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ORIGINAL RESEARCH PAPER

PHYSICAL AND MECHANICAL CHARACTERIZATION OF MICROWAVE AND ULTRASOUND-MODIFIED ZEIN-CORN STARCH EDIBLE FILMS

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Abstract

Edible films and coatings are the biodegradable layers of edible components that are used to coat the food surface or wrap the food in the form of film. In this paper, microwave, and ultrasound technologies, at different processing conditions, separately (US, 70M, 30M) or consecutively in different order (US-70M, 70M-US, US-30M, 30M-US) were applied to the film-forming solutions based on zein and corn starch to increase the compatibility between the biopolymers and to improve some of the edible film characteristics. The physical and mechanical properties and the FTIR spectra of the films were determined. The microwave followed by ultrasound treated edible films (70M-US, 30M-US) were found to be smoother and more homogeneous than the other films. Although all films were found to have hydrophilic character, application of microwave and ultrasound technologies together increased the water contact angle values of the films, providing more hydrophobic film surfaces. 70M-US and 30M-US edible films were found to have the highest tensile strength and elongation % values, respectively. The obvious deeper peaks were observed in the amide-III band of FTIR spectra for microwave followed by ultrasound treated films as an indicator of the enhanced compatibility between zein and corn starch in the film matrix.

Keywords: corn starch, edible film, microwave, ultrasound, zein

Introduction

Edible films and coatings are defined as environmentally friendly, cost-effective, innovative, and sustainable packaging alternatives for the food industry applications (Hellebois *et al.*, 2020). These biodegradable materials protect foods from physical, chemical, and biological spoilage, increasing their quality, shelf life and safety

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(Gemeda *et al.*, 2019). The main biopolymers used in edible films are polysaccharides, proteins, and lipids (Chen *et al.*, 2021). Corn starch and zein are two of the plant-based biopolymers used for edible film production. They can behave like a thermoplastic material in the presence of appropriate plasticizers, such as water, glycerol, sorbitol, oleic acid, and others (Corradini *et al.*, 2006). Corn starch has favorable properties, such as biodegradability, renewability, non-toxicity, and affordability that makes it one of the promising components of edible film formulations (Calderón-Castro *et al.*, 2018; Mohamed *et al.*, 2020). Furthermore, films and coatings obtained from corn starch are tasteless, odorless, and transparent and have good gas barrier properties. However, the weak water vapor barrier and mechanical properties limits its use individually in edible film formulations (Argüello-García *et al.*, 2014; Hassan *et al.*, 2018; Teklehaimanot *et al.*, 2020). Zein, a by-product of industrial corn starch extraction, is an amourphous biopolymer that contains hydrophobic amino acids like leucine, proline and alanine which makes it insoluble in water and provides hydrophobic character to the edible films (Yong *et al.*, 2015; Teklehaimanot *et al.*, 2020; Giteru *et al.*, 2021). However, 100% zein films are brittle (Soliman *et al.*, 2009; Teklehaimanot *et al.*, 2020). For this reason, starch and zein have been blended to improve the edible film properties, as indicated by the literature (Teklehaimanot *et al.*, 2020). However, the hydrophilic nature of the corn starch and the relatively hydrophobic nature of zein may result in miscibility problems (Teklehaimanot *et al.*, 2020). Modification of biopolymers with the help of technology-based approaches, such as microwave, ultrasound, etc., may be an alternative to solve these problems.

Ultrasound and microwave treatments separately or in combination (simultaneously or consecutively) have potential to modify biopolymers, such as corn starch, zein, cellulose, soy protein isolate, etc., and their functionality in edible film matrices (Cheng *et al.*, 2010; Wang *et al.*, 2013, 2014, 2016; Sun *et al.*, 2018). High intensity ultrasound treatment decreased the viscosity and increased the solubility of starch film-forming dispersions and provided excellent film characteristics, such as good transparency, improved water barrier properties and tensile strength (Cheng *et al.*, 2010). Wang *et al.* (2016) modified zein with microwave treatment and blended modified zein with soy protein isolate (SPI) at different concentrations to obtain edible protein films. Microwave modification provided improved mechanical properties for edible films in terms of breaking strength and fracturing distance values (Wang *et al.*, 2016). Wang *et al.* (2013) combined ultrasound and microwave treatments to modify microcrystalline wheat-bran cellulose (MWC) and prepared edible films containing different ratios of soy protein isolate (SPI) and modified MWC (MMWC). SPI/MMWC edible films were found to have more homogeneous and compact structure with smoother surface morphology compared to SPI/WC and SPI/MWC films (Wang *et al.*, 2013). In another study by Wang *et al.* (2014), it was stated that ultrasonic/microwave assisted treatment at 500W for 15 min. improved the miscibility, provided more dense structure, and increased the water contact angle values of soybean protein isolate-based oleic acid/stearic acid blend edible films.

Furthermore, use of sequential combination of microwave and ultrasound treatments has been investigated by Prajapat and Gogate (2015) as a novel approach to modify guar gum solutions. They stated that order of application had changed the extent of depolymerization of guar gum solutions and microwave followed by ultrasound treatment was more effective to depolymerize guar gum solutions as compared to the ultrasound followed by microwave treatment (Prajapat and Gogate, 2015).

Therefore, modification of zein- corn starch polymer solutions with ultrasound or microwave treatments separately or consecutively in different order may be an alternative approach to solve miscibility problems between corn starch and zein and to increase the homogeneity and to improve the performance of zein-corn starch edible films.

In this study, microwave and ultrasound technologies were applied to the zein-corn starch film solutions separately and consecutively in different order to increase the compatibility between biopolymers and to obtain homogeneous film matrices. In order to evaluate the effects of applied treatments on edible film characteristics, physical and mechanical characterization and FTIR analyses had been performed.

Materials and methods

Materials

Zein (CAS: 9010-66-6, Acros Organics Code: 179310025), corn starch (CAS: 9005-84-9, Tekkim Code: TK.920086.01002), acetic acid (CAS: 64-19-7, Isolab Code: 901.013.2500), and glycerol (CAS: 56-81-5, Sigma Code: G6279) were purchased from Acros Organics (Belgium), Tekkim Kimya (Turkey), Isolab (Germany) and Sigma-Aldrich (USA), respectively.

Preparation of control (technology not applied) edible films

A quantity of 15 g of zein was added to 105 g of acetic acid and stirred at room temperature for 5 min on a magnetic stirrer. Then, 5 g of glycerol was added into it and stirred for 1 more h at room temperature. 6 g of corn starch and 1.2 g of glycerol were added to 100 g of distilled water and mixed in a magnetic stirrer for 30 min until the solution temperature reached 60°C (Modified from Chen *et al.*, 2019). The obtained starch solution was added to the zein solution and mixed for 5 min at room temperature and the control film solution (technology not applied) was obtained. Temperature of polymer solutions was monitored during processing with the help of fiber optic thermocouple (FTI-10 Fiber Optic Conditioner, Model: FOT-L-LT-C1-F1-M2-R1-ST, Range: -40 - 300 °C, Canada). 30 ml of control edible film solutions were poured into 20 cm glass Petri dish and kept at room temperature for 48 h to dry. At the end of this period, control films were separated from the Petri dishes.

Preparation of microwave-applied edible films

In the polymer modification with microwaves (M), only the microwave heating option of the infrared-microwave combination oven (Advantium oven™, General Electrics, USA) was used.

The control film solution (220 ml) was placed on the turntable of the infrared-microwave combination oven and different powers (30% (30M) and 70% (70M)) and processing times were adjusted in the microwave device. Microwave application conditions, which were determined according to the preliminary experiments, were given in Table 1.

Table 1. Nomenclature used for zein-corn starch edible film solutions modified under different process conditions.

Treatments	Nomenclature of treatments	Process power		Process Time	
		US	M	US	M
Control (Technology not applied)	C	-	-	-	-
Ultrasound applied	US	100%	-	37 min	-
Microwave applied at 70% power	70M	-	70%	-	1 min 50 s
Microwave applied at 30% power	30M	-	30%	-	5 min
Ultrasound followed by microwave applied at 70% power	US-70M	100%	70%	20 min	50 s
Microwave applied at 70% power followed by ultrasound	70M-US	100%	70%	50 s	20 min
Ultrasound followed by microwave applied at 30% power	US-30M	100%	30%	20 min	1 min 50 s
Microwave applied at 30% power followed by ultrasound	30M-US	100%	30%	1 min 50 s	20 min

US- ultrasound; M- microwave

A fiber optic temperature sensor (FTI-10 Fiber Optic Conditioner, Model: FOT-L-LT-C1-F1-M2-R1-ST, Range: -40 - 300 °C, Canada) was used to measure polymer solution temperatures during microwave operation. Initial and final temperatures of edible film solutions, linear regression equations and model rate constants obtained from time-temperature graphs were given in Table 2. 30 ml of microwave applied edible film solutions were poured into 20 cm glass Petri dish and the same procedure given in "*Preparation of control edible films*" section was followed to obtain edible films.

Preparation of ultrasound-applied edible films

Ultrasonication device (JY92-IIDN, 20-25 KHz, Horn:6, 900W, Ningbo Scientz Biotechnology Co., Ningbo, China) was used for polymer modification with ultrasound heating. Throughout the studies, ultrasound (US) was performed at 100% power, with pulses, working for 60 s followed by 10 s stops. The final film solution

was placed in the ultrasound device and ultrasound was applied under the conditions determined according to the preliminary experiments given in Table 1. During processing, the temperature of polymer solutions was monitored with a in-device thermometer of ultrasonication device (JY92-IIDN, Ningbo Scientz Biotechnology Co., Ningbo, China). Initial and final temperatures of edible film solutions, linear regression equations and model rate constants obtained from time-temperature graphs were given in Table 2. 30 ml of ultrasound applied edible film solutions were poured into 20 cm glass Petri dish and the edible films were obtained according to the procedure given in "*Preparation of control edible films*" section.

Table 2. Initial and final temperatures, regression equations and model rate constants of edible film solutions modified under different process conditions.

Treatments		Initial Solution Temperature (°C)	Final Solution Temperature (°C)	Regression equations and model rate constants after first treatment	Regression equations and model rate constants after second treatment
First	Second				
US	-	30.0 ± 2.0	81.5 ± 2.0	T = 1.38t+35.37 R^2= 0.95	-
70M	-		84.5 ± 2.0	T = 30.79t+28.90 R^2= 0.99	-
30M	-		85.0 ± 2.0	T = 10.84t+32.28 R^2= 0.99	-
US	70M		86.0 ± 3.0	T = 1.80t+28.28 R^2= 0.99	T = 24.85t+65.38 R^2= 1.00
70M	US		71.7 ± 4.0	T= 29.40t+28.83 R^2= 0.96	T = 1.66t+39.75 R^2= 0.99
US	30M		84.9 ± 3.0	T = 1.97t+30.30 R^2= 1.00	T = 8.13t+70.02 R^2= 1.00
30M	US		72.8 ± 4.0	T = 10.89t+29.40 R^2= 0.99	T = 1.62t+41.70 R^2= 0.98

T: solution temperature (°C), t: process time (min), R^2: coefficient of determination

Preparation of microwave and ultrasound-applied edible films

The control film solution was placed in the ultrasound and the microwave devices consecutively in different order and the technologies were applied under the conditions determined according to the preliminary experiments given in Table 1. Initial and final temperatures of edible film solutions, linear regression equations and model rate constants obtained from time-temperature graphs were given in Table 2. 30 ml of combined technology applied edible film solutions were poured into 20 cm glass Petri dish and the same procedure given in "*Preparation of control edible films*" section was followed to obtain edible films.

Film thickness

The thickness of the films was measured with the Peacock Model H comparator (Japan) from 5 different points of each 3 films, then the values were averaged.

Film color

The color of the films was determined using the Konica Minolta CR-400 chroma meter (Japan). CIE L*, a*, b* color scale was used. The color values of the resulting films were measured after they were placed on the white plate of the device. Measurements were made from at least 5 different points of each 3 films and the average of the values was recorded. L_0^*, a_0^*, b_0^* values (L_0^*: 100, a_0^*: -0.34, b_0^*: 3.65) of the white plate of the device were used to calculate the color change (ΔE).

$$\Delta E=\sqrt{(L^*-L_0^*)^2+(a^*-a_0^*)^2+(b^*-b_0^*)^2} \quad (1)$$

Water contact angle (WCA)

The water contact angles of the films were tested with the Terra Lab Attension (USA) device, using distilled water. Measurements were made from at least 3 different points of each 3 films and the values were averaged.

Mechanical properties

The films were stored in a hermetically sealed box for 24 h at room conditions (23±2°C, 60±5% relative humidity) prior to the tensile test. Then film samples were cut into 2 cm wide and 8 cm long pieces. They were tested with an Instron (Model 3345, USA) tensile tester at a tensile speed of 10 mm/min according to the ASTM D 882 standard (ASTM, 2018). Totally 12 samples were tested from each film and the values were averaged.

Fourier transform infrared spectroscopy (FTIR)

A FTIR (Perkin Elmer Spectrum 100, USA) device containing a diamond ATR crystal scanning in the range of 4000-650 cm^{-1} was used to characterize the chemical groups in the film content. The test was repeated twice for each film sample and the values were averaged.

Statistical analysis

The analysis of variance (One-way ANOVA) and Tukey' s multiple range tests (MINITAB, version 16) were performed to determine the significant differences in edible film properties ($p<0.05$).

Results and discussion

The photos of edible films were given in Figure 1.

Figure 1. Photos of zein-corn starch edible films.

It was observed that the appearance and spreadability of control edible films was not homogeneous. The accumulation of control film solution at the center of the film, which can be related to the lack of integrity and miscibility, caused darker color at the center.

According to Figure 1, US edible films had aggregates which could be related to the non-homogeneous dispersion of the components in the polymer matrix under the applied process conditions. No aggregates were observed for 30M, 70M, 30M-US, US-30M, 70M-US and US-70M edible films, resulting in homogeneous appearance.

Moreover, it was observed that the films obtained by microwave followed by ultrasound treatment (70M-US and 30M-US) were smoother and more homogeneous than those obtained by ultrasound followed by microwave treatments (US-70M and US-30M). The application of microwave treatment in the first order provided more structural integrity in the polymer matrix. Since the action mechanisms of ultrasound and microwave technologies on biopolymers were different, their application order had changed the properties of biopolymers and films obtained from those modified polymers. Prajapat and Gogate (2015) stated that when ultrasound and microwave technologies were applied to guar gum solutions in different order, depolymerization degree of guar gum solution had changed, and microwave followed by ultrasound treatment was found to be more effective than the ultrasound followed by microwave treatment in depolymerizing guar gum.

Film thickness and color

Since the thickness of edible films and coatings greater than 0.3 mm limits their use (Pavlath and Orts, 2009; Díaz-Montes and Castro-Muñoz, 2021), researchers try to keep the thickness of the films below this value. In this study, edible films could be successfully produced with a thickness of 0.102-0.125 mm by wet process. The thickness data was shown in Table 3.

Considering the color change values of the edible films, it was found that the color change values of the control film was the highest, whereas 70M-US and 30M-US applied films were the lowest, resulting in the darkest and the lightest colors, respectively (Figure 1 and Table 3). The color values of rest of the films were not statistically different from each other ($p>0.05$) (Table 3). Film photos also supported that result (Figure 1).

Water contact angle (WCA)

Considering the water contact angle values, although it was observed that all the films had hydrophilic ($\theta<90°$) character (Table 3), ultrasound and microwave treatments, applied separately (except 70M) or consecutively, increased the water contact angle values of the edible films significantly ($p<0.05$), i.e., decreased the surface hydrophilicity. Maintenance of structural integrity and continuity in the polymer matrix and improving polymer-polymer interactions through ultrasound and microwave treatments may have resulted in more hydrophobic film surfaces. Wang *et al.* (2014) reported that ultrasound/microwave assisted treatment (at 500W for 15 min) increased the water contact angle values of soybean protein isolate (SPI) edible films containing different ratios of oleic acid/stearic acid. Improving the integration

of stearic acid into the SPI-lipid matrix with the help of ultrasound/microwave assisted treatment had been associated with that result.

Table 3. Physical and mechanical properties of zein-corn starch edible films.

Application	Thickness (mm)	ΔE	Water Contact Angle (°)	Tensile Strength (MPa)	Elongation Values (%)
C	0.107±0.001^{cd}	27.92±0.30^{a}	44.48±0.76^{c}	7.39±0.14^{b}	2.42±0.03^{c}
US	0.103± 0.002^{d}	26.23±0.55^{ab}	50.59±0.69^{ab}	5.61±0.10^{c}	3.19±0.19^{ab}
70M	0.117±0.002^{ab}	25.84±1.17^{ab}	43.50±0.50^{c}	5.14±0.14^{c}	1.56±0.06^{e}
30M	0.112±0.001^{bc}	24.99±0.58^{ab}	49.40±0.20^{ab}	4.94±0.16^{c}	2.38±0.11^{c}
US-70M	0.115±0.002^{ab}	26.26±0.32^{ab}	48.84±0.71^{b}	4.96±0.15^{c}	2.00±0.07^{d}
70M-US	0.102±0.002^{d}	23.70±0.52^{b}	52.94±0.93^{a}	9.41±0.26^{a}	2.90±0.12^{b}
US-30M	0.125±0.001^{a}	24.74±0.39^{ab}	50.15±0.61^{ab}	3.99±0.14^{d}	2.30±0.03^{cd}
30M-US	0.108±0.002^{cd}	23.81±0.64^{b}	50.39±0.11^{ab}	7.31±0.22^{b}	3.32±0.06^{a}

All values shown are means ± standard error. Data marked by different superscript letters in a column indicate significant difference ($p<0.05$).

Mechanical properties

Tensile strength and elongation values are important textural parameters for films that can reflect the internal structure and intermolecular forces and give idea about capability of films to maintain their integrity and withstand external stress (Sun *et al.*, 2018; Zhang *et al.*, 2019). Ultrasound and microwave treatments obviously affected the mechanical properties of the edible films (Table 3).

The effects of ultrasound and microwave treatments on the tensile strength of the films were depicted in Table 3. The 70M-US applied films had the highest tensile strength values as an indicator of compact and strong structure (Table 3). That result can be explained as high power microwave application in the first order easily broke the intra- and inter-chain bonds in a short time with the help of rapid heating and favored the formation of new bonds by increased zein-corn starch interactions. The high model constants related to the temperature rise in solutions modified by 70M-US can be seen in Table 2. High values for the slob of the linear regression equations represent rapid heating and rapid temperature rise in film solutions. Furthermore, the tensile strength values of the 30M-US applied films were found to be not significantly different ($p>0.05$) compared to the control film.

The tensile strength values of US, 30M, 70M, US-30M, US-70M films were found to be statistically similar ($p> 0.05$) and lower than that of the control film (Table 3). The different temperature profiles (Table 2) obtained during heating of polymer solutions under different process conditions may change the interactions between

zein and corn starch, which affects the integrity and the continuity of the polymer matrix.

When the application sequence of the technologies was compared, the application of microwave followed by ultrasound caused a significant increase in the tensile strength values of the films ($p<0.05$) (Table 3). In combined treatments, application of microwave treatment firstly and at high power (70M-US), increased the compatibility between zein and corn starch and ensured the polymer matrix integrity and continuity, resulting in the most durable films. The results of the FTIR analyses also support that result (Figures 2c,d,e,f). Higher absorption peaks of amide-I, amide-II and amide-III had been observed in FTIR spectra of 70M-US films. It can be concluded that in combined treatments, the application of microwave treatment first than US, is more effective in breaking the intra-chain and inter-chain bonds, due to its ability of rapid heating (Table 2), than the application of ultrasound treatment in first, followed by the microwave treatment.

Table 3 demonstrates the effects of ultrasound and microwave treatments on elongation % values of the films. The elongation % values of US, 70M-US and 30M-US applied films were found to be higher than the elongation % values of the control film (Table 3). The increase in microwave power decreased the elongation % values of the films. The lowest elongation % value was found in 70M applied films. Rapid heating provided by high microwave power might have reduced the polymer chain flexibility by breaking the interchain bonds and disrupting the structural continuity. Sun *et al.* (2018) reported that high ultrasound and microwave application reduced the elongation values of corn starch-cellulose films. On the other hand, when the sequence of the treatments was compared, films obtained by applying microwave followed by ultrasound ensured significant increase in the elongation % values of the films ($p<0.05$). 70M-US and 30M-US treatments had been more successful to provide homogeneous distribution of the components in the polymer matrix, better placement of the plasticizer between the polymer chains, and to increase chain flexibility.

Fourier transform infrared spectroscopy (FTIR)

The vibration type and related wavenumbers of all the edible films were shown in Figure 2a. The absorption peak in the range of 3700-3000 cm^{-1}, was due to the stretching vibration of free and bound O-H and N-H groups. Amide I (C=O stretching/hydrogen bonding coupled with COO-), amide II (bending vibration of N-H and stretching vibrations of C-N) and amide III (vibrations in plane of C-N and N-H groups of bound amide or vibrations of CH_2 groups) absorption bands lie between wavenumbers of 1720–1590 cm^{-1},1480–1590 cm^{-1} and 1240 cm^{-1}, respectively (Chen *et al.*, 2019). The (C=O) peak in the 1700 cm^{-1} region is commonly used to examine the secondary structure of the polypeptides, and proteins (Wang *et al.*, 2016; Sun *et al.*, 2018; Chen *et al.*, 2019). The characteristic peaks at 1200-1000 cm^{-1} were connected to the C-O stretching of starch (Chen *et al.*, 2019).

Considering the FTIR spectra of US, 70M and 30M treated films, it was seen that the amide-I, amide-II and amide-III absorption peaks were deeper for 70M films than the others (Figure 2b).

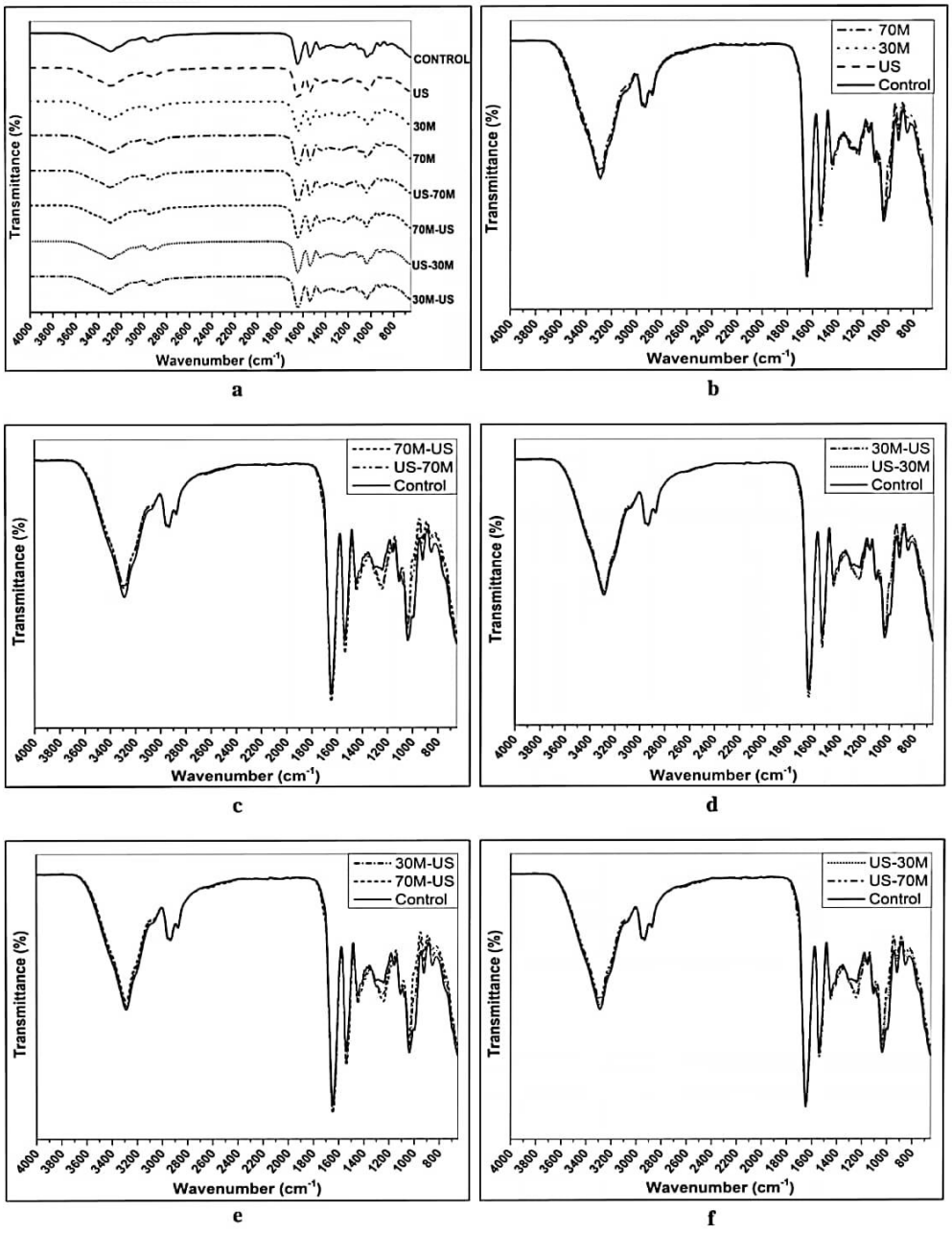

Figure 2. a) FTIR spectra of control and modified films b) FTIR spectra of C, US, 30M and 70M edible films c) FTIR spectra of C, 70M-US and US-70M edible films d) FTIR spectra of C, 30M-US and US-30M edible films e) FTIR spectra of C, 30M-US and 70M-US edible films f) FTIR spectra of C, US-30M and US-70M edible films.

When the combined treatments were compared, it was seen that amide-I, amide-II and amide-III absorption peaks were deeper in microwave followed by ultrasound

treatments and high-power microwave treatment (70M-US) gave stronger peaks (Figures 2c,d,e,f). The peaks in the amide-I, amide-II and amide-III bands observed for combined treatments, were found to be stronger than the control film, indicating the interaction between functional groups of polymers, zein and corn starch. Furthermore, the obvious deeper peaks observed in the amide-III (vibrations in plane of C-N and N-H groups of bound amide or vibrations of CH_2 groups) band for microwave followed by ultrasound treated films (70M-US and 30M-US) had been related to the enhanced compatibility between zein and corn starch in the film matrix (Chen *et al.*, 2019).

Conclusions

In this study, zein-corn starch edible films were successfully obtained from microwave and/or ultrasound modified film solutions by casting method. The treatment type, treatment conditions (power, time) and order of ultrasound and microwave treatments changed the physical and mechanical properties and the FTIR spectra of the edible films obtained from modified film solutions. The microwave followed by ultrasound treatment provided more structural integrity in the polymer matrix and improved the homogeneity, tensile strength, and surface hydrophobicity values of the edible films. 70M-US edible films were found to have the highest tensile strength and water contact angle values. According to the FTIR results, it can be concluded that the sequence of microwave followed by ultrasound treatment improved the compatibility between zein and corn starch in the polymer film matrix.

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