International Journal of Advances in Applied Sciences (IJAAS)
Vol. 14, No. 1, March 2025, pp. 164~174
ISSN: 2252-8814, DOI: 10.11591/ijaas.v14.i1.pp164-174

Development of antioxidative edible film from red dragon fruit peel extract with the addition of CMC and soy protein isolate

Triana Lindriati, Sih Yuwanti, Asmak Afriliana, Aji Sukoco, Ivan Rivaldy Budianto, Wafiq Azizah, Umrotus Shofiyatul Fadhiyah
Department of Agricultural Product Technology, Faculty of Agricultural Technology, University of Jember, Jember, Indonesia

Article Info

Article history:

Received Feb 12, 2024
Revised Nov 7, 2024
Accepted Dec 26, 2024

Keywords:

Antioxidative edible film
Fourier transform infrared
Mechanical properties
Peroxide number
Red dragon fruit peels

ABSTRACT

The red dragon fruit peels (RDFP) have a high content of pectin and total phenolic compounds. This research studied the development of RDFP be an antioxidative edible film. The RDFP was extracted by microwave to obtain high pectin and polyphenol content, and then the red dragon fruit peel extract (RDFPE) was used as a based material. The RDFPE was added with 5% (w/v) of carboxymethyl cellulose (CMC) and 10% (w/v) of soy protein isolate (SPI) to increase their tensile strength. The result showed that RDFPE potential to develop as an antioxidative edible film. There are different effects of CMC and SPI. The addition of CMC had a positive effect on total polyphenol and antioxidant properties but SPI had a negative effect. Against the peroxide number of peanut oils, all RDFPE films can inhibit. The effect of CMC and SPI on physical and mechanical properties were increasing thickness, and tensile strength decreasing transparency, solubility, also elongation. The FTIR showed a difference in macromolecule interaction between RDFPE with CMC and SPI. The interaction between RDFPE with CMC occurred with pectin while SPI interacted both with pectin and polyphenol. Thus, macromolecule interaction affected on physical, mechanical, and antioxidative properties of RDFPE edible films, and revealed that CMC was more suitable to add to RDFPE edible film.

Corresponding Author:

Triana Lindriati
Department of Agricultural Product Technology, Faculty of Agricultural Technology, University of Jember
Kalimantan Street Number 37, Jember, 68121, Indonesia
Email: lindriatitriana@unej.ac.id

1. INTRODUCTION

Increased concern for environmental conversation and food safety has led to the development of edible films. Edible films are packaging that is safe to eat and easy to degrade. Usually, the materials of edible film are food sources such as starch [1]–[3], and protein [4]–[6], which can interfere with food security. Therefore, developing edible films from wastes of food industrial processing is a better solution.

One of the agricultural wastes that has not been widely utilized is red dragon fruit peels (RDFP). Red dragon fruit (Hylocereus polyrhizus) is a tropical fruit that is widely cultivated around Southeast Asia including Indonesia. The peels which are about 22% of the whole fruit considered a waste although has a high content of polyphenols and pectin [7]. According to several researches, the pectin content of red dragon fruit peels were 6.27% [8], 14.96-20.14% [9], 18.53% [10], 10.79% [11], and 23% [12]. The high pectin content makes red dragon fruit peels possible to develop as edible film [13]–[15]. The total phenolic content of RDFP was 7.95 (mg of GAE/gr of dm) in fresh peels and 7.84 dry peels [11], which showed high antioxidant activity [16]. Previous studies showed that the addition of dragon fruit peel extract to edible films provides antimicrobial and antioxidative properties [17]–[19].

Improving the physical and mechanical properties of pectin-based edible films can be done by interaction with other macromolecules such as carboxymethyl cellulose (CMC) or soy protein isolate (SPI). The combination of pectin-CMC has a good influence on the physical and mechanical characteristics of edible film [20]–[22]. Similarly, the interaction between pectin and SPI can increase the physical and mechanical properties [20], [23].

This study used red dragon fruit peel extract (RDFPE) as the main material of edible film, where all parts of the extract are used. This provides many advantages because it simplifies the manufacturing process, in addition to the content of dragon fruit peel extract which has a high pectin and also polyphenols. Strengthening the edible film matrix, CMC and SPI were incorporated. So, this research aimed to study how the addition of CMC and SPI affected on physical, mechanical, and antioxidative properties of edible film from RDFPE. The research also studied the interaction of macromolecules in RDFPE with CMC and SPI using Fourier transform infrared (FTIR).

2. MATERIALS AND METHOD

2.1. Materials

The red dragon fruit was obtained from a "Banyuwangi" farmer. Banyuwangi is an area in the province of East Java, Indonesia that is a farming area of red dragon fruit. The dragon fruit peels were cut into ±2 cm in width and ±0.4 cm in thickness and then were dried using a cabinet dryer at 50 °C for 24 hours. The RDFP was ground and then sifted in 30 mesh (Taylor sieve) to obtain dragon fruit peel powder. Then the powder was kept at 0 °C before processing. Other ingredients to make edible films are CMC, SPI (4.81% ±0.64 moisture content, 88.21% ±0.53 crude protein, 4.21% ±0.36 carbohydrate, 0.02% ±0.01 crude fat, and 2.75% ±0.11 ash), and glycerol, the materials were obtained from a local shop. Chemicals for other analyses were obtained from Sigma-Aldrich.

2.2. Preparation of red dragon fruit peels extract

Preparation of RDFPE used a microwave with a power of 450 watts and a duration of 5 minutes [10]. The ratio of dragon fruit peel powder to water was 1:20 (w/v). The slurry obtained was filtered using a filter cloth to separate the pulp from the filtrate. The RDFPE obtained was stored at 4 °C for a maximum of two days to be used as edible film-based material.

2.3. Preparation of dragon fruit peels extract edible films

An aqueous stock solution of 10% w/v CMC was made by dissolving CMC powder in distilled water at room temperature using a magnetic stirrer (SH-2 digital lab thermostatic hot plate magnetic stirrer) that operated at low speed for 15 minutes to obtain a clear gel. An aqueous stock solution of 10% w/v SPI was prepared by adding SPI powder in distilled water and then heated using a hot magnetic stirrer at 80 °C for 15 minutes at low speed.

Film solutions were prepared by mixing appropriate amounts of CMC and SPI solutions with 75 ml RDFPE. The mixture was heated while stirring at 80 °C. After 10 minutes, 1.5% glycerol was added then heated and stirred for 10 minutes. Edible film gel of 90 gr was poured on a 22×17 cm^2 tray and dried using a cabinet dryer (selecta) at 50 °C for 16 hours. The dried films were removed and wrapped using plastic clips and were stored in a box containing silica gel at 4 °C until analysis.

2.4. Transparency of edible films

Measurement of edible film transparency referred to ASTM D1746-15 with modifications. Transparency was observed using the color reader CR-100 (Minolta Japan). Transparency can be calculated by (1).

$$Transparency = ((a - b)/a) \times 100\% \quad (1)$$

Where a is the whiteness value of white paper and b is the whiteness value of edible film placed on white paper.

2.5. Thickness of edible films

The edible film thickness was measured based on ASTM F2251 using a thickness meter (Mitutoyo Japan) with an accuracy of 0.001 mm. The measurement was at three different positions and the thickness value was the average in units of millimeters (mm). The three positions of measurement are described in Figure 1.

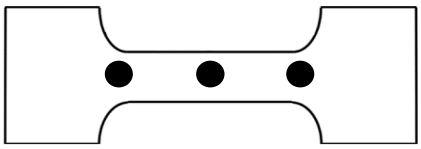

Figure 1. Description of three positions on sample specimen for thickness measurement

2.6. Solubility of edible films

Measurement of edible film solubility referred to research [24] with modifications. The film was cut 2×2 cm^2 and was put in a bottle dried at 105 °C for 24 hours and then weighed. The sample was soaked in 30 ml of distilled water in a tightly closed container and stored at 25 °C for 24 hours. Then the solution was filtered and the residue dried at 105 °C for 48 hours and then weighed. Percent solubility was calculated by comparing the weight of the soluble edible films with the initial weight. The film solubility in water was determined in triplicate for each formulation based on the percentage of dry matter solubilized in water.

2.7. Tensile strength and elongation of edible films

Tensile strength and elongation measurements were using a universal testing machine (Shimadzu) based on ASTM D882. The films were cut to 80 mm length and 10 mm width. The tensile strength was then calculated by comparing the value of the tensile force with the cross-section area of the specimen. The elongation was calculated by dividing the addition in the length of increment at the break by the initial length of the film before stretching. Measurement was replicated 5 times for each type of film.

2.8. Total polyphenol content

The assay of total polyphenol content referred to research [25] with modifications. The first step was making a standard curve of gallic acid. Then, 1.2 gr of edible film (dry basis) was extracted in 50 ml of distilled water to obtain a clear filtrate. After that, the filtrate was measured for total polyphenol content using the Folin-Cealcetau method. In this test, the absorbance of the sample solution was measured using a spectrophotometer at a wavelength of 765 nm. The total polyphenol content (mg GAE/gr edible film) of samples was calculated based on the gallic acid standard curve.

2.9. Antioxidant activity

The assay of total antioxidant content referred to research [26] with modifications. Edible film 1.2 gr was extracted in 50 ml distilled water. After that, the filtrate was tested for antioxidant activity using the 2,2-diphenyl-1-picrylhydrazyl (DPPH) method. The sample solution was measured for absorbance using a spectrophotometer with a wavelength of 517 nm. Antioxidant activity was expressed as a percentage of radical inhibition because antioxidants can inhibit the formation of free radicals, which was calculated using the (2).

$$\%\ free\ radical\ inhibition = \frac{(A_b - A_s)}{A_b} \times 100\% \quad (2)$$

Where A_s is the absorbance of the sample and A_b is the blank absorbance.

2.10. Peroxide value

Peroxide value testing referred to research [27] and SNI 01-3555-1998. Edible films were cut into 10×10 cm^2 and then soaked in 20 ml peanut oil in the petri dish. Then the petri dish is stored in an incubator at 50 °C for 14 days. Furthermore, the peroxide number of peanut oils was measured on 4, 7, 9, 11, and 14 days. Measurement of peroxide value used the iodometric titration method.

2.11. Fourier transform infrared

Fourier transform infrared (FTIR) observation referred to research [28]. At first, the sample was made in the form of infrared (IR) pellets by mixing 2 mg of the sample with 100 mg of KBr. FTIR scanning was then carried out with a range of wave numbers of 4000-600 cm^{-1}. The FTIR test was used to identify the functional groups of samples in the form of spectra as well as the position of the absorption bands expressed as wave numbers in cm^{-1} units.

2.12. Statistical analysis

Data was analyzed using the Minitab version 17.1.0 program. The sample size was three replicates; the data was presented as mean and standard deviation. The variance was analyzed using analysis of variance (ANOVA) to evaluate differences between groups, ensuring that the variations observed were statistically significant. A 5% level ($P<0.05$) was used to assess the significance, and the Tukey-test was applied to identify specific group differences.

3. RESULTS AND DISCUSSION

The physical and mechanical properties of edible film from RDFPE with the addition of CMC and SPI are presented in Table 1. The physical characteristics were transparency, thickness, and solubility, whereas the mechanical properties were tensile strength and elongation. Table 1 shows the CMC and SPI significantly affected the physical and mechanical properties of edible film from RDFPE.

Table 1. Physical and mechanical properties of edible films from RDFPE with the addition of CMC and SPI

Sample (RDFPE +10% w/v CMC +10% w/v SPI)	Transparency (%)	Thickness (mm)	Solubility (%)	Tensile strength (MPa)	Elongation (%)
P1 (75 ml+0 ml+0 ml)	67.02±0.88 c	0.09±0.01 a	80.83±6.99 c	3.54±0.19 a	20.83±2.20 e
P2 (75 ml+10 ml+0 ml)	64.18±0.74 b	0.11±0.03 ab	76.71±6.93 c	3.95±0.49 ab	18.33±0.83 d
P3 (75 ml+20 ml+0 ml)	63.07±0.89 b	0.14±0.03 bc	71.25±4.96 ab	4.49±0.59 b	16.67±0.83 c
P4 (75 ml+10 ml+50 ml)	59.99±0.53 a	0.18±0.01 c	62.18±1.69 ab	6.36±0.16 c	12.50±1.67 b
P5 (75 ml+20 ml+50 ml)	58.21±0.74 a	0.20±0.01 c	58.92±4.39 a	10.29±3.42 d	10.83±0.83 a

3.1. Thickness of edible films

The thickness value of RDFPE edible films was less than 0.25 mm (Table 1), this met the quality requirements of the Japanese Industrial Standard, 1975 [29] as well as based on the American Society for testing and materials, 1985 [30]. Table 1 shows, that there was a significant difference ($p=0.05$) in thickness although the weight of film-forming gel per unit area of the plate was controlled during edible film preparation. These differences are because of the increasing of solid materials which influences film thickness. The film thickness increased as more components were added to the edible film matrix [31].

Nur Hazirah *et al.* [30] showed the more polymers that make up the edible film matrix, the thicker the film. However, Table 1 shows that although there was an increase in material volume, there are no significant differences in thickness value between P2 to P3 and P4 to P5. It appeared that thickness is not only influenced by the total solid components but also by the macromolecular interactions therein. Generally, the thickness of the edible film will affect other parameters such as transparency, tensile strength, elongation, and solubility as well as barrier properties.

3.2. Transparency value of edible films

Table 1 shows that the increasing volume of 10% w/v CMC gel from 10 to 20 ml, did not significantly lower the transparency value because of the high transparency of CMC gel. However, the addition of 10 ml of 10% w/v of CMC gel to RDFPE edible film decreased the transparency value significantly. The SPI addition to RDFPE edible film containing CMC significantly decreased transparency value. This is because SPI gel has a yellowish color that is affected by decreasing transparency. The same result was shown when an edible film from SPI had lower transparency than the film from pectin [23], [32]. Besides, the lower transparency value of the edible film from this research is due to the magenta color of RDFPE. In addition to the color of the raw material, the transparency of edible film is also affected by the thickness. The data in Table 1 shows a correlation between thickness and transparency, where the greater the thickness value, the smaller the transparency value.

3.3. Solubility of edible films

Solubility in water is one of the parameters to assess the film's resistance to water. Of course, a film that does not easily disintegrate when interacting with water is desired. The solubility of RDFPE edible films decreased significantly along with the increase of CMC and SPI (Table 1). The highest value of solubility was sample P1 and there was no significant decrease between samples P1, P2, and P3. The addition of CMC in RDFPE edible film did not significantly affect the solubility value. After SPI was added to edible film from RDFPE the decrease was significant. There was a significant decrease between samples P1, P4, and P5. The presence of amino acids from SPI probably formed an ionic bond with pectin that increases molecular density and results in lower solubility [20]. In addition, cross-linking between CMC and SPI might occur due

to the Maillard reaction during the formation of edible film gel [33], where it can increase the water resistance properties [34]. The interaction between CMC and pectin from RDFPE may happen, but the interaction is usually strengthened by Ca^{2+}. The research of reference showed that there is no significant effect of pectin addition on the water solubility of CMC films [22] without Ca^{2+} as a linker agent.

Solubility is not only influenced by the interaction of the macromolecules but also by RDFPE edible film thickness. The more polymers that composed edible film matrix resulted in thicker film [30], [31]. Although not always, the thicker the edible film, the less soluble it is.

3.4. Tensile strength of edible films

The tensile strength value of edible films from RDFPE in this research was higher than the edible film from dragon fruit peel pectin alone from the study of [14], which was 0.05-0.14 MPa. The edible films in this research were added with CMC and SPI, beside the whole extract was used where the extract contained not only pectin but all components of RDFP. The main macromolecular components in RDFPE are pectin and polyphenols [19]. Polyphenols in RDFPE also play a role in pectin-SPI-CMC macromolecular interactions. There has been no research that developed the RDFPE as a whole be an edible film raw material. The mechanical properties of films can be enhanced by phase compatibility generated by macromolecule interactions [35].

Table 1 shows, that the addition of 5 ml of 5% w/v CMC solution to RDFPE edible film did not increase tensile strength significantly, but 10 ml CMC solution significantly increased tensile strength. The RDFPE film contained SPI, there was a significant increase as a response to CMC addition. The increase in tensile strength was 41.6% for 10 ml CMC solution and 129.2% for 20 ml CMC. It seems macromolecular interactions between RDFPE with CMC and SPI strengthen the edible film matrix.

3.5. Elongation of edible films

The elongation value of edible film from RDFPE with the addition of CMC and SPI ranged from 10.83±0.83 to 20.83±2.20%. There is no elongation data from previous research in edible film from pectin of dragon fruit peel. The addition of CMC and SPI has significantly decreased elongation. Table 1 shows that the decrease in elongation was significant for all levels of CMC and SPI addition. The elongation value was inverse to tensile strength because the more difficult-to-break films are usually less flexible [30]. Many researchers have similar results [32], [36], [37].

3.6. Total polyphenol content and antioxidant activity of edible films

Measurement of total polyphenols related to antioxidant activity. According to Chia and Chong [11], the dominant antioxidant compound in RDFPE is total polyphenol. The CMC and SPI were added to RDFPE films and then heated for gel formation. The mixing and heating process during edible film preparation possibly made polyphenol degraded. According to Volf *et al.* [38], phenolics are generally not resistant to high temperatures and may undergo thermal degradation and polymerization with macromolecules. The total polyphenol content and antioxidant activity of edible film from RDFPE with variations of CMC and SPI were presented in Table 2.

Table 2. The total polyphenol and antioxidant activity of edible film from RDFPE with the addition of CMC and SPI

Sample (RDFPE+10% w/v CMC+10% w/v SPI)	Total polyphenol (mg GAE/gr)	Antioxidant activity (%)
P1 (75 ml+0 ml+0 ml)	14.99±0.11 b	24.25±0.57 b
P2 (75 ml+10 ml+0 ml)	16.16±0.47 c	26.02±0.88 c
P3 (75 ml+20 ml+0 ml)	17.85±0.29 d	28.07±0.41 d
P4 (75 ml+10 ml+50 ml)	13.84±0.07 a	18.19±0.65 a
P5 (75 ml+20 ml+50 ml)	14.21±0.14 ab	18.84±0.73 a

The total polyphenol content of edible films ranged from 13.84±0.07 to 17.85±0.29 mg GAE/gr. There was a significant increase in the polyphenol content and antioxidant activity when CMC was added to RDFPE edible film. On the opposite, there was a significant decrease in polyphenol and antioxidant activity on RDFPE films after SPI was added. The decrease in total polyphenol content was likely due to the interaction between polyphenols and proteins, where according to Le Bourvellec and Renard [39], the protein and polyphenols will form an insoluble ligand. This may also relate to a large decrease in solubility (Table 1). Table 1 also shows that the interaction not only affects solubility but also increases tensile strength and decreases elongation. The total polyphenol content in RDFPE-only edible film was 14.99±0.11 mg GAE/gr,

indicating a high total polyphenol content. Based on the research of Kim *et al.* [40], the RDFPE had a total polyphenol content of 18.16±0.48 mg GAE/gr. Edible films from RDFPE are possible to develop as antioxidative packaging because of their high polyphenol content. Extracts of red dragon fruit peel are categorized as strong antioxidants [41].

The antioxidant activity of edible films was 18.19±0.66 to 28.07±0.41%. Table 2 shows the antioxidant data was in line with total polyphenol where 10% w/v of CMC significantly increased antioxidant activity. However, there was a significant decrease in antioxidant activity after the addition of SPI.

According to Rosiana *et al.* [42], the RDFP had antioxidant activity of 83.48±1.02%. There was a high decrease in antioxidant activity from the RDFPE-to-RDFPE films. This may be due to the degradation of antioxidant components. The are many other antioxidant components than polyphenol in RDFP. The antioxidant components in RDFPs are vitamin C, vitamin E, vitamin A, terpenoids, thiamine, niacin, pyridoxine, cobalamin, phenolic, carotene, and phyto albumin.

3.7. The measurement of peroxide number of peanut oils

The inhibition of peanut oil oxidation was another method to evaluate the antioxidative properties of edible films. The peanut oil has many unsaturated fatty acids that are susceptible to oxidation [43]. Peroxide number is one of the parameters that describe the oxidation level. Table 3 shows the result of the peroxide number measurement.

Table 3 shows that RDFPE edible film samples could inhibit the peroxide number of peanut oils. After 14 days, the peanut oil without RDFPE film had the highest peroxide number, while the lowest was peanut oil with P3 (RDFPE edible film added with 20 ml of 5% w/v CMC). On the 11th storage day, peanut oil without RDFPE films had peroxide number values exceeding the threshold. The maximum peroxide number allowed by the Indonesian National Standard Bureau is 10 meq O_2/kg. However, on the 14th storage day, all peanut oil samples had peroxide numbers exceeding 10 meq O_2/kg.

The P3 RDFPE edible film samples had the highest total polyphenol and antioxidant activity. This is the reason P3 had the highest ability to inhibit the peroxide number of peanut oils. The ability of dragon fruit peel antioxidants to inhibit oxidation has been proven in food products for example pastries [44] and chicken nuggets [45]. The results of previous research [27] had also shown the ability of edible films with tea extracts that are rich in polyphenols to inhibit peanut oil oxidation. The result of the measurement of peanut oils peroxide number showed that RDFPE edible films had the potential to be applied in high-fat food products.

Table 3. Peroxide number of peanut oils during 14 days of storage at 50 °C

Sample	Peroxide number of peanut oil meq O_2/kg				
	Day 4	Day 7	Day 9	Day 11	Day 14
Peanut Oil (control)	4.05 f	5.94 f	8.97 d	14.28 d	23.22 d
Peanut Oil with P1	2.99 c	3.44 c	5.33 c	8.21 c	14.72 bc
Peanut Oil with P2	2.63 b	3.02 b	4.90 b	7.86 b	13.88 ab
Peanut Oil with P3	2.40 a	2.71 a	4.09 a	6.93 a	13.67 a
Peanut Oil with P4	3.86 e	4.37 e	5.62 c	8.48 c	15.54 c
Peanut Oil with P5	3.48 d	4.10 d	5.37 c	8.26 bc	15.19 c

3.8. Identification of functional groups of edible film based on FTIR

FTIR was conducted to identify the functional groups of the compounds in the edible film. Each peak in the FTIR spectrum shows the absorption of infrared radiation by certain functional groups [46]. The FTIR spectrum of RDFPE edible film samples can be seen in Figure 2.

The absorption bands of CMC were in the ranges 3480-3440, 3260-3270, and 2960-2878 cm^{-1} are peaks of the -hydroxide (OH), -carboxylate anion (COO), and the -hydrocarbon (CH) stretching. The FTIR spectrum of pure SPI powder showed the absorption band at 3294, 1636-1680, and 1533-1559 cm^{-1} revealed from hydrogen bonding between protein chains and moisture in the protein, -nihonium (NH) bands of amide I and amide II respectively [33]. The spectrum of pure pectin showed the broad and intense absorption peak around 3400, 2940, 1737, and 1604 cm^{-1} which was the stretching vibration of -OH groups due to the inter- and intra-molecular hydrogen bonding, stretching vibrations of CH groups, ester stretching vibrations and stretching vibrations of the -COO [22].

The spectra of edible film from RDFPE (P1) showed a different spectra pattern compared with the pectin-only edible film in the study [22], where there was a shift of -OH vibrational modes to the lower values 3340 probably because the based materials of edible films in this research was not neat pectin but the hole RDFPE that was rich in pectin and polyphenol. The -OH group of polyphenol plays a role in shifting the peak of FTIR in the -OH group region. In addition, the P1 spectrum shows the absence of peaks in the 1737 and 1604 cm^{-1} regions which are peaks of ester stretching vibrations and stretching vibrations of the COO of

the pectin molecule. However, it shows intensive peaks at wavelengths of 1598 and 1033.56 cm^{-1}. The peak at a wavelength of 1033.56 cm^{-1} of the P1 spectrum probably is the peak of the glycerol molecule. According to Guerrero [47], the spectrum of glycerol will appear in the wavelength region 800-1150 cm^{-1}. Wherein the peak at a wavelength of 1598 cm^{-1} is stretching vibrations of the COO of the pectin molecule which shifted to a lower wavelength. The disappearance of the peak at a wavelength of 1737 cm^{-1} is probably because pectin from dragon fruit peels has a low degree of esterification [48] which will interact with other macromolecules in RDFPE.

The spectra of edible film from RDFPE with 10 ml addition of 5% (w/v) of CMC (P2) show a new peak at 2887.74 cm^{-1}, which was indicated -CH stretching of CMC molecule. Compared to the P1 spectrum, there are two missing peaks at wavelength 1142.81 and 888.13 cm^{-1}, which were indicated by the symmetric stretching vibrations of C-O-C and C-OH of the pectin molecule. This indicates that the interaction of CMC and pectin macromolecules strengthens the -CH stretching of CMC molecule and eliminates the symmetric stretching vibrations of C-O-C and C-OH of pectin molecule. Different interactions are shown in the addition of 20 ml of 5% (w/v) of CMC, which was shown in the P3 spectrum. There are missing peaks at wavelengths of 2887.74 and 1371.44 cm^{-1} which indicated -CH stretching and -OH group of the CMC molecule. This indicates different macromolecular interactions at different compositions.

The spectrum patterns of samples P4 and P5 are the same, where both RDFPE edible film samples are added with 50 ml of SPI 10% w/v, but both have different additions of CMC. The addition of CMC in sample P4 was 10 ml while sample P5 was 20 ml. Compared with a spectrum of P2 and P3, there was a new peak at the area of 2855, 1743, 1633, and 1556 cm^{-1}. According to Xu and Liu [49], the primary characteristic peaks of SPI were 1654 cm^{-1} (amide I, C=O stretching) and 1538 cm^{-1} (amide II, N−H bending). The peak at 1633 and 1556 cm^{-1} is probably the peak of SPI that shifts because of macromolecule interaction in the RDFPE. The peak at the area 2855 cm^{-1} is the -CH stretching of the CMC molecule. According to Zhao *et al.* [50], the peak at 1743 cm^{-1} indicates C=O stretching vibration which can be used to characterize changes in hydrogen bonding. Compared with a spectrum of P2 and P3 there are missing peaks in the spectrum of P4 and P5 at the area of 1592, 953, and 645 cm^{-1} that indicate stretching vibrations of the COO, stretching of C-H of alkane and vibration of C=C-H bend respectively.

Comparison of the spectra of P1, P2, P3, P4, and P5 showed a consistent peak shift towards smaller wavelengths in the hydroxyl group region. This shows that the more components of the edible film matrix, the more OH groups, and the more interactions between OH groups that cause the peak to shift to the left. According to Turhan *et al.* [51], the presence of a hydrogen-bound hydroxyl group may cause a shift of absorption to lower frequencies. The interaction of RDFPE macromolecules with CMC probably at -CH stretching and -OH of CMC molecule and with pectin at C-O-C and C-OH of pectin molecule. After the addition of SPI, there are different mechanisms of macromolecule interaction. The interaction is more complex suggesting between SPI, CMC, and pectin molecule. The interaction between polyphenol and SPI causes the peak vibration of benzene to disappear. The interaction between pectin and SPI causes the peak of the COO group to disappear.

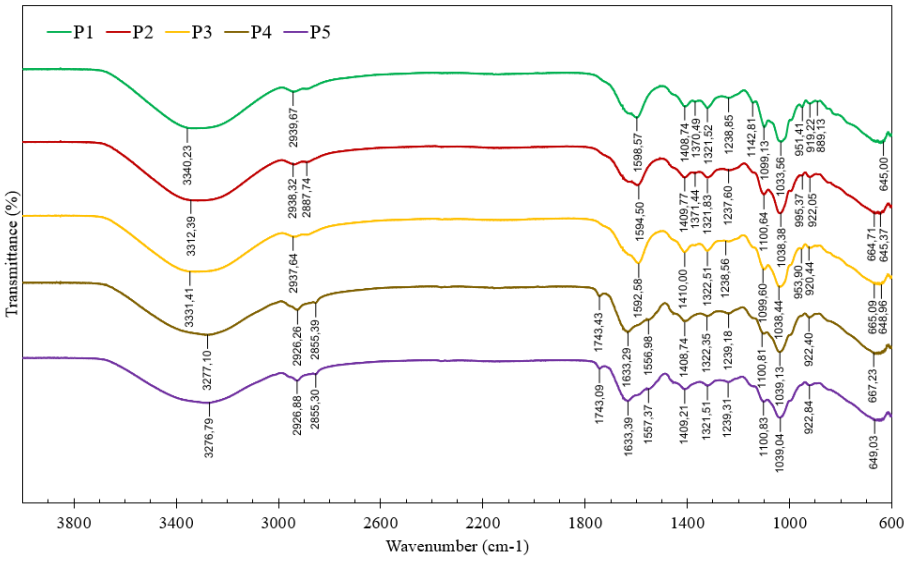

Figure 2. FTIR spectra of active edible film from RDFPE with a variation of CMC addition and SPI

4. CONCLUSION

The RDFPE has the potential to be explored as an antioxidative film because has a high content of pectin and polyphenol. The films had total polyphenol content of 14.99±0.11 mg GAE/gr and antioxidant activity of 24.25±0.57%. It was revealed that CMC addition increased total polyphenol content and antioxidant activity, as well as increased thickness, and tensile strength but decreased transparency, solubility, and elongation. A different effect was shown with SPI addition, it decreased polyphenol and antioxidant activity although increased thickness, tensile strength, and decreased transparency, solubility, and elongation. All of the edible films showed an inhibition effect on increasing the peroxide number of peanut oils. The different effects of CMC and SPI are because, of the differences of macromolecule interaction with pectin or polyphenol in RDFPE. The interaction of RDFPE macromolecules with CMC was between pectin CMC, which strengthens the -CH stretching and -OH of the CMC molecule and eliminates the symmetric stretching vibrations of C-O-C and C-OH of the pectin molecule. There were different interactions when CMC had different compositions. The interaction of RDFPE with SPI was between polyphenol and pectin. The interaction of SPI and polyphenol causes the peak vibration of benzene to disappear, while pectin and SPI cause the peak of the COO group to disappear.

ACKNOWLEDGEMENTS

Our research was supported financially by the University of Jember internal grant through Keris-Dimas's grant program. The number of contracts is 4241/UN25.3.1/LT/2023.

REFERENCES

[1] H. Chen *et al.*, "Developing edible starch film used for packaging seasonings in instant noodles," *Foods*, vol. 10, no. 12, p. 3105, Dec. 2021, doi: 10.3390/foods10123105.

[2] T. Rahmadi Putri, A. Adhitasari, V. Paramita, M. Endy Yulianto, and H. Dwi Ariyanto, "Effect of different starch on the characteristics of edible film as functional packaging in fresh meat or meat products: a review," *Mater. Today Proc.*, vol. 87, pp. 192–199, 2023, doi: 10.1016/j.matpr.2023.02.396.

[3] P. Singh, A. S. Chatli, and H. K. Mehndiratta, "Development of starch based edible films," *Int. J. Dev. Res.*, vol. 8, no. 10, pp. 23501–23506, 2018, [Online]. Available: https://www.journalijdr.com/sites/default/files/issue-pdf/14451.pdf

[4] T. Bourtoom, "Review article edible protein films: properties enhancement," *Int. Food Res. J.*, vol. 16, pp. 1–9, 2009, [Online]. Available: https://scientificpdflibrary.amb-wellness.com/wp-content/uploads/2024/01/Edible-protein-films-properties-enhancement.pdf

[5] V. Mihalca *et al.*, "Protein-based films and coatings for food industry applications," *Polymers (Basel).*, vol. 13, no. 5, p. 769, Mar. 2021, doi: 10.3390/polym13050769.

[6] A. S. Yashwant, P. Kashyap, and G. Goksen, "Recent advances in the improvement of protein-based edible films through non-thermal and thermal techniques," *Food Biosci.*, vol. 55, p. 103032, Oct. 2023, doi: 10.1016/j.fbio.2023.103032.

[7] A. Z. Hashim, "Extraction and characterization of pectin from dragon fruit (hylocerens polyrhizus) peel using different concentration of ammonium oxalate," *Basrah J. Agric. Sci.*, vol. 31, no. 1, pp. 12–19, Apr. 2018, doi: 10.37077/25200860.2018.70.

[8] B. L. Chua, S. F. Tang, A. Ali, and Y. H. Chow, "Optimisation of pectin production from dragon fruit peels waste: drying, extraction and characterisation studies," *SN Appl. Sci.*, vol. 2, no. 4, p. 621, Apr. 2020, doi: 10.1007/s42452-020-2415-y.

[9] N. S. M. Ismail, N. Ramli, N. M. Hani, and Z. Meon, "Extraction and characterization of pectin from dragon fruit (hylocereus polyrhizus) using various extraction conditions," *Sains Malaysiana*, vol. 41, no. 1, pp. 41–45, 2012.

[10] S. Rahmati, A. Abdullah, E. Momeny, and O. L. Kang, "Optimization studies on microwave assisted extraction of dragon fruit (hylocereus polyrhizus) peel pectin using response surface methodology," *Int. Food Res. J.*, vol. 22, no. 1, pp. 233–239, 2015, [Online]. Available: http://www.ifrj.upm.edu.my/22 (01) 2015/(33).pdf

[11] S. L. Chia and G. H. Chong, "Effect of drum drying on physico-chemical characteristics of dragon fruit peel (hylocereus polyrhizus)," *Int. J. Food Eng.*, vol. 11, no. 2, pp. 285–293, Apr. 2015, doi: 10.1515/ijfe-2014-0198.

[12] N. Tongkham, B. Juntasalay, P. Lasunon, and N. Sengkhamparn, "Dragon fruit peel pectin: microwave-assisted extraction and fuzzy assessment," *Agric. Nat. Resour.*, vol. 51, no. 4, pp. 262–267, Aug. 2017, doi: 10.1016/j.anres.2017.04.004.

[13] R. V. Listyarini, P. R. Susilawati, E. N. Nukung, and M. A. T. Yua, "Bioplastic from pectin of dragon fruit (hylocereus polyrhizus) peel," *J. Kim. Sains dan Apl.*, vol. 23, no. 6, pp. 203–208, Jun. 2020, doi: 10.14710/jksa.23.6.203-208.

[14] N. N. T. Tien, H. T. Nguyen, N. L. Le, T. T. Khoi, and A. Richel, "Biodegradable films from dragon fruit (hylocereus polyrhizus) peel pectin and potato starches crosslinked with glutaraldehyde," *Food Packag. Shelf Life*, vol. 37, p. 101084, Jun. 2023, doi: 10.1016/j.fpsl.2023.101084.

[15] X. Zhang, X. Chen, J. Dai, H. Cui, and L. Lin, "Edible films of pectin extracted from dragon fruit peel: effects of boiling water treatment on pectin and film properties," *Food Hydrocoll.*, vol. 147, p. 109324, Feb. 2024, doi: 10.1016/j.foodhyd.2023.109324.

[16] R. Hendra, L. Masdeatresa, R. Abdulah, and Y. Haryani, "Red dragon peel (hylocereus polyrhizus) as antioxidant source," in *The 8th International Conference of The Indonesian Chemical Society (ICICS) 2019*, AIP Conf. Proc. 2243, 2020, p. 030007. doi: 10.1063/5.0001391.

[17] A. R. Arif, F. B., and H. Natsir, "Development of antioxidant edible films based on chitosan enriched with dragon fruit (polyrhizus hyloroceus) peel extract," *J. Akta Kim. Indones.*, vol. 15, no. 1, 2022, doi: https://doi.org/10.20956/ica.v15i1.22004.

[18] W. Homthawornchoo, N. F. S. M. Hakimi, O. Romruen, and S. Rawdkuen, "Dragon fruit peel extract enriched-biocomposite wrapping film: characterization and application on coconut milk candy," *Polymers (Basel).*, vol. 15, no. 2, p. 404, Jan. 2023, doi: 10.3390/polym15020404.

[19] K. Sipayung, H. Sinaga, and D. Suryanto, "Edible coating made of taro starch and red dragon fruit peel extract," *IOP Conf. Ser. Earth Environ. Sci.*, vol. 782, no. 3, p. 032101, Jun. 2021, doi: 10.1088/1755-1315/782/3/032101.

[20] M. Akbarian, B. Ghanbarzadeh, M. Sowti, and J. Dehghannya, "Effects of pectin-CMC-based coating and osmotic dehydration pretreatments on microstructure and texture of the hot-air dried quince slices," *J. Food Process. Preserv.*, vol. 39, no. 3, pp. 260–269, Jun. 2015, doi: 10.1111/jfpp.12229.
[21] S. Panahirad, N. Mahna, R. Naghshiband-Hassani, B. Ghanbarzadeh, and F. Zaare-Nahandi, "Plum shelf life enhancement by edible coating based on pectin and carboxymethyl cellulose," *J. Biodivers. Environ. Sci.*, vol. 7, no. 1, pp. 423–430, 2015, [Online]. Available: https://www.innspub.net/wp-content/uploads/2022/12/JBES-V7-No1-p423-430.pdf
[22] S. Šešlija, A. Nešić, M. L. Škorić, M. K. Krušić, G. Santagata, and M. Malinconico, "Pectin/carboxymethylcellulose films as a potential food packaging material," *Macromol. Symp.*, vol. 378, no. 1, Apr. 2018, doi: 10.1002/masy.201600163.
[23] L. R. Amado, K. de S. Silva, and M. A. Mauro, "Effects of interactions between soy protein isolate and pectin on properties of soy protein-based films," *J. Appl. Polym. Sci.*, vol. 137, no. 21, Jun. 2020, doi: 10.1002/app.48732.
[24] N. Gontard, C. Duchez, J. Cuq, and S. Guilbert, "Edible composite films of wheat gluten and lipids: water vapour permeability and other physical properties," *Int. J. Food Sci. Technol.*, vol. 29, no. 1, pp. 39–50, Feb. 1994, doi: 10.1111/j.1365-2621.1994.tb02045.x.
[25] Y. Yap, M. A. Jamil, H. Hashim, and M. Y. Maskat, "Effect of different methods of deacidification on the properties of noni (morinda citrifolia) extract," *Sains Malaysiana*, vol. 51, no. 6, pp. 1811–1820, Jun. 2022, doi: 10.17576/jsm-2022-5106-17.
[26] M. Jouki, M. Rabbani, and M. J. Shakouri, "Effects of pectin and tomato paste as a natural antioxidant on inhibition of lipid oxidation and production of functional chicken breast sausage," *Food Sci. Technol.*, vol. 40, no. suppl 2, pp. 521–527, Dec. 2020, doi: 10.1590/fst.26419.
[27] T. Lindriati, S. B.W, and Y. Yunianta, "Edible film from jack bean flour for use as an antioxidative packaging incorporating extract of green tea," *Int. J. Adv. Sci. Eng. Inf. Technol.*, vol. 5, no. 4, p. 344, 2015, doi: 10.18517/ijaseit.5.4.547.
[28] A. Sarı and A. Biçer, "Preparation and thermal energy storage properties of building material-based composites as novel form-stable PCMs," *Energy Build.*, vol. 51, pp. 73–83, Aug. 2012, doi: 10.1016/j.enbuild.2012.04.010.
[29] B. Santoso, R. Hazirah, G. Priyanto, D. Hermanto, and D. Sugito, "Utilization of uncaria gambir roxb filtrate in the formation of bioactive edible films based on corn starch," *Food Sci. Technol.*, vol. 39, no. 4, pp. 837–842, Dec. 2019, doi: 10.1590/fst.06318.
[30] M. A. S. P. Nur Hazirah, M. I. N. Isa, and N. M. Sarbon, "Effect of xanthan gum on the physical and mechanical properties of gelatin-carboxymethyl cellulose film blends," *Food Packag. Shelf Life*, vol. 9, pp. 55–63, Sep. 2016, doi: 10.1016/j.fpsl.2016.05.008.
[31] C. Valenzuela, L. Abugoch, and C. Tapia, "Quinoa protein–chitosan–sunflower oil edible film: mechanical, barrier and structural properties," *LWT - Food Sci. Technol.*, vol. 50, no. 2, pp. 531–537, Mar. 2013, doi: 10.1016/j.lwt.2012.08.010.
[32] J. Han, S.-H. Shin, K.-M. Park, and K. M. Kim, "Characterization of physical, mechanical, and antioxidant properties of soy protein-based bioplastic films containing carboxymethylcellulose and catechin," *Food Sci. Biotechnol.*, vol. 24, no. 3, pp. 939–945, Jun. 2015, doi: 10.1007/s10068-015-0121-0.
[33] J.-F. Su, Z. Huang, X.-Y. Yuan, X.-Y. Wang, and M. Li, "Structure and properties of carboxymethyl cellulose/soy protein isolate blend edible films crosslinked by Maillard reactions," *Carbohydr. Polym.*, vol. 79, no. 1, pp. 145–153, Jan. 2010, doi: 10.1016/j.carbpol.2009.07.035.
[34] P. Tahaei, M. Abdouss, M. Edrissi, A. M. Shoushtari, and M. Zargaran, "Preparation of chelating fibrous polymer by different diamines and study on their physical and chemical properties," *Materwiss. Werksttech.*, vol. 39, no. 11, pp. 839–844, Nov. 2008, doi: 10.1002/mawe.200800364.
[35] K. S. Silva, M. A. Mauro, M. P. Gonçalves, and C. M. R. Rocha, "Synergistic interactions of locust bean gum with whey proteins: effect on physicochemical and microstructural properties of whey protein-based films," *Food Hydrocoll.*, vol. 54, pp. 179–188, Mar. 2016, doi: 10.1016/j.foodhyd.2015.09.028.
[36] M. A. Hamdan, N. A. Ramli, N. A. Othman, K. N. Mohd Amin, and F. Adam, "Characterization and property investigation of microcrystalline cellulose (MCC) and carboxymethyl cellulose (CMC) filler on the carrageenan-based biocomposite film," *Mater. Today Proc.*, vol. 42, pp. 56–62, 2021, doi: 10.1016/j.matpr.2020.09.304.
[37] X.-Y. Wang and J.-F. Su, "Biodegradation behaviours of soy protein isolate/carboxymethyl cellulose blend films," *Mater. Sci. Technol.*, vol. 30, no. 5, pp. 534–539, May 2014, doi: 10.1179/1743284713Y.0000000460.
[38] I. Volf, I. Ignat, M. Neamtu, and V. Popa, "Thermal stability, antioxidant activity, and photo-oxidation of natural polyphenols," *Chem. Pap.*, vol. 68, no. 1, Jan. 2014, doi: 10.2478/s11696-013-0417-6.
[39] C. Le Bourvellec and C. M. G. C. Renard, "Interactions between polyphenols and macromolecules: quantification methods and mechanisms," *Crit. Rev. Food Sci. Nutr.*, vol. 52, no. 3, pp. 213–248, Mar. 2012, doi: 10.1080/10408398.2010.499808.
[40] H. Kim, H. Choi, J. Y. Moon, Y. S. Kim, A. Mosaddik, and S. K. Cho, "Comparative antioxidant and antiproliferative activities of red and white pitayas and their correlation with flavonoid and polyphenol content," *J. Food Sci.*, vol. 76, no. 1, Jan. 2011, doi: 10.1111/j.1750-3841.2010.01908.x.
[41] I. Fidrianny, N. Ilham, and R. Hartati, "Antioxidant profile and phytochemical content of different parts of super red dragon fruit (hylocereus costaricensis) collected from west Java-Indonesia," *Asian J. Pharm. Clin. Res.*, vol. 10, no. 12, p. 290, Dec. 2017, doi: 10.22159/ajpcr.2017.v10i12.21571.
[42] N. M. Rosiana, A. L. Suryana, and Z. Olivia, "Polyphenol content and antioxidant activity of beverage from dragron fruit peel powder and soy powder," *IOP Conf. Ser. Earth Environ. Sci.*, vol. 672, no. 1, p. 012055, Mar. 2021, doi: 10.1088/1755-1315/672/1/012055.
[43] S. Akhtar and T. Ismail, "Flaxseed - a miraculous defense against some critical maladies," *Pak. J. Pharm. Sci.*, vol. 26, no. 1, pp. 199–208, 2013.
[44] S. Chumroenvidhayakul, T. Thilavech, M. Abeywardena, and S. Adisakwattana, "Dragon fruit peel waste (hylocereus undatus) as a potential ingredient for reducing lipid peroxidation, dietary advanced glycation end products, and starch digestibility in cookies," *Antioxidants*, vol. 12, no. 5, p. 1002, Apr. 2023, doi: 10.3390/antiox12051002.
[45] P. Madane *et al.*, "Dragon fruit (hylocereus undatus) peel as antioxidant dietary fibre on quality and lipid oxidation of chicken nuggets," *J. Food Sci. Technol.*, vol. 57, no. 4, pp. 1449–1461, Apr. 2020, doi: 10.1007/s13197-019-04180-z.
[46] S. F. Sim and W. Ting, "An automated approach for analysis of Fourier transform infrared (FTIR) spectra of edible oils," *Talanta*, vol. 88, pp. 537–543, Jan. 2012, doi: 10.1016/j.talanta.2011.11.030.
[47] P. Guerrero, A. Retegi, N. Gabilondo, and K. de la Caba, "Mechanical and thermal properties of soy protein films processed by casting and compression," *J. Food Eng.*, vol. 100, no. 1, pp. 145–151, Sep. 2010, doi: 10.1016/j.jfoodeng.2010.03.039.
[48] B. M. N. Nguyen and T. Pirak, "Physicochemical properties and antioxidant activities of white dragon fruit peel pectin extracted with conventional and ultrasound-assisted extraction," *Cogent Food Agric.*, vol. 5, no. 1, p. 1633076, Jan. 2019, doi: 10.1080/23311932.2019.1633076.

[49] Y.-T. Xu and L. Liu, "Structural and functional properties of soy protein isolates modified by soy soluble polysaccharides," *J. Agric. Food Chem.*, vol. 64, no. 38, pp. 7275–7284, Sep. 2016, doi: 10.1021/acs.jafc.6b02737.
[50] Y. Zhao, L. Shi, Z. Ren, Q. Liu, Y. Zhang, and W. Weng, "Physicochemical and antimicrobial properties of soy protein isolate films incorporating high internal phase emulsion loaded with thymol," *Food Chem. X*, vol. 22, p. 101251, Jun. 2024, doi: 10.1016/j.fochx.2024.101251.
[51] K. N. Turhan, F. Sahbaz, and A. Güner, "A spectrophotometric study of hydrogen bonding in methylcellulose-based edible films plasticized by polyethylene glycol," *J. Food Sci.*, vol. 66, no. 1, pp. 59–62, Jan. 2001, doi: 10.1111/j.1365-2621.2001.tb15581.x.

BIOGRAPHIES OF AUTHORS

Triana Lindriati is an Associated Professor with a focus working in the Department of Agricultural Product Technology, Faculty of Agricultural Technology, University of Jember, Indonesia. Skilled in food processing and engineering especially in food packaging. Strong education professional with a Bachelor's degree focused in B.E. (Chemical Engineering) from the Institute of Technology Sepuluh November (ITS). She can be contacted at email: lindriatitriana@unej.ac.id.

Sih Yuwanti is an Associated Professor with a focus working in the Department of Agricultural Product Technology, Faculty of Agricultural Technology, University of Jember, Indonesia. Skilled in food processing and engineering especially in food processing. Strong education professional with a Bachelor's Degree focused in B.Sc. (Processing of agricultural products) from Gadjah Mada University (UGM). She can be contacted at email: sihyuwanti.ftp@unej.ac.id.

Asmak Afriliana is a Doctoral with a focus working in the Department of Agricultural Product Technology, Faculty of Agricultural Technology, University of Jember, Indonesia. Skilled in processing plantation crops, especially coffee processing. Strong education professional with a Bachelor's Degree in Bachelor of Science (B. Sc) Scholarship program based on Coffee and Cocoa, Ministry of Research, Technology and Higher Education, Indonesia. She can be contacted by email: Asmak.ftp@unej.ac.id.

Aji Sukoco is an Assistant Professor with a focus on working in the Department of Agricultural Product Technology, Faculty of Agricultural Technology, University of Jember, Indonesia. Skilled in food processing and engineering especially in the development of edible film. Strong education professional with a Bachelor's Degree in Animal Science (B. Sc) from the University of Brawijaya. He can be contacted by email: ajisukoco@unej.ac.id.

Ivan Rivaldy Budianto is a fresh graduate of the Department of Agricultural Product Technology, Faculty of Agricultural Technology, University of Jember, Indonesia. He has a great interest in food processing and packaging. Strong educational background with a Bachelor's degree in Agricultural Technology from the University of Jember. He can be contacted by email: 181710101075@mail.unej.ac.id.

Wafiq Azizah is a fresh graduate from the Department of Agricultural Product Technology, Faculty of Agricultural Technology, University of Jember, Indonesia. She is very interested in food processing and packaging. She completed her Bachelor's degree in Agricultural Technology at the University of Jember. She can be contacted by email: wafiq771@gmail.com.

Umrotus Shofiyatul Fadhiyah is a fresh graduate of the Department of Agricultural Product Technology, Faculty of Agricultural Technology, University of Jember, Indonesia. She has a great interest in food processing and packaging. Strong educational background with a Bachelor's degree in Agricultural Technology from the University of Jember. She can be contacted by email: umrotusshofiya@gmail.com.