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Curcumin-Cellulose Film for Visual Detection of Fish Spoilage

Film Selulosa-Kurkumin untuk Deteksi Kerusakan Ikan secara Visual

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ABSTRACT

Fish spoilage can be monitored visually through a responsive film to freshness and pH changes. This study aims to produce a film that is responsive to pH changes in the fish environment from curcumin, a safer natural dye. The chemical, physical, and functional characteristics as well as the film response to pH change and fish freshness during storage were studied here. Cellulose-curcumin films were fabricated by impregnation of curcumin into cellulose films. The chemical characteristics such as functional groups and surface morphology were determined by FT-IR and SEM respectively. FT-IR presents an interaction between curcumin and cellulosic polymer. The impregnation of curcumin into the cellulose film caused the segregation on the film surface observed on the SEM photos and decreased the swelling index. Cellulose-curcumin films are highly responsive to both acidic and alkaline pH. At an acidic pH, the film is yellow while at an alkaline pH the film changes to a red-brown color. The film also presented a highly color change from orange to reddish brown with increasing of fish storage days. A higher antioxidant activity of 5.54% was presented by curcumin film than the cellulose pure film. Therefore, cellulose-curcumin film can be used to detect fish spoilage through direct visual inspection.

Keywords: responsive film, fish spoilage, curcumin, cellulose, visual detection

ABSTRAK

Kerusakan ikan dalam kemasan dapat dipantau secara visual melalui film yang responsive terhadap perubahan pH. Penelitian ini bertujuan untuk menghasilkan film yang responsive terhadap perubahan pH lingkungan ikan dari pewarna alami yang lebih aman yaitu kurkumin yang diembankan pada polimer selulosa. Karakteristik film yang dikaji yaitu karakteristik kimia, fisika dan fungsional serta respon film terhadap perubahan pH dan perubahan kesegaran ikan selama penyimpanan. Film selulosa-kurkumin difabrikasi melalui impregnasi kurkumin ke dalam film selulosa. Selanjutnya ditentukan karakteristik kimia seperti gugus fungsi dengan FT-IR, morfologi permukaan dan penampang film dengan SEM, kadar air, kelarutan air dan swelling indeks serta karakteristik fungsional berupa aktivitas antioksidan. Respon film kurkumin diuji terhadap perubahan pH lingkungan dan kesegaran ikan selama penyimpanan dalam kemasan. Hasil IR menunjukkan ada interaksi antara kurkumin dan polimer selulosa. Impregnasi kurkumin ke dalam film selulosa menyebabkan terjadinya segregasi pada permukaan film yang teramati pada foto SEM, serta menurunkan swelling indeks. Film selulosa-kurkumin cukup responsive baik pada pH asam maupun basa. Pada pH asam film berwarna kuning sedangkan pada pH basa film berubah menjadi warna merah kecokelatan. Warna film mengalami perubahan dari orange pada penyimpanan ikan di hari pertama menjadi warna merah kecokelatan di hari ke-3. Aktivitas antioksidan sebesar 5,54% ditunjukkan oleh film kurkumin. Oleh karena itu, film selulosa-kurkumin dapat digunakan untuk memantau kerusakan ikan melalui inspeksi visual langsung.

Kata kunci*:* film responsif, kerusakan ikan, kurkumin, selulosa, deteksi visual

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1. INTRODUCTION

Fish is one of the marine products, rich in essential compounds for human health. The compounds such as ω3, protein, vitamins, and minerals have high nutritional value but are easily degraded. Fish are damaged by extrinsic factors such as temperature and gaseous or intrinsic factors like chemical reactions, enzyme activity, and microbial spoilage during handling, transportation, and storage [1]. Fish spoilage can be in the form of shape, texture and color changes, the presence of mucus, and causes odor [2]. The level of fish freshness can be determined through sensory analysis, chemical experiments, and total vial count tests. Sensory analysis methods are less objective, while chemical experimental methods such as Total Volatile Basic Nitrogen (TVBN) and microbial population testing are complex procedures that also require a lot of time and money [3] [4].

Intelligent packaging has been largely developed to overcome the such problems. It can monitor and inform consumers about food freshness in real-time by displaying special signs on the packaging without open the food package [5]. To detect the changes in food freshness, intelligent packaging is developed with interactive indicators that can monitor temperature and pH change, oxygen concentration, or microorganisms activity in food during storage and distribution [6][7]. The freshness indicator that detects pH as a freshness level parameter is known as a pH indicator, based on dyes whose color is influenced by the environmental pH of packaging [8]. The pH indicator has a relatively simple working principle, dye molecules interact with produced metabolites from food products during storage and cause color changes that can be visually observed [9] [10].

The pH indicator can be fabricated using synthetic or natural dyes. Some synthetic dyes that are highly sensitive to pH change such as bromothymol blue, bromocresol green, bromocresol purple, methyl red, cresol red, chlorophenol, and xylenol can be used to produce pH indicators [11] [6]. However, these dyes are not suitable for food application due to their toxicity and environmental risk [12]. To avoid those toxic compounds, non-toxic pH-sensitive dyes are preferable[13] [14] [15]. Curcumin is a non-toxic and pH-sensitive dye that can track product freshness based on pH changes [16] [17]. Curcumin presents a visual change from yellow to reddish-orange color with increasing pH [18][19][20]. Apart from being a freshness detector, curcumin has antimicrobial and antioxidant activity, so it has the potential as an active agent to extend the shelf life of food products [21].

In order to immobilize curcumin, several biopolymers have been prepared such as agar-PVA [20], pectin [22], gelatin [19], pectin-chitosan[23], and k-carrageenan [24]. In addition to these biopolymers, another biopolymer, cellulose is considered as an appropriate solid support for curcumin pigment. Cellulose is non-toxic, renewable, biocompatible, and has a good film-forming ability [25]. Cellulose also exhibits excellent mechanical properties [12] [26][27]. The crystalline phase of cellulose is complex with a highly ordered network structure and consists of many hydroxyl groups that form hydrogen bonds [26] [28]. The cellulosic film was previously synthesized by incorporating curcumin into the film [25], but the use of this film for food freshness detection in intelligent packaging is still limited. Hence, we present a cellulose-curcumin-based smart colorimetric film as a

fish freshness detector. Our proposed hypothesis was that cellulosic films containing curcumin could be used to both protect and monitor fish spoilage during storage. For this reason, we investigated the visual respons, physico-chemical, and protective characteristics of the fabricated films.

2. MATERIALS AND METHODS

2.1. Materials

Curcuma longa was collected from local market in Kefamenanu City. Ethanol 96%, HCl 37%, 1-allyl-3-methylimidazolium chloride (AmimCl), cellulose, glycerol, methanol, DPPH and buffer pH 2-10 were supplied by Merck.

2.2. Methods

2.2.1. Curcumin Extraction

200 gr of turmeric powder was extracted by maceration using 2L 96% ethanol for 3 days. The crude extract was filtered and evaporated with a rotary evaporator.

2.2.2. Film Fabrication

AmimCl was weighed in three beakers and added with a certain amount of cellulose (4 wt% of the total solution) while stirring at 80 °C for 1 hour. Then 10% glycerol was added to the third beaker, stirring was continued for 30 minutes. The solution was poured with a thickness of about 0.1 cm on a glass plate using a glass rod. The film was washed with distilled water for 30 minutes to obtain a cellulose film. Each film was labeled with FC, FCCur, and FCCurG. Impregnation was carried out by immersing the FCCur and FCCurG films in curcumin extract, then shaking them at 30 °C for 4 hours. The film was washed and dried at ambient conditions for 20 hours.

2.2.3. Characterization Methods

The fabricated films were characterized using Fourier Transform-Infrared (FT-IR) Spectroscopy to determine their functional groups. FT-IR spectra were scanned by a Shimadzu Infrared Prestige 21 spectrophotometer in the wavenumber range of 4000–400 cm^{-1}. The morphology and cross-section of the films were observed using Inspect-S50 FEI Scanning Electron Microscopy (SEM).

Moisture content (MC) was determined at 105°C in the oven. MC was calculated using the equation:

$$\text{MC}\ (\%) = \frac{(\text{Mi}-\text{Mf})}{Mi} \text{ x } 100 \quad (1)$$

Mi: initial weight of film (g); Mf: final weight of the film dried at 105°C (g).

Water Solubility (WS) was determined by cutting the film into squares (2 × 2 cm) and drying in an oven at 70 °C for 24 hours to determine the initial dry weight (W0). Then the sample was placed in 50 mL of distilled water at room temperature for 24 hours, while stirring at 100 rpm. Then the sample was taken from the container and the surface water was removed with filter paper. The wet weight of the sample was immediately weighed (W1), then the wet film sample was dried in an oven and the dry weight (W2) was assesed. Each treatment was repeated 3 times to get the mean value. Swelling Indeks (SI) and WS (%) were calculated by the equation:

$$\text{WS}\ (\%) = \frac{(\text{Wo}-\text{W2})}{\text{Wo}} \text{ x } 100 \quad (2)$$

$$\text{SI}\ (\%) = \frac{(\text{W1}-\text{Wo})}{\text{Wo}} \text{ x } 100 \quad (3)$$

2.2.4. pH Changes of Fish During Storage

10 g of fresh fish samples were prepared and placed on sterilized petri dishes. Fish samples were stored at room temperature and analyzed periodically (up to 3 days) to detect pH changes. Each fish sample was mixed with 90 mL of distilled water for 3 minutes and then the pH value was recorded using a pH meter.

2.2.5. Visual Evaluation of Colour Changes

The films were cut into 2 x 2 cm, then placed on 10 g of fish samples stored in petri dishes (diameter 90 mm) during the test period (3 days). The prepared samples were stored in an incubator under a controlled temperature (25 ^{o}C). During the experiment, the pH meter was used to monitor the pH of the fish samples that were in contact with the film. When a color change in the film occurs, both the color change and the pH value were recorded and documented.

2.2.6. Antioxidant Activity

The film was cut into 20 × 20 mm, then put into a test tube containing 4 mL of methanol. The mixture was stirred for two hours at 25 °C. Next 3 mL of the supernatant, 1 mL of methanol solution, and 150 M of DPPH solution were mixed. The absorbance of the solution was measured at 517 nm (A1), while 3 mL of the supernatant sample and 1 mL of 150 M methanol solution were mixed and the absorbance of the solution was measured at 517 nm (A0). The rate of scavenging DPPH radicals was determined by:

$$\text{SR}\ (\%) = \frac{(\text{A0}-\text{A1})}{A0}\ \text{x}\ 100 \qquad (4)$$

3. RESULTS AND DISCUSSION

3.1. Physico-Chemical Characteristics

The fabricated film from this work was displayed in Figure 1. Visually, the cellulose film (FC) has a colorless and transparent appearance. The cellulose-curcumin film (FCCur) possesses a yellow color and transparent appearance with a very stiff film texture. A similar appearance is exhibited by the cellulose-curcumin-glycerol film (FCCurG), but it is more flexible than FCCur due to the presence of glycerol as a plasticizer.

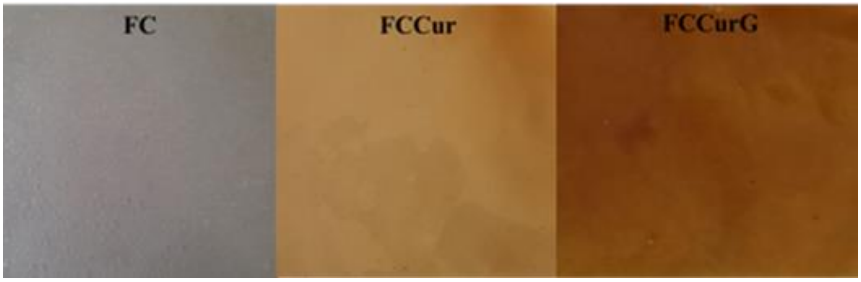


Figure 1. Visual Appearance of Fabricated Films

The physical characteristics of the cellulosic films are summarized in Table 1. It shows an increase in film weight when incorporated with curcumin and glycerol. Previous research has reported that curcumin content can change the interior structure of the hydrogel-forming matrix and increase the spatial distance between curcumin and film-forming cellulose polymers [18]. This process increases the thickness and mass of the film. Another physical characteristic, the moisture content is an important factor affecting the stability and brittleness of the film. The moisture content decreased by 1.16% when added to curcumin. This behaviour could be related to the hydrophobic nature of curcumin [29], whereas the glycerol addition provides more polar sites to absorb moisture from surroundings, which may have contributed to the moisture content increase.

Table 1. Physical Properties of The Cellulose Film

Sample	Film Weight (g)	Water Content (%)	Water Solubility (%)	Swelling Index (%)
FC	1.37	19.80	9.78	251.94
FCCur	2.57	18.64	7.56	238.38
FCCurG	5.44	21.72	15.11	227.42

Water solubility (WS) is a crucial characteristic in food packaging and is generally considered as an indicator of water tolerance. Films with low WS levels can effectively protect food against physical and microbial spoilage. On the other hand, a high WS value can increase the biodegradability of the film. The WS value of the fabricated film slightly decreased when added with curcumin and contrary increased when added with glycerol. These conditions may be attributed to the hydrophilic groups of glycerol that can easily attract more water molecules. Previous researchers have reported a decrease in WS values due to increased concentrations of curcumin in chitosan [5] and agar films [30].

The swelling index (SI) parameter is expressed as the percentage of water absorption by the film until it is saturated. SI of a film can affect the efficiency of the film colour response [31]. A high swelling index will lead to faster dye release, which is not conducive to the indicator colour response [13]. In our work, curcumin caused a decrease in swelling index value, which can be associated with a more compact structure of the films. Intermolecular interactions are increased between the filler matrix and cellulose polymer. This interaction reduced the binding sites of the film to absorb water, as a result, the total water molecules occupying the microstructure network decreased. This behaviour was in agreement with the other researchers [32] [13].

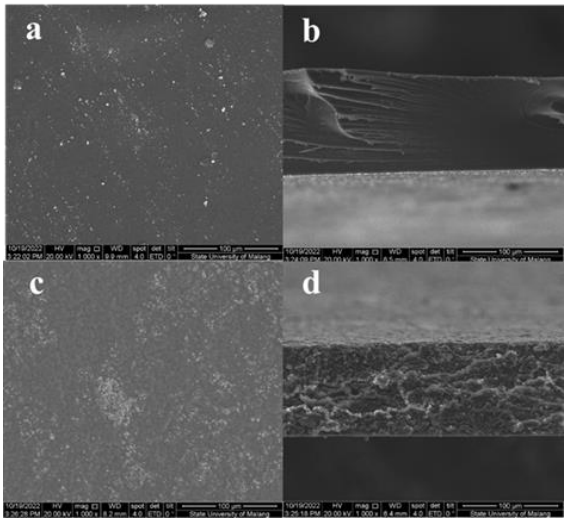


Figure 2. Surface Morphology of (a) FC and (b) FCCur and Cross-section of (b) FC and (d) FCCur

The morphological properties of the film surface and the cross-section were observed by SEM. The SEM photos was given in Figure 2. The surface of FC was compact, indicating the good film formability of cellulose, but it had some white spots implying unwell-dispersed of cellulose particles on the surface. The cross-section of the pure cellulose film is relatively smooth and compact, however, there are still cellulose particles that are not evenly dispersed. Also noticed in the cross-sectional photos that the addition of curcumin reduced the particles uniformity of the films and caused aggregation of curcumin particles in the matrix. Increasing curcumin concentration has been previously reported to increase the agglomeration of curcumin with polymers [24] [25].

FT-IR spectroscopy is generally used to determine the impact of additive incorporation on the chemical structure of a polymer-based film. The film structure was studied by observing the changes

that occur in the absorption frequency of certain functional groups. FT-IR spectra depicting changes in the cellulose structure after immobilization of curcumin into the film are shown in Figure 3.

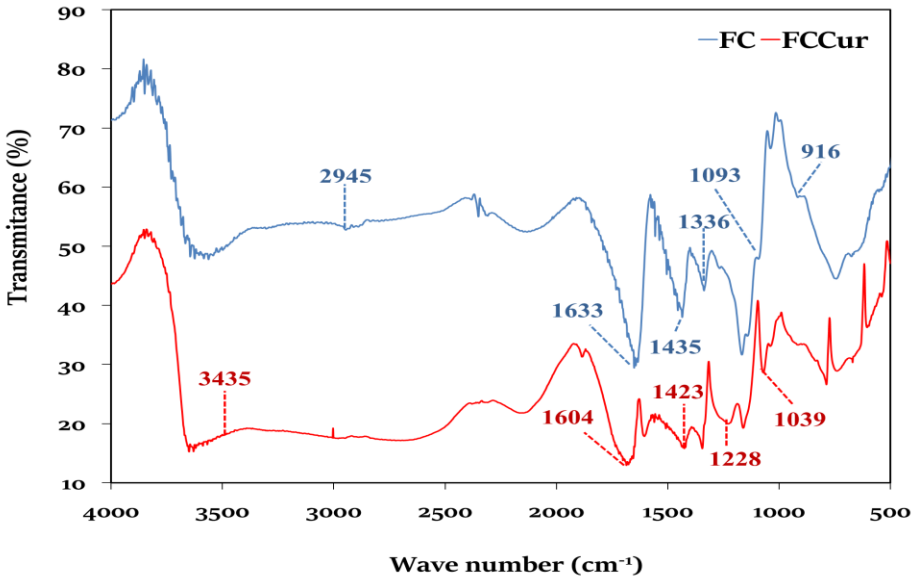


Figure 3. IR Spectra of FC and FCCur Films

Several characteristic peaks for the FC and FCCur were identified. An observed peak at a wave number of 2945 cm^{-1} is attributed to C-H stretching, at a wave number of 1093 cm^{-1} there was a -C-O-C bond vibration on the pyranose ring and at a wave number of 916 cm^{-1} there was α-glycosidic bond. A similar characteristic peak has also been reported by Chen [33]. The other peaks are also identified and typical for cellulose, the wave number of 1633 cm^{-1} is for water molecules that are absorbed in cellulose, 1435 cm^{-1} indicates a -CH_2 bending, meanwhile the C-H bending is found at a wave number of 1336 cm^{-1}. Another investigator has reported that the FT-IR spectra of cellulose showed absorption peaks at 1633, 1428, and 1367 cm^{-1} for water molecules, bending -CH_2 and C-H [34]. The FCCur film showed specific absorption of curcumin at wave numbers 3435 cm^{-1} for phenolic -OH stretching, 1604 cm^{-1} for stretching C=C benzene bonds, 1039 cm^{-1} for -C-O-C stretching, 1228 cm^{-1} for C-O aromatics, and 1423 cm^{-1} for bending CH. Identical absorption of curcumin has also been reported by previous investigators [35] [18].

3.2. Antioxidant activity

Antioxidant activity is one of the most important functional characteristics of active packaging. Antioxidant activity is needed to prevent oxidative reactions that cause changes in taste, rancidity, and color in packaged foods. The antioxidant activity of FC and FCCur was measured based on the activity of scavenging DPPH radicals. The measurement results are shown in Figure 4. FC film counteracts DPPH radicals by 3.53%, while FCCur has an antioxidant activity of 5.54%. The addition of curcumin to the cellulose film increased the free radical scavenging properties by 2.01%. The higher DPPH radical scavenging activity of FCCurs is attributed to the phenolic hydroxyl groups of curcumin, which form phenoxy groups to scavenge free radicals [12]. The previous researcher stated that the free radical scavenging activity of a film is generally influenced by various factors such as the film's microstructure, the active compound and polymer matrix interaction, and the release rate of active ingredients [36]. The higher radical scavenging activity in the DPPH method was due to the high release rate of alcohol-soluble curcumin using methanol as the solvent in the present method.

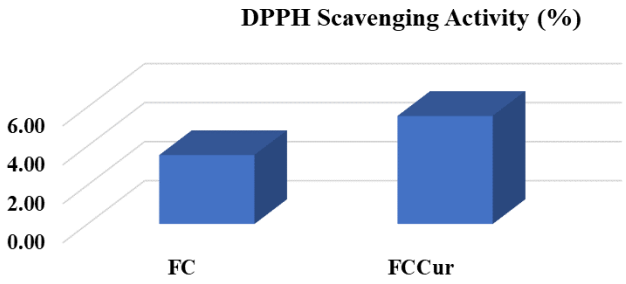


Figure 4. DPPH Scavenging Activity of The Films

3.3. Film Responses to pH Changes and Fish Freshness

Figure 5 shows the response of curcumin extracts and films (FCCur and FCCurG) to pH changes. Both the extract and curcumin film at acidic pH exposed yellow to orange and then the color changed to reddish-orange when the pH was increased. The color change of curcumin is caused by changes in the dominant structure of curcumin at certain pH conditions [9]. Curcumin has a regular crystalline structure composed of seven carbon chains consisting of α, ß-unsaturated ß-diketone moiety bonded to two aromatic rings having a phenolic ortho-methoxy hydroxy group. At neutral and alkaline pH, the α, ß-unsaturated ß-diketone bond breaks and acts as a hydrogen donor site which causes hydrolysis and degradation of curcumin. Intramolecular hydrogen atom transfer in the ß -diketone chain occurs in the structure of curcumin, which is in the form of a keto-enol tautomer depending on the nature of the solvent. The bis-keto form is found at acidic or neutral pH, and the enolic form predominates at alkaline pH [37]. In a solution pH range of 1−7, the predominant bis-keto form of curcumin displays a yellow color with very low water solubility.

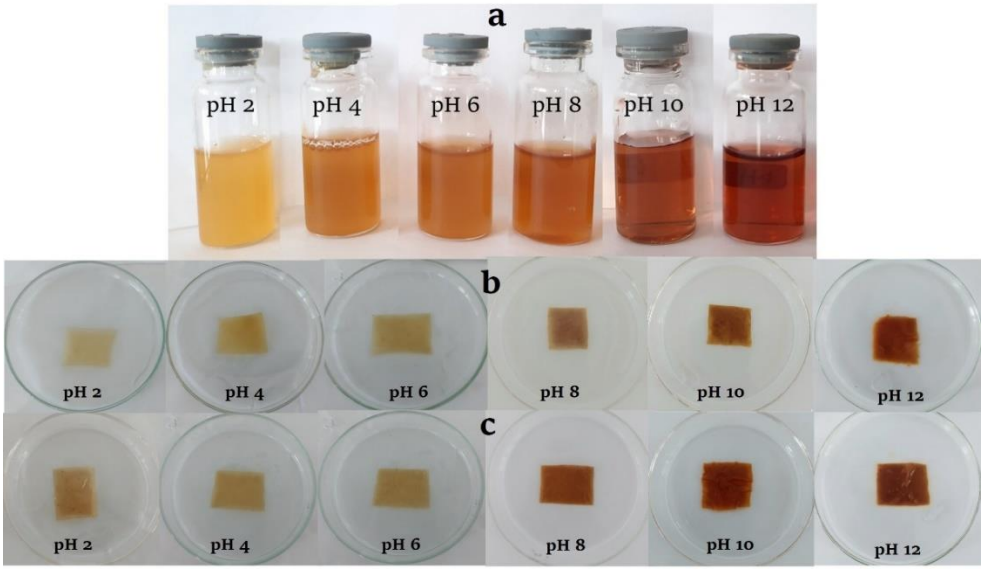
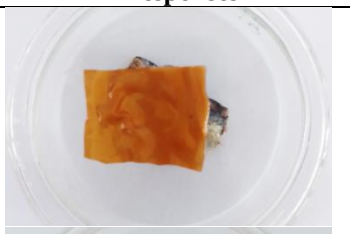


Figure 5. Responses of a) Curcumin Extract, b) FCCur, and c) FCCurG at different pH

Under neutral or basic conditions, curcumin rapidly decomposes due to the loss of protons from the phenolic group hence the enolate form predominates. An increase in pH will degrade curcumin causing the formation of trans-6-(4-hydroxy-3-methoxyphenyl)-2,4-dioxo-5-hexenal as the main degradation product, while feruloyl methane, ferulic acid, and vanillin are minor degradation products that causes a color change to red [38].

The spoilage of most seafood products, especially fish, is caused by microorganisms. Microbial growth and metabolism produce amines, and biogenic amines (putrescine, histamine, and cadaverine) that can increase environmental pH [39]. Glycogen (stored carbohydrate) or lipid is oxidized by the tissue enzymes in a series of reactions which ultimately produce carbon dioxide (CO_2), water and the energy-rich organic compound adenosine triphosphate (ATP). As shown in Table 2, the film color was orange on the first day of storage, changing to a brownish-red color on the 3rd day due to an increase in pH. The color change is supported by higher ambient humidity, thus facilitating the reaction between NH_3 and H_2O to form NH_4^+ and OH^-. This results in the formation of an alkaline environment on the film surface. Under alkaline conditions, the phenolic hydroxyl group can easily form an acid-base reaction with OH- to form a phenolic oxygen anion in the structure of curcumin. The formation of phenolic oxygen anions causes color changes as shown in Table 2. The increase of pH in shrimp products due to the formation of NH_3 and other volatile amines has been monitored with curcumin films by previous researchers [40]. Therefore, curcumin-cellulose films can be used to monitor fish spoilage through direct visual inspection.

Table 2. Film Responses During Fish Storage

Storage Time (Day)	pH	Film Responses
1	8,99	
2	10,48	
3	11,44	

4. CONCLUSIONS

The smart film fabricated from cellulose-curcumin is responsive to pH changes and also changes in fish freshness during storage. At an acidic pH, the film is yellow while at an alkaline pH the film changes to a reddish-brown color. The film color changed from orange on the first day of storage to brownish red on the third day. Cellulose-curcumin films can be used to monitor fish spoilage through direct visual inspection.

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