Antimicrobial properties colorimetric film of Damask Rose and freshness monitoring: A review

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Abstract. This work aims to investigate the antimicrobial properties of colorimetric film using different types of anthocyanins on different types of microorganisms such as Escherichia coli, Staphylococcus aureus, Bacillus cereus, Listeria innocua, Pseudomonas aeruginosa and other types of microorganisms. Most of colorimetric film with addition of anthocyanins exhibits the antimicrobial properties which could prolong the shelf life of food products. Moreover, the methods to extract anthocyanins using different kinds solvent such as ethanol, methanol, tetrahydrofuran and others solvent also been studied. Finally, the effect of colorimetric films such as colour changes to monitor the freshness on different kinds of food products also been reported.

1. Introduction
According to (Pineros-Hernandez et al., 2017), the world production of plastic has shown the continuous growth for more than 50 years. It is estimated that the production of plastic increased by 300 million tons. 39.6% of the total of plastic demand comes from the packaging application and contemplate as the biggest market for the plastic industry. The usage of plastic from petroleum derivation can pose threat to environment and safety of consumer (Zheng et al., 2019). In order to overcome these problems, the researchers have developed edible packaging and recently smart packaging have been attract the attention of the researcher because of it provides information to manufacturers, retailers and consumers with safe, simple and fastest manner (Dong et al., 20). PH based colorimetric sensing film is one of part of smart packaging which deliver the information on the freshness of the products by showing the color changes on the films. The application of colorimetric film to monitor the freshness of shrimp (Kang et al., 2020, Dong et al., 2020, Merz et al., 2020), fish (Jiang et al., 2019, Zhai et al., 2017, Huang et al., 2019), meat (Ezati et al., 2019, Zhai et al., 2020) and milk (J. Liu et al., 2019, B. Liu et al., 2017) has been developed.

In order to develop the colorimetric film, the anthocyanins from flower, fruits and vegetables are widely used because of a few factors such as safe to consumer and pH sensing characteristic such as those from red cabbage (Pourjavaher et al., 2017, Freitas et al, 2020, Chu et al., 2020), purple sweet potato (Jiang et al., 2020, Chen et al, 2020, Choi et al., 2017), mulberry (Jiang et al., 2019), roselle (Zhai et al., 2017), and black rice bran (Ge et al., 2020). Our main concern is anthocyanins that extracted from rose petals (damask rose) to make the colorimetric film for monitoring purpose and there is limited research on the colorimetric film from rose anthocyanins. (Kang et al., 2020) stated that rose is rich with anthocyanins that it have a potential as pH sensing indicator for the development of colorimetric film.

According to (Park et al., 2016), rose petal and roots contains organic molecules that can resist the microbial attack in wild environments. The extracts of methylene chloride or methanol from red,
white and yellow rose petals can display antimicrobial properties to certain bacteria and fungi. Besides that, anthocyanins not only exhibit attractive color in nature but it also shows benefits in physiological health and prevent from diseases. This is because it was revealed that the anthocyanin compounds have have high radical scavenging capacity which can prevent the cardiovascular disease, diabetes, cancer and many more (Q. Ge & Ma, 2013).

In this study, the main objectives is to compare the anthocyanins from rose extract with other types of anthocyanins in terms of method use to extract the anthocyanins because this steps is very important to determine the function of colorimetric film. The antimicrobial properties of colorimetric film of different types of anthocyanins also been compared and lastly the effect on film in application of colorimetric film on certain products such as shrimp, fish, meat and milk to monitor the freshness are also being studied.

2. Methods of anthocyanin extraction
Extraction process is a crucial steps which can determine the antioxidant activity in the films. The extraction of anthocyanins from flowers has been done by several steps, including using different solvents, temperature, extraction methods and times. Generally, ethanol has been used in the making of film to extract the anthocyanins. The solvent selection affected by the particular nature of bioactive compound measured and other solvent such as methanol, ethanol, acetone, ether, isopropanol, water and the mixture of solvent also been used for extraction (Fernandes et al., 2017). Basically, temperature is crucial parameter that affect the extraction process whether it is use high temperature or low temperature. The extraction process usually been done at room temperature which requires at least 12 hours while requires less amount of time when extract in high temperature.

According to (Vankar & Bajpai, 2010), a favorable method to extract the anthocyanins from rose flowers is using methanolic solution and consist 3% of citric acid that gives better amount of anthocyanins extraction than using the methanolic solution containing 0.1% of hydrochloric acid. It was shown that the the pH changing in the extract was shown from pH 2-9 and the color changes from dark pink to green. Because of its variation change of colour due to different pH it is considered as the indicator in the acid-base titration.

2.1. Anthocyanin extraction using ethanol
The most common method to extract anthocyanins from plants are by using absolute ethanol. For example, the extraction of anthocyanins from rose was shown by (Kang et al., 2020). Firstly, the rose was dried at temperature of 40 °C for 16 hours and pounded into powder. In dark condition, the powder was mixed with absolute ethanol at temperature of 40 °C. Then, the mixture was filtered and the filtrate was dried at 40 °C under dark condition to obtain the rose anthocyanins powder after the 10 hours of extraction. After that, the filter residue was extracted again and lastly the rose anthocyanins powder was dissolved with water at 30 °C in dark condition. After stirring for six hours, the solution was filtered, diluted and stored at temperature of 4 °C. This method also was performed by several researchers to extract the anthocyanins from plants (Dong et al., 2020). Aqueous ethanol acidified with HCl (Jiang et al., 2020) and acetic acid (J. Liu et al., 2019) also was used to extract the anthocyanins.

2.2. Anthocyanin extraction using methanol
This method was performed by (Mohammadalinejad et al., 2020) by using aqueous methanol to extract E. amoenum with a few modifications. The flower was ground into powder before added into aqueous methanol and stirred for 30 minutes at 30 °C. After that, the methanolic extract containing E. amoenum was filtered and concentrated by a rotary drier. Then, the extract was stored in dark condition and refrigerated for next analysis.

2.3. Anthocyanin extraction using tetrahydrofuran
Certain extraction using tetrahydrofuran as solvent as performed by (Li et al., 2014). The flower was ground into fine particles and extracted with tetrahydrofuran at 30 °C for 30 minutes in a shaking water bath. The sample was centrifuged and the supernatant was collected as well as extracted twice for further analysis. The mixture was acidified with HCl and to remove fatty acid the mixture is extracted five times with hexane. Then, the combined extract was evaporated to dryness and kept at -20 °C.

3. Anthocyanin as pH indicator in colorimetric film

Anthocyanins is the most common and available acid or base indicator. It is a plant pigment which makes the plants becomes the color that they are now become such as red cabbage is purple and poppies is red. The indicator change color from red to purplish in alkaline solution and to green in alkaline solution. There are a few types of colorant which present in the anthocyanin which are cyanidin, pelargonin and peonidin. Figure 1 shows the colorant that present in rose flowers. The application of anthocyanins to monitor the freshness of products is shown in Table 1.

![Image of anthocyanin colorants]

**Figure 1.** Types of colorant present in rose flower
Figure 2. The color of rose anthocyanin at different pH

Table 1. Application of anthocyanins to monitor the freshness of products

| Polymer used             | Source of anthocyanins | Types of products       | Reference                      |
|--------------------------|------------------------|-------------------------|--------------------------------|
| PVA/okra mucilage polysaccharides | Rose                   | Shrimp                  | Kang et al., 2020              |
| Pulp                     | Arnebia euchroma       | Shrimp and pork         | Dong et al., 2020              |
| PVA/chitosan             | Syzygium cuminii       | Shrimp                  | Merz et al., 2020              |
| Bacterial cellulose      | Echium amoenum         | Shrimp                  | Mohamadalinejhad et al., 2020  |
| PVA/gelatin              | Mulberry               | Fish                    | Jiang et al., 2019             |
| PVA/starch               | Roselle                | Fish                    | Zhai et al., 2017              |
| Agar                     | Arnebia euchroma root  | Fish                    | Huang et al., 2019             |
| PVA/starch               | Purple sweet potato    | Fish                    | Chen et al., 2020              |
| Carboxymethyl-cellulose/starch | Purple sweet potato   | Fish                    | Jiang et al., 2020             |
| Gelatin/oxidized chitin nanocrystal | Black rice bran     | Fish                    | Ge et al., 2020                |
| Cellulose-chitosan       | Alizarin               | Minced beef             | Ezati et al., 2019             |
| Agar/gellan gum/TiO$_2$ nanoparticles | Rose                  | Pork and fish           | Zhai et al., 2020              |
| K-carrageenan            | Lycium ruthenicum      | Milk and shrimp         | J. Liu et al., 2019            |
| PVA/starch               | NA                     | Milk                    | B. Liu et al., 2017            |

NA - Data not Available
3.1. Monitor the freshness of seafood

Anthocyanins from flowers, fruits and vegetables are commonly used to develop colorimetric film due to several factors such as safety factors and pH sensing properties. Recently, many indicators based on flowers, fruits and vegetables have been developed to observe the product’s freshness such as aquatic products dairy product and meat foods. The previous study have found that the presence of rose anthocyanine (RA) incorporated with polyvinyl alcohol/okra mucilage polysaccharide (PVA/OMP) composite film decrease crystalline of PVA and networks by hydrogen bonds among RA, thus enhance the mechanical and barrier properties of the films. The RA exhibit significant color changes in different pH value which range from 2 to 12. This application on shrimps has discovered the prospective of PVA/OMP-RA film in the application of food packaging for monitoring the freshness of aquatic and meat products. From the results, the colour changes on the film was observed. It is found that the colour turn from purple to yellow in 32 hours and the film will turn into dark green indicating that the shrimp is already spoiled (Kang et al., 2020).

Besides that, the freshness monitoring for shrimp and pork are conducted by (Dong et al., 2020). The colour changes in products were observed for five days with varying temperature conditions which is 20, 4 and -20 °C. For shrimp samples, the color changes from magenta to slight purple after day one at 20 °C and at 4°C the color changes from magenta to purple after passing third day. Nevertheless, after passing day five there was no notable changes in color for condition of -20 °C which indicate that the sample are well preserved and no spoilage detected. Similar to shrimp sample, the pork sample change color under 20 and 4 °C conditions. After three days, the film at 20 °C became bluish violet color but condition at 4 °C only change color to purple after five days of storage. Other previous study such as (Merz et al., 2020) also using various anthocyanin from fruits, flower (Mohammadalinejhad et al., 2020), and vegetable (Kang et al., 2020) to observe the freshness of shrimp.

Other than aquatic product like shrimp the the freshness of fish also being monitored. The anthocyanins from mulberry was used in order to evaluate the freshness of fish. The fish was sealed in jar and the film inserted inside the cap and the temperature environment is 25 °C. At the beginning, the colorimetric indicator film exhibit a purple colour and after 18 hours of storage, the film change color to grey purple and finally displayed dark green after 24 hours of storage (Jiang et al., 2020). The color changes of colorimetric films also being displayed by (Zhai et al., 2017). At the beginning the film displayed purple color then change to green color at 90 hours of storage and finally yellow color after 135 hours of storage. A colorimetric film using Arnebia euchroma root extracts (Huang et al., 2019), purple sweet potato (Chen et al., 2020, Jiang et al., 2020) and black rice bran (Ge et al., 2020) also exhibits the color changes in order to evaluate the pH indicator properties on the films.

3.2. Monitor the freshness of meats

In addition, the colorimetric films for meat products are also developed such as (Ezati et al., 2019) which use alizarin to observe the freshness of minced beef. From the results the film changes color from brown to purple after four days of storage and at 4°C which indicate that the meat started to spoiled. On the final day of storage, the highest pH reading was observed because the increase in pH in meat could be due to the release of microbial metabolites or endogenous enzymes that speed up the meat spoilage which cause protein hydrolysis and formation of volatile compounds (ammonia and trimethylamine). The increasing of pH not only cause the acceleration of microbial growth but can affect the quality of products such as the meat becomes darker in color and the textures becomes firm which is not acceptable standard for consumer. The color changes in colorimetric film using rose (Zhai et al., 2020) and roselle (Zhang et al., 2019) also being detected to monitor the freshness of meat.

3.3. Monitor the freshness of milk
The detection of spoilage on dairy products such as milk also can be detected using colorimetric film. (J. Liu et al., 2019) develop colorimetric film using Lycium ruthenicum Murr. (red seaweed) to monitor the freshness of milk and shrimp. Generally, when the pH of milk decreases it indicates contamination by microbial which lead to accumulation of lactic acid by the microbial metabolism. The colorimetric film was grey when expose to fresh milk and there is no obvious changes in colour after 24 hours of storage at temperature of 25 °C. After storage of 36 hours the color change to grayish purple and dark pink after 48 hours which indicates the milk already spoiled. Other research to monitor the freshness of milk also being conducted by (B. Liu et al., 2017).

4. Antimicrobial properties in colorimetric film

Antimicrobial film play a crucial role in food packaging that could guarantee the food preservation and extend shelf life. Antimicrobial packaging is prepared by incorporating it with different biopolymers such as starch, chitosan, PLA, PHB and other types of materials that compatible and acts as antimicrobial agents for the food packaging. If the film is not incorporated with other biopolymer materials it will leads to poor mechanical and barrier properties which lead the shelf life of food packaging is shorter compared to conventional food packaging (Al- Tayyar et al., 2020).

According to (Fernandes et al., 2017), the antimicrobial activity is related to the plants species because of the inhibitory substances that are present for the certain microorganisms. For example, the ayurvedic plants extract was tested on Escherichia coli and Staphylococcus aureus for evaluation of antimicrobial activity. From the results, the antimicrobial activity for both microorganism roughly same although gram positive (Staphylococcus aureus) much higher than gram negative (Escherichia coli) (Floroian et al., 2017). Also the antimicrobial activity was conducted using several plants extract such as guarana seeds, rosemary leaves, cinnamon barks and leaves of boldo-do-chile. The plant extracted by using ethanolic extract and the microorganism tested was Escherichia coli and Staphylococcus aureus (Bonilla & Sobral, 2016). These films showed the antimicrobial properties because of the presence of polyphenol components such as diterpenes, carnosol, carnosid acid. Besides that flavonoids alkaloids and tannins also show the inhibition towards microorganisms.

The tannins from oak bark, red peel grape and white peel grape was used with the proteins in order determine the antimicrobial activity. The inhibitory activity of Escherichia coli and Listeria innocua was found in this study when the proteins was incorporated with the plants extracts while no inhibitory was found in the pure protein (Cano et al., 2020). The tannin can also be obtained using crude rice straw by separation and purification method to obtain tannin (Shi et al., 2020). Other studies such as (Srisod et al., 2018, Zhu et al., 2019) also using tannin to evaluate the antimicrobial activity. The films shows remarkable inhibition towards microorganism due to the presence of flavonoids (Ibarguren et al., 2015) and alkaloids (Nguyen et al., 2020). The application of rose extract into the edible film is still a little and not much research have been conducted in order to determine the antimicrobial properties or rose extract into the film. Nonetheless, several studies have been conducted to determine the antimicrobial properties of rose into several applications. In research conducted by (Yi et al., 2019), the antimicrobial properties were evaluated using several bacteria which are Staphylococcus aureus, Bacillus subtilis, Escherichia coli, Pseudomonas aeruginosa as well as three fungi Aspergillus niger, Rhizopus nigricans and Blastocladiad pringsheimii on rose essential oil (EO), rosa damascene (RD), rosa centifolia (RC), rosa pomponia (RP) and rosa chinensis “crimson glory”(CG). RCEO and RPEO manifested the significant inhibitory activity against all tested microorganisms. RDEO clearly inhibited all the tested microorganisms except P. aeruginose but CGEO only showed significant inhibitory towards S. aureus and B. subtilis. Other previous research also exhibit the inhibition of microorganisms and it is proven that rose plant has the ability to act as antimicrobial materials for a better film (Cantelli et al., 2020, VT et al., 2018, Boukhatem et al., 2013). Table 2 shows the effects of colorimetric film on different types of microorganism.

| Colorimetric | Plant used | Microorganism | Effect on the film | Reference |
|--------------|------------|---------------|-------------------|-----------|
|              |            |               |                   |           |
| film | sm used | The film tested on Escherichia coli Staphylococcus aureus show more than 90% antimicrobial efficiency while E. coli only show 50% antimicrobial efficiency. | Floroian et al., 2017 |
|------|---------|----------------------------------------------------------------------------------------------------------------------------------|----------------------|
| Polymer (PMMA) + bioactive glass (BG) | Ayurvedic extract | Gelatin + Guarana seeds, leaves of boldo-do-chile, cinnamon barks and rosemary leaves | An increase chitosan proportion with incorporation of plant extract increase inhibition zone than pure chitosan while pure gelatin does not show inhibition activity. | Bonilla & Sobral et al., 2016 |
| Caseinate+ gelatin | Oak bark, red peel grape, and white peel grape | Polysaccharide biofilms | The biofilms with the incorporation of crude rice straw shows the significant effect of inhibitory activity on Staphylococcus aureus because there are a few dead cells found in the biofilms and culture medium. | Shi et al., 2020 |
| Whey protein isolate (WPI) + silver nanoparticles | Xylocarpus granatum bark extract (XGBE) | Chitosan (CS) | Untreated cotton and cotton treated with WPI and XGBE shows no antimicrobial activity against either Escherichia coli and Staphylococcus aureus. Only with addition of silver nanoparticles exhibit the antimicrobial activity. | Srisod et al., 2018 |
| Chitosan (CS) | Natural extracts gallnut tannis (GT) | Gelatin + enterocin solution (ES) | The bacterial reduction of CS fiber against Staphylococcus aureus only display 49% which is difficult to meet the requirement of antibacterial textiles standards. But with CS/GT composite film can meet the requirements by high bacterial reduction to 99.7% against Staphylococcus aureus. | Zhu et al., 2019 |
| Polysaccharide biofilms | Crude rice straw | Gelatin | Pure gelatin has no inhibitory effect towards all bacteria but the films added with ES and PL | Ibarguren et al., 2015 |
Bacillus cereus exhibit the inhibition effect on each bacteria. L. monocytogenes clearly inhibited by the films prepared with ES but adding PL did not support the inhibition. Meanwhile, the film that only added ES do not show the inhibition activity towards S. aureus and B. cereus but clearly inhibit similar inhibition by adding the PL into the films.

Staphylococcus aureus

Chitosan Sonneratia caseolaris (L.) Engl. leaf extract (SCELE) Staphylococcus aureus Pseudomonas aeruginosa Chitosan film without SCELE has not displayed significant antibacterial activity. It was found that SCELE revealed stronger antibacterial activity against P. aeruginosa compared to S. aureus.

5. Conclusion
As a conclusion, the antimicrobial properties of colorimetric film from rose extract was discussed and compared with other source of anthocyanins such as ayurvedic extract, crude rice straw, guarana seed, okra and many more. It is proven that with the addition of these anthocyanins can exhibit the antimicrobial activity of the films thus prolong the shelf life of products. Besides that, application of colorimetric films to monitor the freshness of products such as shrimp, fish, meat and milk show positive results by the obvious color changes of the films and can estimate the duration of these products until it is spoiled. Lastly, the method to extract the anthocyanins using different kinds of solvent also been investigated because it is an important steps to evaluate the physical and chemical properties of colorimetric film.

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