Dielectric Properties and Heating Properties of Aniseed Powder with Radio Frequency Heating

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Abstract-Microbial contamination is currently a major safety issue in 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, which require long time heating, leading to quality deterioration with significant loss of colour and flavour. Radio Frequency (RF) heating has great potential in pasteurization of low moisture foods and offers the possibility to rapidly inactivate microorganisms while maintaining the food quality. Aniseed powder as one of typical low moisture foods possesses significant risk of microbial contamination. This study was to develop RF heating process to inactivate microorganisms in aniseed powder. The dielectric properties (i.e. dielectric constant (ε') and dielectric loss factor (ε'') of aniseed powder, the influences of processing conditions on heating properties and the inactivation of microorganisms in aniseed powder were all studied in this research. Results showed that ε' and ε'' of aniseed powder decreased with an increase in frequency from 6.78 MHz to 47.46 MHz but then fluctuated between 300 and 2745 MHz. The overall change of ε' and ε'' ultimately exhibited a downward trend. Under a constant frequency treatment, ε' and ε'' of aniseed powder increased with an increase in moisture content, whereas the penetration depth decreased. Particle size showed no significant effects on the dielectric properties, heating rate and penetration depth. With an increase in RF processing time, the heating rate of the sample increased and adding the rolling-over operation step helped to improve the uniformity of RF heating. After processing for 100 s, the temperature of aniseed powder could increase to 60 °C, and

the number of microorganism in aniseed powder was reduced by 6.18 logs when time extended to 150 s.

Index Terms—aniseed powder, radio frequency, moisture content, dielectric properties, penetration depth, heating rate, heating uniformity, microbial inactivation

I. INTRODUCTION

As a low-moisture food, spice has generally been considered microbiologically safe and can inhibit the growth of microorganisms because of its low water activity (a_w) . However, spice is apt to be contaminated by microorganisms at all stages through the production and supply chain which including growing, harvesting, processing, packing, handling and transportation. Some pathogens that exhibit relatively high heat tolerance and resistance to environment would land on the spices and remain viable for a certain period of time and become the uppermost source of foodborne outbreak [1]-[6]. Escherichia coli and Salmonella, which have been identified as major pathogens of spices and also have been associated with a number of outbreaks [7]-[10]. Therefore, pasteurization is an important processing step in the processing of low-moisture foods. As an important spice and a typical low-moisture food, star anise (Illicium verum, Hooker f.) is widely used to enhance the flavor of food and mainly distributed in East Asia, Southeast Asia and the Americas, with China as the most, followed by Vietnam, Cambodia and other countries and regions [11]. However, more attention has focused on incidents such

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as adulteration, poisoning by accidental eating similar species of star anise, little attention has been paid to its microbial contamination. After harvesting, solarizing, drying and other processes, star anise is mechanically milled into aniseed powder, which is easily contaminated by foodborne pathogens.

Several methods have been widely used to inactivate foodborne pathogens. These techniques include chemical fumigation, high-temperature steam, irradiation, and microwave heating, which have been shown to act faster and more efficiently, compared with conventional heat treatment methods (pasteurization and sterilization) [12], [13]. However, a large number of problems associated with these approaches cannot be ignored. Chemical fumigation was banned by the European Union because of its adverse environmental effects [14]. Owing to the conduction modes of high-temperature steam in heat transfer, samples treated with high-temperature steam vary in flavor and color changes and show severe quality loss [15]. Irradiated foods have not been accepted by consumers because of health concerns [3]. Microwaves involve higher frequencies (915 and 2450 MHz) and shorter wavelengths, resulting in shallower penetration [16]-[18]. Thus, these approaches could be applied to inactivate pathogens without significantly reducing the quality of low-moisture food products [19], [20].

Radio frequency (RF) is a type of high-frequency alternating current electromagnetic wave with a frequency of 3 kHz to 300 MHz [21]. As a method of volumetric heating, rapid and volumetric heating favors the great potential of RF technology for pasteurization of low-moisture foods [22]-[24]. Compared with traditional methods, RF heating ensures more uniform heating and deeper penetration in solid and semi-solid low-moisture food because of the lower frequency range and longer wavelengths, particularly 13.56, 27.12 and 40.68 MHz, which provides longer wavelength and deeper penetration than those of microwaves at 915 or 2450 MHz [25]-[27]. Recently, RF has been explored for its potency in the inactivation of foodborne pathogens in low-moisture food products. RF processing of chili peppers in 35 s and black pepper in 40 s decreases the E. coli O157: H7 and Salmonella typhimurium populations in the sample below the limit of detection $(1 \log CFU/g)$ [28]-[29].

Numerous studies have reported that RF is used to process a variety of spices such as pepper and cinnamon, but few studies have reported that RF is used to process aniseed powder. And still few studies have reported the dielectric properties of aniseed powder and RF heating rate and uniformity when processing aniseed powder. Dielectric properties of food directly affect RF heating rate and uniformity. The dielectric properties are mostly influenced by physical properties such as moisture content, temperature and frequency [30]-[32]. These properties are essential for developing an effective RF pasteurization procedure for aniseed powder. Moisture content is a critical indicator of food quality, safety, and shelf life. Moisture content influences the taste, texture, weight, appearance, and shelf life of foodstuffs. Food safety and stability are extremely related to moisture content [33]. In addition to the food sector, lower moisture content is also desirable property for the oil methyl esters to be used as a fuel additive in the internal combustion engines [34], [35]. In order to more comprehensive gain insight into the feasibility of using RF to process aniseed powder, it is also necessary to consider the influences of particle size of aniseed powder on various factors. Therefore, this study aimed 1) to measure the dielectric properties, penetration depth, and heating rate of aniseed powder with different moisture contents and particles of different sizes; 2) to evaluate the heating uniformity of RF-treated aniseed powder; and 3) to evaluate the inactivation effect of RF treatment on microorganisms when aniseed powder has better heating conditions.

II. MATERIAL AND METHODS

A. Sample Preparation

Aniseed powder samples were purchased from Ruisheng Food Co., Ltd. (Xinghua, China) with initial moisture content of 6.75% (w.b.).

To study the effect of particle size, > 0.355 mm, 0.3-0.355 mm, 0.2-0.3 mm, 0.15-0.2 mm, 0.125-0.15 mm and < 0.125 mm, six ranges of particle size of aniseed powder were separated by sample sieve with different aperture.

To study the effect of moisture content, the moisture content of white pepper was adjusted by adding distillated water to the sample to reach 8.84%, 11.44%, 12.12%, 13.11%, 14.48%, 18.12%, and 24.79% (w.b.). And aniseed powder was stored in polyethylene bags for 48 h at 4 $^{\circ}$ C, and shaken twice in a day to obtain a uniform moisture distribution throughout the sample.

B. Cultivation, Inoculation, and Enumeration of Bacterial Strains

The Escherichia coli ATCC 25922 strains was obtained from the School of Food Science and Technology, Jiangnan University (Wuxi, China). Microbes from glycerol frozen stocks were allowed to thaw at room temperature and then were streaked onto Luria-Bertani (LB, Beijing Land Bridge, Beijing, China) agar to incubate in an incubator (Shanghai Sumsung Laboratory Instrument Co., Ltd, Shanghai, China) for 24 h at 37 °C. After 24 h, a loop of single E. coli colony was transferred into 150mL LB broth and cultured at 37 $\,^{\circ}$ C for 24 h. Then the culture was centrifuged at 8000 rpm (SC-3610, Anhui USTC Zonkia Scientific Instruments Co., Ltd. China) for 5 min at 4 °C, and the resulting pellets were washed twice in 150 mL of sterile 0.1% w/w buffered peptone water (BPW; Difco, Sparks, MD). The pellets were re-suspended in 3 mL sterile 0.1% buffered peptone water to obtain the suspension which was placed in a vacuum desiccator for 3 d to ensure aw below 0.10. The dried suspension was ultimately ground into powder by a mortar and pestle and sealed in a polyethylene bottle.

Aniseed powder were autoclaved for 20 min at 120 °C before inoculation to remove pre-existing microorganisms. About 0.99 g of aniseed powder and

0.01 g of dried inoculum were thoroughly mixed up and placed in a sterilized polyethylene bags and then vacuum packaged. It was supposed that the addition of E. coli had no influence on aw of the aniseed powder because the dose was too small to be influential. The total number of colonies in aniseed powder after inoculation was 8.72 logs.

The number of microorganisms before and after RF processing for aniseed powder was determined by plate count method. All samples were respectively dissolved in 9 mL of sterile saline and then homogenized for 1 min. After homogenization, 1 mL of the dissolved sample was serially diluted by using sterile test tubes containing 9 mL of sterile saline to obtain the appropriate degree of dilution. Subsequently, 1 mL of appropriate dilution was transferred to a nutrient agar medium and then enumerated after incubation at 37 $^{\circ}$ C for 24 h. The effect of inactivation was expressed by microbial lethality, as provided in (1)

Microbial Lethality =
$$lg\left(\frac{N_o}{N}\right)$$
 (1)

where No is the number of microorganisms in the initial sample (cfu/g), and N is the number of microorganisms remaining in the sample after RF treatment (cfu/g).

C. RF Heating System

The RF apparatus used in this study was developed by State Key Laboratory of Food Science and Technology and Key Laboratory of Advanced Process Control for Light Industry, Ministry of Education, Jiangnan University, Wuxi. The conditions were as follows: frequency, 27.12 MHz; power, 0-15 kW, and current debugging power, 3 kW. The RF device mainly consists of a transmission device and an RF processing cavity. The RF processing cavity is composed of 2 parallel aluminum electrode plates (800 mm \times 600 mm with a thickness of 5 mm). The output power of the system could be controlled by changing the distance of electrode plates. Samples in a plastic container between electrodes were moved on a conveyor belt during RF heating to continuous processes. simulate The real-time temperatures in the center of the samples during RF heating were measured by using a fiber optic temperature sensor (SR-C, Skyray Photoelectricity Science & Technology Co., Ltd, Fuzhou, China) with a temperature signal conditioner.

D. Determination of Dielectric Properties

The dielectric properties of foods and biological products are essential parameters in food engineering and technology and can influence the distribution of electromagnetic energy on food products, thereby affecting the energy absorption and conversion [36]-[38]. The dielectric properties involve the dielectric constant ε' and the dielectric loss factor ε'' , which represent real and imaginary components respectively and he relative complex permittivity ε can be expressed as (2) [32]

$$\varepsilon = \varepsilon' - j\varepsilon'' \tag{2}$$

where $j = \sqrt{1}$.

The dielectric properties measurement system consisted of the ENA-L RF network analyzer (E5062A, Agilent Technologies), coaxial cable, open-ended coaxial probe computer, and Dielectric Probe Kit software (85070E, Agilent, the United States). In this study, the frequency ranges of the RF band and the microwave band were 6.78-47.46 MHz and 300-2745 MHz, respectively. Specially, 13.56, 27.12 and 40.68 MHz commonly used on RF equipment in the food industry and then 915 and 2450 MHz commonly used on microwave equipment in the food industry. The dielectric properties of aniseed powder with different moisture content at 20 $^{\circ}$ C were measured. The relationship between dielectric properties of aniseed powder with different particle size and frequency (9.04 - 47.46 MHz) also was obtained. ɛ' and ε" were calculated by the Dielectric Probe Kit software.

E. Determination of Penetration Depth

The penetration depth mainly depends on the dielectric properties of samples, which can provide a guide for the thickness of the samples during RF processing and help improve the uniformity of heat distribution in samples. The penetration depth is expressed as (3) [39]

$$d_{p} = \frac{c}{2\pi f \sqrt{2\varepsilon' \left[\sqrt{1 + \left(\frac{\varepsilon''}{\varepsilon'}\right)^{2} - 1}\right]}}$$
(3)

where dp is the penetration depth (m), c is the speed of light under vacuum (m/s) with a value of 3.0×108 , and f is the frequency (Hz). According to the dielectric properties of aniseed powder at a specific frequency, the penetration depth of the electromagnetic wave in different samples were accurately obtained by using Matlab (Version 2018a).

F. RF Heating Rate Curve

Seven kinds of anise powder samples with different moisture content were obtained by following the method described in A. Appropriate amount of samples was packed into polyethylene bags then flattened to 10 mm thickness and cuboid shape by using a glass plate. Seven samples were separately treated on the condition of 40 mm spacing between electrode plates of RF. The probe of the optical fiber sensor (Skyray Technology Co., Ltd, Fuzhou, China) was inserted into the center of samples and the temperature of the sample was recorded every 5 s. The curve depicting the influence of moisture content on the heating rate of aniseed powder can be obtained. Six kinds of anise powder samples with different particle size were obtained by following the method described in A and the thickness of each sample was flattened to about 20 mm. The effect of particle size on RF heating rate of aniseed powder was finally determined.

G. Heating Uniformity

Considering the actual production process of aniseed powder, it was inconvenient to uniformly mix up samples when the sample was pre-packaged. Thus, the effect of adding a rolling-over operation on improving the RF heating uniformity in aniseed powder was only studied. Before the processing, aniseed powder was heated at constant temperature (25±1) °C for 12 h and 37 g powder was packed into a polyethylene bag and flattened to 5 mm thickness and rectangular shape by using a glass plate. A model consisting of four symmetrically stacking bags of samples (140 mm \times 100 mm \times 20 mm) was considered as a whole sample as shown in Fig. 1. During the RF processing (27.12 MHz, 5 min), the RF device was turned off every 30 s to remove and mix up sample once. After the processing completed, the surface temperature of each layer of samples was collected by using an infrared thermometer (Ti95 visible infrared thermometer, Fluke, the United States). Then the average and standard deviation of the surface temperature of each layer of samples were calculated. Samples treated by RF without any auxiliary operation were used as control group.



Figure 1. Packaged model of aniseed powder and distribution of test locations. (A, B, C, D, and E represent the sample layer)

Heating uniformity is an important indicator for measuring the quality of RF processing. Wang [40] defined the uniformity index (UI) as the ratio of the rise in the standard deviation of the sample temperatures to the rise in the average sample temperatures during RF heating. It can be calculated by using (4) [40]

$$\lambda = \frac{\Delta\sigma}{\Delta\mu} \tag{4}$$

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

H. Inactivation Dynamics Curve

With electrode plates spacing of 40 mm and sample thickness of 20 mm, the inoculated aniseed powder was treated with RF for 20, 40, 60, 80, 100, 120 and 150 s. After the RF processing completed, the aniseed samples

were immediately experienced cold treatment (-18 $^{\circ}$ C, 48 h). The heating and inactivation dynamics curves of the aniseed powder were finally obtained.

I. Data Processing and Analysis

Each experiment was run in triplicate and the variance of means was analyzed using SPSS (Version 17.0). P =0.05 was used to determine statistical significance in all tests. Matlab (Version 2018a) was used to formula editing and mathematical operations, and Origin (Version 9.0) was used for mapping, editing model equations, and curve fitting.

III. RESULTS AND DISCUSSION

A. Effect of Frequency, Moisture Content and Particle Size on Dielectric Properties of Aniseed Powder

Fig. 2 shows the variation of the dielectric constant (ε') and dielectric loss factor (ε ") of aniseed powder with frequency in the range of radio frequency (6.78~47.46 MHz) and microwave (300~2745 MHz). Overall, ɛ' and ε " of aniseed powder decreased with increasing frequency and the decreasing trend was more intuitive especially in the RF frequency range. With the frequency increased from 6.78 to 47.46 MHz, ε' and ε'' of three kinds of aniseed powder with different moisture contents (8.84%, 12.12%, 14.48%) both clearly decreased. When the moisture content of aniseed powder increases to 24.79%, ε' and ε'' significantly decreased and then gradually stabilized. In the microwave frequency range, as the frequency increased, both ε' and ε'' fluctuated greatly than in the RF frequency range and the fluctuation amplitude of ε' was larger than that of ε'' . When the microwave frequency is lower than 1000 MHz, ε' fluctuated drastically at the beginning and gradually decreased slowly. ε " decreased within the entire measurement frequency range and this trend became more pronounced under high moisture content. The frequency of RF had an evident influence on ε' of aniseed powder, but the frequency of microwave had both significant influence on ε' and ε'' . Therefore, RF frequency had a stable effect on the dielectric properties of aniseed powder.





Figure 2. Frequency-dependent dielectric properties of the aniseed powder with different moisture content. ((a) and (b) are the relationship between ε' and ε'' of aniseed powders with different moisture contents and the RF frequency range of 6.78 to 47.46 MHz; (c) and (d) are the relationship between ε' and ε'' of aniseed powders with different moisture contents and the MW frequency range of 300 to 2745 MHz)

Fig. 3 illustrates the influence of moisture content on the dielectric properties of aniseed powder. At frequency of 13.56, 27.12, 40.68, 915 and 2450 MHz, ε' and ε'' of aniseed powder increased with an increase in moisture content. When the moisture content of aniseed is below 18.12%, the dielectric properties of aniseed powder increased slowly at five frequencies. However, when moisture content is more than 18.12%, ε' increased at both five frequencies, whereas ε'' only significantly increased at RF frequencies of 13.56, 27.12 and 40.68 MHz. For example, when the moisture content of aniseed powder increases from 18.12% to 24.79% and the frequency is 27.12MHz and 2450MHz, ε " increased from 0.4725 and 0.4568 to 1.8168 and 0.7710 respectively, with an increase amplitude of 284.5% and 68.8% respectively. This indicates that the effect of moisture content on ε " of aniseed powder is more significant in the range of RF frequency than in the range of microwave frequency.



Figure 3. Relationship between dielectric properties and moisture content of aniseed powder at specific frequencies. ((a) effect of moisture content on ε' of aniseed powder; (b) effect of moisture content on ε'' of aniseed powder)

As shown in Fig. 4, when the particle size of aniseed powder is less than 0.3 mm, ε' and ε'' increased with the range of 9.04 MHz to 47.46 MHz. When the particle size of aniseed powder exceeds 0.3 mm, ɛ' decreased with the increase of frequency and the decreasing amplitude of ε' is more significant with the increase of particle size. However, ε'' exhibited a different trend and increased with increasing frequency. When at specific frequency of 13.56, 27.12 and 40.68 MHz, ε' of aniseed powder increased firstly and decreased next and then increased slowly with the decrease of particle size (Fig. 5 (a)-6 (c)). During the increase of frequency from 13.56 MHz (Fig. 5 (a)) to 27.12 MHz (Fig. 5 (b)), the increasing amplitude of ε' increased in the increasing interval and the decreasing amplitude of ε' decreased in the decreasing interval with the change of particle size. When the frequency increases to 40.68 MHz (Fig. 5 (c)), the changing tendency of ε' was more significant and when

the particle size is less than 0.125mm (No. 5), ε' appeared to decrease again. ε'' of aniseed powder exhibited a similar trend—increased firstly and decreased next and then increased slowly with the decrease in particle size at 13.56 MHz (Fig. 6(a)). However, at the frequency of 27.12 and 40.68 MHz, the trends of increasing and decreasing occurred alternately. As the frequency increases, the magnitude of increase and decrease became larger and larger (Fig. 6 (a)–6 (c)).



Figure 4. Frequency-dependent dielectric properties of aniseed powder with particles of different sizes. ((a) effect of variation in the particle size of aniseed powder on the relationship between ε ' and frequency; (b) effect of variation in the particle size of aniseed powder on the relationship between ε '' and frequency)





Figure 5. Relationship between the dielectric constant and particle size of aniseed powder at specific frequencies. (The frequencies of a, b, and c are 13.56, 27.12, and 40.68 MHz, respectively; No. 1-6 represent the following average particle sizes: 0.355, 0.3–0.355, 0.2–0.3, 0.15–0.2, 0.125–0.15, and <0.125 mm)

B. Effect of Frequency, Moisture Content and Particle Size on Penetration Depth of Electromagnetic Waves in Aniseed Powder

The penetration depths of electromagnetic waves in aniseed powder with different moisture content were listed in Table I. Based on the measured dielectric properties at 20 °C, this value was calculated according to (3). As Table I shown, when the moisture content of the sample is constant, the penetration depth of the electromagnetic wave significantly decreased with frequency increased. When the moisture content is 8.84%, the frequency increased from 40.68 MHz to 915 MHz, and the penetration depth of the electromagnetic wave in the aniseed powder abruptly decreased from 1954.08 cm to 44.5 cm. Thus, when other conditions are consistent, the penetration depth of electromagnetic wave in aniseed powder was markedly deeper in the RF frequency range than in the microwave frequency range. At a constant frequency, the penetration depth of the electromagnetic wave was significantly reduced with an increase in moisture content and the amplitude of reduction was larger in the RF frequency range than in the microwave frequency range.



Figure 6. Relationship between the dielectric loss factor and particle size of aniseed powder at specific frequencies. (The frequencies of (a), (b), and (c) are 13.56, 27.12, and 40.68 MHz, respectively; No. 1-6 represent the following average particle sizes: > 0.355, 0.3–0.355, 0.2–0.3, 0.2, 0.125–0.15, and < 0.125 mm)

TABLE I. PENETRATION DEPTHS OF ELECTROMAGNETIC WAVES IN ANISEED POWDER WITH DIFFERENT MOISTURE CONTENTS

Frequenc	<i>dp</i> / cm	Moisture content / %							
y / MHz		8.84	11.44	12.12	13.11	14.48	18.12	24.79	
13.56	d_p	12128.76	4124.58	2949.24	2690.15	2374.58	1393.71	363.40	
27.12	d_p	4140.62	1034.58	972.74	895.61	801.51	597.81	193.50	
40.68	d_p	1954.08	685.64	601.06	484.95	408.09	361.74	135.93	
915	d_p	44.50	28.71	25.89	23.80	20.30	15.09	10.82	
2450	d_p	23.08	12.57	11.74	10.69	10.09	7.07	4.89	

Table II shows the penetration depth of the electromagnetic waves under three specific RF frequencies in aniseed powder with different particle sizes at 20 °C. When the particle size of the sample is constant, the depth of penetration of electromagnetic waves significantly decreased with the increase of frequency. For example, when the particle size range of aniseed powder is 0.3-0.355 mm and the frequency increased from 13.56 MHz to 40.68 MHz, the penetration depth of the electromagnetic wave in the aniseed powder

significantly decreased from 959.68 mm to 134.25 mm. This result showed that the frequency exerted a significant effect on penetration depth. When the frequency is constant, the depth of penetration of electromagnetic waves also fluctuated with the change of particle size. Especially, when the particle size range of aniseed powder is 0.2-0.3mm, the penetration depth of electromagnetic wave in aniseed powder was higher than that of other particle sizes.

TABLE II.	PENETRATION DEPTHS OF ELECTROMAGNETIC WAVES IN ANISEED POWDER WITH DIFFERENT PARTICLE SIZE

E	Dp /cm	Particle size/mm						
Frequency / MHz		>0.355	0.3-0.355	0.2-0.3	0.15-0.2	0.125-0.15	< 0.125	
13.56	dp	1848.20	959.68	3513.79	3178.30	2538.04	2232.47	
27.12	dp	338.63	293.66	421.17	321.05	359.37	249.94	
40.68	d _p	180.90	134.25	214.34	140.06	163.37	116.45	

C. Effect of Moisture Content and Particle Size on Heating Rate

As shown in Fig. 7, with the increase in RF processing time, the temperature of aniseed powder continuously increased and when the moisture content of aniseed powder increases, the heating rate of aniseed powder also increased significantly, which might be correlated with the ε " of aniseed powder. As shown in Fig. 3 (b), the ε " of aniseed powder increased with the increase in moisture content and ε " became an important factor affecting the heating rate of RF. In addition, with other conditions remaining the same, when the sample moisture content does not exceed 12.12%, the temperature of the sample increased slowly and the temperature curve increased in linear. When the moisture content exceeds 13.11%, the heating rate of aniseed powder increased with a large increase in RF treatment. Fig. 8 shows that with the increase of RF processing time, the heating rate of aniseed powder increased continuously and this is consistent with what Fig. 7 described. However, that the particle size exerted no significant effect on the heating rate of the RF-treated aniseed powder.



Figure 7. Effect of moisture content on heating rate of aniseed powder during RF treatment.



Figure 8. Effect of particle size on heating rate of aniseed powder during RF treatment.

D. Heating Uniformity of RF Treatment Combined with Rolling Over

After sample model (140 mm×100 mm×20 mm) processed by radio frequency alone or by radio frequency combined with rolling over, the temperature distribution of each layer of aniseed powder was shown in Fig. 9. The results indicated that the temperature of the central aniseed powder were significantly higher than the temperatures of the surface and the edge of aniseed powder and also indicated the phenomenon of RF uneven heating. As shown in Figs. 9 ((a)-(e)), when no auxiliary measures are added, the central temperature of each layer was significantly higher than the temperature of the sample edge and when the central temperature of aniseed powder increases to 90 °C and higher, the temperature of the edge just rose to about 70 °C. Meanwhile, the maximum temperatures of the sample surfaces ((a) and (e)) were 77.8 $^{\circ}$ C and 76.9 $^{\circ}$ C respectively, which were significantly lower than the maximum temperatures of the central section ((b), (c), (d)). This finding is contrary to conventional heating and several subsequent reasons may cause that. Firstly, it may be attributed to the small thickness of fringe aniseed powder after aniseed powder

placed in a polyethylene bag. Secondly, when the surface of the packaged sample is heated, the polyethylene bag expanded and generated steam to absorb heat, which reduced surface heat. The heating process of aniseed powder was related to the generation of a large amount of heat when the water dipole rotated at a high speed in the dielectric field. External moisture of aniseed powder was quickly reduced during the RF processing and the large decrease of water dipole caused a significant reduction on the temperature rising rate of the surface layer of aniseed powder. Thirdly, this results may be correlated with the dielectric loss factor between the sample and the packaged bag. Fourthly, partial heat loss occurred during the transfer process of aniseed powder from RF to the visible infrared thermometer after finishing RF processing, which became one of the reasons for the significant differences between the edge temperature of aniseed powder and the central temperature of aniseed powder.





Figure 9. Temperature distribution on the surface and cross-section of each aniseed powder sample after RF treatment. ((a)–(e) represents the temperature distribution in each layer of the powder from top to bottom after separate RF treatment; (a')–(e') represent the temperature distribution in each layer of the powder from top to bottom after RF treatment combined with flipping operation, respectively. The highest temperature point has been marked in the figure)

In addition, as shown in Table III, the temperature of the surface layer ((a) \rightarrow (a'), (e) \rightarrow (e')) of aniseed powder increased by adding the rolling over operation and the temperature of the central section ((c) \rightarrow (c')) slightly increased. The temperature difference between the surface layer and the central section of aniseed powder decreased from 17.6 °C to 6.7 °C. It also found that the uniformity index value after adding the rolling over operation was lower than the heating uniformity without the rolling over operation. The results showed that the heating uniformity of aniseed powder could be improved by adding rolling over operation during the heating process of RF treatment.

TABLE III. TEMPERATURE AND HEATING UNIFORMITY INDEXES OF ANISEED POWDER AFTER RADIO FREQUENCY (RF) TREATMENT WITH ROLLING OVER

Layer	а	b	С	d	e
Only RF treatment / °C	77.8±4.2	91.7 <u>±</u> 4.7	94.5±4.8	93.5±4.7	76.9±4.1
λ_0	0.0758 ± 0.0064	0.0675 ± 0.0056	0.0662 ± 0.0052	0.0657 ± 0.0057	0.0752 ±0.0045
Layer	a´	b´	c´	d´	e´
Combination of RF treatment and rolling over / °C	85.9±4.3	86.1±4.3	90.3±4.4	86.2±4.2	83.6±4.2
λ_{I}	0.0673±0.0038	0.0671±0.0059	0.0643±0.0046	0.0654 ± 0.0061	0.0683 ±0.0034

E. Inactivation Kinetics Curve of Microorganisms in Aniseed Powder by Radio Frequency Treatment

After comparing several pre-experiments, the heating curve as Fig. 10 shown of aniseed powder was measured by selecting the combination of electrode plates spacing of 40 mm and sample thickness of 20 mm and can be aware of the temperature of aniseed powder at different times. Within 100 s, the inactivating temperature of microbial inactivation was quickly increased by adjusting the RF power and it didn't take long time for microbial inactivation. As shown in Fig. 11, the number of microorganisms decreased significantly with the increase in RF processing time. When the RF processing time is less than 60 s, the temperature of aniseed powder did not exceed 50 $\,^{\circ}$ C and the lethality of microorganisms was considerably slow and when the RF processing time continues, the temperature of aniseed powder gradually increased. With the heating rate gradually increases, the logarithm of microbial residual rate sharply dropped

when at the lethal temperature of E. coli. And when the RF processing time extends to 150s, the number of microorganisms in aniseed powder was reduced by 6.18 logs.







Figure 11. Inactivation kinetics curve of microorganisms in aniseed powder by RF heating.

IV. CONCLUSION

The dielectric constant and dielectric loss factor of aniseed powder decreased with an increase in frequency between 6.78 and 47.46 MHz. At a constant frequency, the dielectric constant and dielectric dissipation factor of aniseed powder increased with an increase in moisture content, whereas the penetration depth of the electromagnetic wave decreased gradually. In addition, particle size showed no significant effect on the dielectric properties and penetration depth of aniseed powder. With an increase in frequency, the penetration depth of the electromagnetic wave in aniseed powder was reduced significantly and the penetration depth of the electromagnetic wave was noticeably greater than that of the microwave under the RF treatment. At a constant frequency, with an increase in the moisture content of aniseed powder, the penetration depth of the electromagnetic wave markedly decreased. With increased RF processing time, the heating rate of the sample also increased and the significant effect of moisture content was determined. The particle size also exhibited no significant effect on the heating rate of aniseed powder. When the sample temperature fell below 50 $^{\circ}$ C, the lethality of microorganisms was determined to be considerably low. As RF processing continued, both the temperature and heating rate of the sample gradually increased; when the lethal temperature of E. coli was reached, the logarithm of microbial residual rate sharply decreased.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

Hong Cao, Yurong Hua, Feng Zhang and Fang He conducted the research; Xiaojin Zhou and Wei Zhao analyzed the data; Feng Zhang, Fang He, Xiaojin Zhou and Wei Zhao wrote the paper; all authors had approved the final version.

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