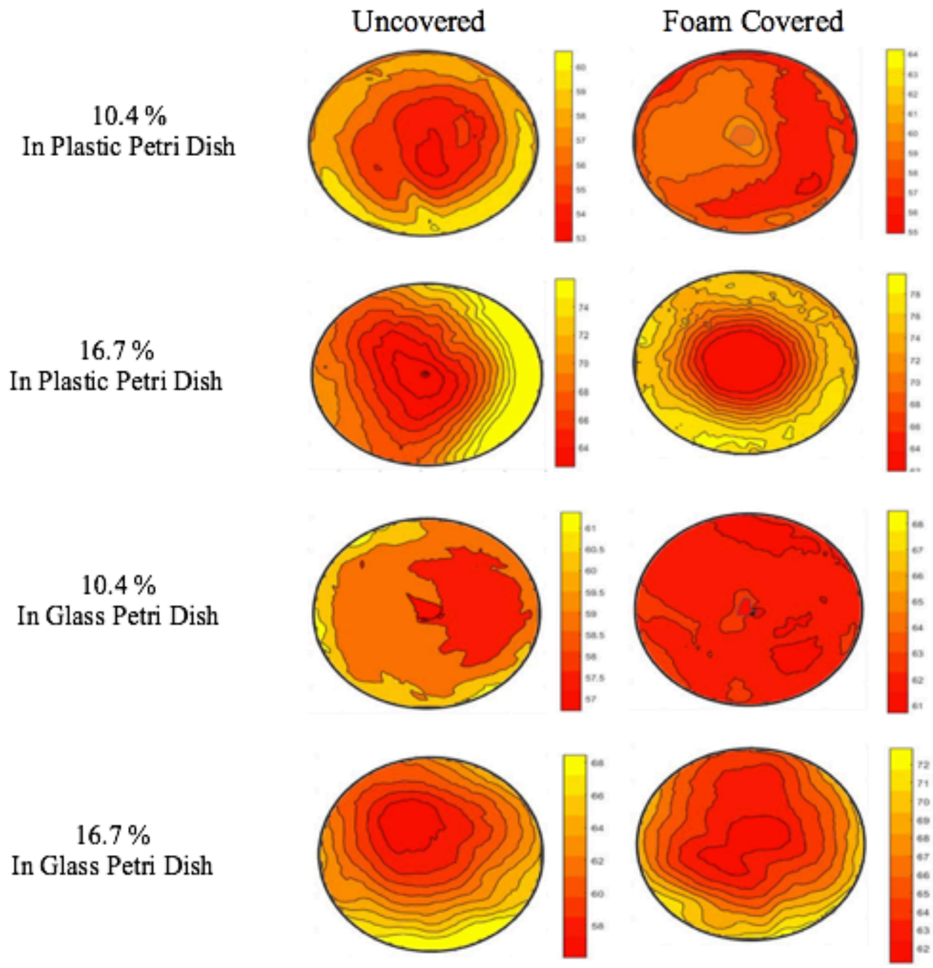

Figure 5.6. Top surface temperature distribution (°C) of corn flour (50 g) in polyester and glass petri dish with two MC content (10.4 and 16.7 % (w.b.)). The container was placed in the middle of two parallel electrodes of the RF oven with an electrode gap of 15 cm

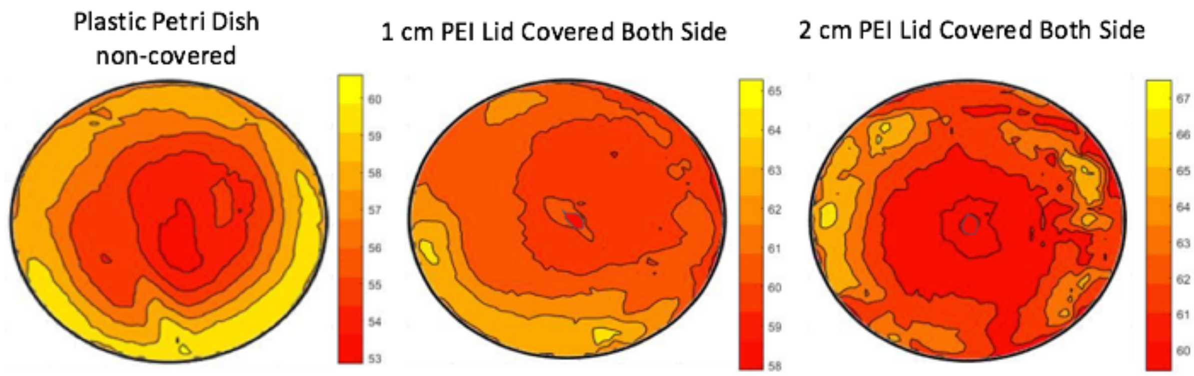

Figure 5.7. Top surface temperature distribution (°C) of corn flour (50 g) with 10.4 % (w.b.) in a polyester petri dish covered with PEI disc on both sides. The sample was placed in the middle of two parallel electrodes of the RF oven with electrode gap of 15 cm

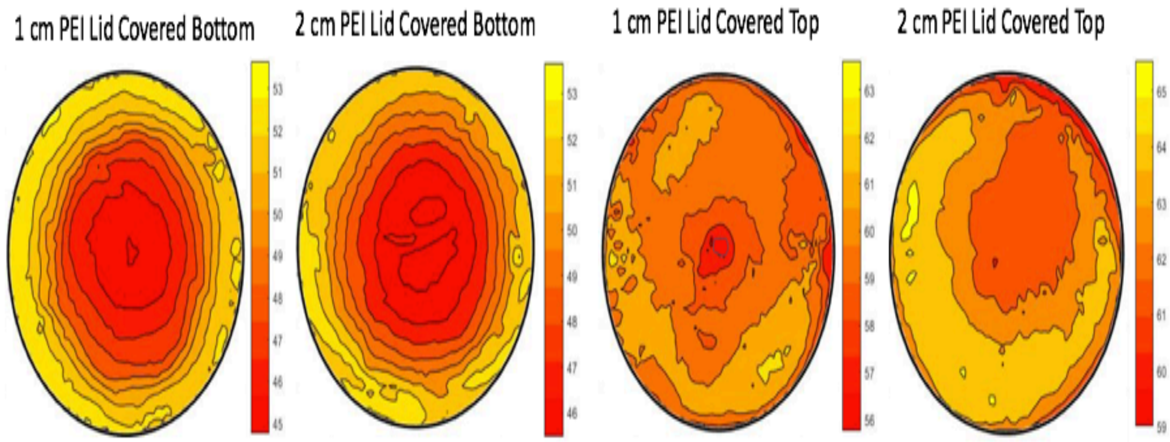

Figure 5.8. Top surface temperature distribution (°C) of corn flour (50 g) with 10.4 % MC (w.b.) in a polyester petri dish covered with PEI cylindrical blocks on only bottom or top surface. The container was placed in the middle of two parallel electrodes of the RF oven with electrode gap of 15 cm

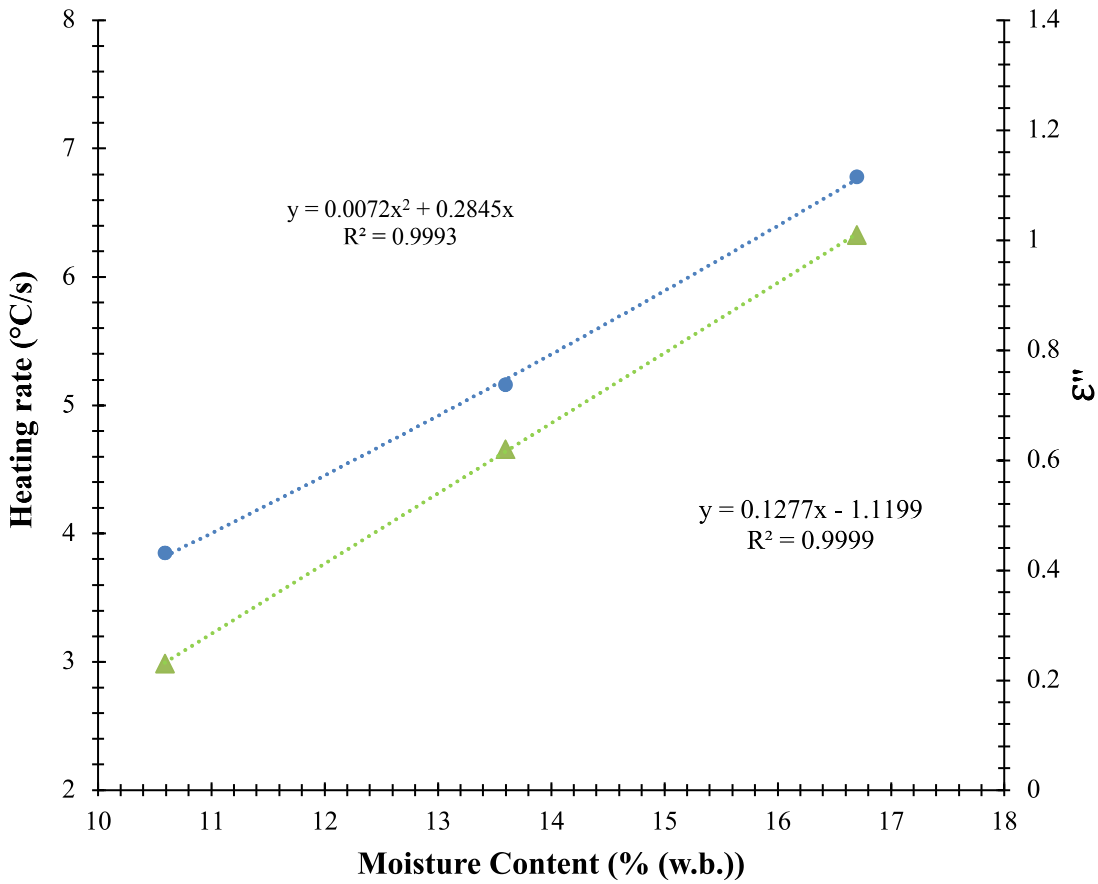

Figure 5.9. The relationship among moisture content, heating rate and dielectric loss factor of corn flour as exposed to the RF heating

Table 5.1. Thermal conductivity and dielectric properties of glass, polystyrene (PS), polyetherimide (PEI) and polyurethane foam

	Glass[a]	(PS)[b]	(PEI)[b]	Polyurethane Foam[c]
Thermal Conductivity ($Wm^{-1}K^{-1}$)	1.05	0.032	0.122	0.03
Dielectric Constant	4.6	3.0	3.15	2.1
Dielectric Loss Factor	0.001	0.003	0.0025	0.01

(Bansal & Doremus, 1986)[a]
(Lampman, 2003)[b]
(Domeier & Hunter, 1999)

Table 5.2. Dielectric and thermal properties of corn flour as influenced by moisture content, temperature and frequency*

		10.3 % (w.b.)		13.6 % (w.b.)		16.7 % (w.b.)	
		13.56 MHz	27.12 MHz	13.56 MHz	27.12 MHz	13.56 MHz	27.12 MHz
ε'	20 °C	3.86 ± 0.55	3.68 ± 0.71	4.19 ± 0.78	4.02 ± 0.87	4.94 ± 0.51	4.77 ± 0.59
ε''		0.16 ± 0.73	0.13 ± 0.77	0.18 ± 0.33	0.15 ± 0.62	0.21 ± 0.86	1.7 ± 0.81
d_p (m)		27.37 ± 0.73	18.83 ± 0.5	14.65 ± 0.94	12.74 ± 0.42	9.97 ± 0.6	7.18 ± 0.65
ε'	80 °C	9.11 ± 0.92	8.43 ± 0.75	12.48 ± 0.75	11.35 ± 0.62	21.27 ± 0.77	18.51 ± 0.82
ε''		1.11 ± 0.74	0.74 ± 0.81	2.1 ± 0.71	1.34 ± 0.86	6.14 ± 0.87	4.23 ± 0.83
d_p (m)		4.04 ± 0.33	2.89 ± 0.77	2.67 ± 0.87	1.80 ± 0.72	1.88 ± 0.78	1.27 ± 0.88
Thermal Conductivity (W/m °C)	20 °C	0.113 ± 0.33		0.132 ± 0.55		0.151 ± 0.51	
Thermal Diffusion ($m^2 s^{-1}$)		$14.9 \times 10^{-8} \pm 0.78$		$15.3 \times 10^{-8} \pm 0.86$		$15.7 \times 10^{-8} \pm 0.75$	
Specific Heat (kJ/kg.K)		2.12± 0.95		2.79 ± 0.62		3.12 ± 0.81	

*Reported are average and standard deviation of three measurements.

Table 5.3. Comparisons of average temperature and heating uniformity index of the corn flour with 10.4 % (w.b.) moisture content after RF heating with different electrode gaps*

	Location	11 cm	13 cm	15 cm	Foam Covered at 15 cm
Temperature (°C) Mean ± SD	Top Surface	61.97 ± 2.2	57.84 ± 1.8	58.19 ±1.76	59.79 ± 2.5
	1 cm Depth	69.08 ± 1.9	66.94±2.18	70.36 ± 1.88	70.47 ± 1.72
	Middle Surface	70.22 ± 2.56	70.54 ± 2.67	71.39 ± 2.4	71.79 ± 2.87
	6 cm Depth	67.81 ± 2.23	66.29 ± 2.22	66.32 ± 2.48	68.58 ± 2.99
Heating Uniformity Index (λ) Mean ± SD	Top Surface	0.050 ± 0.040	0.044 ± 0.001	0.042 ± 0.016	0.037 ± 0.020
	1 cm Depth	0.049 ± 0.016	0.050 ± 0.013	0.044 ± 0.011	0.041 ± 0.06
	Middle Surface	0.044 ± 0.006	0.043 ± 0.009	0.033 ± 0.013	0.029 ± 0.021
	6 cm Depth	0.052 ± 0.008	0.051 ± 0.001	0.049 ± 0.012	0.046 ± 0.009
Heating Rate (°C min^{-1})	Geometric Center of Container	12.71 ± 1.84	6.53 ± 0.1	5.52 ± 0.32	5.91 ± 0.54

*Reported are average and standard deviation of three measurements.

Table 5.4. Effect of thickness of PEI material on the surface layer heating uniformity and average temperature in corn flour (50 g) in polystyrene petri dish during the RF heating*

	Petri Dish non-covered	1 cm PEI Lid covered both side	2 cm PEI Lid covered both side
Temperature (°C)	59.5 ± 3.3	62.3 ± 2.3	64.3 ± 2.7
Heating Uniformity Index (λ)	0.051 ± 0.02	0.044 ± 0.01	0.045 ± 0.03
Heating Rate (°C min^{-1})	3.85 ± 0.6	5.58 ± 0.8	6.69 ± 0.3

*Reported are average and standard deviation of three measurements.

Table 5.5. Effect of MC and bulk density on the RF heating uniformity and rate, and average temperature of the corn flour in polyester petri dish *

			Surrounding Material: Surface layer in Polystyrene (PS) Petri Dish
Bulk Density (g cm^{-3})	0.42	Temperature (°C)	59.5 ± 2.4
		Heating Uniformity Index (λ)	0.051 ± 0.05
		Heating Rate (°C min^{-1})	3.85 ± 0.07
	0.53	Temperature (°C)	62.8 ± 3.1
		Heating Uniformity Index (λ)	0.0416 ± 0.01
		Heating Rate (°C min^{-1})	3.46 ± 0.45
	0.61	Temperature (°C)	64.7 ± 3.3
		Heating Uniformity Index (λ)	0.0377 ± 0.025
		Heating Rate (°C min^{-1})	3.16 ± 0.81
Moisture Content (w.b.(%))	13.6	Temperature (°C)	63.85 ± 2.1
		Heating Uniformity Index (λ)	0.0691±0.015
		Heating Rate (°C min^{-1})	5.16 ± 0.91
	16.7	Temperature (°C)	68.7 ± 2.9
		Heating Uniformity Index (λ)	0.094±0.03
		Heating Rate (°C min^{-1})	6.28 ± 1.2

*Reported are average and standard deviation of three measurements.

Table 5.6. Effect of RF heating and hot air heating on moisture (M.C), water activity (a_w) distribution and color values (L, a, and b) of the corn flour at five different locations of a PEI container as influenced by electron gap and foam coverage*

	L	a	b	T_1		T_2		T_3		T_4		T_C	
				M.C (%)	a_w	M.C (%)	a_w	M.C (%)	a_w	M.C (%)	a_w	M.C (%)	a_w
Control	93.5 ± 0.16	4.22±0.07	32.37±0.14	10.4	0.509	-	-	-	-	-	-	-	-
Hot Air	95.41±0.36	3.21±0.04	28.70±0.36	5.62	0.201	-	-	-	-	-	-	-	-
11 cm	92.7± 0.41	4 ± 0.08	32.6 ± 0.36	8.12	0.337	8.59	0.383	8.42	0.375	8.38	0.397	9.47	0.437
13 cm	92.5 ±0.07	3.9±0.11	32.4±0.41	8.22	0.381	8.57	0.372	8.89	0.387	8.49	0.379	9.54	0.444
15 cm	93.1±0.16	3.89±0.07	32.1±0.22	8.5	0.364	8.41	0.372	8.89	0.379	8.78	0.374	9.66	0.453
15 cm with Foam Sheet	93.24±0.28	4.16±0.07	32.21±0.22	9.26	0.413	9.28	0.417	9.34	0.432	9.42	0.431	9.76	0.469

*Reported are average and standard deviation of three measurements.

CHAPTER 6

INACTIVATION OF *SALMONELLA* AND *ENTERACOCCUS FAECIUM* NRRL B-2354 IN PACKED CORN FLOUR BY RADIO FREQUENCY HEATING WITH AND POST-FREEZING STORAGE[4]

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Abstract

The study aimed to (1) validate radio frequency (RF) heating in inactivating *Salmonella enterica* Enteritidis PT30 (*S*. Enteritidis PT30) and evaluate the feasibility of using *Enterococcus faecium* NRRL B-2354 (*E. faecium*) as a surrogate of *Salmonella* in bulk corn flour, and (2) study the effect of post-freezing storage treatment in enhancing the inactivation efficiency of RF heating. Corn flour with water activity (a_w) at 25°C ($a_{w,25°C}$ = 0.45 ± 0.05) was homogeneously inoculated with *S*. Enteritidis PT30 and *E. faecium* separately at 8.5±0.23 log CFU/g. Inoculated samples (0.85 g) were sealed in a closed system, and subjected to isothermal treatment in a water bath at 75, 80, and 85°C to obtain information of thermal resistances (D and z-values) information. For validation study, corn flour (3.18 kg) was loaded into a polyetherimide (PEI) container (inner dimension 7×24×30 cm^3) with pre-packaged inoculated sample (5 g) placed in the geometric center. Samples were subsequently subjected to RF heating to reach temperatures of 75, 80, or 85°C, respectively, and held for 10 min before bacteria survivors were enumerated. The RF treated samples were stored at -20°C for 96 h and taken in a 24 h time intervals to evaluate the effect of post-freezing storage treatment on microbial survival. Results showed that thermal resistance of *E. faecium* was higher than *S*. Enteritidis PT30. RF heating to 75, 80 and 85 °C resulted in 0.91±0.07, 1.84±0.13, and 2.49±0.11 log CFU/g reductions in *S*. Enteritidis PT30 population, and 0.39±0.09, 0.94±0.05, and 1.72±0.18 log CFU/g reduction in *E. faecium* population, respectively. Samples heated at 85°C were held for 10 min and stored at -20°C for 48 h resulting in reduction of *S*. Enteritidis PT30 (6.59±0.21 log CFU/g) and *E. faecium* (4.79±0.17 log CFU/g), but no further decrease in survival was observed with longer than 48 h freezing storage. This study demonstrated that RF heating can effectively inactivate *S*. Enteritidis PT30, and *E. faecium* could be used as its surrogate for validation studies in packaged corn flour.

Additionally, RF heating combined with post-freezing storage treatment could significantly reduce the survival of both microorganism in the inoculated corn flour.

Key words; Radio frequency pasteurization; Post-freezing storage; Corn flour; *Salmonella*; *Enterococcus faecium*

Introduction

Low-moisture foods with $a_w < 0.6$ have been historically considered as safe in terms of scare outbreaks of microbial contamination, and limited growth of either vegetative or spore-forming bacteria should occur as in high-a_w foods (Blessington et al., 2013; Farakos and Frank, 2014). However, a number of foodborne outbreaks were linked to low moisture foods in the recent years (Beuchat et al., 2013; Podolak et al., 2010), which brought a great attention of scientific community and the food industry to the safety of dry foods and ingredients for ready-to-eat products that are not heat treated before packaging or consumption. Heat resistance of pathogens such as *Salmonella* has been shown to increase with the decrease of a_w in foods (Podolak et al., 2010). Therefore, it is critical to develop an effective pasteurization process with reliable performances in eliminating foodborne pathogens in foods with low a_w. However, pathogens are not normally allowed to directly use for process validation in production environment due to strict safety consideration. To accomplish this task, it is covetable to define a surrogate of pathogens to conduct in-plant process validation. Thus, FDA (2015) suggested the use of non-pathogenic surrogate microorganisms with equal or slightly higher thermal resistance as an alternative approach to conduct microbial validation. *Entorococcus faecium* NRRL B-2354 has been reported as a *Salmonella* surrogate in various low-moisture foods including, wheat flour (Liu et al., 2018; Tiwari et al., 2011), and various spices (Rachon et al., 2016). The organism has showed a great potential as a surrogate of *Salmonella* with a thermal resistance higher than *Salmonella* under the same environmental conditions. The feasibility of using *E. faecium* as a surrogate for validation of microbial inactivation has been evaluated in different pasteurization techniques including extrusion (Bianchini et al., 2012), moist-air convection heating (Jeong et al., 2011), and infrared pasteurization (Bingol et al., 2011). Several decontamination methods such as steam treatment, fumigation with ethylene oxide, and irradiation have been developed to reduce the microbial load of low-moisture foods (Lee et al., 2006). However, ethylene oxide is regarded as a carcinogen, and irradiated foods are not readily accepted by consumers due to safety concerns (Farkas, 2006; Schweiggert et al., 2007; Waje et al., 2008).

Radio frequency (RF) heating is dielectric heating with frequency range of 3 kHz - 300 MHz, which generates heat inside the low-moisture foods through ionic conduction and dipole rotation, providing a fast and volumetric heating throughout the foods (Marra et al., 2009). Although RF heating provides a better heating uniformity than microwave heating, the heating uniformity is still a major challenge for inactivating foodborne pathogens while maintaining product quality (Jeong and Kang, 2014; Kim et al., 2012). The effect of RF heating on inactivation of pathogenic bacteria has been studied for various foods including shell eggs (Geveke et al., 2017), meat lasagna (Wang et al., 2012), almonds (Gao et al., 2012; Gao et al., 2011; Gao et al., 2010), flour (Villa Rojas, 2015; (Tiwari et al., 2011), peanut butter (Villa Rojas, 2015), shell almond (Li et al., 2017), and spices (Kim et al., 2012). There is, however, still an essential need for improving RF heating uniformity and systematic studies to achieve a wide application of RF pasteurization for low-moisture foods.

Even though a few studies had investigated the effect of RF heating in inactivation of pathogenic bacteria and its surrogate in low-moisture foods, very limited studies have been reported on RF inactivation kinetics of pathogen and its surrogate with combination of RF heating and post-freezing storage treatment. A recent study showed that storing sample at freezing temperature following decontamination by pulsed electric field in green tea infusions successfully reduced the survival population of pathogenic microorganisms (Zhao et al., 2009). In this study, corn flour was selected as a model food, and *E. faecium* was used as a surrogate to validate the efficacy of thermal inactivation. The objectives of this study were to (1) evaluate the feasibility of *E. faecium* as a surrogate of *Salmonella* to validate RF heating pasteurization of *Salmonella* in corn flour, and (2) determine the effect of holding and freezing storage (-20 °C for 96 h) on enhancing the level of inactivation of RF treatment by comparing survivor kinetics.

Material and Methods

Corn flour was purchased from Georgia Spice Company (Atlanta, GA USA). The water activity at 25 °C ($a_{W,\ 25°C}$) was determined using an a_w meter (AQUA PRE, Decagon Devices, Pullman, WA, USA), and adjusted to keep a_w level the same for each trial by using a Hotpack 435315 humidity chamber (SP Industries, Inc., Warminster, PA, USA) set with relative humidity (45%) at room temperature (25°C).

Bacterial inoculation of corn flour

Salmonella Enteritidis PT30 (*S*. Enteritidis PT30) and *Enterococcus faecium* NRRL B-2354 (*E. faecium*) were obtained from Dr. Mark Harrison's laboratory at the University of Georgia. Both bacterial strains were cultured in 9 ml of tryptic soy broth (TSB) supplemented with 0.6 % (wt/vol) yeast extract (TSBYE) at 37 °C for 24 h and then 1 ml evenly spread on a plate (150 x 15 mm) of TSAYE. The bacterial lawn on TSAYE were harvested into 20 ml of sterile 0.1% peptone water and centrifuged for 30 min at 2,600 *g*. Then, the supernatant was discarded and the pellet was re-suspended in 3 ml 0.1% peptone water as described by (Liu et al., 2018). One ml of concentrated pellet of *S*. Enteritidis PT30 or *E*. faecium was mixed into 10 g corn flour in a sterile stomacher bag until an even mixture was obtained. After mixing, inoculated corn flour samples (10 g) were used to further inoculate 90 g each sample, which was mixed and stomached (Seward Stomacher, 400 Lab System, Norfolk, United Kingdom) at 260 rpm for 5 min. Then, 10 samples (1 g each) were randomly selected and enumerated on tryptic soy agar (TSA) plates as described subsequently to confirm the uniformity of inoculum distribution.

Equilibration of water activity

To avoid the effect of a_w on thermal resistances of *S*. Enteritidis PT30 and *E. faecium* in corn flour, inoculated samples were placed in sterile trays and then put into a Hotpack 435315 humidity chamber (SP Industries, Inc., Warminster, PA, USA) (Liu et al., 2018) for four days to ensure equilibrium with the target $a_{w,25°C}$ = 0.45 ± 0.05 (Hildebrandt et al., 2016).

Isothermal Treatment

Thermal resistances of *S*. Enteritidis PT30 and *E. faecium* in corn flour with $a_{w,25°C}$ = 0.45 ± 0.05 were determined at 3 different temperatures (75, 80, and 85°C) using aluminum thermal-death-time (TDT) cells (Washington State University, Pullman, WA, USA) (Chung et al., 2008; Villa-Rojas et al., 2013). The cells were fully filled with inoculated corn flour (0.85 g, 4 mm thickness), and subjected to isothermal heat treatment in a water bath (Model: SWB-10L-2, Saratoga, CA USA) maintained at 75, 80, and 85°C. To obtain thermal death curves, each isothermal treatment was performed at the same time intervals starting after the come-up times (CUT) ended. The CUT was defined as the time to reach target temperature (75, 80, and 85 °C) in the water bath and determined using a T-type thermocouple inserted in the center of cell with non-inoculated corn flour sample (0.85 g, $a_{w,25°C}$ = 0.45 ± 0.05). Isothermal treated cells were removed from the water bath and immersed immediately in an ice-water bath for 90 s to stop thermal inactivation in sample. Triplicates of each set of conditions were performed.

Enumeration of microbial survival

Thermally treated corn flour in TDT cells was transferred into sterile stomacher bags and diluted 1:10 with 0.1 % peptone water. The samples were firstly mixed by hand and then homogenized for 2 min at 260 rpm with a Seward Stomacher (Seward, London, UK) (Harris et al., 2012). The proper tenfold serial dilutions were spread-plated in duplicate onto modified mTSA for *S*. Enteritidis PT30 or eTSA for *E. faecium* plates (mTSA; TSA agar, Yeast Extract, 0.05% Ammonium Iron (Ш) Citrate, and 0.03% Sodium Thiosulfate Pentahydrate ($5H_2O$), and eTSA; TSA agar, Yeast Extract, 0.05% Ammonium Iron (Ш) Citrate and Esculin Hydrate) to enumerate both *S*. Enteritidis PT30 and E. *faecium* survivors in corn flour, respectively. The plates were incubated aerobically 37 °C for 24-48 h, then the colonies were enumerated, and the populations were converted to log CFU/g. Log reductions were calculated by subtracting the survivor counts from the initial population before thermal treatment.

Inactivation kinetics

The first order kinetics (D-value) Eq. (1), and the Weibull model Eq. (2) were used to express inactivation kinetics (Peleg, 2006)

$$log\ \frac{N}{N_0} = -\frac{t}{D} \qquad \text{Eq. (1)}$$

$$log\ \frac{N}{N_0} = -\left(\frac{t}{\delta}\right)^{\alpha} \qquad \text{Eq. (2)}$$

where N_0 is the initial concentration of microorganism (CFU/g), and *N* is the population of survival (CFU/g) at time (*t*), the isothermal treatment time (min) after CUT; D is the time to reduce microbial population by 10-fold at the inactivation temperature (°C); δ indicates the overall steepness of survival curve; α is the survival curve factor which indicates whether it is linear (α=1) or non-linear (α≠1) with a decreasing (α < 1) or increasing (α > 1) inactivation rate with time.

Data were used to fit the model to the microbial survival curves and estimate the model parameters. Root mean square error (RMSE) was used to evaluate the performance of the model.

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}\left[log\frac{N}{No}_{data,i} - log\frac{N}{No}_{model,i}\right]^2}{n-p}} \qquad \text{Eq. (3)}$$

where $log\frac{N}{No}_{data,i}$ is measured log CFU/g reduction, and $log\frac{N}{No}_{model,i}$ is predicted log CFU/g reduction from model, n is the total number of observations, and p is the number of model parameters. 1stOpt software was used to estimate its fitness and provide RMSE directly. ANOVA in Minitab 14 (Minitab Inc., State College, PA) was used to evaluate differences between obtained and predicted D-values among samples. To obtain the z-value, the log of D-values was plotted against temperature, in which the slope refers to the -1/z, where z is the change in temperature to alter the thermal-death-time (D-value) by one log-cycle (Gaillard et al., 1998).

Radio frequency heating of inoculated corn flour

A 27.12-MHz, 6-kW pilot scale RF system (COMBI 6-S, Strayfield International, Wokingham, UK) was used in this study. The RF system had two parallel electrodes, top electrode position can be adjusted to change the gap. It has been reported that the gap between parallel electrodes affects the distribution of electromagnetic field throughout the system, which directly influence RF heating rate and uniformity in the sample (Liu et al., 2013; Tiwari et al., 2011; Uyar et al., 2014; Uyar et al., 2016). The gap between the two parallel electrodes was fixed as 15 cm in this study to obtain better heating rate and uniformity based on our previous study (Ozturk et al., 2017). Detailed information about heating uniformity and design were described by Ozturk et al. (2017) and Liu et al. (2018). To obtain temperature distribution and to determine cold spot area in the middle layer of PEI container, a cheese cloth was used to divide the PEI container in two equal layers as described by Ozturk et al. (2017). Uninoculated corn flour (3.18 kg) was loaded into a PEI container ($7 \times 24 \times 30$ cm^3) and heated by RF heating system in between the upper and lower electrodes to reach target temperatures (Figure 6.1). Additionally, the PEI container surface was covered by foam sheet acts as an insulator to enhance electromagnetic area around the container, and to prevent significant heat loss while maintaining more uniform heating and better moisture distribution. The change in temperature during the RF heating was recorded using a fiber optic temperature sensor with accuracy of ± 1°C (Fiso Tech. Inc., Quebec, Canada) connected to a data logger. After heating, the RF system was turned off, and the PEI container was removed immediately to take thermal images of middle layer using an infrared camera (FLIR T440, FLIR Systems, Inc., North Billerica, MA, USA).

To evaluate the inactivation efficiency of RF heating on both *S*. Enteritidis PT30 and *E. faecium* in corn flour, inoculated corn flour (5 g) in a sterile plastic bag (50 x 50 x 10 mm^3) was sealed and placed in cold spot area, where was in the geometric center of the PEI container as determined by infrared camera. Additionally, to reduce the survival populations of both *S*. Enteritidis PT30 and *E. faecium* in inoculated corn flour, treated sample packs were held in the PEI container for 10 min.

To determine the effect of sample plastic bag on heating rate of sample, a fiber optic sensor (Fiso Tech. Inc., Quebec, Canada) was inserted in the center of the same size plastic bag but with non-inoculated sample as described by Liu et al. (2018). Another fiber optic sensor connected to a data logger was also used to monitor the real-time temperature history as a control during the RF heating process without inserting bag in the PEI container. The results indicated no significant impact on heating rate from the plastic bag.

The PEI container filled uninoculated corn flour (3.18 kg) with inoculated sample bag were subjected to the RF heating to reach temperatures of 75, 80, and 85°C, respectively, and held for 10 min in the PEI container after the RF system was turned off. In another test, the same experimental process was followed without holding treated sample bags in the PEI container. Then RF heated sample packages were immediately removed from the container and inserted in an ice-bath for 90 s to stop further thermal inactivation process in the sample. One-gram RF treated sample with or without holding was transferred in sterile bags. The samples were serially diluted and plated for surviving bacterial population as described in enumeration section.

Combination of RF heating and post-freezing storage on microbial survival

The RF treated corn flour samples (with or without holding) were placed in a freezer (-20 °C) for up to 96 h. Samples were taken in a 24 h interval to determine the effect of cold-shock on the survival populations of *S*. Enteritidis PT30 and *E. faecium*. One gram of sample from each treatment combination was collected randomly, tenfold serially diluted and plated as described previously to enumerate the survivors.

Results and Discussion

D- and z- values of *S*. Enteritidis PT30 and *E. faecium* in corn Flour

Fig. 2 shows the inactivation kinetics of *S*. Enteritidis PT30 and *E. faecium* in corn flour at $a_{w,25°C}$ = 0.45 ± 0.05. As seen in Table 6.1, survival data of both *S*. Enteritidis PT30 and *E. faecium* shows good fit to the primary models with similar RMSE values. The D-values for *S*. Enteritidis PT30 at 75, 80, and 85°C were 14.6 ± 0.51 min, 6.11± 0.45 min, and 2.03 ± 0.27 min, respectively, and with z-value of 11.9 °C. Vancauwenberge et al. (1981) reported that to obtain a 2 log reduction in the population of *S. Seftenberg*, corn flour with moisture content from 10 to 15% had to be heated at 49°C for 5.8 and 2.2 h, respectively. In this study, as inactivation temperature increased, thermal resistance of both microorganisms in corn flour decreased proportionally (Vancauwenberge et al. (1981). Smith et al. (2016) and Syamaladevi et al. (2016) reported the $D_{80°C}$ for the same *Salmonella* serotype in wheat flour with the same levels of water activity as 6.9 ± 0.7 and 5.51 ± 0.22 min, respectively. Additionally, the obtained D-values for *S*. Enteritidis PT30 in corn flour were also in good agreement with organic wheat flour (Liu et al.,

2018) who reported 17.65 ± 1.58, 7.17 ± 0.35, and 2.92 ± 0.35 min at 75, 80 and 85°C, respectively. The slight difference in the D-value of the same serotype of *Salmonella* might be due to differences in food matrix, inoculation method, moisture content, and isothermal treatment method. In addition, as seen in Figure 6.2, *E. faecium* shows significantly ($p < 0.05$) higher heat resistance than *S*. Enteritidis PT30 in corn flour at inactivation temperatures and has a similar z-values as compared to *S*. Enteritidis PT30 in the range of inactivation temperatures (75 to 85°C). For example, $D_{80°C}$ values of *S*. Enteritidis PT30 and *E. faecium* in corn flour (aw=0.45) were 6.11 ± 0.35 min and 10.71 ± 0.85 min, respectively. Several studies reported that *E. faecium* NRRL B-2354 is a valid surrogate for pathogenic bacteria in various foods including high moisture food (dairy products, juices and meat) and low-moisture foods (almonds, walnuts, peanut butter, extruded products) in thermal processing (Annous and Kozempel, 1998; Bianchini et al., 2012; Blessington et al., 2013; Ma et al., 2007; Pan et al., 2012b; Piyasena et al., 2003). It has been used as a surrogate of *Salmonella* to validate thermal process conditions for 4 to 5-log reduction of *Salmonella* in almonds (Bingol et al., 2011; Kopit et al., 2014). The present study indicates *E. faecium* is a conservative, but suitable surrogate for *S*. Enteritidis PT30 for corn flour in the tested conditions with higher heat resistance.

RF heating and uniformity

In the light of our previous study (Ozturk et al., 2017), the PEI container filled with corn flour (3.18 kg) was subjected to RF heating at 15 cm electro gap in between two parallel electrodes till to reach the target temperature (75, 80, and 85°C) with or without holding for 10 min. Figure 6.3 shows the typical temperature-time profiles of heated sample, and the un-inoculated sample package in the center of the PEI container. As seen in the Figure 6.3, the increase in temperature the surface of packed corn flour was close to the central temperature of unpacked sample. The difference between temperature of packed sample and surface of sample pack in the cold spot area was 1.0 ± 0.4 °C, which was also in good agreement of packaged wheat flour heated by RF system (Liu et.al, 2018; Xu et al., 2018). In addition, the corner and edge heating were observed in the middle layers, and the cold spot area were located in the center (Figure 6.4). Recent studies also reported similar observations for RF heated coffee bean (Pan et al., 2012a), rice (Zhou et al., 2015), and wheat flour (Tiwari et al., 2011). The average temperature of the target layer after RF heating was determined to be 72.2 ± 2.4, 78.4 ± 2.9, and 81.9 ± 3.2°C at 75, 80, 85°C, respectively. Our previous study showed that sample heated in the PEI container surrounded by foam sheet had achieved better heating uniformity in the middle layer with higher average temperature (Ozturk et al., 2017). As the focus of this study was to determine the pasteurization effect of RF heating on pathogen bacteria and its potential surrogate, the cold spot area in the middle layer was used as the least heating zone to validate the RF process for packaged inoculated corn flour. Moreover, holding the sample in PEI container for 10 min after RF system was turned off resulted in a sample temperature decreasing in the temperature of sample around 4.1 ± 1.2 °C due to heat conduction throughout the PEI container and heat loss to the air.

Effect of post-freezing storage treatment on survival after RF heating with or without holding

This study was conducted using various combinations to enhance the inactivation effect of RF heating on both *S.* Enteritidis PT30 and *E. faecium* in corn flour with $a_{w,25°C} = 0.45 \pm 0.05$. Inactivation effect of RF heating on various foodborne pathogens and their potential surrogates have been reported for different low-moisture foods such as wheat and almond flour (Liu et al., 2018; Villa Rojas, 2015), peanut butter (Villa Rojas, 2015), black and red peppers (Jeong and Kang, 2014). In this study, RF heating during CUT resulted in 0.91 ± 0.07, 1.84 ± 0.13, and 2.49

± 0.11 log CFU/g reductions in *S*. Enteritidis PT30 population, and 0.39 ± 0.09, 0.94 ± 0.05, and 1.72 ± 0.18 log CFU/g reduction in *E. faecium* population, respectively. Additionally, holding RF heated sample in PEI container for 10 min also helped to increase the reduction levels of both microorganisms in corn flour (Figures 6.5-6). The effect of holding sample on the inactivation of both *S*. Enteritidis PT30 and *E. faecium* in corn flour after RF heating shows similar trend with various low-moisture foods including wheat flour (Liu et, al. 2018; Xu et, al. 2018), red pepper (Hu et. al, 2018), corn grain (Zheng et al., 2017), and shell almond (Li et al., 2017).

As seen in Figures 6.5-6, holding sample after reaching 80°C in the PEI container resulted in 1.92 ± 0,17 and 1.24 ± 0.09 log CFU/g additional reductions, respectively in *S*. Enteritidis PT30 and *E. faecium* populations. However, the bactericidal action of RF treatment with 10 min holding for 80°C was not enough to achieve 4-5 log reduction in corn flour (3.76 ± 0.24 log CFU/g) for *Salmonella*. To enhance the efficiency of RF heating on bacterial inactivation in low moisture foods, it is necessary to consider the hurdle effect of RF heating with other technologies.

In this study, post-freezing storage treatment (-20°C) for 96 h was applied following RF heating with or without holding sample. The reason to apply post-freezing storage treatment to RF heated samples was to inactivate sub-lethally injured microorganisms in corn flour. Mattick et al. (2001) reported that the inactivation of bacteria in foods with high-a_w (close to 1) is based on protein denaturation which requires sufficient water to hydrate protein. On the other hand, microbial inactivation in foods with low-a_w (less than 0.6) is mostly associated with injured cell membrane through lipid fluidization because sufficient water is not presented for protein denaturation in low-moisture foods (Rychlik and Barrow, 2005). Additionally, Russell et al. (2000) reported that for healthy cells, lower temperatures promotes redeeming changes in fatty acid composition which increases the fluidity of cell membrane to acclimate freezing temperature conditions. However, RF heating can discompose the membrane stability, and as an effect of that lipid change may not be effective in acclimating damaged bacterial cell to growth at cold temperatures. Bacteria in low moisture food as subjected to RF heating might be sublethally injured especially in the cold spot areas (Figure 6.4). When environmental conditions such as water, temperature or nutrient permit, microorganisms with sublethally injured cell membrane can recover and become metabolically active. Therefore, RF heating with or without holding combined with post-freezing storage treatment may enhance inactivation level of *S*. Enteritidis PT30 and *E. faecium* in corn flour through inhibition of repair process and recovery of sublethally injured microorganisms. The effect of various treatment combinations on survival is presented in Figures 6.5-6 for both microorganisms. As represented in Figures 6.5-6, post-freezing storage treatment (-20 °C) for 24 h caused reduced survival of *S*. Enteritidis PT30 and *E*. *faecium* populations in RF heated sample with or without holding; however, no further decrease in survival was observed with longer than 48 h holding in cold temperature. Therefore, extended cold storage time beyond the 24 h is not necessary. In addition, no any significant change ($p<0.05$) in the initial load of both un-treated *S*. Enteritidis PT30 and *E*. faecium (approximately 8.5 ± 0.23 log CFU/g) was observed in corn flour during post-freezing storage treatment (Figures 6.5-6). The results evidenced that post-freezing storage treatment following RF heating with holding significantly enhanced the inactivation efficiency of RF heating. The combination of RF heating at 85°C with holding and post-freezing storage treatment for 48 h resulted in a 6.59 log CFU/g reduction in *S*. Enteritidis PT30 population and 4.79 log CFU/g reduction in *E. faecium* population in corn flour. The effect of post-freezing storage treatment on the survival of microorganisms has also been reported for green tea infusions as treated by pulsed electric field

(Zhao et al., 2009). We assumed that RF heating can generate sublethally injured microorganisms which were not able to recover or repair themselves in freezing temperature while un-treated healthy cells could survive during cold storage at -20°C for 96 h. Thus, RF treatments combined with post-freezing storage treatment successfully enhanced microbial inactivation in a synergistic manner, possibly through inhibition of repair process and recovery of sublethally injured microorganisms. Further studies are needed to find optimum storage time to develop protocol for industrial application, and to understand the survival mechanism of sublethally injured cells treated by RF heating and stored at cold temperatures.

Conclusions

The results from this study suggest that *E. faecium* can be used as a surrogate of *Salmonella* in corn flour to validate RF heating for pasteurization of low-moisture foods due to comparable thermal resistances of *S.* Enteritidis PT30 and *E. faecium* at target temperatures in inoculated corn flour. A combination of RF heating with holding and post-freezing storage treatment enhanced the inactivation level of pathogenic microorganisms in corn flour. The results also proved that *E. faecium* can be used to validate designed RF heating system for pasteurization of corn flour. Further studies are needed to optimize conditions in combination of RF heating with holding and post-freezing storage treatment to achieve the safety requirements while maintaining food quality in low-moisture foods.

Acknowledgment

This study was funded by the USDA-NIFA AFRI. Grant Number: 2013-67005-21295. We also would like to acknowledge Dr. Mark A. Harrison and Dr. Abhinav Mishra for their help on this project. The authors also thank the Turkish Ministry of National Education Council for providing the chance to Samet Ozturk for his Ph.D. program.

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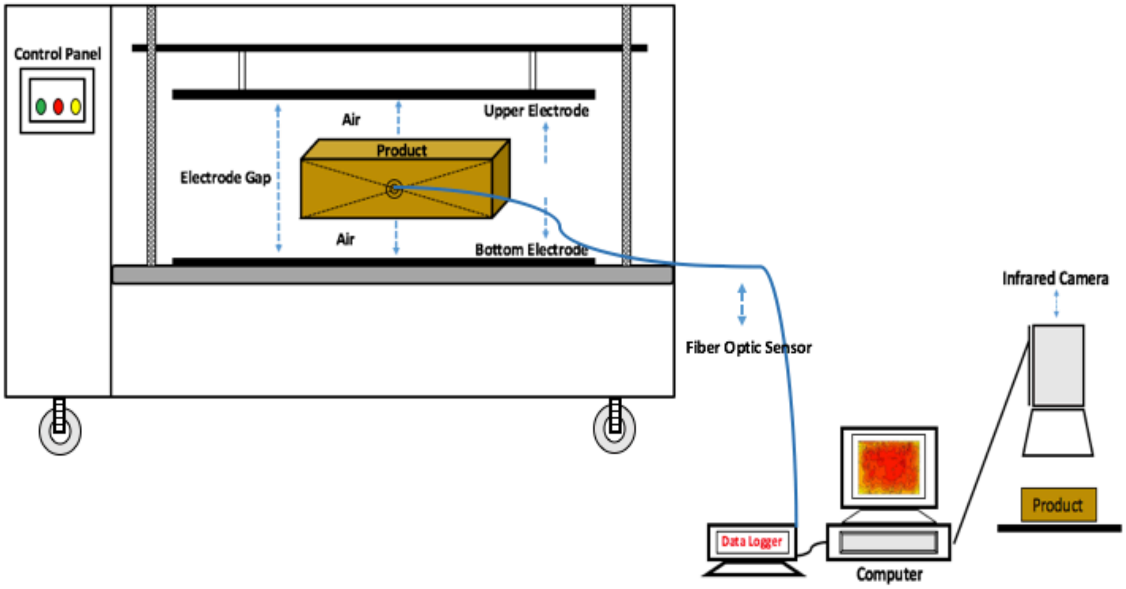

Figure 6.1. Schematic diagram of corn flour filled PEI rectangular container placed in the middle of two parallel electrodes in a 6 kW 27.12 MHz radio frequency heating (Ozturk et al., 2017)

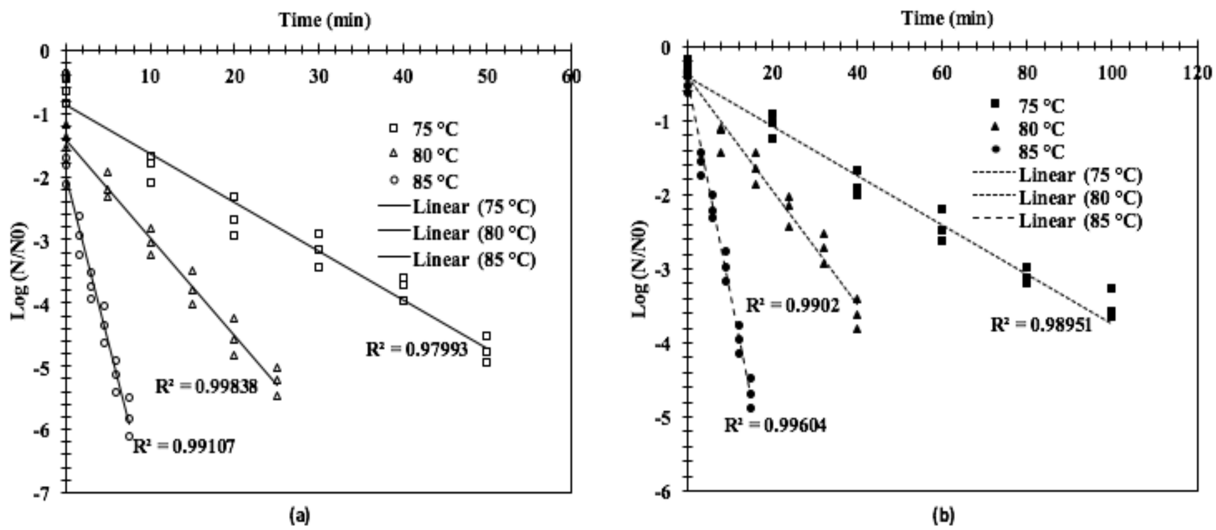

Figure 6.2. Inactivation kinetic curves of *S.* PT30 (a) and *E. faecium* NRRL B-2354 (b) in corn flour (water activity 0.45 ± 0.05) at 75, 80 and 85ºC

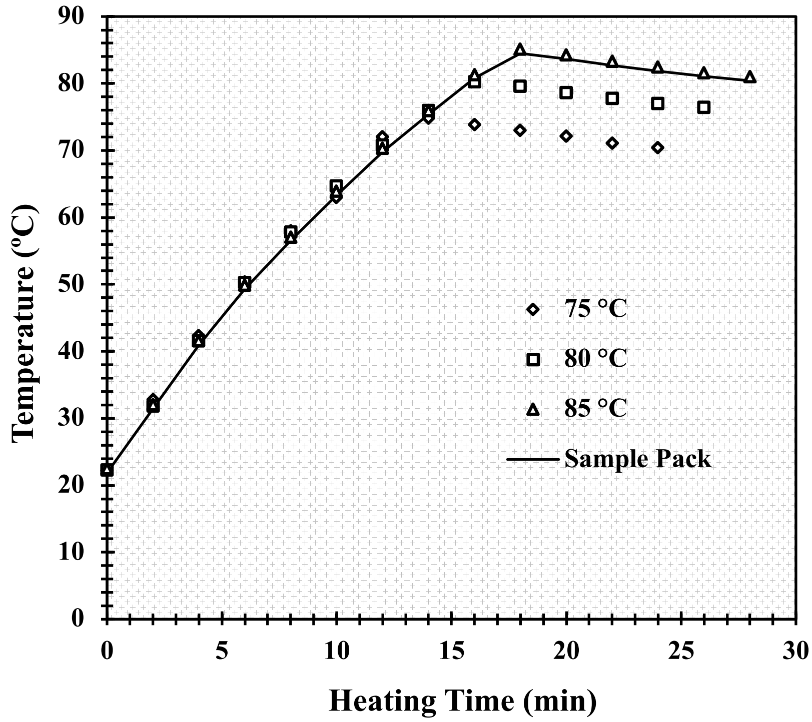

Figure 6.3. A typical measured temperature–time curve of corn flour in a PEI container subjected to RF heating at 15 cm electrode gaps with 10 min holding

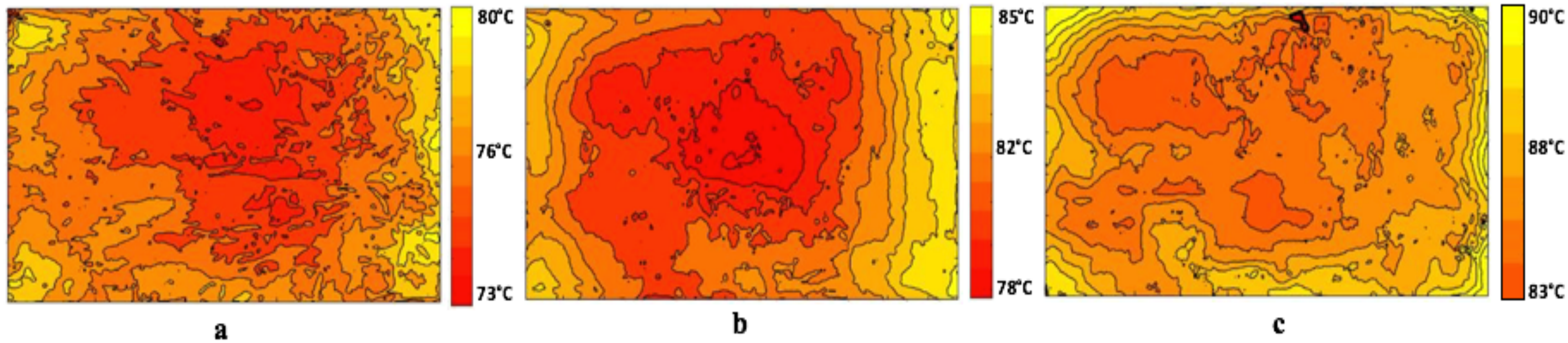

Figure 6.4. Temperature distribution in the middle layer of RF heated corn flour during CUT at 75 (a), 80 (b), and 85ºC (c) at 15 cm electrode gap

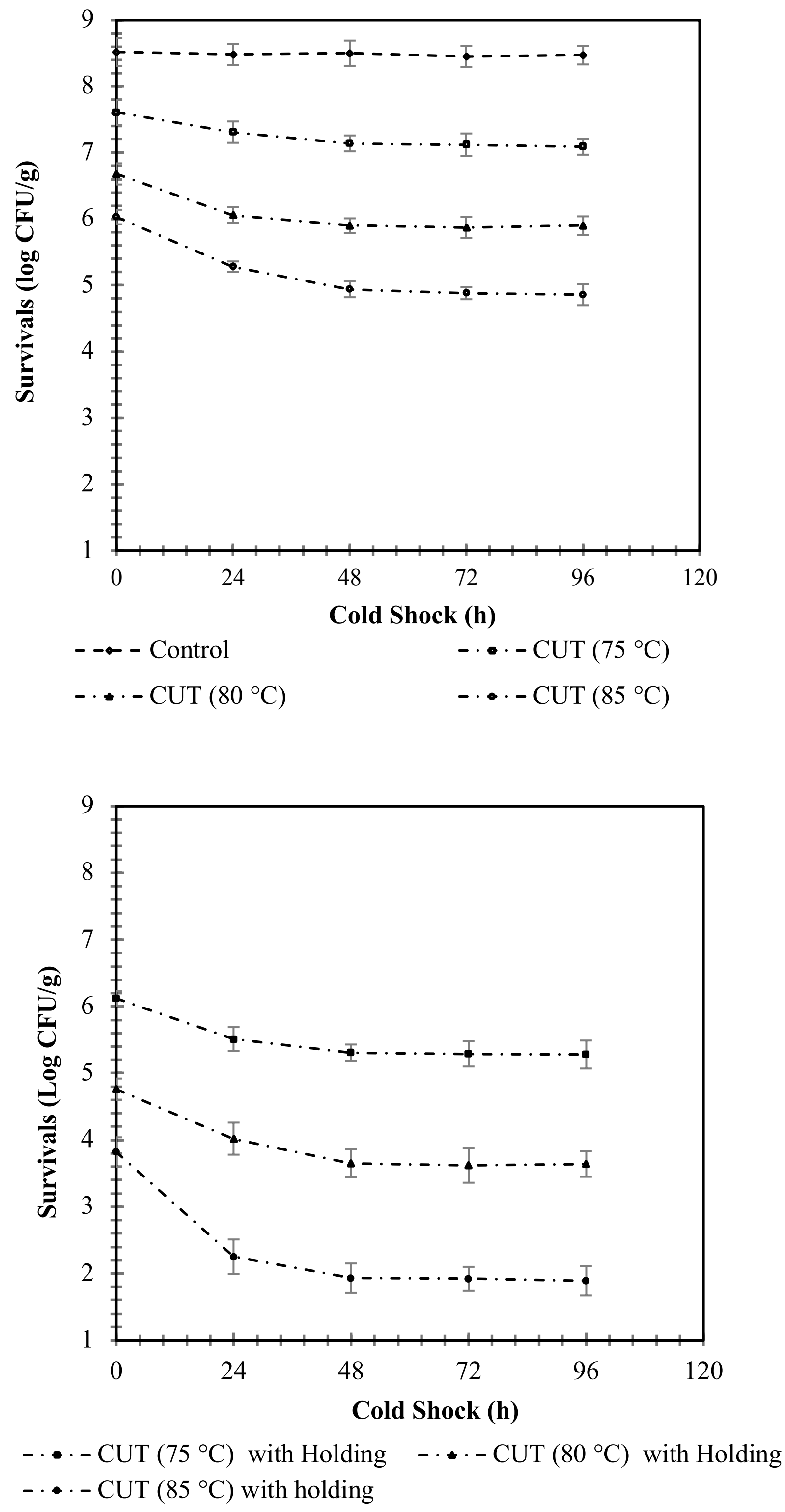

Figure 6.5. Effect of post-freezing storage (-20°C) on survivals of *S.* PT30 in untreated and RF treated corn flour after CUT with or without holding

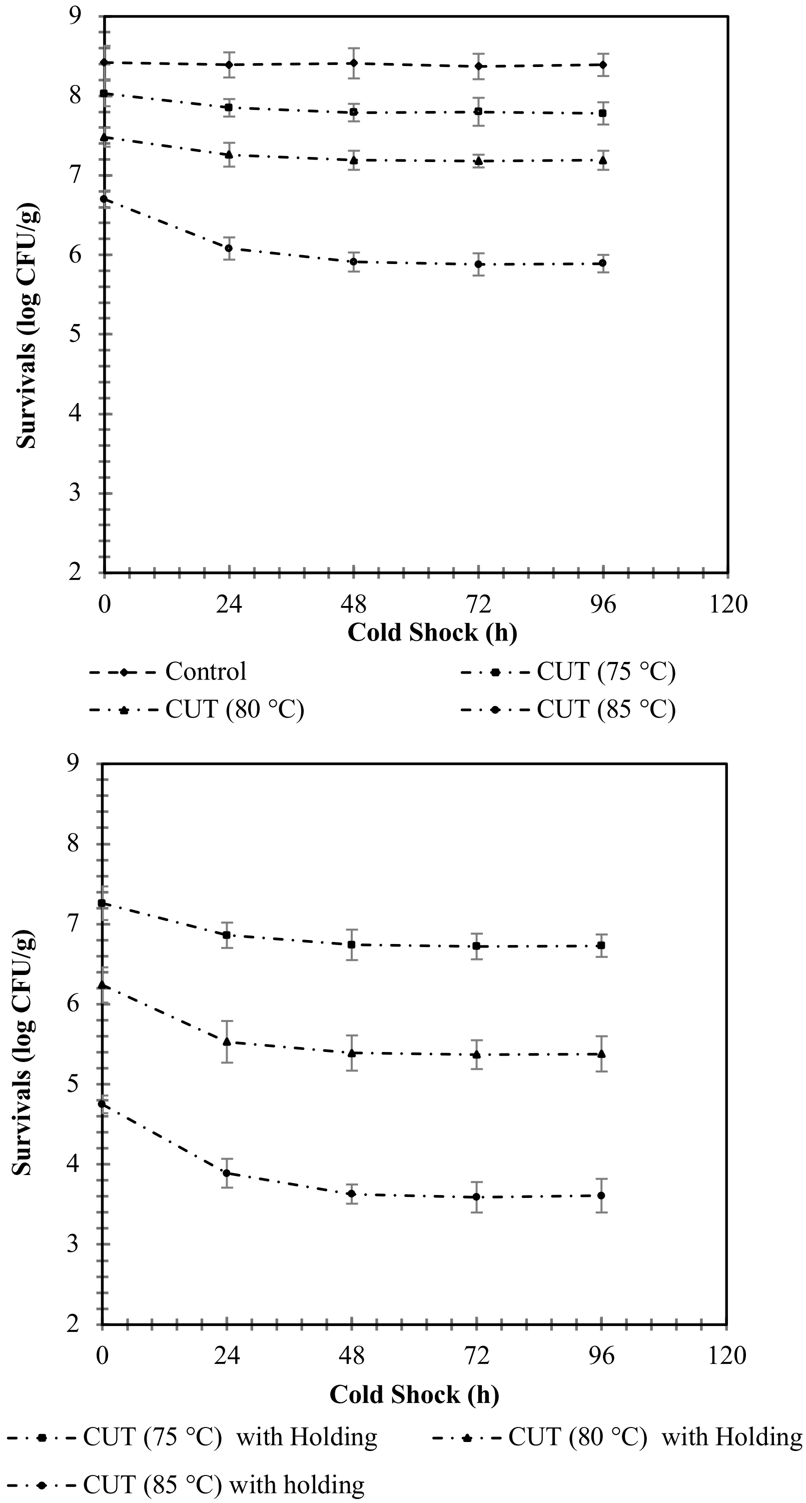

Figure 6.6. Effect of post-freezing storage (-20°C) on survivals of *E. faecium* in untreated and RF treated corn flour after come-up time (CUT) with or without holding

Table 6.1. Parameter estimates for the primary models

		Linear Model		Weibull Model		
	Temperature (ºC)	D-value (min)	RMSE (log CFU/g)	δ(min)	α	RMSE (log CFU/g)
S. PT30	75	14.18 ± 1.24	0.53	15.22 ± 3.42	1.42 ± 0.14	0.62
	80	6.11 ± 0.59	0.24	7.13 ± 0.64	0.72 ± 0.09	0.12
	85	2.02 ± 0.31	0.19	1.67 ± 0.46	0.66 ± 0.04	0.19
E. faecium	75	22.41 ± 1.43	0.42	24.67 ± 4.35	1.02 ± 0.26	0.42
	80	10.32 ± 0.82	0. 33	9.12 ± 0.87	0.84 ± 0.16	0.11
	85	3.17 ± 0.26	0.12	2.24 ± 0.52	0.92 ± 0.08	0.17

* The root mean square error (RMSE) was determined by 1stOpt software using triplicate experimental data.
* Values are means ± standard errors. Parameters were estimated separately for each data set. Smaller RMSE values indicate a better fitness of the model.

CHAPTER 7

INACTIVATION KINETICS OF *SALMONELLA* AND *ENTERACOCCUS FAECIUM* NRRL B-2354 IN PACKED SPICES AS SUBJECTED TO RADIO FREQUENCY HEATING[5]

[5]S. Ozturk, R. K. Singh, F. Kong. To be submitted to the *Journal of Food Engineering*.

Abstract

Salmonella contaminations on various seasoning spices occurred in the last decade. Radio frequency (RF) heating has potential to be an alternative inactivation method for pathogens in low moisture foods (LMF) including spices. This study aimed to (1) determine thermal resistances (D and z values) of *Salmonella*, as well as *Enterococcus faecium* NRRL B-2354 (*E. faecium)* as a potential surrogate for thermal validation studies in paprika, white pepper and cumin powders, and (2) investigate inactivation effect of RF on both microorganisms in packaged spice samples. Initial water activity ($a_{w,25°C}$) of each sample was adjusted as 0.45 ± 0.05 using a humidity chamber. The samples were inoculated with *Salmonella* cocktail (*S.* Typhimurium, *S.* Agona, *S.* Montevideo, and *S.* Tennessee) and *E. faecium* at approximately 8.5 log CFU/g. Inoculated samples were loaded in the thermal death time (TDT) cells and subjected to isothermal treatment in a water bath at 70, 75 and 80°C to evaluate thermal resistances. In addition, inoculated samples (20 g) were put in a small polystyrene petri dishes and subjected to RF heating in a 27.12-MHz, 6-kW pilot scale RF unit with 10.5 cm electrode gap until the temperature at the geometric center of the sample reached 80°C. The survival of bacteria was enumerated and converted to log CFU per gram to evaluate inactivation effect. The change in color of RF heated samples was also evaluated to assess the impact of RF heating on the product quality. Results show that thermal resistance of *E. faecium* at inactivation temperatures is significantly higher than *Salmonella* in the three spices. Both microorganisms showed less thermal resistance in paprika than the others. $D_{80°C}$ values of *Salmonella* and *E. faecium* were determined as 1.21 and 1.82 min in paprika, 2.85 and 5.27 min in white pepper, and 4.47 and 9.53 min in cumin powder, respectively. During RF heating, the come-up times (CUT) to reach 80°C were determined as 2.15, 4.2 and 6.1 min for paprika, white pepper and cumin powders, respectively. RF heating during the CUT resulted in 4.59, 3.65 and 2.83 log CFU/g reductions of *Salmonella*, and 2.44, 1.76 and 0.96 log CFU/g reductions of *E. faecium* in paprika, white pepper and cumin powders, respectively. Results shows that *E. faecium* is a potential surrogate for *Salmonella* to validate thermal processes, and RF heating is efficient to control foodborne pathogens in spices.

Key Words: *Salmonella*, *Enterococcus faecium* NRRL B-2354, Radio Frequency (RF) Heating, Pasteurization, Spices

Introduction

Reduced water activity (a_w < 0.6) limits the growth of microorganism and extends the shelf stability of low moisture foods (LMF) including dehydrated vegetable powders and spices; therefore, these foods have been considered historically as safe ingredients for foodborne pathogens. However, such ready to eat foods do not undergo any further thermal processing after packaging; thus, they might be potentially contaminated by pathogenic microorganisms during the all stages through the processing and supply chain. Although the antimicrobial effects of spices are widely known to reduce the microbial load of product during the storage (Arora and Kaur, 1999; Billing and Sherman, 1998; Ceylan and Fung, 2004; Dorman and Deans, 2000; Weerakkody et al., 2011), it is not clear that whether inhibitory effect of spices are sufficient or not to inhibit the growth of foodborne pathogens. In addition, the decrease in a_w causes the increase in heat resistance and survivability of *Salmonella* in LMFs during processing and the extended storage time (Beuchat et al., 2013; Burnett et al., 2000; Mattick et al., 2000). Furthermore, the heat resistance and survivability of *Salmonella* in LMFs are also influenced by other factors such as ingredient composition (fat, sugar, carbohydrate, salt and protein etc.), pH, food matrix and the physiological state of the foodborne microorganism cells (Juven et al., 1984; Podolak et al., 2010; Shachar and Yaron, 2006). Recent studies showed that the presence of foodborne pathogen in spices could lead to severe food-borne illnesses imposing threat to human public health around the world ((EFSA), 2010; Rico et al., 2010; Waje et al., 2008). In the last decade, *Salmonella* outbreaks were found associated with contamination of various spices including white pepper (CDC, 2010), paprika (Lehmacher et al., 1995), cumin powder (Moreira et al., 2009), and other spices in the United States (Zweifel and Stephan, 2012). The increase in pathogenic outbreaks and recalls associated with spices indicates that the current processing methods and regulations to eliminate or control foodborne pathogens are not sufficient. Therefore, the food industry and scientific community need to take a responsibility to improve the processing techniques and to evaluate the survival ability of foodborne pathogen in dehydrated foods and ingredients with low a_w during the process and storage time. Current pasteurization methods for *Salmonella* in LMFs include steam treatment, fumigation, ethylene oxide and irradiation. But due to the various reasons such as processing cost, quality problem, and public health concern, they are not widely accepted or allowed to use (Lee et al., 2006). As an alternative novel technology, radio frequency (RF) heating in the range of 3 kHz - 300 MHz shows a great potential with better heating uniformity and shorter processing time to inactivate *Salmonella* in LMFs in which heat is generated in dielectric material through molecular fraction caused by ionic conduction and dipole rotation. The inactivation efficiency of RF heating on foodborne pathogens in various LMFs including almonds (Gao et al., 2012; Gao et al., 2011; Gao et al., 2010), flour (Tiwari et al., 2011; Villa-Rojas et al., 2017), peanut butter (Villa Rojas, 2015) and pepper spices (Kim et al., 2012) has been investigated.

Process validation is critical to establish an effective RF heating protocol for commercial applications. There are strict safety limitations to use foodborne pathogens directly in industrial food processing plants for validation studies (Niebuhr et al., 2008). Consequently, to establish reliable regulations and to validate processing conditions, it is necessary to use nonpathogenic microorganisms with slightly higher thermal resistance as surrogate of foodborne pathogens in-plant process validation (Enache et al., 2015). As heat resistant nonpathogenic bacteria, *Entorococcus faecium* NRRL B-2354 (*E. faecium*) has been used in thermal process validation of LMFs including almond, wheat flour and various species as a potential surrogate for *Salmonella* (Bingol et al., 2011; Liu et al., 2018; Rachon et al., 2016; Tiwari et al., 2011).

Although *E. faecium* have shown a potential for using as surrogate of *Salmonella* (California, 2007; Enache et al., 2015; Jeong et al., 2011; Jeong and Kang, 2014), it also has limitations as a surrogate depending on types of foods and processing methods (Rachon and Gibbs, 2015). So far, there has no study conducted to evaluate the applicability of using *E. faecium* as a surrogate for *Salmonella* in spices as subject to RF heating. Furthermore, the log-linear kinetic is commonly used to estimate the efficiency of conventional thermal processing on inactivation of microorganisms using obtained survival curves; however, recent studies shows that the inactivation kinetics of novel treatment methods such as RF heating, microwave and ohmic heating on microorganisms do not always follow traditional linear survival curves (Bermudez-Aguirre and Corradini, 2012).

This study was focused to investigate inactivation kinetic of *Salmonella* and its potential surrogate, *E. faecium*, in selected spices as subjected to RF heating. Specifically, the objectives of aimed study were (1) to evaluate the suitability of *E. faecium* as a potential surrogate of *Salmonella* in paprika, white pepper and cumin powder with $a_{w,25°C} = 0.45$ at target temperatures (70, 75 and 80°C) and investigate the inactivation kinetics using the First order kinetic and Weibull models, (2) to investigate the inactivation effect of RF heating at 80°C and the quality changes of RF heated samples using color as an indicator.

Material and Methods

Paprika, white pepper and cumin powder were purchased from a local store (Athens, GA USA). The initial water activity at 25 °C ($a_{W,\ 25°C}$) of each sample was measured with a water activity meter (AQUA PRE, Decagon Devices, Pullman, WA) as 0.71, 0.68 and 0.66, respectively. The water activity of the samples was adjusted as $a_{w,25°C} = 0.45 \pm 0.05$ using the Hotpack 435315 humidity chamber (SP Industries, Inc., Warminster, PA).

Bacterial inoculation of paprika, cumin and white pepper

A cocktail of four *Salmonella* strains was used in this study consisting of *S*. Typhimurium, *S*. Agona, *S*. Montevideo and *S*. Tennessee. Selections of each strain was based on strains from outbreaks linked to LMFs. Additionally, *Entorococcus* faecium NRRL B-2354 (*E. faecium*), which is used as a potential surrogate of *Salmonella*, was used as a single strain of *Enterococcus*. All strains were obtained from Dr. Harrison's laboratory (University of Georgia, Athens GA), and cultured in 9 ml of tryptic soy broth (TSB) supplemented with 0.6 % (wt/vol) yeast extract (TSBYE) at 37 °C for 24 h and then 1 ml evenly spread on a plate (150 x 15 mm) of TSAYE. Growth cells on TSAYE were harvested into 20 ml of sterile 0.1% peptone water, and centrifuged for 30 min at 2,600g. Subsequently, suspended pellets of each strain of the four-pathogen species were combined to make culture cocktails. Then, the supernatant was discarded and the pellet was re-suspended in 3 ml 0.1% peptone water as described in (Liu et al., 2018). To obtain more uniform inoculation in sample, 10 g of each spices were put in small sterile bags and inoculated with 1 ml of concentrated pellet. Inoculated samples were hand-mixed until the pellet was visibly mixed in a safety cabinet. After mixing, inoculated spices in sterile bags were used to further inoculate 90 g sample of each spice separately, which were mixed and stomached (Seward Stomacher, 400 Lab System, Norfolk, United Kingdom) at 260 rpm for 5 min. To confirm the uniformity of bacterial inoculation in samples, ten of 1 g each spice was randomly selected and enumerated on mTSA (TSA agar, Yeast Extract, 0.05% Ammonium Iron (Ш) Citrate, and 0.03% Sodium Thiosulfate Pentahydrate ($5H_2O$)) and eTSA (TSA agar, Yeast Extract, 0.05% Ammonium Iron (Ш) Citrate and Esculin Hydrate) plates for *Salmonella and E. faecium*, respectively. The concentration level of inoculated spices was determined as approximately 8.5 log CFU/g for isothermal and RF treatments.

Equilibration of water activity

To avoid the effect of water activity on thermal resistances of *Salmonella* cocktail and *E. faecium* in sample, inoculated spice samples were placed in sterile trays and then put into a Hotpack 435315 humidity chamber (SP Industries, Inc., Warminster, PA) (Liu et al., 2018) to ensure equilibrium with the target water activity ($a_{w,25°C}$ = 0.45 ± 0.05) (Hildebrandt et al., 2016)

Isothermal treatment

To investigate thermal resistances (D and z values) of *Salmonella* and *E. faecium* in paprika, white pepper and cumin powder with $a_{w,25°C}$ = 0.45 ± 0.05, inoculated samples were filled in the aluminum test cells (Washington State University, Pullman, WA) with around 0.6-0.8 g and 4 mm thickness, and isothermally treated in a water bath (Model: SWB-10L-2, Saratoga, CA USA) with maintained inactivation temperatures (70, 75 and 80°C) (Chung et al., 2008; Villa-Rojas et al., 2013). To obtain thermal death curves of *Salmonella* and *E. faecium* in the sample, isothermal treatments was applied for different time intervals after the predetermined CUT was reached. The CUTs for each spice were separately determined using an aluminum test cell with a T-type thermocouple inserted in the center of cell filled with non-inoculated spice samples (around 0.6-0.8 g, $a_{w,25°C}$ = 0.45 ± 0.05). Isothermally treated cells were immediately removed from the water bath and placed in an ice-water bath for 90 s to deactivate thermal inactivation of bacteria. Each trial set up was triplicated.

Enumeration of microbial survival

Isothermally treated samples in the test cells at the selected time intervals were transferred into sterile stomacher bags containing 0.1 % peptone water to dilute 1:10, and homogenized for 2 min 260 rpm with a Seward Stomacher (Seward, London, UK) (Harris et al., 2012). After homogenization, 1 ml of each diluent was serially diluted in 9 ml of 0.1 % peptone water, and

0.1 ml of final diluent was spread-plated in duplicate onto mTSA for *Salmonella* or eTSA for *E. faecium* to enumerate survivals in selected spices, respectively. All plates were incubated at 37 °C for 24-48 h, and counted populations were converted to log CFU per gram. Log reductions were calculated by subtracting the survivor counts from the initial population.

Microbial inactivation kinetics

To describe the inactivation kinetics and calculate heating times of *Salmonella* and *E. faecium* in paprika, white pepper and cumin powder as treated by isothermally, the first order kinetics (D-value) Eq. (1), and the Weibull model Eq. (2) were applied using the following equations below (Peleg, 2006);

$$log\ \frac{N}{N_0} = -\frac{t}{D} \qquad \text{Eq. (1)}$$

$$log\ \frac{N}{N_0} = -\left(\frac{t}{\delta}\right)^{\alpha} \qquad \text{Eq. (2)}$$

where N_0 was the initial load of microorganism (CFU/g), and *N* was the population of survivals (CFU/g) at time (*t*), the isothermal treatment time (min) after CUT; D is the time to reduce microbial population by 10-fold at the inactivation temperature (°C); δ indicates the overall steepness of survival curve; α is the survival curve factor which indicates whether it is linear (α=1) or non-linear (α≠1) with a decreasing (α < 1) or increasing (α > 1) inactivation rate with time.

Obtained data after isothermal treatment was applied to fit the two models to the survival curves of both microorganisms and to estimate the model parameters. Root mean square error (RMSE) (log CFU/g) were applied to interpret the performance of the models.

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}\left[log\frac{N}{No}_{data,i} - log\frac{N}{No}_{model,i}\right]^2}{n-p}} \qquad \text{Eq. (3)}$$

where $log \frac{N}{No}_{data,i}$ is measured log CFU/g reduction, and $log \frac{N}{No}_{model,i}$ is predicted log CFU/g reduction from model, n is the total number of observations, and p is the number of model parameters. To estimate fitness of model and calculate RMSE, 1stOpt software was used for analyzing the results. To evaluate differences between D-values among samples, the ANOVA in Minitab 14 (Minitab Inc., State College, PA) was also performed. The thermal death time curves was obtained by plotting the log of D-values against temperature, in which the slope refers to the -1/z, where z is the change in temperature to alter the thermal-death-time by one log-cycle (Gaillard et al., 1998).

Radio frequency heating: uniformity and effect of microbial inactivation

To investigate temperature time profiles during the RF treatment, and the inactivation effect of RF heating on *Salmonella* and *E. faecium* in inoculated paprika, white pepper and cumin powder, samples (20 g) were filled in a small cylindrical polystyrene petri dishes with diameter 50 mm and height 15 mm. Recent studies showed that placing dielectric material at the center of parallel electrodes provides better heating rate with improved heating uniformity through sample as subjected to RF heating (Ozturk et al., 2017; Tiwari et al., 2011). To determine come-up time (CUT) till 80°C from initial temperature (23 ± 2 °C) of each sample, un-inoculated samples filled in petri dishes and sealed with polystyrene plastic (1 mm thickness) (Figure 7.1) was placed between two parallel electrodes with gap of 105 mm, and then subjected to RF heating in a 27.12-MHz, 6-kW RF system (COMBI 6-S, Strayfield International, Wokingham, UK) (Figure 7.2). The change in temperature of samples during RF heating was recorded using a fiber optic temperature sensor with an accuracy of ± 1 °C (Fiso Tech. Inc., Quebec, Canada) connected to a data logger to determine the CUTs at the geometric center of the cylindrical petri dishes. The obtained CUTs were applied to evaluate the efficiency of RF heating on pasteurization of inoculated samples with *Salmonella* and *E. faecium*. The heating uniformity of RF heated samples were also evaluated using an infrared camera (FLIR T440, FLIR Systems, Inc., North Billerica, MA, USA) with an accuracy of ± 2 °C. Only top surface thermal images were used to determine heating uniformity index and average temperature of RF heated sample due to the small thickness of sample holder petri dishes (15 mm). The heating uniformity of RF heated sample was evaluated using uniformity index (IU), which has been applied to determine heating uniformity of various RF heated LMFs (Hou et al., 2014; Jiao et al., 2012; Pan et al., 2012; Wang et al., 2008; Wang et al., 2010; Wang et al., 2005). The UI value of RF heated spices was calculated by following equation; (Wang, 2005):

$$\lambda = \frac{\Delta\sigma}{\Delta\mu} \qquad \text{Eq. (4)}$$

where $\Delta\sigma$ is identified as the increase in the standard deviation (SD) of RF heated sample (°C), and $\Delta\mu$ is the increase in the average temperature (°C) during the RF heating. The lower UI values indicates better heating uniformity in the heated sample.

To determine efficiency of RF heating on inactivation of both microorganisms, sealed cylindrical petri dishes filled with inoculated samples (20 g) were treated by RF system during predetermined CUTs to reach 80°C with different time intervals. Then RF heated sample packages were immediately removed from container and inserted in ice-water bath for 90 s to stop further thermal inactivation of microorganism in sample. Each treated 20 g of sample were transferred in a sterile stomacher bags containing 180 ml of 0.1 % peptone water and homogenized for 3 m in a 260 rpm with a Seward Stomacher (Seward, London, UK). The microbial survival was enumerated following steps described above.

Color measurement

To evaluate the effect of RF heating on food quality, the change in color values (L*, a*, and b*) of RF heated sample was measured using a Minolta colorimeter (model CR300, Minolta Co., Osaka, Japan). L*, a*, and b* values indicate the lightness of color, whiteness, and yellowness of the sample, respectively. The color meter was calibrated before each measurement by using black and white calibration plates. RF treated samples at 80°C were spread into a plastic tray and top surface was flattened for taking measurement at randomly selected five points. The average of five measurements was used to evaluate the effect of RF on food quality. Color date analyzed by analysis of variance using the ANOVA procedure with Duncan's multiple range test of SAS (SAS Institute, Cary, NC, USA). Value of $P < 0.05$ was used to indicate significant difference between treated and non-treated samples.

Results and Discussion

D- and z- values of *Salmonella* and *E. faecium* in paprika, white pepper and cumin powder

The CUTs of paprika, white pepper and cumin powder, defined as the time to reach targeted temperature ± 0.5°C, were around 120-180 s for isothermal treatment in the water bath. Inactivation kinetics of *Salmonella* and *E. faecium* in paprika, white pepper and cumin powder with $a_{w,25°C}$ = 0.45 ± 0.05 were represented in the Figures 7.3, 7.4 and 7.5, respectively. Survival data of both *Salmonella* and *E. faecium* in each sample fit well the primary models and showed similar RMSE values (Table 7.1). The first order kinetic (log-linear) model was applied to describe thermal resistances of both microorganisms at the inactivation temperatures. As seen in the Table 7.1, the D values at 70, 75 and 80°C of *Salmonella* in white pepper were determined as 13.79 ± 1.42, 5.27 ± 1.05 and 2.85 ± 0.79 min, respectively, with z value being 14.16°C. As a potential surrogate for *Salmonella, E. faecium* has significantly ($p < 0.05$) higher thermal resistance in each spice sample at the inactivation temperatures (Table 7.1). For example, $D_{75°C}$ of *Salmonella* and *E. faecium* in cumin powder are 8.01 ± 1.12 and 18.64 ± 1.72 min, respectively; but for paprika, $D_{75°C}$ values of both microorganism were 3.09 ± 0.82 and 4.51 ± 0.68, respectively. The z-values of *Salmonella* and *E. faecium* in cumin paprika, white pepper and cumin powder were also determined as 13.63 and 13.82°C, 14.61 and 15.48°C, and 17.55 and 19.14°C, respectively. Fig. 6 shows a comparison among the $D_{80°C}$ values of *Salmonella* and *E. faecium* in paprika, white pepper and cumin powder. As seen in the Figure 7.6, both microorganisms have the least thermal resistance in paprika among the three spices. The different heat resistance and survivability of the same serotype in different spices with the same a_w may be due to different composition. Rachon et al. (2016) reported the $D_{80°C}$ of *Salmonella* and *E. faecium* NRRL B-2354 as 1.85 and 8.66 min in culinary powder with $a_{w,25°C}$ = 0.65, and 8.93 and 23.75 min in chicken meat powder with $a_{w,25°C}$ = 0.38. Recent studies also showed that *E. faecium* can be used as a sufficient surrogate for *Salmonella* to validate thermal studies to achieve 4 to 5-log reduction in almond contaminated with *Salmonella* (Bingol et al., 2011; Kopit et al., 2014), and various LMFs including wheat flour (Liu et al., 2018; Smith et al., 2016; Syamaladevi et al., 2016), pet food and confectionery (Rachon et al., 2016). The present study specified that *E. faecium* has significantly ($P<0.05$) higher heat resistance (D-value) and survivability than *Salmonella* at all inactivation temperatures with an equivalent z-value in the 70-80°C, thus can be used as a valid surrogate for *Salmonella* to validate pasteurization of spices.

Radiofrequency heating and uniformity

Figure 7.7 shows the temperature time profiles of paprika, white pepper and cumin powder with $a_{w,25°C}$ = 0.45 ± 0.05 as subjected to RF heating to reach 80°C. The CUTs during the RF heating in each spice sample were determined as 100 s for paprika with 0.43°C s^{-1} heating rate, 250 s for white pepper with 0.22 °C s^{-1} heating rate, and 370 s for cumin powder with 0.15 °C s^{-1}. The obtained CUTs were applied to inactivate both *Salmonella* and *E. faecium* in inoculated samples. The heating profiles of these spices were also in good agreement with our previous study based on the relationship between dielectric properties and heating rates of various seasoning spices (Ozturk et al., 2018). Dielectric loss factors for the three spices were paprika > white pepper > cumin powder (Ozturk et al., 2018). Furthermore, similar temperature time profiles have been reported for different LMFs subjected to RF heating such as broccoli powder (Ozturk et al., 2016), coffee bean (Pan et al., 2012), egg white powder (Boreddy and Subbiah, 2016), legume flours and wheat flours (Guo et al., 2008; Guo et al., 2010; Jiao et al., 2011; Nelson and Trabelsi, 2006), chili powder (Li et al., 2016), black and red peppers (Jeong and Kang, 2014)..

Heating uniformity of each RF treated sample at 105 mm electrode gap is presented in the Figure 7.8, and average temperature of the surface of each sample was determined as 76.2 ± 2.3°C for paprika with 0.064 (UI), 73.6 ± 1.4°C for white pepper with 0.057 (UI), and 72.8 ± 2.1°C for cumin powder with 0.042 (UI). Due to the heat conduction throughout the cylindrical petri dish and heat loss to the air, the average temperature of the surface area was less than the target temperature at the geometric center of the container. As seen the Figure 7.8, the faster increase in temperature in paprika as subjected to RF heating resulted overheating and less heating uniformity on the top surface as compared with white pepper and cumin powder. Results shows similar observations with previous studies related to RF heating uniformity in chili powder (Li et al., 2016), black pepper and white pepper (Ozturk et al., 2018). According to our previous studies, heating samples at 105 cm electrode gap provides a linear increase in temperature with better heating uniformity. Also, from the Figure 7.8, the hot spots were located around the edges, and the cold spots in each sample were located at the center of the top surface. It is important to improve the overall heating uniformity throughout the heated sample; but due to the amount of the treated sample in this study, we believed that RF heating could be efficiently applied for pasteurization of selected spices in small petri dishes with a good heating uniformity.

Inactivation kinetic of *Salmonella* and *E. faecium*

The survival curves of *Salmonella* and *E. faecium* in paprika, white pepper and cumin powder with $a_{w,25°C}$ = 0.45 ± 0.05 as heated in the RF system with different time intervals to reach 80°C were represented in the Figure 7.9, where less log reduction was attained for *E. faecium* than *Salmonella* in each sample. From the Figure 7.9, it can be clearly seen that the first kinetic order model (log-linear model) did not fit well the survival curves of both microorganisms in each spice sample as subjected to RF heating. When paprika, white pepper and cumin powder were heated in the RF system, increased heating times are correlated with increased reduction levels of both microorganisms. The inactivation kinetics of *Salmonella* and *E. faecium* in samples as treated by RF heating can be described by the weibull model. Farakos et al. (2013) and Hu et al. (2018) also used the weibull model to evaluate the survival kinetics of *Salmonella* in various LMFs under conventional thermal treatments. In the light of obtained thermal resistance values of selected spices, a reduction of 4.59 log CFU/g of *Salmonella* was achieved in paprika while only 2.44 log CFU/g reduction of *E. faecium* was obtained after 100 s of RF heating (Figure 7.9). Also, RF heating the white pepper for 250 s showed that the level of reduction for *E. faecium* was always lower than *Salmonella* reduction (Figure 7.9). The inactivation effect of RF heating has been investigated for various LMFs by predominantly thermal effect, including denaturation of enzymes, proteins and nucleic acids, and as well as disruption of membranes (Datta and Davinson, 2000; Heddleson and Doores, 1994; Kim et al., 2012). Furthermore, Jeong and Kang (2014) reported that *S*. typhimurium associated with red and black pepper can be effectively inactivated without any subthermally injured cells, which can recover during the storage, as heated by RF system. In addition to recent studies, results of this study also agreed that RF heating can be applied to inactivate *Salmonella* in paprika, white pepper and cumin powder with reduced a_w (0.45 ± 0.05). Previous studies also showed that a reduced a_w in food resulted in a decrease in the overall log reduction of foodborne pathogens (Hu et al., 2018; Kim et al., 2012; Rachon et al., 2016). Furthermore, *E. faecium* as a potential surrogate of *Salmonella* was also investigated for almond and wheat flour; but not many studies reported its potential for *Salmonella* in spices as subjected to RF heating. The results from this study prove that *E. faecium* with higher survival and heat resistance can be applied as a valid surrogate of *Salmonella* to validate RF heating process for pasteurization of paprika, white pepper and cumin powder. Furthermore, the color of paprika, white pepper and cumin powder after RF treatment were measured and are summarized in the Table 7.2. As can be seen, L*, a*, and b* values had no significant change during the RF heating treatment, indicating the short time RF heating effectively reduced degradations in food quality

Conclusions

This study proved that *E. faecium* can be used as a surrogate of *Salmonella* to validate RF heating for pasteurization of paprika, white pepper and cumin powder. The inactivation kinetic of *Salmonella* and *E. faecium* as isothermally treated can be described by the two primary models with similar RMSE values, but the Weibull model can fit better the survival curves of *Salmonella* and *E. faecium* under RF heating. Results indicate that the RF heating is highly effective in reducing foodborne pathogens in paprika, white pepper and cumin powder without affecting the color. RF heating could be applied as an alternative pasteurization method to control foodborne pathogens in spices over conventional decontamination methods.

Acknowledgment

This study was funded by the USDA-NIFA AFRI. Grant Number: 2013-67005-21295. We would also like to acknowledge Dr. Mark A. Harrison and Dr. Abhinav Mishra for their help on this project. The authors also thank the Turkish Ministry of National Education Council for providing the chance to Samet Ozturk for his Ph.D. study.

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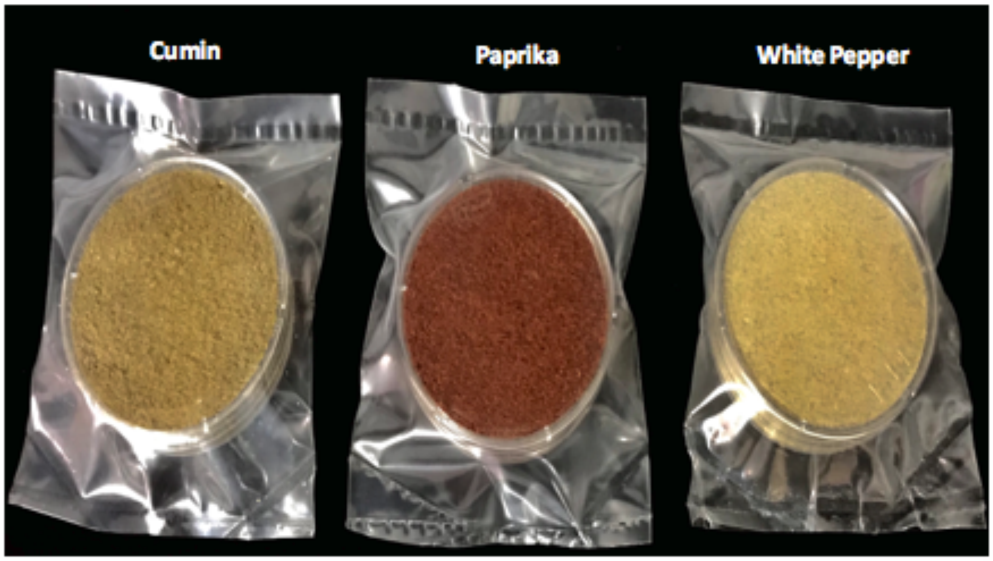

Figure 7.1. Samples in sealed small petri dishes before subjected to RF heating

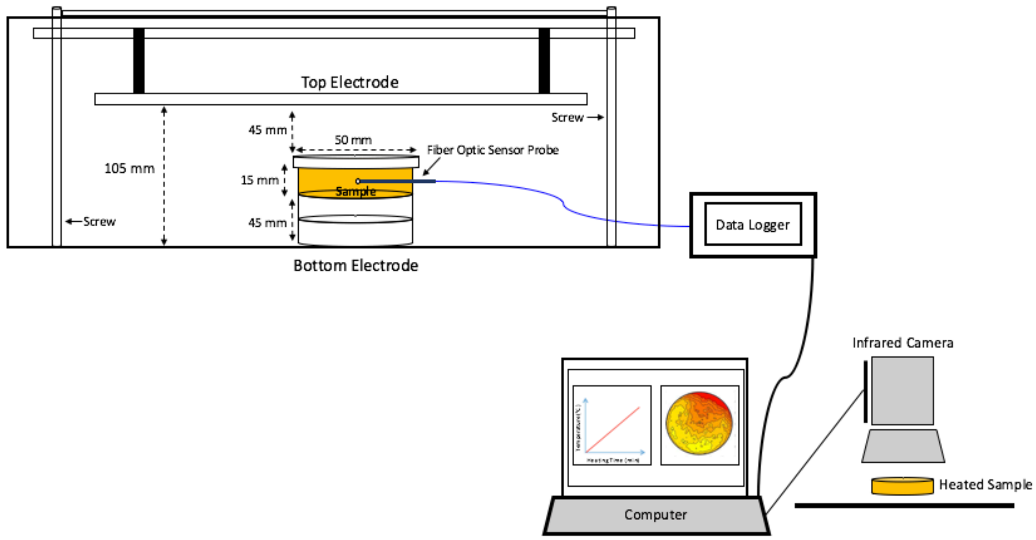

Figure 7.2. Experimental set up of RF heating system adapted from (Ozturk et al., 2018)

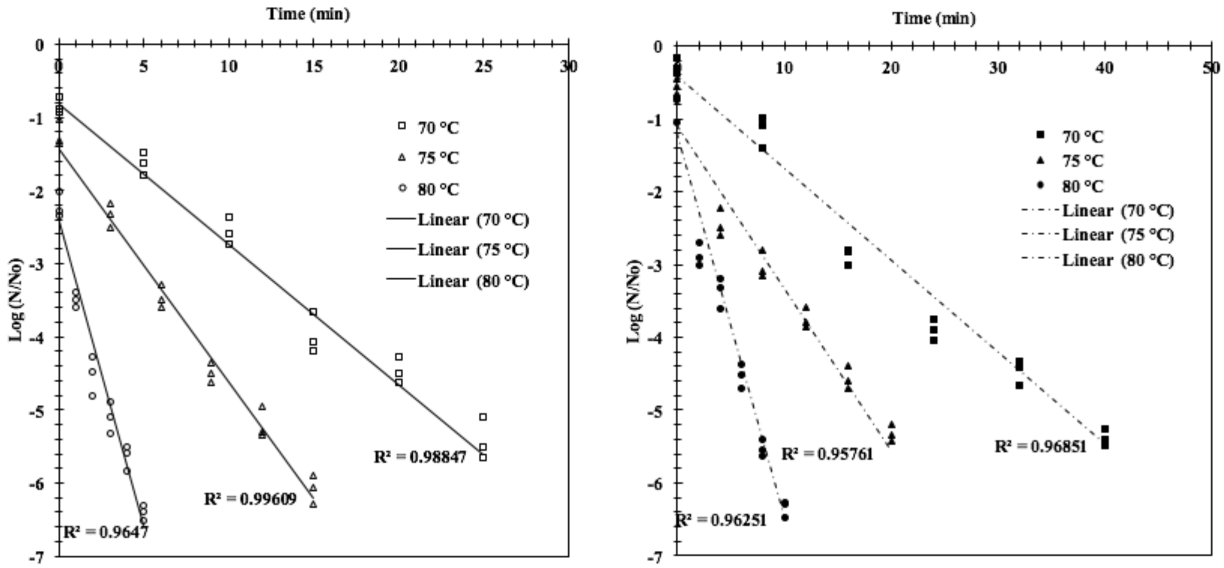

Figure 7.3. The inactivation kinetic of *Salmonella* (left) and *E. faecium* (right) in paprika with $a_{w,25°C}$ = 0.45 ± 0.05 at 70, 75 and 80°C

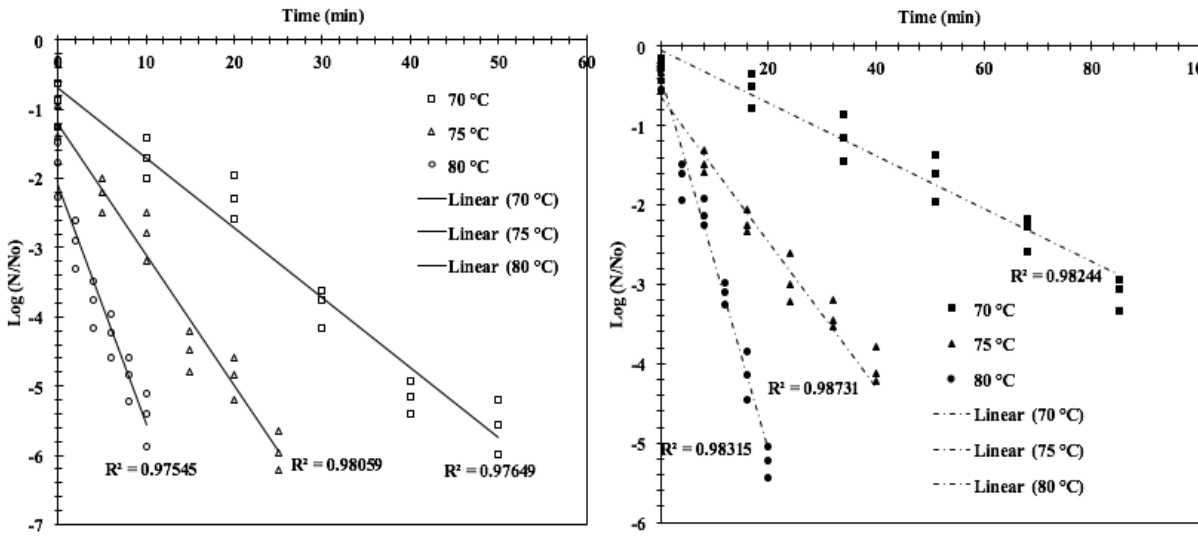

Figure 7.4. The inactivation kinetic of both *Salmonella* (left) and *E. faecium* (right) in white pepper with $a_{w,25°C}$ $= 0.45 \pm 0.05$ at 70, 75 and 80°C

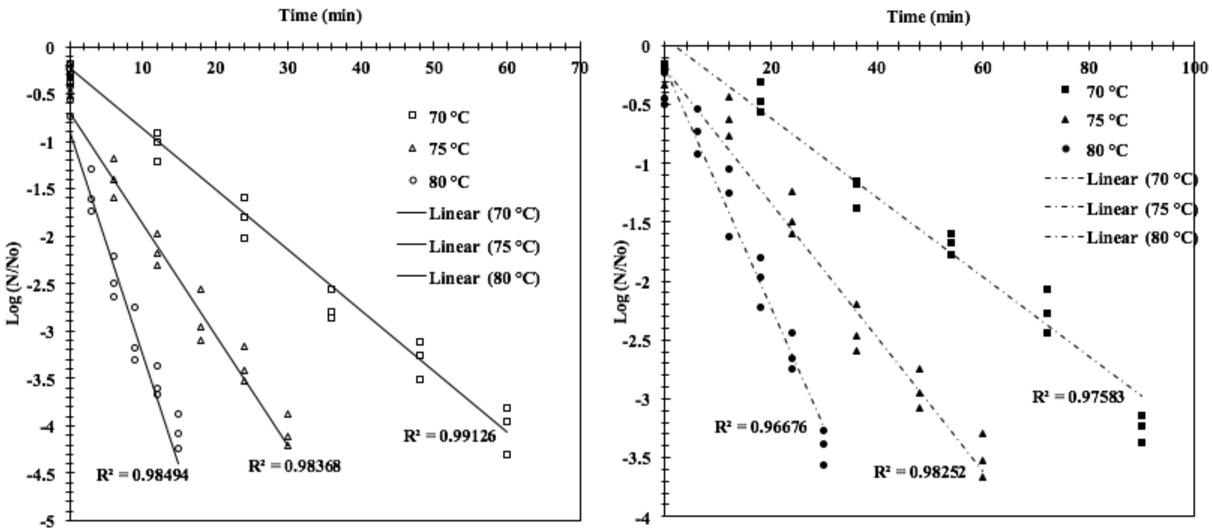

Figure 7.5. The inactivation kinetics of both *Salmonella* (left) and *E. faecium* (right) in cumin powder with $a_{w,25°\mathrm{C}} = 0.45 \pm 0.05$ at 70, 75 and 80°C

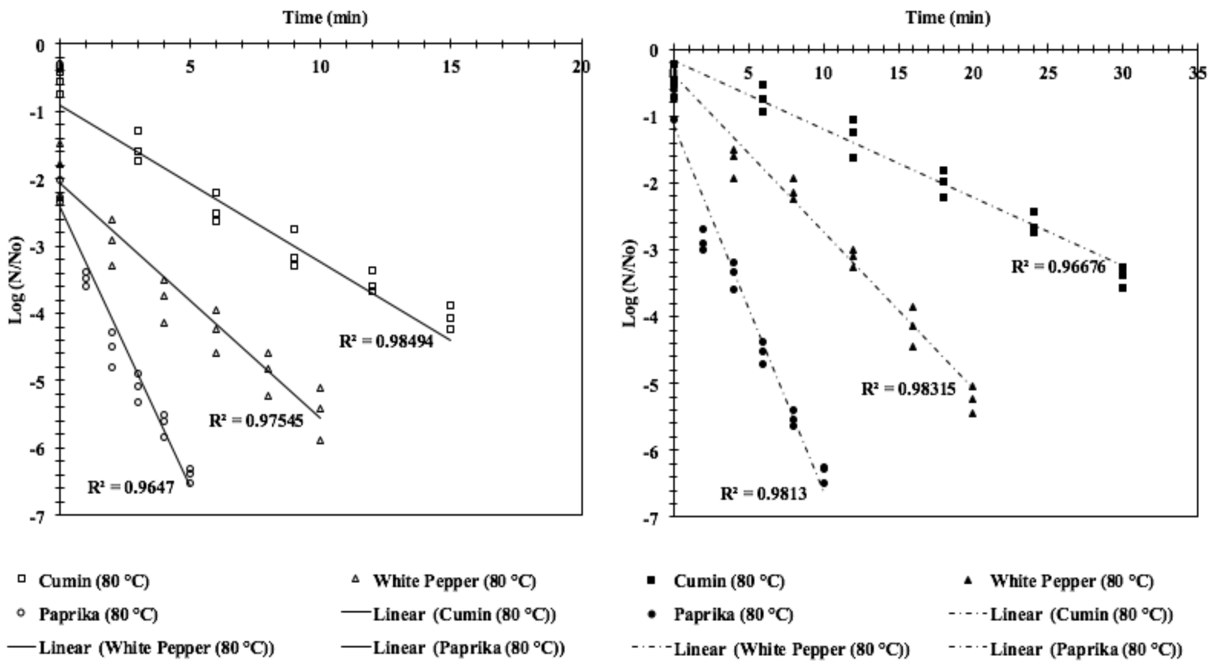

Figure 7.6. A comparison among the inactivation kinetics of *Salmonella* (left) and *E. faecium* (right) in paprika, white pepper and cumin powder with $a_{w,25°C}$ $= 0.45 \pm 0.05$ at 80°C

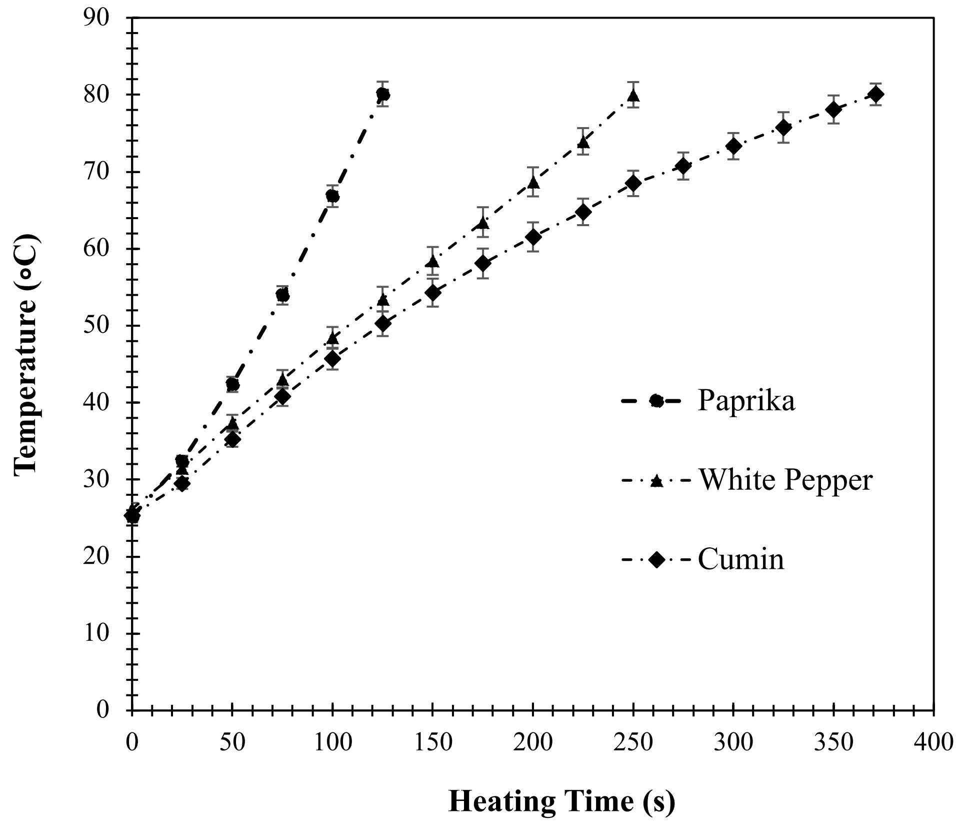

Figure 7.7. The temperature time profiles of paprika, white pepper and cumin powder with $a_{w,25°C}$ $= 0.45 \pm 0.05$ during come-up times to reach 80°C during RF heating with 10.5 cm electrode gap

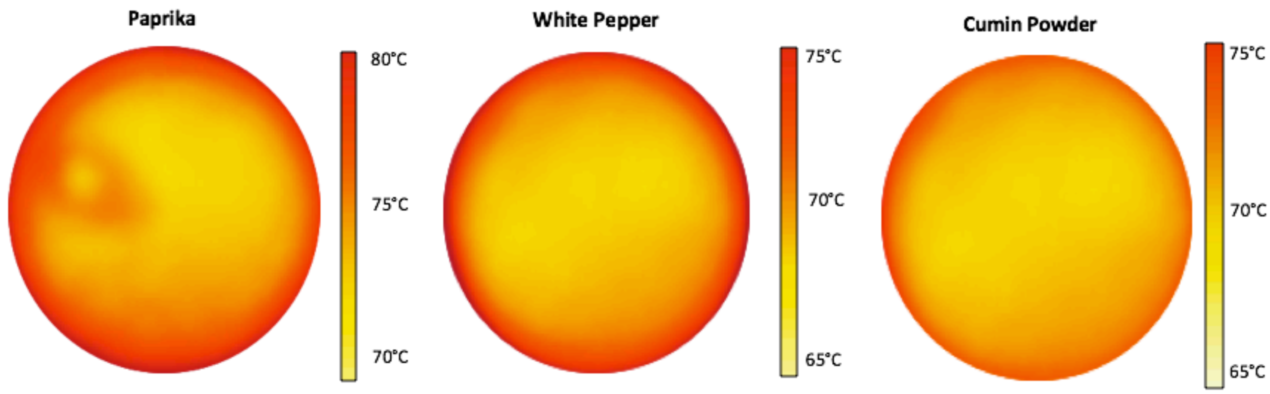

Figure 7.8. Temperature distribution on surface layer of petri dishes filled with paprika, white pepper and cumin powder as subjected to RF heating after come-up time to reach 80°C

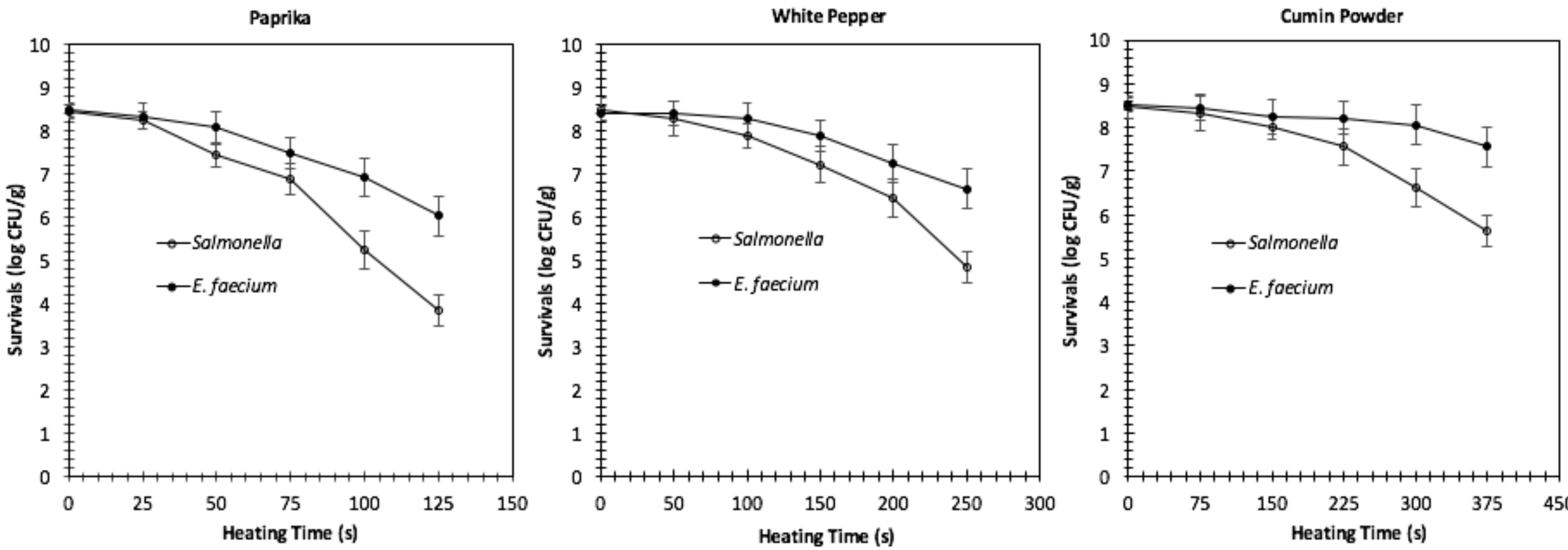

Figure 7.9. The inactivation kinetics of *Salmonella* (open circle) and *E. faecium* (close circle) in paprika, white pepper, and cumin powder with $a_{w,25°C} = 0.45 \pm 0.05$ as subjected to RF heating with different time intervals to reach 80°C

Table 7.1. Parameter estimates for the primary models of thermal resistances of *Salmonella* and E. *faecium* in selected samples

			Linear Model		Weibull Model		
Sample	Bacteria	Temperature (ºC)	D-Value (min)	RMSE (log CFU/g)	δ(min)	α	RMSE (log CFU/g)
		70	6.55 ± 1.12	0.46	7.72 ± 1.42	1.16 ± 0.14	0.56
	Salmonella	75	3.09 ± 0.82	0.23	4.56 ± 1.12	0.69 ± 0.09	0.32
		80	1.21 ± 0.46	0.17	1.78 ± 0.39	0.72 ± 0.04	0.21
Paprika							
		70	9.62 ± 0.96	0.21	12.86 ± 1.26	1.09 ± 0.26	0.32
	E. faecium	75	4.51 ± 0.68	0.35	5.16 ± 0.52	0.74 ± 0.16	0.41
		80	1.82 ± 0.26	0.52	2.56 ± 0.42	0.89 ± 0.06	0.49
		70	13.79 ± 1.42	0.37	16.12 ± 1.69	1.24 ± 0.17	0.42
	Salmonella	75	5.27 ± 1.05	0.19	7.09 ± 1.45	0.92 ± 0.11	0.21
		80	2.85 ± 0.79	0.44	4.21 ± 1.09	0.76 ± 0.08	0.39
White Pepper							
		70	23.32 ± 1.86	0.53	26.19 ± 2.42	1.12 ± 0.36	0.63
	E. faecium	75	12.67 ± 1.32	0.24	14.52 ± 1.63	0.81 ± 0.13	0.19
		80	5.27 ± 0.86	0.41	6.51 ± 1.06	0.96 ± 0.09	0.38
		70	16.61 ± 1.24	0.58	16.61 ± 1.24	1.52 ± 0.11	0.64
	Salmonella	75	8.01 ± 1.12	0.35	8.01 ± 1.12	0.83 ± 0.07	0.46
		80	4.47 ± 0.89	0.24	4.47 ± 0.89	0.78 ± 0.06	0.19
Cumin Powder							
		70	28.31 ± 2.24	0.73	33.19 ± 2.56	1.26 ± 0.18	0.82
	E. faecium	75	18.64 ± 1.72	0.52	21.04 ± 2.14	0.92 ± 0.26	0.61
		80	9.53 ± 1.18	0.29	11.42 ± 1.18	0.86 ± 0.07	0.42

* The root mean square error (RMSE) was determined by 1stOpt software using triplicate experimental data.
* Values are means ± standard errors. Parameters were estimated separately for each data set. Smaller RMSE values indicate a better fitness of the model.

Table 7.2. Color values[a] of RF treated paprika, white pepper and cumin powder

		Control	RF Heated
Paprika	L*	34.71 ± 1.23[a]	34.36 ± 1.42[a]
	a*	26.31 ± 1.33[a]	26.16 ± 1.56[a]
	b*	29.10 ± 3.44[a]	29.66 ± 4.29[a]
White Pepper	L*	70.74 ± 0.58[a]	71.19 ± 0.47[a]
	a*	1.92 ± 0.11[a]	2.09 ± 0.17[a]
	b*	21.64 ± 0.27[a]	22.06 ± 0.54[a]
Cumin	L*	48.36 ± 1.22[a]	49.34 ± 1.58[a]
	a*	7.75 ± 0.32[a]	7.72 ± 0.23[a]
	b*	33.23 ± 2.18[a]	32.98 ± 2.34[a]

[a]Mean of five replications ± standard deviation. Values followed by the same letters within the row per parameter are not significantly different (P>0.05).
[b]Color parameters are L* (lightness), a* (redness), and b* (yellowness)

CHAPTER 8

OVERALL CONCLUSIONS AND FUTURE RESEARCH RECOMMENDATIOS

Overall Conclusions and future research recommendations

In this study, dielectric properties (DP) of the selected dehydrated food powders were determined. Results showed that dielectric constant and loss factor of samples decreased with frequency and compaction density and increased with increasing moisture content or temperature. The changes in dielectric properties of low-moisture samples with increased moisture content and temperature were greater at lower frequencies (<10 MHz) than at higher frequencies. The greater penetration depth of RF waves at lower frequencies in foods indicated that RF heating can be used to treat food powders with larger package sizes as compared with microwave heating. Additionally, the LE and LLLE equations provided a better prediction for DP values of spice mixtures than that of the CRIME equation. The mass fraction and volume fraction of each spice and salt content affected the dielectric properties of seasoning mixtures. RF heating rates of spices and their mixtures were linearly related to moisture content and the mass fraction of individual spices. The heating rates were in agreement with measured dielectric properties. In practice, RF heating uniformity and rate of food powders need to be optimized using industrial scale sample sizes and plastic pouches or other shape containers to improve their effectiveness.

For the sample heated at the central position between the two parallel electrodes, the middle layers were hotter than bottom and top layers, and the hot spot and cold spot were located in edges and center of the sample for all experimental setup, respectively. Adding and increasing the thickness of cylindrical PEI blocks on top and bottom of polystyrene container increased RF heating uniformity, rate and average temperature of corn flour. RF heating uniformity was also improved by addition of surrounding polyurethane foam sheet leading to decreased difference between hot and cold spot temperatures. The uniformity improvement methods developed in this study could be also applied for plastic pouches to optimize RF heating and minimize quality degradation in food product. Computer simulation could be developed in the future to predict heating behavior in food powders.

In this study, an effective RF heating process has been developed to pasteurize *Salmonella* in corn flour, paprika, white pepper and cumin powder without resulting in significant changes in color or moisture distribution of sample. Results suggest that *E. faecium* can be used as a surrogate of *Salmonella* in food powders for validation of RF heating. Furthermore, results showed that food matrix also plays an important role on survivability of foodborne pathogens and its surrogate in addition to a_w and temperature. A combination of RF heating with holding and post-freezing storage treatment enhanced the inactivation level of *Salmonella*. In future studies, it will be helpful to determine effect of post-freezing storage with shorter time intervals like 1 or 2 hours on survival of foodborne pathogens and food quality changes after RF heating. Moreover, although the antimicrobial effect of various spices on reduction of microorganisms at room temperature has been studied, it would be interesting to determine the change in antimicrobial effect of spices at high temperatures and evaluate their effect on inactivation of pathogenic microorganism during RF heating.

Based on this study, RF heating can be combined with other technologies such as freeze storage to enhance pasteurization effect on low-moisture foods. *E. faecium* can be used to validate designed RF heating protocol for pasteurization of *Salmonella*. RF heating has a great potential to be used in food industry to achieve the safety requirements while maintaining food quality in low-moisture foods.