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Research article

# Fabrication of freshness indicators based on methylcellulose-containing color indicator solutions for monitoring the quality of coconut water

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A R T I C L E I N F O

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A B S T R A C T

This is the first study to apply intelligent packaging to coconut water. The purpose of this study was to determine the best color indicator solution for making freshness indicator labels based on methylcellulose along with the color change profile of coconut water during storage at room temperature. Three color indicator solutions were used, namely phenol red, bromothymol blue, and methyl red, which were then continued with the fabrication of freshness indicator labels based on methylcellulose from each of these color indicator solutions and applied to coconut water at 25 $^{\circ}C$ room temperature storage for 24 h with observations every 4 h in the form of pH, total dissolved solids, total acid, turbidity, total microbes, $CO_2$ gas, $O_2$ gas, and freshness indicator label color changes. The values of pH, total soluble solids, and $O_2$ gas decreased with storage time, whereas the values of total acid, turbidity, total microbes, and $CO_2$ gas continued to increase. The methylcellulose-based phenol red freshness indicator label provides the best color change profile that matches the freshness condition of coconut water, namely purplish red (fresh), orange (immediately consumed), and yellow (damaged) so that it can be used as intelligent packaging to monitor the quality of coconut water.

## 1. Introduction

Packaging is generally used in food products to protect them from damage due to external factors such as environmental conditions. In addition, packaging has other functions as a storage medium and facilitates the use of products, making them more practical. The main function of other packaging is as a medium of communication to consumers regarding the product itself, such as nutritional content or consumption instructions [1]. However, the function of packaging cannot provide information to consumers about the quality and freshness of the product directly, so consumers need to re-confirm the quality of the product after opening the packaging [2]. This causes the use of packaging to continue to develop, particularly with the existence of intelligent packaging as a packaging technology to complement the shortcomings of the general packaging function.

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Intelligent packaging can monitor and provide information about changes in product quality directly and at any time during transportation and storage from suppliers to consumers [3]. One type of intelligent packaging is a freshness indicator that has the form of a label or film, accompanied by a color indicator that is sensitive to changes in pH [2]. The mechanism of action of the freshness indicator is the change in the color of the indicator from one color to another according to the color indicator solution used, which indicates that the packaged product has undergone microbial growth or other chemical reactions that indicate that the freshness or quality of the product has decreased owing to the appearance of metabolite compounds such as organic acids (acetic acid and lactic acid), ethanol, carbon dioxide, and nitrogenous bases such as trimethyl amine and sulfur [4]. The use of freshness indicators in packaging can make it easier for consumers to determine product quality while providing more accurate results than expiration dates without the need to open the packaging so that it can help reduce food waste problems [2,5].

The fabrication and application of freshness indicators for intelligent packaging have been carried out for various products such as beef [6–10], chicken [11,12], fish [13–16], shrimp [17–19], fruit and vegetables [20–23]. However, based on existing literature, there is no freshness indicator for coconut water. Coconut water is a functional drink with a variety of health benefits, including antioxidant, cardioprotective, antithrombotic, anticholesterol, antibacterial, antidiabetic, and hepatoprotective [24]. However, its relatively short shelf life is accompanied by signs of deterioration that are difficult to detect directly by consumers. Commonly used methods to monitor coconut water quality are based on sensory evaluation such as aroma and color [25]. The formation of acidic carbon dioxide gas due to the activity of microorganisms that carry out anaerobic and aerobic fermentation, this makes the use of freshness indicators important to apply so that it can make it easier for consumers to know the quality of the coconut water [26].

Based on these problems, this research will be carried out on the fabrication of freshness indicators based on methylcellulose biopolymers containing color indicator solutions accompanied by LDPE plastic layers that are sensitive to carbon dioxide gas to monitor the freshness of coconut water. The color indicator solution acts as a provider of information on the quality of coconut water which is characterized by changes in color due to the accumulation of acidic carbon dioxide gas, several studies use synthetic color indicator solutions in the fabrication of freshness indicators such as bromothymol blue and methyl red in golden drop dessert and mango fruit [23,27], phenol red and bromothymol blue in chicken meat [12]. However, the difference in the results of the best type of color indicator solution for each type of product applied necessitates further research on the type of color indicator solution that is well used in making freshness indicators to be applied to coconut water. Therefore, this research will focus on determining the best color indicator for making freshness indicators that have clear color changes visible to the eye to monitor the quality of coconut water.

## 2. Materials and methods

### *2.1. Coconut water*

Coconut (*Cocos nucifera* L.) water was purchased from the traditional market of Perintis Independence IV (Makassar). The coconut water used comes from green coconuts that have a maturity level of 7–9 months, are free from defects, and have a volume of 500–700 mL of coconut water. Coconut water was used directly from freshly opened fruits and packaged in plastic cups with freshness indicator labels.

### *2.2. Fabrication of freshness indicator label*

The fabrication of freshness indicator labels begins with the preparation of color indicator solutions in advance for phenol red, bromothymol blue, and methyl red based on Hidayat et al. the color indicator solution was made in a concentration of 1% (w/v) using 50% ethanol as a solvent [28]. The pH of each color indicator solution was adjusted using NaOH and $CH_3COOH$ (phenol red = 8.0, bromothymol blue = 7.6, methyl red = 6.0). Furthermore, the freshness indicator label was made based on Obaidi et al. with the modification that 2% (w/v) methylcellulose was dissolved in distilled water (55 °*C*) until homogeneous [12]. Then, 0.5% (v/v)

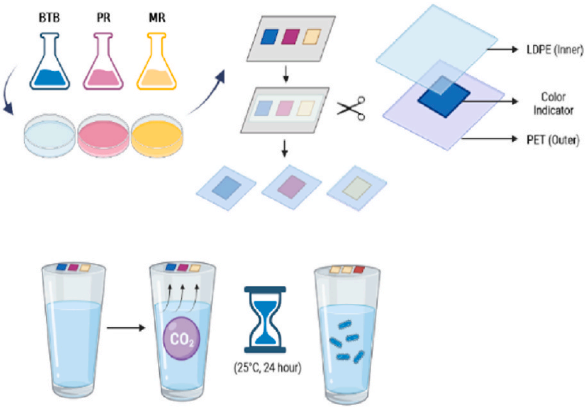



**Fig. 1.** Fabrication and application of freshness indicator labels.

polyethylene glycol 400 and 2% (v/v) color indicator solution were added. Furthermore, 30 mL of the homogeneous freshness indicator label mixture was printed on a Petri dish that had been coated with polyethylene terephthalate (PET) plastic and dried at 40 °C in an oven. After drying, the freshness indicator label was coated with LDPE plastic and cut to a size of 1.5 × 1.5 cm (Fig. 1).

### 2.3. Application of freshness indicator label for coconut water

Fresh coconut water taken directly from the fruit was packed in up to 200 mL of 12 Oz PP plastic cups using plastic wrap as a wrapper that has been added with a freshness indicator label with the surface of the LDPE plastic layer directly facing the coconut water. Coconut water that had been packaged with a freshness indicator label was stored at room temperature (25 ± 3°*C*) for 24 h and observed for changes in the color of the indicator label and the freshness quality of coconut water at 0, 4, 8, 12, 16, 20, and 24 h (Fig. 1).

### 2.4. Color analysis of the freshness indicator label

The freshness indicator labels were photographed using a digital camera (Sony A6000). The color change data were measured in the form of L*, a*, and b* values based on Hunter's Lab Colorimetric System scale using a colorimeter (AMT 507), which is then expressed in the form of ° Hue and Δ E [6].

### 2.5. Chemical structure of the freshness indicator label

The chemical structure of the freshness indicator label was determined using Fourier transform Infrared/FTIR spectroscopy (Shimadzu IRSpirit) in the form of spectra in the wavelength range 4000–500 $cm^{-1}$ [7]. The FTIR spectrum obtained can be used to observe changes in the chemical structure of the freshness indicator label after interacting with coconut water.

### 2.6. Chemical analyses

Chemical testing is divided into two indicator solutions that are tested for sensitivity before use by adding $CH_3COOH$ and NaOH to produce several variations in the pH [23]. Furthermore, the coconut water was tested for pH using a pH meter (Phs-3C), total acids [29], total dissolved solids with a digital refractometer (Hanna HI 96801), turbidity with a turbidity meter (Lutron Tu-2016), $CO_2$ and $O_2$ gases using a $CO_2$ analyzer (Sentry ST 303), an oxygen detector (Smart sensor AR8100) expressed in %, and total microbes [30].

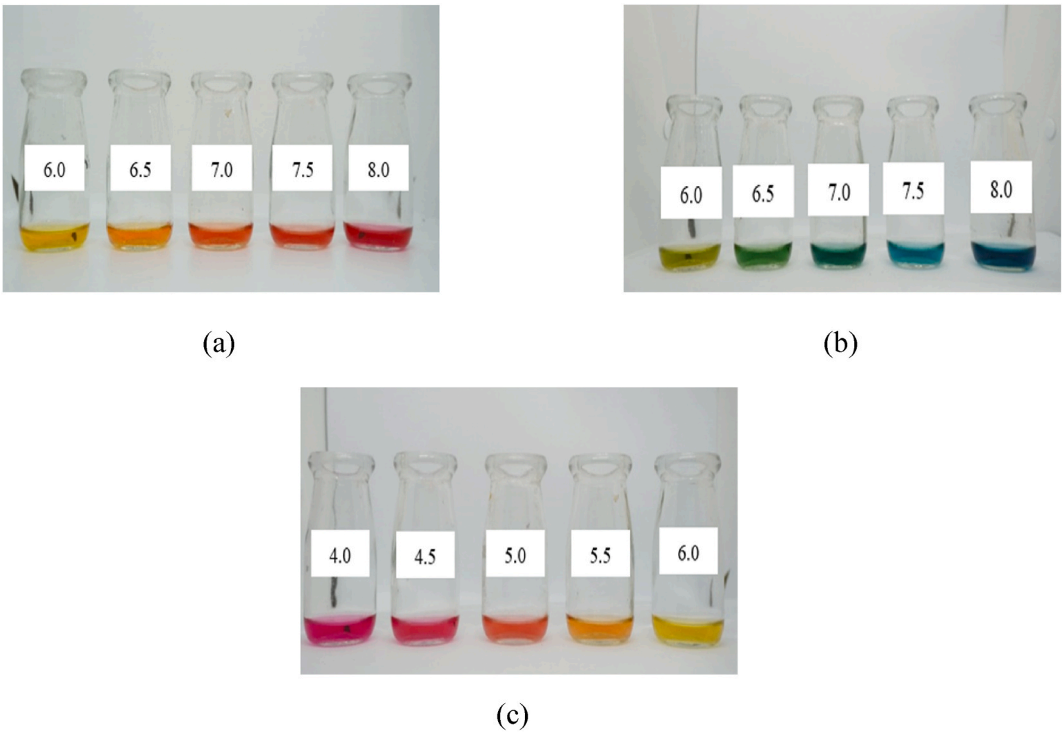



**Fig. 2.** Color indicator solution sensitivity results phenol red (a), bromothymol blue (b), methyl red (c).

### 2.7. Statistical analysis

This study used a completely randomized design, each of which used three replicates ($n = 3$). The results were processed using SPSS software to conduct one-way analysis of variance (ANOVA), followed by Duncan's multiple range test ($p < 0.05$).

## 3. Results and discussions

### 3.1. Sensitivity of color indicator solution

Sensitivity testing of color indicator solutions was carried out with the aim of determining the color change profile of each color indicator solution under different pH conditions before being applied to the fabrication of freshness indicators. This sensitivity test was carried out by adding 0.01 N $CH_3COOH$ acidic compounds and 0.01 N NaOH base compounds at different concentrations. Determination of the pH of each color indicator solution, namely phenol red, bromothymol blue, and methyl red, is based on the initial pH settings on the freshness indicator label, as well as the conditions under which the freshness indicator label indicates damage to the product to be packaged. Based on the results shown in Fig. 2, the phenol red indicator solution under alkaline conditions will have a purplish-red color, which will then turn orange and yellow under acidic conditions (Fig. 2a). The bromothymol blue indicator solution produces a purplish-blue color under alkaline conditions, which then turns green and yellow under acidic conditions (Fig. 2b), as well as the methyl red indicator solution, which changes color from yellow to purplish red under alkaline to acidic conditions (Fig. 2c). The color changes produced by the three indicator color solutions were in accordance with the statement [31].

### 3.2. Coconut water quality

Based on Table 1, the pH of coconut water decreased with storage time, starting from pH 5.22 at the beginning of storage to pH 4.64 after storage for 24 h. According to (SNI 4268:2020) the pH standard of coconut water is in the range of 5.0–6.0. Based on the results obtained, it can be concluded that coconut water at a storage time of 24 h was damaged because it had a pH of 4.64, which is below the minimum standard of pH 5.0. This is supported by statistical results that showed a very significant effect at the 5% level ($p < 0.05$), and Duncan's further test at the 5% level showed a significant difference between pH at 0 h storage and 24 h. The decrease in pH of coconut water is caused by the sugar content that undergoes a fermentation process, both homofermentative and heterofermentative, due to the creation of anaerobic conditions during storage, producing organic acid compounds [32].

During storage, coconut water continued to experience an increase in total acid, from 0.1072% at storage time 0 h to 0.2702% at storage time 24 h. The statistical results show that the total acid content of coconut water at a storage time of 0–12 h is not significantly different; therefore, it can be said that it still has fresh quality, but the quality decreases after the 12-h storage time. The acidity level of a solution may be low even when in contact with a significant amount of dioxide because of the limited solubility of carbon dioxide in water and the low acid dissociation constants of the conjugated species of carbonic acid. This phenomenon is particularly notable at low partial pressures of $CO_2$, a situation commonly encountered in the release of this gas from coconut water [33]. The increase in total acid content indicates that the freshness of coconut water continues to decrease as storage time progresses, which can occur because coconut water contains sugars such as glucose, sucrose, and fructose, which can be degraded by microbes both aerobically and anaerobically to produce organic acid compounds such as lactic acid and acetic acid [32].

Coconut water at the beginning of storage had a total soluble solids content of 5.43°Brix, which continued to decrease constantly to 5.10°Brix after storage for 24 h. According to Yin, coconut water is good if it contains total soluble solids ranging from 5.5 to 8.0°Brix [32]. Based on these standards, the coconut water at the beginning of storage (0 h) was not met, which can be caused by various external factors from the coconut fruit itself, such as planting location and fruit maturity level [34]. The decrease in total soluble solids of coconut water during storage indicates that damage occurs because sugar compounds, such as fructose, glucose, and sucrose, are broken down by microbes into energy in the form of ATP for cell needs, accompanied by the formation of by-products such as carbon dioxide gas, water, and organic acids [26].

The results showed that the turbidity of the coconut water increased as the storage time increased from 28.83 NTU 290.17 NTU.

**Table 1**
Chemical parameters of coconut water quality during 24-h storage.

| Storage time (hour) | 0 | 4 | 8 | 12 | 16 | 20 | 24 |
|---|---|---|---|---|---|---|---|
| pH | 5.20 $\pm 0.00^{a}$ | 5.22 $\pm 0.00^{a}$ | 5.20 $\pm 0.10^{b}$ | 5.16 $\pm 0.10^{c}$ | 5.14 $\pm 0.00^{c}$ | 5.09 $\pm 0.17^{d}$ | 4.64 $\pm 0.10^{e}$ |
| Total Acidity (%) | 0.1072 $\pm 0.00^{a}$ | 0.1139 $\pm 0.00^{a}$ | 0.1117 $\pm 0.00^{a}$ | 0.1385 $\pm 0.03^{ab}$ | 0.1608 $\pm 0.02^{b}$ | 0.2032 $\pm 0.00^{c}$ | 0.2702 $\pm 0.01^{d}$ |
| Total Soluble Solids (°*Brix*) | 5.43 $\pm 0.05^{c}$ | 5.40 $\pm 0.00^{c}$ | 5.37 $\pm 0.05^{bc}$ | 5.33 $\pm 0.05^{bc}$ | 5.30 $\pm 0.17^{bc}$ | 5.20 $\pm 0.05^{ab}$ | 5.10 $\pm 0.05^{a}$ |
| Turbidity (NTU) | 28.83 $\pm 1.31^{a}$ | 29.26 $\pm 0.66^{a}$ | 31.48 $\pm 0.99^{a}$ | 38.67 $\pm 0.91^{a}$ | 80.50 $\pm 6.87^{b}$ | 153.83 $\pm 27.49^{c}$ | 290.17 $\pm 39.93^{d}$ |
| Total Microbes (Log CFU/mL) | 3.46 $\pm 0.08^{a}$ | 4.30 $\pm 0.08^{b}$ | 5.78 $\pm 0.01^{c}$ | 7.00 $\pm 0.04^{d}$ | 7.97 $\pm 0.15^{e}$ | 8.29 $\pm 0.05^{f}$ | 8.53 $\pm 0.07^{g}$ |

A-G: Means with different letters in the same row during the storage period are significantly different at $P < 0.05$.

According to Yin, coconut water is classified as fresh if its turbidity value is < 50 NTU [32]. Based on these standards, it can be said that at 12 h of storage, coconut water is still fresh and suitable for consumption, but after 12 h, it is not suitable for consumption as in 16 h with 80.50 NTU, 20 h with 153.83 NTU, and 24 h with 290.17 NTU. This is supported by statistical results that showed no significant difference in storage time from 0 to 12 h. The cause of the increase in turbidity in coconut water is that the coconut water itself contains polyphenol oxidase (PPO) and peroxidase (POD) enzymes, which, when in contact with oxygen continuously, change the clear color to cloudy, then yellow to brown [35]. There is also a white precipitate that results from protein denaturation [36]. This increase in turbidity indicates a decrease in freshness as well as deterioration of coconut water because the higher the turbidity value, the more pungent the aroma and sour taste.

The results showed that there was a significant increase in total microbes every 4 h of storage starting from 3.46 Log CFU/mL to 8.53 Log CFU/mL. Based on the SNI (4268:2020), the maximum limit of total microbial contamination in coconut water was 6 Log CFU/mL. coconut water stored for 8 h can be categorized as fresh or safe for consumption because the total microbial contamination is still below the standard, but at a storage time of 12–24 h it is not suitable for consumption. The increase in total microbes is because coconut water contains many carbohydrates, especially simple sugars such as glucose, fructose, and sucrose, which are growth substrates for microbes, especially groups of lactic acid bacteria and yeasts that grow in coconut water. Storage for 0–12 h showed a significant increase in total microbes due to microbial growth substrates, but when entering storage for 16–24 h the increase in total microbes was not as significant as before because the sugar substrate began to decrease accompanied by acidic conditions, indicated by a decrease in pH and an increase in total acid. Reduced substrate conditions and increased acidic conditions inhibit microbial growth to be inhibited [24].

Food spoilage is characterized by the fermentation of glucose by lactic acid bacteria that produce organic acids such as lactic acid and acetic acid [27]. $CO_2$ gas is a by product of food deterioration due to increased microbial growth and respiration [37]. Based on this, carbon dioxide ($CO_2$) and oxygen ($O_2$) gas testing were performed to prove that the damage to coconut water during storage was caused by microbial fermentation reactions. The results obtained are shown in Fig. 3, showing an increase in $CO_2$ gas every 4 h of storage up to 24 h of storage, ranging from 0.0622% to 0.4398%. The increase in $CO_2$ gas concentration followed a logarithmic equation curve characterized by the highest increase in the first 4 h of storage. This is because, at the beginning of storage during preparation, coconut water had direct contact with oxygen gas so that it would produce higher $CO_2$ gas than other storage when it was in a sealed or anaerobic state. This is supported by the statement Shaalan et al. that the high concentration of $CO_2$ gas formed is a sign that a product is spoiled [38]. The results of the $O_2$ gas measurement decreased as the storage time increased from 25% to 0% after 16, 20, and 24 h of storage. This indicates that during the storage of coconut water, there is no contact with oxygen gas, and the color change on the freshness indicator label is influenced only by changes in the $CO_2$ gas concentration.

### 3.3. Freshness indicator label color change profile

Freshness indicator labels were applied to coconut water packaging using three different indicator color solutions: phenol red, bromothymol blue, and methyl red. The three indicator color solutions had different color changes depending on the pH conditions of the surrounding environment. This color change can provide direct information about the freshness of and damage to stored coconut water. Determination of the color change of the freshness indicator label using a colorimeter that provides results in the form of L*, a*, and b* values according to Hunter's Lab Colorimetric System standards, and then converted to °Hue values to determine the resulting color (Fig. 4).

The °hue values obtained can be described by color using the table below [39]. The color change on the phenol red freshness indicator label at the time of application in coconut water was characterized by the formation of three phases of color change, namely purplish red to orange and then to yellow (Fig. 4a). These three phases of color change can be seen with the °Hue value, which was

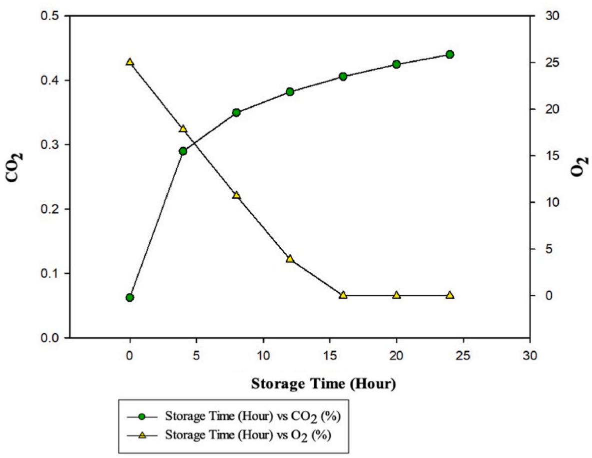



**Fig. 3.** $CO_2$ & $O_2$ gas concentration of coconut water during 24 h storage.

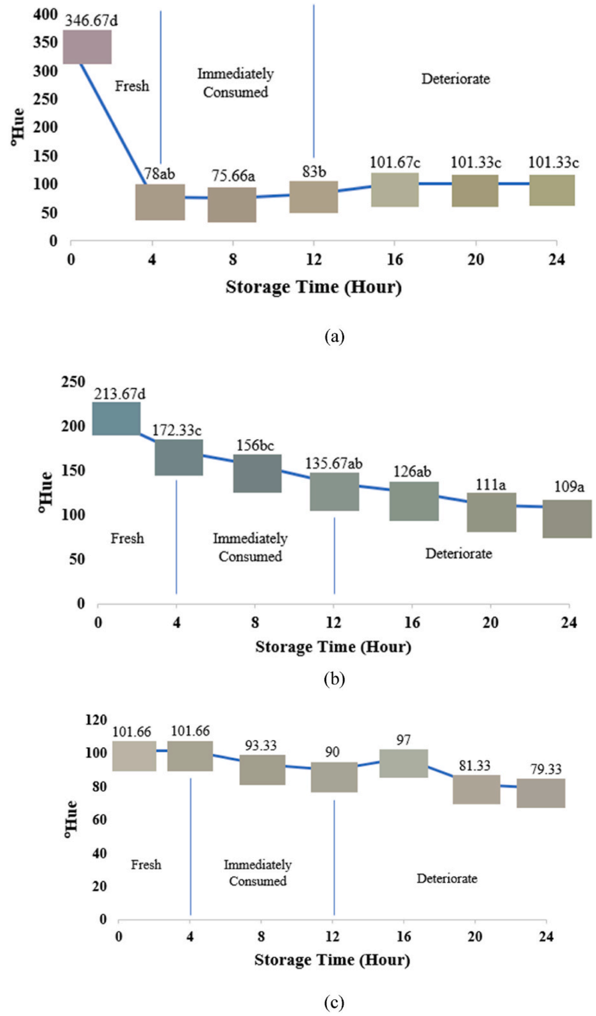



**Fig. 4.** Results of °Hue label freshness indicator on coconut water during 24 h storage phenol red (a), bromothymol blue (b), methyl red (c).

initially 346.67 at 0 h storage, then decreased to 75.66–83 at 4–12 h storage and increased again to 101.33 at 24 h of storage. For the freshness indicator label, bromothymol blue provides a color change of four phases in coconut water during storage (Fig. 4b). The initial color formed is greenish blue with °Hue 213.67, then turns green with °Hue 172.33 at 4 h storage which then turns greenish-yellow with °Hue decreasing from 156-126 at 8–16 h storage and ends with a yellow color that has °Hue 109 at 24 h storage. The methyl red freshness indicator label only gives two phases of color change (Fig. 4c), namely yellow with °Hue values of 90–101.66 during storage for 0–16 h and reddish yellow color with °Hue values of 79.33–81.33 during storage for 20–24 h. The formation of several phases of color change on the freshness indicator label during storage indicates that the freshness indicator label is sensitive to changes in the condition of damaged coconut water. According to Donsingha & Assatarakul, microbial and biochemical parameters, such as turbidity, are the main parameters that can be used to determine the freshness and acceptability of coconut water to consumers [40]. The results obtained on the total microbial parameters and turbidity show that coconut water can still be consumed up to 12 h of storage, while at 16–24 h of storage, it is declared damaged or unsuitable for consumption. Therefore, the phenol red freshness indicator label has the best color because three phases of color change can show that the purplish red color at 0 h storage indicates that the coconut water is still fresh, orange color indicates that the coconut water is immediately consumable at 4–12 h storage, and yellow color at 16–24 h storage indicates that the coconut water has been damaged.

Discoloration of the freshness indicator label occurs because of changes in air composition in the headspace of coconut water packaging due to the accumulation of $CO_2$ gas along with the length of storage time. The formation of $CO_2$ gas during storage is an indicator of deterioration in the quality of coconut water. As in previous research by Nopwinyuwong et al. [27], the indicator label changes color from light green to bright red as the $CO_2$ gas concentration increases from 0 to 2.5%. $CO_2$ gas is soluble in water (1449 mg/L) and can form carbonic acid compounds ($H_2CO_3$) [27,41]. Carbonic acid ($H_2CO_3$) is a diprotic acid compound that can release two acid ions, hydrogen ions ($H^+$) and bicarbonate ions ($HCO_3^-$), with a pKa value of 6.36 (25 °$C$) [37]. Hydrogen ions as proton compounds can react with water molecules to form hydronium ions ($H_3O^+$), which can react with color indicators to form acidic conditions (HIn), thereby changing the color of the indicator label [42,43].

Determination of the best freshness indicator label is based on not only the °hue value but also the conversion of L*, a*, and b* values in the form of total color change (ΔE). ΔE is a value that can directly determine the difference between two colors, which is based on the color change of the freshness indicator label every 4 h–24 h of storage against the control color, namely the freshness indicator label at 0 h of storage. Based on the ΔE values obtained from the three types of freshness indicator labels during storage (Fig. 5), the phenol red freshness indicator label had the highest value at each storage time ranging from 14.07 to 26.14 compared to the bromothymol blue freshness indicator label, which had ΔE values ranging from 8.89 to 20.58, and the methyl red freshness indicator label with ΔE values of 6.89–11.82. The perception standard of the ΔE value uses five scales namely:1.0 (invisible to the human eye), 1–2 (visible to focused observation), 2–10 (visible at a glance different), 11–49 (visible different from the initial color), and 100 (visible color opposite to the initial color) [6]. Furthermore, Fig. 6 illustrates how the color change of the indicator label on coconut water during 24-h storage correlates with the obtained ΔE value. Thus, the phenol red freshness indicator label can be used as an intelligent packaging that can easily distinguish the freshness level of coconut water during storage. This is supported by a statement from Zhang & Lim, 2018, that the color indicator with the best level of sensitivity in response to $CO_2$ gas must have a pKa value of $CO_2$ soluble in water greater than 6.35 [37]. Phenol red has a pKa of 7.4; methyl red has a pKa of 4.95; bromothymol blue has a pKa of 7.0 [12,37]. In addition, color indicators with sulfonephthalein structures were better for detecting $CO_2$ gas.

### 3.4. Chemical structure of the freshness indicator label

Chemical characterization of freshness indicator labels was carried out using an FTIR instrument to determine changes in the chemical groups of the indicator labels during the storage process. The FTIR spectra of the phenol red freshness indicator labels at the beginning of storage and after storage are shown in Fig. 7. At the beginning of storage, the phenol red freshness indicator label showed a broad peak at 3360 $cm^{-1}$, indicating the presence of OH groups. In addition, peaks also appeared at wavelengths of 1586 $cm^{-1}$ and 1051 $cm^{-1}$ indicating the presence of C═C and C–O groups. The clusters that appear in the initial conditions of the freshness indicator label are in accordance with the results of this study by Oliveira et al. that the basic component of the label is methylcellulose [44]. After storage for 24 h, the phenol red freshness indicator label showed a spectrum that was almost the same as that at the beginning of storage, characterized by the presence of peaks at wavelengths of 3350 $cm^{-1}$ (OH group), 1582 $cm^{-1}$ (C═C group), and 1061 $cm^{-1}$ (C–O group) with different intensities. However, there are differences in the peaks that are clearly visible on the phenol red freshness indicator label that has been stored based on the results of the FTIR spectrum, namely at a wavelength of 2356 $cm^{-1}$ indicating the presence of O═C═O groups, especially carbon dioxide. The existence of a sharp peak at a wavelength of 2356 $cm^{-1}$ indicates that the color change of the freshness indicator label is caused by the binding of carbon dioxide gas which has weak acidic properties as a result of damage to coconut water during storage.

### 3.5. Correlation of freshness indicator label color with coconut water quality

The correlation between the color change of the freshness indicator label and all testing parameters of coconut water, such as pH, total acid, total dissolved solids, turbidity, total microbes, and carbon dioxide gas, was conducted to determine the sensitivity of the freshness indicator label in detecting the freshness of coconut water. This aims to determine the synchronization of changes in the color of the indicator label with the decrease in freshness of packaged coconut water during storage.

Based on Fig. 8a–c, it can be seen that pH and total dissolved solids decreased during storage, while total acid, turbidity, total microbes, and carbon dioxide gas continued to increase. These changes were captured by the decrease and increase in the degree of the freshness indicator labels. Coconut water is a beverage with various nutritional contents, such as vitamins, minerals, and sugars, such as sucrose, fructose, and glucose [45]. The high sugar content in coconut water reduces its freshness of coconut water during storage

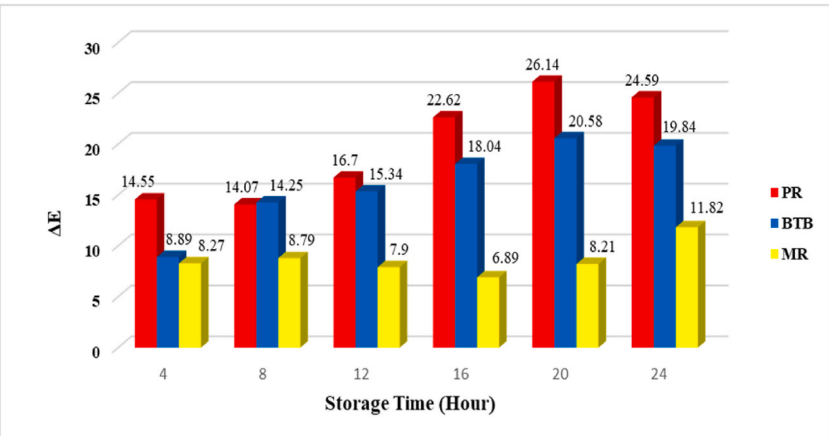



**Fig. 5.** Results of ΔE freshness indicator label on coconut water during 24 h storage.

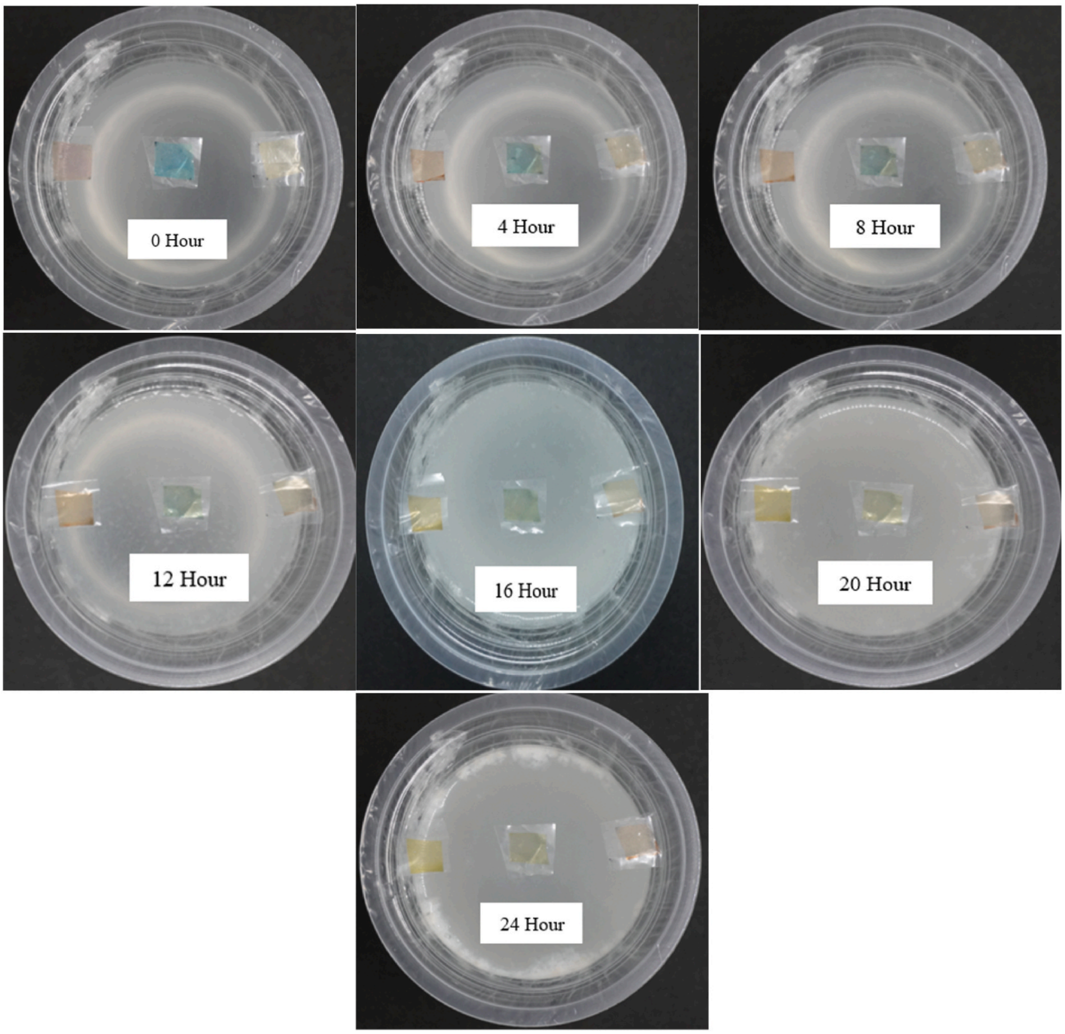



**Fig. 6.** Visualization of freshness indicator label phenol red (left), bromotymol blue (center), and methyl red (right) on coconut water during 24 h storage.

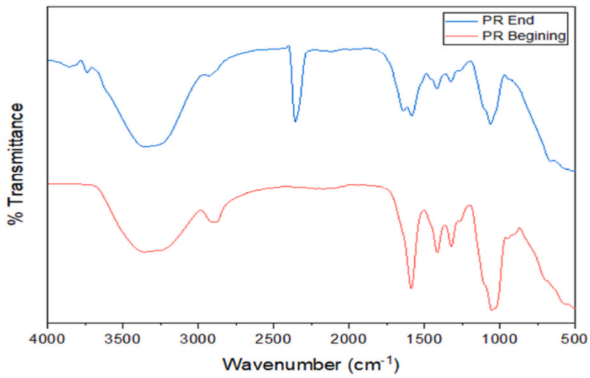
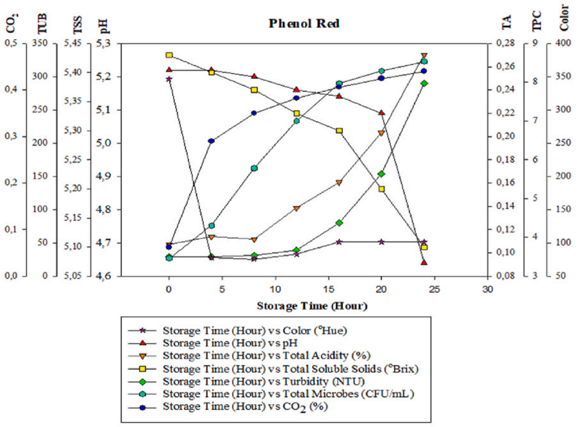
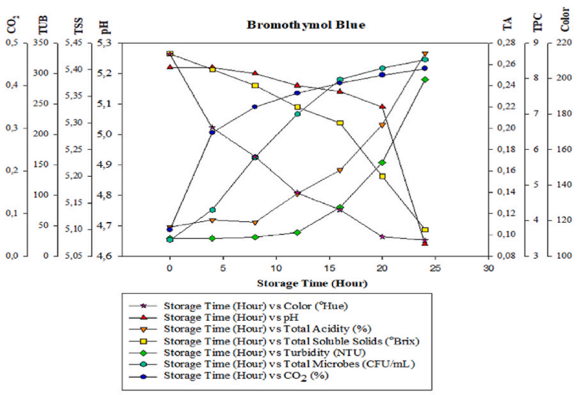



**Fig. 7.** Phenol red freshness indicator label spectrum results.

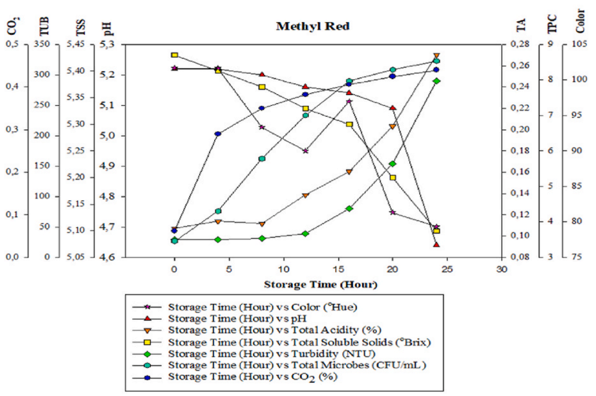
because it can be a good growth medium for microbes, which can be seen with the anaerobic fermentation process that causes carbon dioxide gas as a by-product of coconut water damage. During the fermentation process, the pH decreases to acidic due to the formation of acidic compounds from fermentation, such as lactic acid and acetic acid, as well as total soluble solids such as glucose, sucrose, and




(a)




(b)




(c)

**Fig. 8.** Correlation between Change in °Hue of Freshness Indicator Label and Overall Parameters of Coconut Water during 24 Hours phenol red (a), bromothymol blue (b), methyl red (c).

fructose, which will decrease due to microbial breakdown so that coconut water will decrease in sweetness during storage. The increase in acid compounds due to the breakdown of sugar compounds causes an increase in total acid content in coconut water during storage, which is also the cause of increased turbidity because acid compounds can damage proteins in coconut water, which causes the formation of white deposits apart from the enzyme activity of polyphenol oxidase (PPO) and peroxidase (POD) [36]. In addition, the total number of microbes that continue to increase in line with the increase in carbon dioxide gas, because at room temperature, the sugar content of coconut water microbes such as yeast and lactic acid bacteria are easy to grow because they fulfill their energy sources so that there will be a breakdown of these sugar compounds into simple compounds such as organic acids and by-products in the form of carbon dioxide gas, as evidence that the decay process occurs in the form of anaerobic fermentation. This is in line with Carmago et al., 2015 that the occurrence of increased microbial growth depends on the composition of sugar present in coconut water, and reduced sugar components and increased acidic conditions can slow microbial growth [24]. Overall, the six parameters can provide information about the deterioration of coconut water, but not all parameters have a match between the value of the standard and the direct reality of coconut water products that have been damaged. The parameters that can be used as the main reference and are in accordance with the standard are turbidity and total microbes, which are still safe up to 12 h of storage. This is supported by its suitability for carbon dioxide gas production, which will be captured by the freshness indicator label.

Based on the change in the °hue value of the freshness indicator label for all parameters, phenol red and bromothymol blue freshness indicator labels showed better changes compared to methyl red. This can be observed from the values as well as the visual colors displayed (Fig. 6). The methyl red freshness indicator label only provides a yellow to reddish yellow color change that is not very clear and does not match the freshness condition of the packaged coconut water. The bromothymol blue freshness indicator label provides a color change from greenish blue to green, greenish yellow as the last limit of safe consumption, and yellow as a sign that coconut water has been damaged. However, the formation of these four-color phases makes the bromothymol blue freshness indicator label less well used because it can cause incorrect perceptions of consumers when compared to the phenol red freshness indicator label, which has three color phases, namely purplish red at 0 h storage, meaning fresh, orange color at 4–12 h storage, meaning immediate consumption, and yellow color at 16–24 h storage, meaning it is damaged. These three-color phases are more easily distinguished by consumers regarding the freshness of coconut water, which is supported by the higher $\Delta E$ value than the other two freshness indicator labels. This is similar to the results of Nopwinyuwong et al. who showed that the three color phases of the indicator label, namely green (fresh), orange (consumed immediately), and red (spoiled), show an increase in carbon dioxide and total microbes in food spoilage [27]. And on Obaidi et al., 2022 also showed that the phenol red indicator label produces three color phases, purple, dark pink, and orange, indicating that food spoilage has occurred due to the accumulation of carbon dioxide gas [12]. Based on the results displayed in one graph, the phenol red freshness indicator label can be used to assist consumers in determining the freshness level of coconut water products with clear visualization without the need to open the packaging or smell the aroma.

## 4. Limitations

The identification of acidity using an indicator does not align with detecting the concentration of $CO_2$ released from coconut water due to the complex interplay of the carbonic acid system's multiple equilibria in water at various pH levels. Moreover, while an indicator can detect acidity, it may not accurately mirror the acidity present in coconut water. Therefore, a direct measurement of coconut water acidity, such as employing a wooden stick infused with the indicator in direct physical contact with the water, would have been a preferable approach.

## 5. Conclusions

Methylcellulose based phenol red freshness indicator labels provide the best three-color change profiles that match the freshness conditions of coconut water, namely purplish red (fresh), orange (consumed immediately), and yellow (damaged); thus, they can be used as intelligent packaging to monitor the quality of coconut water compared to bromothymol blue, which provides four phases of color change, and methyl red, which provides two phases of color change.

## Data availability statement

Available data are presented in the manuscript.

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## Ethics declarations

Not applicable.

## CRediT authorship contribution statement

**Andi Dirpan:** Writing – review & editing, Supervision, Conceptualization. **Jumriah Langkong:** Supervision. **Amran Laga:** Supervision. **Muspirah Djalal:** Writing – review & editing, Writing – original draft, Conceptualization. **Matthew Khosuma:** Visualization, Investigation, Data curation. **Nandita Irsaulul Nurhisna:** Supervision, Resources. **Meysi Azkiyah:** Supervision, Methodology.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:Andi Dirpan reports financial support was provided by Hasanuddin University, Indonesia. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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