Development of Intelligent Food Packaging from Turmeric (Curcuma longa)

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Abstract: In this study, intelligent food packaging in the forms of film and coating were developed based on starch, chitosan and curcumin extracted from turmeric. Solution casting method was applied to develop the film. Both of the film and coating were evaluated and compared by their chemical, physical and biological properties. The film was evaluated in terms of tensile strength measurement, FTIR spectroscopy, antioxidant activity and antimicrobial activity as well as color difference parameters after application on the strawberry. The results obtained showed that the film has a tensile strength of 1.37 MPa, elongation at break of 18.9%, antioxidant activity of 95.65% and high antimicrobial activity as the film had successfully delayed the formation of mould on the strawberry after 5 days of storage. In addition, the stability of both film and coating were evaluated based on their applications on strawberries at two different conditions which are at room temperature and chiller temperature during 5 days storage to identify their potential use as intelligent food packaging. After 5 days, it was found that the film at room temperature had been partially degraded and the coating had caused colour degradation and texture deterioration of the strawberry. In contrast, the film and coating stored at chiller temperature remained the same in terms of physical structure and able to monitor and extend the shelf life of the strawberry. For the evaluation of the film as pH sensing film, the colour of the film changed after 5 days from 53.46 to 48.92 for L*, 26.01 to 22.68 for a* and 42.49 to 44.65 for b* at chiller temperature, while at room temperature, the values of L* changed from 53.96 to 48.96, 25.54 to 20.36 for a* and 46.34 to 44.10 for b*. These showed that the film was able to monitor the freshness of the strawberry by changing its colour in respond to pH changes of the strawberry. The results obtained revealed that both of the film and coating have a greater stability at chiller temperature as compared to storage at room temperature and both have a strong antioxidant activity and strong antimicrobial activity that they delayed the spoilage of the strawberries. Therefore, the film and coating based on starch, chitosan and curcumin can be used to monitor freshness of refrigerated food and have the potential to be used as intelligent food packaging.

Keywords— Chitosan, coating, curcumin, film, freshness, Intelligent food packaging, starch, strawberry.

I. INTRODUCTION

In recent years, general concern on the disposal of conventional synthetic plastic has increases in which the total duration of the plastic degradation requires a very long period. This has results in increase of interest in the study of biodegradable films. Many researches studied biodegradable films such as edible films and coatings from edible materials in a hope to improve the quality of food products as well as extending the shelf-life of the food products [1].

Starch is the most abundant natural polymers and commonly used material in the preparation of edible films and coatings due to its low cost, renewability, biodegradability and edibility [2]. Starch-based films have an excellent oxygen barrier properties but due to its poor moisture barrier properties and low mechanical properties (brittle and low tensile strength), its applications are limited [1]. Chitosan is a natural cationic polysaccharides and deacetylated derivative of chitin which is available mainly from shellfish processing waste. Chitosan has an antimicrobial properties which can inhibit the growth of a wide variety of fungi, yeasts and bacteria. Chitosan film is found to have a strong mechanical strength, flexible, biodegradable and antibacterial [3]. To improve the functional properties of starch and chitosan films, both of these natural biopolymers are combined to form a composite film-forming materials. Previous studies have proven that chitosan/starch composite films have an improved mechanical properties (better strength and flexibility), lower water permeability and antibacterial properties compared to starch based films without chitosan [2].

Intelligent food packaging is a food packaging that has a system which can indicate the actual quality information of packaged food products. Due to low cost and easy to use, colorimetric pH indicators have been widely applied in intelligent food packaging [4]. The colorimetric pH indicators can be incorporated into the food packaging material to monitor changes of the packaged food products' quality conditions by showing visual colour variation [5]. pH indicators are usually used to monitor the freshness of food in storage because the process of food spoilage is always associated with pH change [5]-[7].

Natural pH indicators or pigments are preferred to be incorporated into the intelligent food packaging since they are non-toxic and ideal materials for food packaging. Curcumin is a polyphenolic compound extracted from ground turmeric (curcuma longa) rhizomes. It possesses desirable biological activities such as anticancer, antimicrobial, antioxidant, anti-inflammatory and free radical scavenging activities [8], [9]. Curcumin is a highly sensitive material which reacts by changing its colour in acidic or basic conditions and the colour changes can be seen by naked eyes. In the pH range between 1 to 7, which is acidic and neutral conditions, its colour is yellow while at pH more than 8.5 or at basic condition, the colour of the curcumin changed from yellow to red.

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There are several literatures that have reported on the incorporation of curcumin in film as pH colorimetric indicator such as curcumin incorporated in films based on k-carrageenan [9], curcumin incorporated in films based on gelatin [8], and curcumin incorporated in films based on tara gum/polyvinyl alcohol [10].

In this study, intelligent food packaging in the form of film and coating were prepared by incorporation of curcumin into a composite starch/chitosan film-forming solution. The film and coating were evaluated by their applications on fresh strawberries for freshness monitoring. The prepared film was characterized by Fourier transform infrared (FTIR) spectroscopy, mechanical properties, antioxidant activity and antimicrobial activity.

II. METHODOLOGY

A. Materials

Fresh turmeric (curcuma longa) were obtained from local market. Potato starch (Bendosen), chitosan (Maya Reagent), glycerol (anhydrous, ChemAR) as plasticizer, distilled water, 95% ethanol (HmbG Chemicals) as solvent for extraction, glacial acetic acid (Riendemann Schmidt Chemical) and sodium hydroxide (NaOH) were supplied by the UiTM laboratory. Fresh strawberry was purchased from supermarket.

B. Extraction of curcumin

Fresh turmeric were cleaned, washed and sliced. The drying process of the turmeric was based on [11] with a slight modification. The sliced turmeric were dried in a hot air oven at 50 ºC for 24 hours. The dried turmeric were then powdered using a laboratory blender. In this study, turmeric extract was prepared according to the literature with a slight modification [4] and [9]. 2 g of turmeric powder was added to 30 mL of 95% ethanol and heated for 1 h at 50 ºC to get the curcumin extract. The ethanol-curcumin solution was then filtered using a filter paper into a 250 mL schott bottle.

C. Film preparation

Film was prepared by casting technique. The preparation of starch and chitosan solutions were done separately before combining both solutions. The preparation of the chitosan solution was based on a literature [12]. 2 g of chitosan powder was dissolved in 30 mL of aqueous acetic acid solution, 20 mL of 1 M sodium hydroxide solution and 50 mL distilled water and mixed under magnetic stirring at room temperature. The solution was stirred for 24 hours at 650 rpm until all the chitosan flake disappear in order to achieve a complete dispersion of the chitosan. For starch solution preparation [5], 4 g of starch and 4 mL of glycerol in 100 mL of distilled water and stirred with a magnetic stirrer at 100 ºC for 30 min at 650 rpm until the starch solution become homogenized and gelatinized. After that, both prepared starch and chitosan solutions were mixed together with addition of 10 mL of curcumin extract. To enable the combined solution to be well mixed, the solution was stirred at room temperature for 24 h at room temperature. Then, 20 mL of the filmogenic solution was spread on a plastic petri dish (8 cm in diameter) and dried in a dryer with forced air convection for 48 h at 30 ºC. Dried film was conditioned in desiccator before other tests.

D. Film thickness and mechanical properties

The thickness of the film was measured using a digital micrometer with a precision of 0.1 μm. The mechanical properties of the film (tensile strength and elongation at break) were measured using a Universal Testing Machine (Tinius Olsen) according to ASTM D882-10 method [13], a standard test method for tensile properties of thin plastic sheeting. The film specimen was cut into rectangular strip (20 mm x 60 mm) for the measurement. The initial grip separation was set to 50 mm and operating speed 0.5 mm/s, with 2.5 kN load cell. Tensile strength is the maximum stress the film can sustain before it actually fractures or amount of force necessary to pull a material apart, while strain refers to the amount the material will stretch before breaking.

E. Colour stability

The stability of the pH-sensitive film was identified according to the change of colour. The colour stability of the film was evaluated based on time and temperature. The film was applied on a fresh strawberry which was placed in a petri dish and covered with a piece of plastic film and stored at room temperature and under refrigeration for 5 days. The colour changes of the pH sensing film was measured every day. The colour of the films was determined using a chromameter (CR-400, Konica Minolta Chroma Meter), which uses three colour coordinates, namely L* (lightness), a* (redness/greenness) and b* (yellowness/blueness). The total colour difference (ΔE) was calculated by equation [9],

$$\Delta E = [(L - L^*)^2 + (a - a^*)^2 + (b - b^*)^2]^{0.5} \qquad (1)$$

where L*, a* and b* are colour values of the standard.

F. Fourier transform infrared (FTIR) spectroscopy

FTIR spectroscopy was used to identify the chemical structure of the composite films and possible interactions between their components. The FTIR spectra were conducted on a Perkin Elmer Spectrum Model TGA/ SDTA 851 FTIR spectrometer. Film was directly placed on the reading surface. The scan was performed at a 4 cm^{-1} resolution and the measurement was recorded in the range of 400-4000 cm^{-1}.

G. Antioxidant activity of film

The antioxidant activity of the film sample was measured based on the method reported by [4]. The film was cut into a size of (20 x 20 mm), placed into a test tube containing 4 mL of methyl alcohol and stirred at 25 ºC for two hours. After that, 3 mL of the sample supernatant was mixed with 1 mL of 150 μM DPPH methanol solution. DPPH scavenging rate was measured according to the following equation:

$$DPPH\ scavenging\ rate\ (\%) = \frac{A_{DPPH} - A_S}{A_{DPPH}} \times 100 \qquad (2)$$

where ADPPH is the absorbance value at 517 nm of the methanol solution and AS is the absorbance value at 517 nm of the mixture of DPPH methanol solution and the sample supernatant.

H. Antimicrobial activity of film and coating

The antimicrobial activity of the film and coating was measured by observing the physical appearance of the strawberry such as appearance of mould on the surface of the strawberry. The film was applied to the strawberry by applying the film with size of 30 mm x 30 mm onto the surface of the strawberry and wrapped with plastic film. For coating, the strawberry was fully immersed in the film-forming solution and dried before wrapped with plastic film. The film and coating applied on the strawberries are as shown in Fig. 1.

Fig. 1: Application of film and coating on strawberries

III. RESULT AND DISCUSSION

A. Thickness and mechanical properties of film

Film thickness is measured by using digital micrometer at 3 different measurement points to obtain more accurate data. The thickness measurement is required since it effects the tensile strength of the film. Higher tensile can be achieved with increasing thickness of the film.

The thickness of the starch/chitosan film containing curcumin is 0.185 mm. The mechanical properties of the film is measured by pulling test of the film using a Universal Testing Machine that will give tensile strength and elongation at break values. Tensile is a maximum tension given by the film to retain a load exerted before break, while elongation at break is a maximum length addition after pulling the film until break. A high tensile strength is required since it indicates the strength of the film developed. In food packaging application, especially, the strength of the film plays a major role since it will determine the protection of the food from external forces [14]. The tensile strength of the starch/chitosan/curcumin film is 1.37 MPa while its elongation at break is 18.9 % as shown in Fig. 2. The resulting film shows that it is quite brittle and has a low tensile strength when compared to the composite films that were based only on starch and chitosan in the previous studies in which their tensile strengths were 4.8 MPa and 19 MPa with the same amounts of starch and chitosan compositions but different amounts of glycerol, which were about 0.7% and 2.5% of the total solutions, respectively [2], [3]. The studies clearly showed that the addition of glycerol plays a significant role in increasing the rigidity of the film since the film with high amount of glycerol exhibited greater tensile strength. The low tensile strength of the composite starch/chitosan/curcumin film may be due to high content of curcumin that leads to poor dispersion of agglomerated curcumin [9]. However, the incorporation of curcumin into the composite film helps to increase the elasticity of the film since the curcumin can help to increase the free volume in the composite film [10].

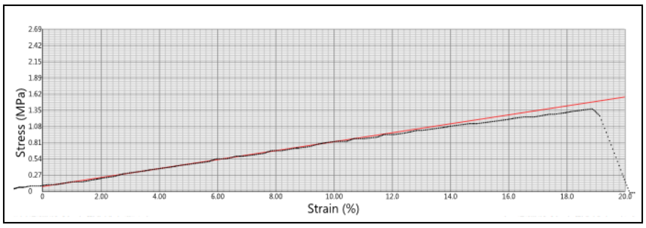


Fig. 2: Graph of stress (MPa) against strain (%) of the starch/chitosan/curcumin film.

B. Fourier transform infrared (FTIR) spectroscopy analysis

FTIR analysis is analyzed to determine the interactions between starch, chitosan and curcumin by observing the changes in the peak positions or wavenumbers of the functional groups that exist in the spectrum.

Fig. 3 shows the spectra of starch/chitosan containing curcumin film. The functional groups present in the composite film which were shown by the formation of the absorption peaks, can be classified based on its raw materials' compounds by referring to the previous study on the spectra of chitosan, starch and curcumin. The absorption peaks at 3285.07 cm^{-1} (O – H stretching and N-H stretching) was found to be contributed by intermolecular hydrogen bonding of starch, chitosan, acetic acid, glycerol and curcumin molecules. The N-H stretching was due to the presence of chitosan [3, 15, 16] in the composite film. At 2933.77 cm^{-1} (CH_2 stretching) represents alkane group that was attributed by chitosan and curcumin compounds [17], 1645.92 cm^{-1} (C = O stretching) represents amide band I of curcumin [17] and chitosan [3, 15], 1410.82 cm^{-1} (CH bending vibrations) represents CH_2 group, 1151.30 cm^{-1} (C – O stretching) represents ester group, 1025.86 cm^{-1} (C - O - C stretching) in ester of curcumin [10] and starch [2], 925.72 cm^{-1} (= C – H bend) represents alkene group.

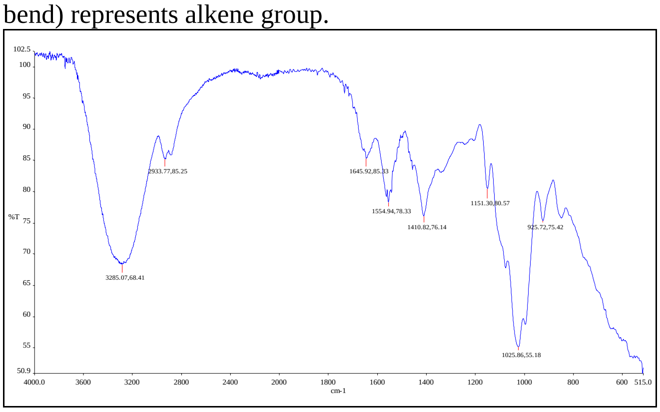


Fig. 3: FTIR spectra of starch/chitosan containing curcumin

C. Antioxidant activity of film

DPPH (2, 2-diphenyl-1-picrylhydrazyl) is a stable free radical scavenging assay that has been widely used as standard method to test the antioxidant activity of composite films [4]. In the presence of antioxidants, the solution of the DPPH solution decolourise which results in reduction of absorbance values [18].

The result obtained from the antioxidant activity of the starch/chitosan/curcumin film showed a free radical scavenging activity of 95.65% which clearly shows that this film contains high antioxidant activity.

The high free radical scavenging activity of the film may be attributed to the higher contents of phenolics in the curcumin as well as the interaction of compound between active components and polymers [4].

D. Antimicrobial activity

The antimicrobial activity of the film and coating were identified by observing the physical appearance of the strawberries samples such as the appearance of mould on the strawberries' surface at two different storage conditions which are at room temperature and chiller temperature. The appearance of the strawberries samples were observed for 5 days both at room temperature (25 ºC) and chiller temperature (5.7 ºC).

Based on the observations that have been made on the strawberries that have been applied with film and coated, it was found that at low chiller temperature, both of the strawberries show a satisfactory appearance during 5 days storage. The strawberries remain in fresh condition and no mould formation was found on the strawberries, except for the part where the film applied was not contacted with the surface of the strawberry showing a small dark spot on the fourth day of storage. This shows that film and the coating that are based on starch/chitosan/curcumin have a high antimicrobial activity that they are effective at controlling the bacterial growth on the strawberries [16]. However, at room temperature, both strawberries that were applied with film and coating show unsatisfactory appearance on the second day of storage. This happened may be due to strawberry is very sensitive to high temperature. For the strawberry with film, there is a dark spot and its colour started to degrade at one spot. For the coated strawberry, the whole colour of the red strawberry degraded and the colour turned from bright red colour to pale brownish-red colour as can be seen from the image in Table 1. The degradation of the strawberries' colour may be due to the high acidity of the films prepared. The physical appearance of the strawberries samples were not observed on the third day of storage since the laboratory was closed due to public holiday.

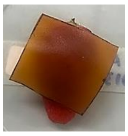
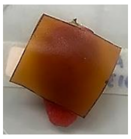
E. Application of film and coating on strawberry

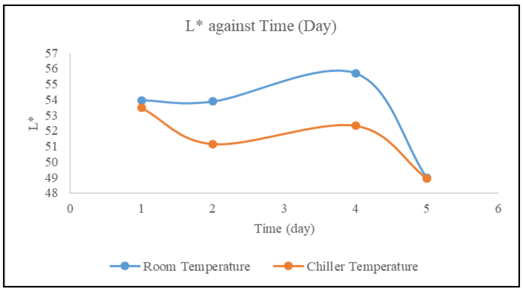
After drying, the films were easily removed from the petri dishes, showed thin and flexible structure. The films were then employed as a pH sensing indicator for food freshness monitoring on strawberry. Strawberry vaporizes volatile compounds which varies in composition and concentration depending on its freshness and maturity [19]. The main components of the volatile compounds were found by some researchers to include esters, alcohols, furans, aldehydes, terpenoids, aromatic compounds, ketones and acids. The changes in the concentration of the volatile compounds may results in the changes of pH value of the medium [9] surrounding the strawberries samples. The results regarding with the applications of films on the strawberries samples at both room temperature and chiller temperature were shown in Table II. The colour parameters as well as condition of the films (1st to 5th days) are shown in Table III and Table IV at two different storage temperatures which are at room temperature and chiller temperature. The colour changes of the applied films at the fifth day of storage were observed and compared with the first day of storage at both conditions. From the observation, the results show that the colour changes of the pH film can be identified by naked eyes since the colour changes values, ΔE are more than 5. A clear colour changes of the pH sensitive films are not much noticeable if compared day to day since the films were prepared in acidic condition in which the original colour of the films is yellow, and when applied to monitor the freshness of strawberry and as the strawberry spoiled, it becomes more acidic. Film with curcumin as pH indicator turns red if reacts with basic environment and remains yellow in neutral condition or if reacts with acidic environment. In addition, it is noticeably nfound that the film applied on the strawberry at room temperature started to degrade on the third day of application. In contrast, at chiller temperature, the condition of the film remained the same. Based on Figure 4, from day 1 to day 2, the colour of the films became darker as the values of L* slightly decreased, started to become lighter starting from day 2 to day 4, and become darker again toward day 5 at both room and chiller temperatures. As for a* which indicates red/green coordinate, it was found that at chiller temperature, the redness of the film remained stable from day 1 to day 4 before started to decrease towards day 5. In contrast, at room temperature, the redness of the film linearly decreased from day 1 to day 5, indicated that the spoilage of the strawberry continuously increase and not delayed as it becomes more acidic. As for b* which indicates the yellow/blue coordinate, both films at room and chiller temperatures show that their yellowness became brighter until the second day of storage before started to decrease toward day 5 of storage.

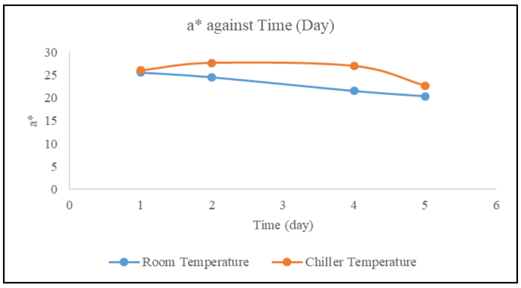
Table I: Appearance of strawberries applied with film and coating at room and chiller temperatures

Storage time (day)	Film		Coating	
	Room temperature	Chiller temperature	Room temperature	Chiller temperature
1				
2				
4				
5				

Table II: Application of sensing film for monitoring freshness of strawberries.

Time (Day)	Room Temperature	Chiller Temperature
1		
2		
4		
5		





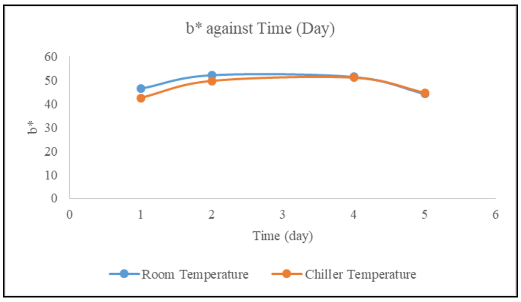


Figure 4: Graphs of L*a*b* coordinates against time (day) for films application

Table III: Colour spectrum of the pH-sensitive films containing curcumin and coated strawberry at room temperature

Storage time (day)	Film					Coating			
	Colour/ condition	L*	a*	b*	ΔE	L*	a*	b*	ΔE
1		53.96	25.54	46.34	0	35.15	36.52	28.77	0
2		53.88	24.49	52.03	5.79	39.75	23.47	17.57	17.80
4		55.70	21.52	51.27	3.57	38.02	17.55	13.02	7.66
5		48.96	20.36	44.10	9.91	36.37	17.46	15.92	3.34

Table IV: Colour spectrum of the pH-sensitive films containing curcumin and coated strawberry at chiller temperature

Storage time (day)	Film					Coating			
	Colour/ condition	L*	a*	b*	ΔE	L*	a*	b*	ΔE
1		53.46	26.01	42.49	0	35.22	33.48	23.4	0
2		51.14	27.66	49.63	7.69	32.08	30.85	19.9	5.39
4		52.32	26.99	50.98	1.91	32.70	29.88	18.26	2.00
5		48.92	22.68	44.65	8.38	32.59	28.57	16.99	1.83

IV. CONCLUSION

The composite film and edible coating prepared based on starch and chitosan incorporated with pH indicator, curcumin extracted from turmeric were developed for freshness monitoring of the strawberry. The film and edible coating exhibited greater stability when stored at chiller temperature as compared to room temperature. This showed that both film and coating were suitable for monitoring the freshness of refrigerated foods. As for the film, according to the results obtained from the characterization tests, the resulting film showed strong antioxidant properties due to the high DPPH scavenging rate which was 95.65%, as well as strong antimicrobial properties that it delayed the formation of mould on the surface of the strawberry. Moreover, the resulting film also responded well to the freshness reduction of the strawberry as the colour of the films became darken at both conditions, although the colour changes of the film was not clearly noticeable since the film was prepared in acidic pH condition same as the acidic environment of the monitored strawberry. The darkness of the films can be measured as the L* coordinate value decreased from 53.96 to 48.96 at room temperature and from 53.46 to 48.92 at chiller temperature. In overall, the film and coating have a great potential to be used as intelligent food packaging as they could provide information about the freshness of the monitored food as well as extending the shelf-life of the food through the material's antioxidant and antimicrobial properties.

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