Utilization of areca nut (Arecha chatechu L.) extract for tannin based colorimetric indicator in smart packaging

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Abstract. Areca nut contains tannin which has a great potential to apply as natural colour agent in food industries. Tannin offers specific colour and alter its colour due to environmental sensitivity. This study aims to fabricate a tannin-based colour indicator from areca nut in smart packaging in the form of a strip type and to characterize the indicator at different storage conditions and at various pH solutions. The indicators were synthesized using filter paper (No.1 and No.42) soaked in a solution with 1, 3, and 5% areca nut ethanolic extract. Then the indicators were stored at room temperature and 4-7°C for 10 days, then their Red Green Blue (RGB) coefficient values were measured. Characterization of the indicators at pH 3-10 were also determined by RGB coefficient. The results showed that the indicator stored at 4-7°C had more stable RGB coefficient than the indicator stored at room temperature indicated the indicator was influenced by the temperature factor. The indicator offered a potential to be used as a sensor on packaging of temperature sensitive foods. The indicator using Whatman paper No.42 with 1% of extract steadily decreased in RGB coefficient and changed its colour to darker in basic pH solution while the indicator with other treatments had unstable alteration RGB coefficient and colour.

1. Introduction
Smart packaging can monitor food freshness and food quality of the product during transportation and storage. Smart packaging has some functions to control packaged foodstuffs and the environment inside and outside the packaging. Smart packaging can be embedded with a colorimetric indicator which is a label form equipped with colour guidelines. The indicator changes its colour due to chemical or biological alteration in the food product. Colour indicators are applied by indirect contact with food and generally sensitive to environmental changes such as temperature and pH [1]. To produce colorimetric indicator, some natural pigments have been utilized as colorimetric sensor compound, such as betacyanin [2] and anthocyanin [3].

As one of natural colour, tannins have been used widely as natural dyes, however rarely applied in food industries. Tannins have a yellowish white to light brown colour and darker colour when exposed to direct sunlight [4]. Tannin extract from Areca nut (Areca catechu L.) has commonly been used as a...
raw material for textile colouring. Moreover, areca nut availability is abundant and spreads almost all over Indonesia, especially on Sumatra Island, becoming a leading plantation commodity in Aceh Province. BPS Aceh [5] stated that the total area of areca nut plantations in Aceh Province was 40,681 ha. Areca nut is widely used as traditional medicine, preservative, cosmetic raw material, and clothing dye [6]. Although areca nut is edible, utilization of tannin extract from areca nut in food industries is still lack and requires more studies to broaden application of areca nut extract in the food industries. According to Wulansari et al. [7], areca nut contains tannin compound that can be used as a natural red colour in food. The tannin content in areca nut reaches 6.45% by water solvent extraction and 8.53% by ethanol solvent extraction [8]. According to its colour properties and abundant source in Indonesia, tannin extracted from areca nut has a great potential to be applied as colorimetric indicator compound in smart packaging for food products.

Some reports showed that colorimetric indicator can be synthesized in simple method by dipping and soaking filter paper, frequently filter paper No.1 and 42, in colour solution [3, 9, 10] resulting a strip-type indicator. However, currently studies have not provided any information about concentration of tannin extract in fabrication of colour indicators. Different concentrations of colourant can produce different characteristic of colour indicators. Therefore, this study aims to synthesize strip-type colorimetric indicators from areca nut extract with adding different concentration of the extract and using a different grade of filter paper. Moreover, characterization of the indicators was carried out at various pH and under different storage conditions.

2. Materials and methods

2.1. Materials

This study used areca nuts obtained from Aceh Besar District, Aceh Province, Indonesia, Whatman filter paper No. 1 and 42, and absolute ethanol (Merck) as extraction solvent.

2.2. Extraction of tannin

The tannin extraction from areca nut was conducted by ethanol solvent extraction. The areca seeds were separated from their skin and shell, then the seeds were dried under sunlight. Further the seeds were crushed and sifted using 16 mesh sieves to produce areca nut powder. Then 50 g of the powder was put into a flask, added 250 ml of ethanol. The extraction was carried out at 60°C and 1500 rpm for 5 hours. The solvent was evaporated by rotary vacuum evaporator, followed by oven drying at 60°C to obtain powder of the tannin extract [8, 11].

2.3. Analysis of areca nut extract

2.3.1. Analysis of tannin content. Tannin content was analysed by titrimetric method titrating the extract with standard potassium permanganate solution [12].

2.3.2. Analysis of phenolic content. The phenolic content was measured using the Folin-Ciocelteau method using UV-VIS spectrophotometer at a wavelength of 750 [13].

2.4. Fabrication of tannin based colorimetric indicators

Strip type colour indicator fabrication was conducted by dipping the filter paper in tannin extract solution. Firstly, the extract powder was dissolved in water at 90°C with respective concentration of 1%, 3% and 5%, then each solution was homogenized using a stirrer for 10 minutes. Further Whatman filter papers (grade No.1 and No.42) were respectively cut into 2 cm x 2 cm and soaked in the tannin extract solutions for 12 hours at room temperature. After 12 hours, the paper were drained and placed in petri dishes, then dried at room temperature for 3 hours [14].
2.5. Characterization of colorimetric indicator.

2.5.1. Storage of the colorimetric indicator. The indicator was placed in a transparent container, then covered with plastic wrap and stored at room temperature and in fridge at 4-7°C for 10 days. The sample at room storage had lamp light exposure during the day but the light was off at night. The fridge was opened only for picking up the sample. The colour change in the indicator was determined by Red Green Blue (RGB) coefficient using Adobe Photoshop.

2.5.2. pH solution. The pH test was aimed to observe colour change of the indicator in base and acidic solution. The test was carried out by preparing each pH solution of 3 to 10, then the indicators were respectively immersed in the solution. Red Green Blue (RGB) coefficient using Adobe Photoshop was applied to determine colour alteration of the indicator.

3. Results and discussion

3.1. Content of tannin and phenolic compounds
Tannin content in this study were 2.41% (Table 1) which is lower than the previous studies [8] and [11]. Tannin content in areca nut seeds have different contents which can be influenced by planting location and climatic factors [8]. Percentage of tannin in areca nut can affect the yield of extraction and the extract properties that the higher the tannin content thus colour of extract is darker [15]. In addition, the phenolic content of the extraction was 128.67 mg GAE/g (Table 1). The content of tannins and phenols in areca nut extract powder effect the solubility level of areca nut extract powder in polar solvents. The higher the tannin then the more soluble areca nut extract powder in polar solvents.

| Parameters                 | Content |
|----------------------------|---------|
| Tannin (%)                 | 2.41    |
| Polyphenol (mg GAE/g)      | 128.67  |

3.2. Characteristic of the colour indicator during storage

Brightness of the colour indicator can be indicated from RGB coefficient value, where the higher the RGB coefficient thus the brighter colour of indicator and vice versa. Figure 1 exhibits different colour of the indicator using different grade of filter papers and concentration of tannin extract solution. It shows that the indicator using paper No.1 is darker than the indicator of paper No.42 at same level concentrations of tannin extract. However, RGB coefficients of the indicator using paper No.1 are close to RGB coefficients of the indicator using paper No.42 (Figure 2, at 0 day). Novitasari and Barik [16] stated that colour of the indicator using Whatman paper No.1 was brighter than the indicator using paper No.42. Colour indicators have different colours and coefficient values at each concentration, where the higher the tannin concentration in solution, the darker the colour solution. Furthermore, if the concentration of tannins in the solution is higher, then filter paper can absorb more tannins. According to their colour in Figure 1 and RGB coefficient value on 0 day (Figure 2 and 3), Whatman paper No.1 absorbed more tannins, consequently it has darker colour and lower RGB coefficient value than the indicator from Whatman paper No.42.

In general, the colour indicator at room temperature during storage for 10 days experiences a decrease in the RGB value at each concentration, although there is an increase in the RGB value on day 3 to day 5 in some concentrations, in further the RGB coefficient values decrease (Figure 2). A few fluctuations of the RGB value indicates unstable alteration of colour from bright to dark. This shows that environment including temperature and room lighting during storage effect the indicator and consequently colour of the indicator changes to darker. In comparison, at low temperature of 4-7°C, RGB coefficients of the indicators are steadier over the storage time (Figure 2). This shows that at
low temperature the indicator more stable and has low impact from environment. The tannin compound in the indicator is sensitive to the environment as light exposure and temperature change, following by the tannin colour alteration to a darker colour which is showed by darker colour of the indicator. In addition, exposure of tannin to direct light and higher temperature can lead to tannin oxidation thus visually the light brown tannin colour turn darker.

Figure 1. The colorimetric indicators in different treatment.

Figure 2. RGB coefficient of the colorimetric indicator during storage at room temperature (G1: Whatman paper No.1, G2: Whatman paper No.42; T1: 1%, T2: 3% and T3: 5%).
3.3. Characteristic of the colour indicator in pH solution

In general, the indicators have fluctuation value of RGB coefficient over pH (Figure 4). Colour of the indicator using Whatman paper No. 1 in concentration of 1% and 5% alter their colour to brighter from pH 3 to 4. However, at pH 4 to 6 there was a colour change to become darker, which is shown by the RGB coefficient decrease. Colour of the indicator at acidic pH is lighter than in base pH, although the colour alteration is not steady. The indicator using Whatman paper No.1 concentration of 1% steadily declines its RGB coefficient value over the pH. The colour of tannins will be darker by increase pH conditions [17]. Colourant can change its colour depends on pH, such as phenolphthalein indicator which became colourless until pH 8 and turned to red colour at pH 10. Then in strong base solution, the phenolphthalein turned back colourless. This colour alteration occurred because of compound reaction by an acid-base reaction [18].
4. Conclusions
The tannin-based colorimetric indicator using paper No.1 is darker than the indicator of paper No.42 at same level concentrations of tannin extract and higher concentration of tannin extract addition resulting in lower RGB coefficient and darker colour of the indicator. The indicator has steadier RGB coefficient value during storage at 4-7°C than at room temperature. The indicator using Whatman paper No.42 with 1% of extract concentration steadily decreased in RGB coefficient. It changed its colour to darker in pH solution while the indicator with other treatments had unstable alteration RGB coefficient and colour. These results showed a great chance to develop a tannin-based colour indicator from areca nut with further application testing in food packaging.

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