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Improvement of heat-sealing properties of composite chitosan films via the use of gelatin and green seaweed blends in film-forming solutions

Sarinya Prateepchanachai*

[1]Department of Industrial Management Engineering, Faculty of Industrial Technology, Rajabhat Rajanagarindra University Chachoengsao 24000

* Corresponding author.

E-mail: sarinya.ptcc@gmail.com; Telephone: 0 8682 07422

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Abstract

Gelatin and green seaweed were used to prepare composite chitosan-based films with improved heat sealing strength. Fourier transform infrared spectroscopy and Differential scanning colorimetry technique were analyzed; the results were used to explain the sealing properties data. Color measurements are also examined. As a result, combined addition of gelatin and green seaweed to treat the film-forming solution led to increased heat seal strength and decreased glass transition temperature, resulting in improved film heat sealing properties. The addition of gelatin and green seaweed resulted in enhanced electrostatic interactions between chitosan chains and improved sealability of the films. The film with the optimum sealing properties and glass transition temperature were obtained from a solution with 10% (w/v) gelatin and 1% (v/v) green seaweed. Films with addition of both gelatin and green seaweed exhibited less lightness and less greenness, but high yellowness, while the total different color of chitosan film was the highest values.

Keywords

gelatin; green seaweed; heat seal strength; composite film

1. Introduction

Since commercial plastic packaging or synthetic plastic are known to be associated with both health and environmental issues, development of biopolymer-based packaging, especially packaging films have attracted significant attention as an alternative to non-biodegradable plastic packaging due to many advantages of production for various applications. Among any kind of biopolymer-based widely used to prepare films, chitosan which is polysaccharide biopolymer-based, has been developed from natural resources and it is the most important material due to non-toxicity, biodegradability and a good film-forming [1]. Although it has a good potential, chitosan films are not thermoplastic and degrade before the melting temperature. Therefore, it cannot be processed by industrial methods such as thermoplastic synthetic polymers and still it cannot be sealed by heat [2]. Therefore, the author would like to increase the sealing ability of chitosan films. Some proposed applications of such films include pouches or sachets to package dry ingredients and ingredient delivery systems to deliver pre-measured ingredients during processing to prevent human error in weighing and handling, sealability of the material and the formation of an adequate seal are important.

To overcome these limitations for excellent sealability of a biopolymer, several schemes have been made by mixing films with an appropriate proportion of copolymer into the polymer-based. Since gelatin is a common copolymer that is added to support the sealability of many base polymers. Gelatin promotes molecular interdiffusion during heat sealed, which stabilizes hydrogen and covalent bonds within the base polymer. As a result, the film has the ability to seal the film has increased. In addition, gelatin also helps form thermo-reversible gel, which in turn assists in formation of sealed joints [3]. Prateepchanachai et al. [4], for example, studied the combined effects of glycerol and gelatin to modify the sealability of chitosan films. The film was known to be more sealable due to increased molecular interdiffusion via hydrogen bonding between the chains that exhibited more melting and hence it can be improved sealing behavior. The film was prepared from a solution with 25% (w/w) glycerol and 10% (w/v) gelatin has the best heat sealing properties compared with only a chitosan solution. Liu et al. [5], studied the addition of gelatin to soluble soybean polysaccharide and also the heat sealing properties of the resulting films. Fourier-transform infrared (FTIR) spectroscopy technique showed that the addition of gelatin affected significant changes in the absorption band at 3,305 cm^{-1} (hydroxyl groups); the bar is an angularity and is shifted to a lower wavelength. The peak of the gelatin amide I, II and III are also shifted from at 1,013 cm^{-1} to higher wavelength at 1,036 cm^{-1} due to interaction between gelatin and soluble soybean polysaccharide as a result of the hydrogen bonds being tightly interfaced. The thermal transition characteristic of the films, as shown by differential scanning calorimetry (DSC) technique, appeared to higher glass transition temperature (T_g), which corresponded to the enhanced thermal stability and increased sealability of the films.

Another interesting option that can be used to achieve the above-mentioned goal is the addition of the phenolic compounds from plant extracts to allow uniform cross-linking of gelatin into the polymer chains. In order to facilitate controlling the functional properties of the composite films. Recently, seaweed is a potential renewable resource in the marine environment, which has received much interest and attention in applications related to energy, food, and tissue engineering due to the highly utilized and inexpensive source of polysaccharide [6]. About 25-30,000 species of seaweed have been identified and are classified into three groups based on pigmentation such as namely brown algae (Phaeophyta), green algae (Chlorophyta) and red algae (Rhodophyta). Seaweed contains water 80-90 percent and their dry weight has 50 percent carbohydrate, 1-3 percent dry weight lipids and 7-8 percent dry weight minerals. The protein content was highly variable in the range of 10-47 percent with a high proportion of essential amino acids [7]. Total global seaweed production of the world in 2004 was greater than 15 million metric tons. However, seaweed in Thailand are still underutilized. Only a small portion has been used as food, animal feed, and for the production of hydrocolloids. Some researchers reported that improving properties of the functional group of gelatin molecules by the addition of seaweed could be a novel cross-linker in gelatin molecules for film preparation and can be used to achieve that purpose [8]. However, the functional properties modification of the gelatin nevertheless depends on the seaweed concentration [9]. Rattaya et al. [8], studied the use of seaweed in combination with gelatin based-film on the thermal property of the resulting films and found that addition of seaweed at 6% (w/v) will be increased transition temperature and the extent of covalent cross-linking between the molecular chains by phenolic compound in seaweed, resulting in improved thermal stability. Although some studies devoted to the investigation of the combined effects of gelatin with seaweed on thermal property of biopolymer, until now no studies have been attempted to produce and test chitosan-based composite films, gelatin and seaweed along with the use of drying of the resulting film-forming solutions.

This objective of this work is evaluating the combined effect of gelatin and seaweeds to prepare a composite chitosan film-forming solution on the properties of the resulting films. Physical and heat sealing properties of the films were determined. Fourier transform infrared spectroscopy (FTIR) and thermal properties by Differential scanning calorimetry (DSC) of the films and film color values were examined and these results were taken to explain the mechanisms leading to heat sealing properties of the films.

2. Materials and methods

2.1 Materials

Chitosan powder with a molecular weight of 900 kDa and degree of deacetylation of 90.2% was purchased from S.K. Profishery Co., Ltd. (Samut Sakhon, Thailand). Analytical grade glacial acetic acid was purchased from Merck (Darmstadt, Germany). Porcine gelatin powder and green seaweed extract were purchased from Ruam Chemical 1986 Co., Ltd. (Bangkok, Thailand).

2.2 Preparation of film-forming solutions

A chitosan film-forming solution was prepared based on the methods of Prateepchanachai et al. [4] with some modification as shown in Figure 1. Firstly, 1.5% (w/v) chitosan powder was dissolved in 1% (v/v) acetic acid under stirring continued at 700 rpm at room temperature for 6 hour by using a magnetic stirrer (Framo Gerätetechnil, M21/1, Eisenbach, Germany). In our previous study, Prateepchanachai et al. [4] studied the effect of adding gelatin solutions at various concentrations in the range of 0-50% (w/v) on the sealing properties of the chitosan films. The concentration of 5% (w/v) did not lead to any improvement in the sealing strength of the films prepared only from a chitosan solution. On the other hand, the gelatin concentrations of higher than 10% (w/v) are affected in agglomeration of gelatin and the films with higher thickness than the desired value of 15 μm. In this work, we therefore decided to compare the use of gelatin solution only at 10% (w/v).

1 g of gelatin powder was added to 10 ml of deionized water and then the mixture was heated at 70 ^{o}C for 30 min. The gelatin solution was added at 10% (w/v) to chitosan film-forming solution. Green seaweed was then added at either 0, 1, 5, 10 or 15% (v/v). The solution was stirred at 500 rpm at room temperature for another 1 hour and centrifuged at 19,414×*g* for 15 min to remove undissolved impurities to hot air drying via the use of a refrigerated centrifuge. The overall experimental procedures are summarized in Figure 1.

2.3 Drying of film-forming solutions

Each prepared film-forming solution (16 g) was poured onto an acrylic plate with the dimensions of 13 × 10 cm to cast a film. Drying was conducted at a hot air temperature of 40 °C and air velocity of 0.25 m/s until the film reached a moisture content of approximately 14% (d.b.). After that, Film was kept for at least 48 hour prior to further characterization in a desiccator containing a saturated solution of sodium chloride, which produces a relative humidity of 75% (this is the average relative humidity in Thailand) [1].

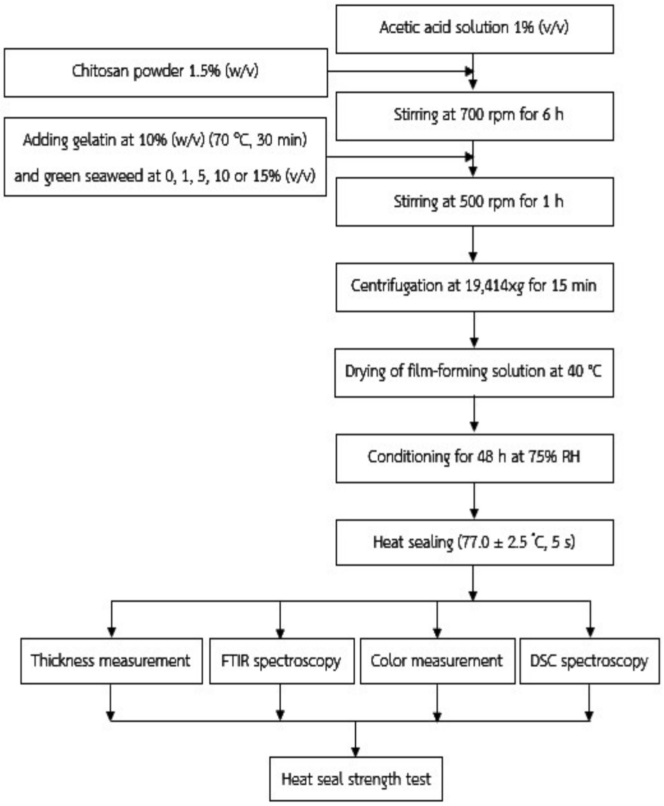

Fig. 1 Overall experimental steps of film preparation

2.4 Characterization of chitosan films

2.4.1 Determination of film thickness

The film thickness was measured by a micrometer (Mitutoyo, 102-309, Tokyo, Japan) with an accuracy of ±2 μm. Each film sample was measured at four different positions along the strip; The average thickness was used to calculate the heat seal properties of the film.

2.4.2 Determination of film moisture content

The moisture content of the film sample was determined using a standard vacuum oven method (AOAC 934.06, 1995) [10]. The film was dried in a vacuum oven (Sanyo, Gallenkamp/ OM-09980, Loughborough, UK) at 70 °C and a vacuum pressure of 900 mbar for 24 hour.

2.4.3 Seal strength determination

A film sample was cut into a 10×2.5 cm strip, then was placed on top of another 10×2.5 cm strip as shown in Figure 2. A 1 cm width of the seal area was heat-sealed at 77.0 ± 2.5 °C and a dwell time of 5 minutes using an automatic heat sealer (AUTOMATIC EXTERNAL PACKING MACHINE, DZ-400T, Bangkok, Thailand). The sealing strength of the heat-sealed film was determined by analysis similar with texture analyser according to the ASTM Standard Test Method F88 (ASTM, 2005) with some modification [11]. Each leg of the sealed film was clamped to the texture analyser and hung perpendicular to the test direction. The maximum forces required to report the cause of sealing damage as the seal strength in N/m:

$$\text{Seal strength} = \text{Peak force/Film width} \qquad (1)$$

2.4.4 Color determination

Color was measured in the CIELAB color space via the use of a Color meter (HunterLab, UltraScan® Vis, Virginia, USA). The measurement was performed in a transmittance mode by using a simulated D65 illuminant and 10° observer angle. Lightness (L^*), greenness (a^*) and yellowness (b^*) were measured. Each film sample was measured at its center and three other positions along the strip; average color values are reported.

The total color difference (ΔE^*) of a film sample, as compared with the color of the commercially available plastic films (M Wrap™), were also determined as:

$$\Delta E^* = (\Delta L^2+\Delta a^2+\Delta b^2)^{1/2} \qquad (2)$$

where $\Delta L = L^*_{plastic} - L^*_{sample}$, $\Delta a = a^*_{plastic} - a^*_{sample}$ and $\Delta b = b^*_{plastic} - b^*_{sample}$. $L^*_{plastic} = 96.93$, $a^*_{plastic} = -0.02$ and $b^*_{plastic} = -0.08$.

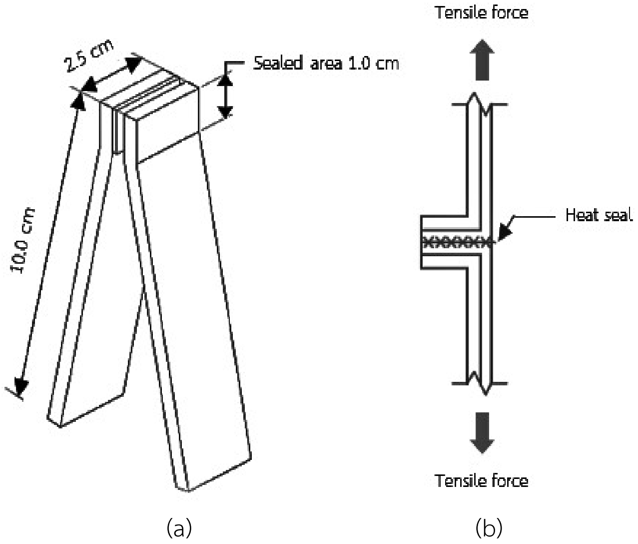

Fig. 2 Simplified illustration of test specimen dimensions and testing direction for peel test. (a) Film sample and (b) Direction of pull is 90° to the seal in peeling test

2.4.5 Determination of film FTIR Spectra

Attenuated total reflectance FTIR (ATR-FTIR) spectroscopy of a film sample was conducted with modification via a FTIR spectrometer (Nicolet is50, Thermo Scientific, Waltham, USA). FTIR spectra was analyzed at a resolution of 4 cm^{-1} and wave range of 4000 to 400 cm^{-1} with a DTGS detector (wavelength accuracy of 0.1 cm^{-1} at 1,600 cm^{-1}) [1].

2.4.6 Determination of differential scanning calorimetry (DSC)

The thermal properties of composite chitosan films were determined using a differential scanning calorimetry (DSC) (DSC 3+, Mettler Toledo, Greifensee, Switzerland). Films (1 mg) were placed in an aluminum pan with light 20 uL. The nitrogen flux was 50 mL/min. First step, the light is heated from 20 °C to 130 °C with a heating rate of 10 °C/min, followed by a 5 min isotherm. After that, films were cooled down from 130 °C to 5 °C at a rate of 20 °C/min followed by a 5 min isotherm in the nitrogen atmosphere. Finally, films were heated up to 300 °C at a rate of 10 °C/min to determine the glass transition temperatures (T_g).

2.5 Statistical analysis

All data were analyzed to the analysis of variance (ANOVA) using SPSS® software (version 17; SPSS Inc., Chicago, IL) and are presented as mean values with standard deviations. Differences between mean values were established using Duncan’s new multiple range tests at a confidence level of 95%. All experiments were performed in triplicate.

3. Results and discussion

3.1 FTIR spectra

FTIR spectroscopy was used to analyze the interactions between the functional groups of chitosan and added gelatin and film combined with gelatin and green seaweed over the wavelength range of 4,000–400 cm^{-1}; the results are shown in Figure 3. The film prepared from the chitosan solution exhibited peak absorption at 1,633, 1,530 cm^{-1}, 1,250 cm^{-1} and 1,020 cm^{-1}, which are related to amide-I band (C=O stretching), amide-II band (N-H bending), aromatic ester (C-O stretching) and amine (C-N stretching), respectively. These spectra are similar to those reported by Prateepchanachai et al. [1].

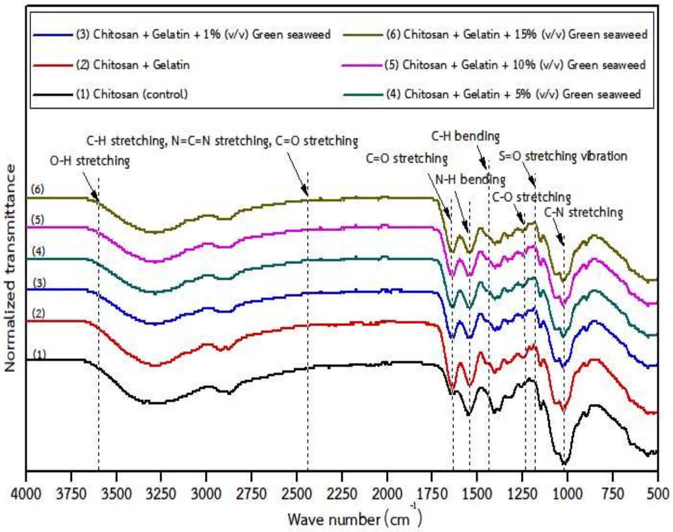

Fig. 3 FTIR spectra of film samples prepared at various concentrations of gelatin (0 and 10% w/v) and green seaweed (0 and 1% v/v)

Considering the FTIR spectrum of the composite chitosan film with gelatin, the peaks exhibited similar absorption peaks of 1,633 cm^{-1} (C=O stretching), 1,530 cm^{-1} (N-H bending), 1,250 cm^{-1} (C-O stretching) and 1,020 cm^{-1} (C-N stretching), which increased spectrum for the same functional groups. Besides, the absorption peaks were also shown in the strong peak intensity at 1,450 cm^{-1}, which is attributed to alkane (C-H bending). This might be due to the ion interactions between the positively charged chitosan and negatively charged gelatin (such as carboxyl, hydroxyl groups and amino groups), thus the appearance of absorption peaks were confirmed the perfect combination of these to polymers [12].

Combined with the addition of gelatin and green seaweed affected in an increase in the peak intensities at 4,000-3,450 cm^{-1} (O-H stretching). Therefore, an increase may be attributed to amino acids of gelatin and phenolic compounds in green seaweed interacting with the chitosan structure through hydrogen bond. A higher peaks absorption was also demonstrated in the wavenumber regions of 3,130-1,700 cm^{-1}, which are related to aldehyde (C-H stretching), carbodiimide (N=C=N stretching) and carboxylic acid (C=O stretching). These peaks represent the electrostatic interactions between peptide hydrogen bonds of gelatin and hydrogen linkages between phenolic of green seaweed increases intermolecular interactions within the chitosan chains by increasing adhesion strength between the films. This results in changes in physical properties, such as increased thickness and seal strength of the films. The last absorption peak area at 1,200 cm^{-1} (SO=stretching vibration) of sulfate esters might be an indication that interactions occur between covalent bond of green seaweed and short-chain peptides of gelatin combined with cationic groups (-OH and NH_2 groups) of chitosan. Moreover, it can be noted that the increase of green seaweed concentration from 5% (v/v) to 15% (v/v) resulted in the stronger peak intensity. The overall results from the FTIR spectrum for absorption and functional group of the films are summarized in Table 1.

3.2 Thickness and Heat seal strength

The thickness was studied and compared with the control film due to thickness characterization can affect the physical properties of edible film by composition of material mixture. As can be seen in Table 2, the result found that addition of gelatin resulted in an increase in thickness, which represent information of chemical hydrogels. However, at the same gelatin concentration, the combined addition of gelatin and green seaweed at higher concentrations (1% and 5% v/v) did not have statistically significant influence on the thickness. At a higher green seaweed concentration of 10% (v/v) and 15% (v/v) increased the film thickness. This might be due to the combined effect of electrostatic interactions and hydrogen linkages as mentioned earlier.

Table 1 Summary of results from the FTIR spectrum of the films

Film type	Absorption (cm^{-1})	Functional group
Chitosan (control)	1,633	amide-I band (C=O stretching)
	1,530	amide-II band (N-H bending)
	1,250	aromatic ester (C-O stretching)
	1,020	amine (C-N stretching)
Chitosan + Gelatin	1,633	amide-I band (C=O stretching)
	1,530	amide-II band (N-H bending)
	1,450	alkane (C-H bending)
	1,250	aromatic ester (C-O stretching)
	1,020	amine (C-N stretching)
Chitosan + Gelatin + 1% (v/v) Green seaweed	4,000-	alcohol (O-H stretching)
	3,130-1,700	aldehyde (C- H stretching), carbodiimide (N=C=N stretching) , carboxylic acid
	1,200	Sulfate esters (SO=stretching vibration)

When sealing was made at temperature substantially lower than the melting point of the sealant material, all samples failed in peeling mode failure as shown in Figure 4. This is expected because disentanglement of polymer molecules occurred which caused the bond to peel apart and probably occurred when the sealing strength of the composite film decreases.

Table 2 Thickness, heat seal strength and glass transition temperature of different films

Film type	Thickness (μm)	Heat seal strength (N/m)	Glass transition temperature (°C)
Chitosan (control)	15.0 ± 0.5^{A}	3.8 ± 1.3^{A}	108.1 ± 1.9^{D}
Chitosan + Gelatin	16.2 ± 0.3^{AB}	13.2 ± 1.0^{BC}	99.8 ± 2.5^{B}
Chitosan + Gelatin + 1% (v/v) Green seaweed	16.3 ± 1.2^{AB}	18.1 ± 1.0^{D}	95.7 ± 1.4^{A}
Chitosan + Gelatin + 5% (v/v) Green seaweed	16.5 ± 1.0^{AB}	14.7 ± 2.5^{C}	99.5 ± 1.5^{B}
Chitosan + Gelatin + 10% (v/v) Green seaweed	17.2 ± 1.2^{B}	10.3 ± 2.9^{B}	101.2 ± 1.3^{BC}
Chitosan + Gelatin + 15% (v/v) Green seaweed	17.7 ± 1.6^{B}	6.1 ± 1.4^{A}	103.3 ± 1.2^{C}

Same letters in the same column mean that the values are not significantly different at 95% confidence level (p>0.05)

Fig. 4 The composite chitosan films separated from the sealing layer

Table 2 shows the maximum achievable heat seal strength values of the film samples. The seal strength value was calculated from equation 1. The films sealed at 77.0 ± 2.5 °C and a 5 second overpressure showed peeling mode failure of the seal, demonstrates optimum seal strength. The control film proved difficult to heat seal. This is attributed to its rigid crystalline nature and the occurrence of strong inter- or intramolecular hydrogen bonding. This results in a restricted mobility of the chitosan chains during thermal degradation, resulting in the lower molecular interdiffusion at the film surface and hence the poor sealability [4].

In general, function groups such as hydroxyl (OH), carboxyl (COOH) and aldehyde (CHO) are responsible for adhesive strength of a film. Addition of gelatin to the film-forming solution increased the seal strength of the film, probably because the ionic interaction and hydrogen bond between -COOH groups of gelatin and -NH_2 group of chitosan that a reaction can lead to electrostatic interactions and intermolecular hydrogen bonds between opposite charge, leading to enhance the molecular interdiffusion between the chains [12]. This led to better melting and hence the improved sealing behavior.

Seal strength of the film prepared from the combined addition of gelatin and green seaweed led to the highest seal strength at 18.1 ± 1.0 N/m. This was possibly caused by the increased repulsion force between gelatin and green seaweed molecules, leading to increase partial denaturation and crystallization, thereby transforming themselves into a more ordered structure, reflecting the lower rigid structure of the film network. This resulted in the increase in thermal stability. Thus, the addition of green seaweed had the impact on the thermal property of resulting films.

The seal strength significantly increased from 3.8 ± 1.3 to 18.1 ± 1.0 N/m with increasing green seaweed concentrations in blend film solution from 0% to 1% (v/v) when compared with only added gelatin. However, further increasing in green seaweed concentration (above 5% v/v) did not produce a significant increase in the seal strength as shown in Figure 5. Thus, the green seaweed concentration was 1% (v/v), leading to the highest sealing strength value when compared with the control film.

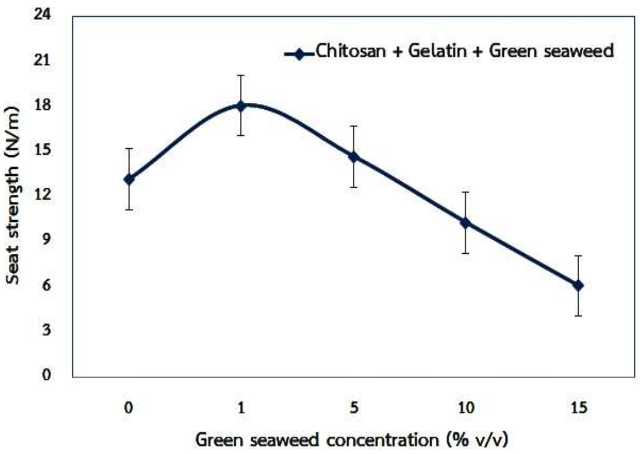

Fig. 5 Graph of heat seal strength versus platen green seaweed at various concentrations

The highest sealing strength obtained from this study was 18.1 ± 1.0 N/m for the composite chitosan film with 10% (w/v) gelatin and 1% (v/v) green seaweeds; this value is lower than heat sealed synthetic polymers (≥ 678 N/m, for LDPE). This research, seal strength is lower than the previous research of Prateepchanachai [4]. Since the author uses an industrial automatic sealing machine, which had temperature sealing and the pressure set at all. Therefore, the effect of seal strength is different. However, this composite chitosan film samples had higher seal strength than the films with sago starch film (10 N/m) [13].

3.3 Thermal properties

In general, thermal transition temperatures typically are used to determine sealing temperatures of polymers by Differential scanning calorimetry (DSC) technique. Table 2 also shows glass transition temperatures (T_g) of composite chitosan films with gelatin and green seaweed. For the control film had the highest value of 108.1 ^{o}C; followed by film with gelatin of 99.8 ± 2.5 ^{o}C. This may be due to the intramolecular interactions between N-H bending and O-H stretching that led to the formation of intramolecular hydrogen bonding, which limited the movement of the chitosan chains. This behavior was reflected on the difficult in heat seal strength. Combined addition of gelatin and green seaweed exhibited the lowest T_g value at 95.7 ± 1.4 ^{o}C than control film. The decrease in T_g value may be attributed to the plasticizing effect of the composite chitosan film with gelatin and green seaweed, which increased the free volume and intermolecular distance in the films by the increasing repulsion force between the chitosan chains. This led to a decrease in the transition temperature of the resulting film. Therefore, the composite chitosan film with gelatin and green seaweed had the different thermal transition characteristics, compared with the sole chitosan film. In the case of film prepared from the solution with added green seaweed concentration in the range of 5% (v/v) to 15% (v/v), T_g value of the film had trends to increase, probably due to more crystalline cellulose structure from a green seaweed. This led to restricted mobility of the chitosan chains during thermal degradation, resulting in the lower molecular interdiffusion at the film surface and hence difficult to heat seal.

3.4 Color

The color of commercial film (M Wrap^{TM}) and that of chitosan films after heat sealing was determined as equation 2, while the results are shown in Table 3. In terms of lightness (L^*), the films with gelatin exhibited decreased lightness when compared to the value of the control film. The lightness of the films with combined gelatin and green seaweed had the lowest lightness value of 31.94. In terms of redness (a^*), the control film had the redness value of -0.89, while films with gelatin had decreased greenness compared to the control films. The films with combined gelatin and green seaweed exhibited the lowest greenness value of -0.27. In the case of yellowness (b^*), the M Wrap^{TM} (PVC) exhibited the highest yellowness value of -1.95; this followed by the control film. The films with gelatin and the films with combined gelatin and green seaweed exhibited significantly increased yellowness by -0.06 and 0.05, respectively, compared with the values belonging to the control film. However, combined gelatin and green seaweed at all concentration was not the color value significantly different.

Table 3 Color and total color difference values of the films

Condition	Color value			ΔE^*
	L^*	a^*	b^*	
Chitosan (control)	47.48 ± 0.32^{C}	-0.89 ± 0.02^{A}	-1.25 ± 0.03^{B}	11.81 ± 0.32^{C}
Chitosan + Gelatin	35.60 ± 0.88^{B}	-0.47 ± 0.04^{B}	-0.06 ± 0.07^{C}	2.01 ± 0.21^{A}
Chitosan + Gelatin + 1% (v/v) Green seaweed	31.94 ± 0.29^{A}	-0.27 ± 0.03^{D}	0.05 ± 0.03^{D}	4.26 ± 0.27^{B}
Chitosan + Gelatin + 5% (v/v) Green seaweed	32.04 ± 0.16^{A}	-0.26 ± 0.01^{D}	0.04 ± 0.01^{D}	4.23 ± 0.03^{B}
Chitosan + Gelatin + 10% (v/v) Green	31.85 ± 0.18^{A}	-0.25 ± 0.01^{D}	0.04 ± 0.01^{D}	4.34 ± 0.16^{B}
Chitosan + Gelatin + 15% (v/v) Green	31.63 ± 0.01^{A}	-0.26 ± 0.01^{D}	0.08 ± 0.01^{D}	4.53 ± 0.01^{B}
M Wrap^{TM} (PVC)	35.70 ± 0.42^{B}	-0.40 ± 0.02^{C}	-1.95 ± 0.09^{A}	N/A

Same number, lowercase and uppercase letters in the same column mean that the values are not significantly different at 95% confidence level ($p > 0.05$) among various green seaweed concentrations.

The total color difference (ΔE^*) of the films increased significantly as a result of the highest a^* and b^* values. ΔE^* of the control films was the highest at 11.81; followed by film with combined gelatin and green seaweed. Adding only gelatin resulted in a film with the lowest ΔE^* value equal to 2.01.

4. Conclusion

Improvement of seal strength of composite chitosan-based films by the combined addition of gelatin at a concentration of 10% (w/v chitosan) and green seaweed at a concentration of 0, 1, 5, 10 or 15% (v/v) was demonstrated. The green seaweed is postulated to disturb interactions between the chitosan structure with O-H stretching, C-H stretching, N=C=N stretching, C=O stretching and SO= stretching vibration. Therefore, green seaweed leads to enhance electrostatic interactions, which is increased mobility of the chitosan chains during thermal degradation, resulting in the higher molecular interdiffusion at the film surface and hence the modified sealability. This represented a decrease in the glass transition temperature by 95.7 °C compared with those of film prepared from sole chitosan. Optimum condition was identified as 10% (w/v) of gelatin and 1 % (v/v) of green seaweed concentration that affected in the highest seal strength of 18.1 N/m. Films with combined gelatin and green seaweed exhibited significantly decreased lightness and greenness and increased yellowness, while the total different color of chitosan film was the highest value.

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Reference

[1] Prateepchanachai S, Thakhiew W, Devahastin S, Soponronnarit S. Mechanical properties improvement of chitosan films via the use of plasticizer, charge modifying agent and film solution homogenization. *Carbohydrate Polymers*. 2017; 174: 253–261.

[2] Priyadarshi R, Rhim JW. Chitosan-based biodegradable functional films for food packaging applications. *Innovative Food Science and Emerging Technologies*. 2020; 62: 1–9.

[3] Prateepchanachai S, Thakhiew W, Devahastin S, Soponronnarit S. Improvement of mechanical and heat sealing properties of chitosan films via the use of glycerol and gelatin blends in film-forming solution. *In Proceeding of the 10th TSAE International Conference (TSAE 2017).* 2017; 30–35.

[4] Prateepchanachai S, Thakhiew W, Devahastin S, Soponronnarit S. Improvement of mechanical and heat-Sealing properties of edible chitosan films via addition of gelatin and CO_2 treatment of film-forming solutions. *International Journal of Biological Macromolecules*. 2019; 131: 589–600.

[5] Liu C, Huang J, Zheng X, Liu S, Lu K, Tang K, Liu J. Heat sealable soluble soybean polysaccharide/gelatin blend edible films for food packaging applications. *Food Packaging and Shelf Life.* 2020; 24: 1–9.

[6] Khalil HPSA, Tye YY, Saurabh CK, Leh CP, Lai TK, Chong EWN, Nurul MRF, Hafiidz JM, Banerjee A, Syakir MI. Biodegradable polymer films from seaweed polysaccharides: A review on cellulose as a reinforcement material. *eXPRESS Polymer Letters*. 2017; 11(4): 244–265.

[7] Banu T, Ramani PS, Murugan A. Effect of seaweed coating on quality characteristics and shelf life of tomato (Lycopersicon esculentum mill). *Food Science and Human Wellness*. 2020; 9:183–176 .

[8] Rattaya S, Benjakul S, Prodpran T. Properties of fish skin gelatin film incorporated with seaweed extract. *Journal of Food Engineering*. 2009; 95: 157–151.

[9] Kadam SU, Pankaj SK, Tiwari BK, Cullen PJ, O' Donnell CP. Development of biopolymer-based gelatin and casein films incorporating brown seaweed Ascophyllum nodosum extract. *Food Packaging and Shelf Life.* 2015; 6: 68–74.

[10] Association of official analytical chemists (AOAC). Moisture in cheese (vacuum oven method). *Official Methods of Analysis of AOAC International*. 14th ed. Washington DC; 1995.

[11] American society for testing and materials (ASTM). Standard test method for seal strength of flexible barrier materials (F88). *Annual Book of ASTM Standards*. Philadelphia PA; 2005.

[12] Gharaie S.S. Habibi S. Nazockdast H. Fabrication and characterization of chitosan/ gelatin/ thermoplastic polyurethane blend nanofibers. *Journal of Textiles and Fibrous Materials.* 2018; 1: 1–8.

[13] Abdorreza MN, Cheng LH, Karim AA. Effects of plasticizers on thermal properties and heat sealability of sago starch films. *Food Hydrocolloids*. 2011; 25: 56–60.