Natural wrapping paper from banana (*Musa paradisiaca* Linn.) peel waste with additive essential oils

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Abstract. The research aimed to produce natural wrapping paper from banana (*Musa paradisiaca* Linn.) peel waste with additive essential oils. The method used in this research was alkalization. The delignification process is done with the use of NaOH 4% at the temperature of 100°C for 1.5 hours. Additive materials in the form of essential oils are added as a preservative and aroma agent, namely cinnamon oil, lemon oil, clove oil and lime oil respectively 2% and 3%. Chemical and physical properties of the produced papers are tested included water content (dry-oven method SNI ISO 287:2010), pH (SNI ISO 6588-1.2010), grammage (SNI ISO 536:2010) and brightness (SNI ISO 2470:2010). Testing results of each paper were compared with commercial wrapping paper. The result shows that the natural paper from banana peel waste with additive essential oil meets the standard of ISO 6519:2016 about Basic Paper for Laminated Plastic Wrapping Paper within the parameter of pH and water content. The paper produced also meet the standard of ISO 8218:2015 about Food Paper and Cardboard within the grammage parameter (high-grade grammage), except the paper with 2% lemon oil. The paper which is closest to the characteristic of commercial wrapping paper is the paper with the additive of 2% cinnamon oil, with pH of 6.95, the water content of 7.14%, grammage of 347.6 gram/m² and the brightness level of 24.68%.

1. Introduction

The development of science and education increase the demand for paper which has been growing every year. It is proved by the demand for paper in 2005 that reaches almost 5.60 tons, and it increased to 6.45 tons in 2009. The paper is used for various needs, for instances, to write, to read, or to wrap the food. One of the paper types which is produced in Indonesia is the wrapping paper. Its production number increases from 88.940 to 90.930 tons during 2009 to 2011 [2]. The paper which is commonly used is made of the wood. It then causes the deforestation to be widely spread. Moreover, the pulping process remains using chemical material that is hard to be degraded naturally. The government currently finds it hard to overcome the scarcity of wood as the raw material of paper production in the significant number. As a result, the paper company produces the paper through illegal logging that destroys the nature [3].

The high number of paper demand causes the need for alternative raw material that is other than wood for wrapping paper industry. The production starts with the process of pulp making. The raw material to produce pulp is the cellulose in the form of fiber in which almost all of the plants contain the cellulose that can be used as the raw material of pulp production. One of the plants containing high cellulose is the banana.
Banana (*Musa paradisiaca* Linn.) is one of the tropical fruit thriving throughout Indonesia. It is one of the useful commodities that lives in a short time, is easy to be cultivated and can be harvested along the year. The most widely used part of banana is the flesh of the fruit, meanwhile, the outer skin is only used for animal feed and organic fertilizer. It indicates that the banana peel waste has not been optimized yet.

The data from Institute for Research and Development of Cellulose Industries shows that the banana peel waste contains more fine fibers than those that are made of wood with high cellulose (60-65%), hemicellulose (6-8%) and lignin (5-10%). Meanwhile, the most commonly used softwood to make the conventional raw material of the paper only contains cellulose (41%), hemicellulose (24%) and lignin (27.8%) [4]. It indicates that the cellulose in the banana peel is far higher than that in softwood. Moreover, banana peel only contains lignin (5-10%) that makes the cellulose separation process easier to do. Thus, the banana peel is potentially used to be the raw material of papermaking to reduce the dependence of wood as the raw material of wrapping paper. The use of banana peel waste also increases the optimization of the waste itself by increasing the economic value.

To produce the pulp, lignin that contains in banana peel should be hydrolyzed and separated through a delignification process. Alkalization method is suitable to make the pulp by using the material which is fine and has the short fiber as banana peel [5]. The cooking solution of NaOH is widely used due to its economical factor [6]. Sodium hydroxide is the strong alkali compound. In the process of paper production, the addition of NaOH solution is functioned to dissolve lignin in the pulping process. It also speeds up the process of fiber separation and extraction. Based on the previous research, banana peel cooked in NaOH of 4% for 1.5 hours in 100°C results the natural paper in which the chemical characteristics (pH and water content) resemble the commercial paper very closely [8].

The qualified papers contain not only the fiber but also the additional material [9]. This research uses the additional additive material which is tapioca and essential oil consisting of cinnamon oil, lemon oil, clove oil, and lime oil. Tapioca acts as the sizing agent and adhesive [10]. Meanwhile, the essential oil is functioned to preservative and aroma agent due to the unpleasant smell produced by the alkalization process. The essential oil is proved to be the antibacterial, antifungal, insecticidal properties and antiviral antioxidants. Some of them have also been being used as cancer treatment, food preservative, aromatherapy and perfume [11].

2. Methods

2.1 Tools and Materials

The research used a blender, oven, filter paper, baking pan, rotary evaporator, electric cooker, pipettes, pot, balance scales, sample test, covered cup, furnace, analytical balance, cup clamp, glassware and pH Meters. The materials of this research were Kepok banana peel, NaOH, tapioca, essential oil consisting cinnamon oil, lemon oil, clove oil as well as lime oil, potassium chloride as well as distilled water.

3. Research Procedure

3.1 Pulp Making

The pulp making was carried out by alkalization with NaOH solution of 4% in a 6:1 ratio to banana peel mass. The cooking is done at 100°C for 1.5 hours. After being cooked, it then should be filtered and cleaned with distilled water. The bleaching process using H₂O₂ of 10%.

3.2 Addition of Additive Substance

The addition of additive includes tapioca of 10% and essential oil, consisting cinnamon oil, lemon oil, clove oil and lime oil with the concentration of 2% and 3% of the pulp mass.

3.3 Printing

The process of pulp printing was done in glass plate of 20 x 20 cm. Afterward, the drying process was done in the oven of 40-50°C for 24 hours.
3.4 Chemical Characterizations
The water content test was conducted by the dry-oven method based on SNI ISO 287: 2010. The pH test was done based on SNI ISO 6588-1: 2010 on how to test pH in the water extract-part 1: Cold extract.

3.5 Physical Characterization
Physical characterization was conducted at The Institute of Pulp and Paper. Grammage test was performed based on SNI ISO 536: 2010; meanwhile, the brightness test was done based on SNI ISO 2470: 2010.

4. Result and Discussions
4.1 Paper from Banana Peel Waste
Banana peel can be used as raw material in the making of paper because of its high concentration of cellulose. Mashing process of banana peel is essential as banana peel should be brushed, lacerated, and pushed to expand the fibers. While expanding, the outer layer of the fiber will be decomposed, and the fibrils come out. The process which causes the fiber surface to get larger is necessary to tighten the fiber of the paper. In the process of cooking, the temperature should not exceed 100°C since the high temperature in cooking process will decrease the pulp quality. It then causes the pulp not to last for the long period. Moreover, cooking process will shade the color of banana peel from yellow to dark brown indicating the process of lignin extraction from the cellulose.

The bleaching process was done to remove the lignin contained both in pulp and fiber. It then results in the high and stable brightness. In this process, the $\text{H}_2\text{O}_2$ solution was used due to the characters of hydrogen peroxide which has strong oxidation properties with the bleaching concept of Totally Chlorine Free [12]. Also, hydrogen peroxide also has a weak acid, is easily decomposed when heated, and releases the oxygen. Due to its ability to release the oxygen, hydrogen peroxide is effective as the bleaching material. The $\text{O}_2$ will act as the oxidizer to bleach the material.

In bleaching process, the pulp shaded from dark brown to yellow; this color shows that lignin has been removed. Moreover, the heating process generated bubbles from the decomposition reaction of the hydrogen peroxide into oxygen and water.

4.2 The Process of Adding Additives
The resulted pulp is added by 10% of tapioca and essential oil consisting of cinnamon oil, lemon oil, clove oil, and lime oil with the concentration variation of 2% and 3%. Tapioca is functioned as the sizing agent and the adhesive; meanwhile, the essential oil is functioned as preservative and aroma agent. It is due to the unpleasant smell of the paper which is caused by alkalization process. After being added by additive substance, the pulp became lighter, and the resulted aroma was based on the added essential oil.

4.3 Paper Printing
This is the finishing process in which the formed pulp was printed to produce the paper. The form of the paper is shown in Figure 1 and Figure 2. Physically, all of the papers were yellowish white in which they have the different level of color and slightly rough texture.
Figure 1. The Paper with The Addition of 2% Essential Oil
  a. Cinnamon, b. Lemon, c. Clove, d. Lime

Figure 2. The Paper with The Addition of 3% Essential Oil
  a. Cinnamon, b. Lemon, c. Clove, d. Lime

4.4 Chemical Characterization
The chemical characteristics of paper, include water content and pH, are shown in Table 1.

| No | Essential Oil Variation      | Water Content (%) | pH   |
|----|------------------------------|-------------------|------|
| 1. | No essential oil             | 5.470             | 7.95 |
| 2. | Cinnamon oil 2%              | 7.140             | 6.95 |
| 3. | Cinnamon oil 3%              | 7.295             | 6.90 |
| 4. | Lemon oil 2%                 | 7.351             | 6.85 |
| 5. | Lemon oil 3%                 | 7.642             | 6.75 |
| 6. | Clove oil 2%                 | 7.600             | 6.90 |
| 7. | Clove oil 3%                 | 7.725             | 6.85 |
| 8. | Lime oil 2%                  | 7.402             | 6.75 |
| 9. | Lime oil 3%                  | 7.647             | 6.70 |
| 10.| Commercial wrapping paper    | 6.093             | 7.55 |
| 11.| SNI                          | ≤ 8               | 6 - 9|
Figure 3. Water Content of Paper with Additive Essential Oils

SNI ISO 6519:2016 states that the water content of the basic paper for Laminated Plastic Wrapping Paper should not exceed 8%. All of the various papers of banana peel with essential oil meet the standard of water content stated by SNI. The result of water content test indicates that the concentration of essential oils affects the water content of the paper produced. The higher the concentration of essential oils added the higher water content of the paper produced (Figure 3.). Essential oil as additive agent increases the amount of bond between fiber, so it reduces the amount of pore water evaporation as a path. Physically, it can be seen from the surface of the paper with additives that are more subtle than the paper without additives due to filling the empty spaces on the bond between fiber. Thus the water content of the paper produced is also increasing. Moreover, there may be another factor affecting the water content, namely drying process, room humidity, air flow, air pressure, the numbers of sample and the thickness of the sample.

Figure 4. pH Value of Paper with Additive Essential Oils

The pH value is essential to be noticed since it affects the skin response when contacting the paper. SNI ISO 6519:2016 states that the Basic Paper for Laminated Wrapping Food Plastic must have pH value in the range of 6-9. All of the variation of banana peel paper with additive essential oil meet the standard quality. The result showed that the concentration of essential oils affects the pH value of the paper produced. Essential oils decrease the pH value of paper produced. The higher concentrations of essential oils added the higher acidity of the produced paper, which was marked by a decrease in pH values (Figure 4.). This relates to the chemical content of essential oils used. Cinnamon oil and clove oil has a high concentration of eugenol while lemon oil and lime oil has a high concentration of
limonene. Both eugenol and limonene are acids so that the addition of the essential oils into the pulp could increase the acidity of the paper produced.

The result of the research on chemical characteristics parameters shows that banana peel paper with additive essential oil meet the standard of ISO 6519:2016 about Basic Paper for Laminated Plastic Wrapping Paper on the parameter of pH and water content.

4.5 Physical Characterization
The physical characteristics of paper, include grammage and brightness, are shown in Table 2.

Table 2. Physical Properties of Natural Wrapping Paper

| No | Essential Oil Variation | Grammage (g/m²) | Brightness (%) |
|----|-------------------------|-----------------|---------------|
| 1. | No essential oil        | 487.4           | 28.80         |
| 2. | Cinnamon oil 2%         | 347.6           | 24.68         |
| 3. | Cinnamon oil 3%         | 356.6           | 54.43         |
| 4. | Lemon oil 2%            | 651.6           | 47.22         |
| 5. | Lemon oil 3%            | 319.2           | 34.34         |
| 6. | Clove oil 2%            | 474.8           | 26.99         |
| 7. | Clove oil 3%            | 275.2           | 21.96         |
| 8. | Lime oil 2%             | 375.6           | 27.53         |
| 9. | Lime oil 3%             | 238.2           | 38.01         |
| 10.| Commercial wrapping paper | 83.0          | 22.82         |
| 11.| SNI                     | 255 - 500       | -             |

Grammage is the mass of paper of a particular unit area. According to SNI, paper and cardboard grammage for food packaging range from 255-500 g / m². Grammage is essential to be measured due to the function of parameter value on the tensile index and tear measurement. All variations of banana peel paper with the additive essential oil are meet the standard of SNI 8218:2015 about Paper and Cardboard for Food Packaging on the parameter of grammage (Food packaging with high grammage) except the paper with 2% lemon oil (651.6 g/m²). Grammage related to the strength of the tear. Paper with high grammage shows the high number of the fibers contained. The amount of fiber will play a role in the process of tearing because each fiber will draw up a sheet of pulp on the particular grammage that contribute to the overall energy needed to tear.

Table 2 shows that the brightness level of banana peel with additive essential oil tends to decrease as the concentration of essential oil increases. However, it will not occur in the paper of additive cinnamon oil. It indicates that the concentration of essential oil affects the brightness level of the produced papers. The more the concentration of essential oil is used, the less the brightness level of the paper is produced.

In addition to being adapted to ISO standards, the produced paper is also compared to the characteristics of commercial wrapping paper. All of the natural wrapping paper produced have higher water content and lower pH value than commercial wrapping paper. The results show that natural wrapping paper from banana peels with 2% cinnamon essential oil additives has the closest characteristics of commercial wrapping paper. Natural wrapping paper with additive 2% cinnamon oil has pH of 6.95, the water content of 7.14%, grammage of 347.6 gram/m² and the brightness level of 24.68%.

4. Conclusions
The natural paper from banana peel waste with additive essential oil meet the standard of ISO 6519:2016 about Basic Paper for Laminated Plastic Wrapping Paper within the parameter of pH and water content and ISO 8218:2015 about Food Paper and Cardboard within the grammage parameter (high grade grammage), except the paper with 2% lemon oil. The result which is closest to commercial
wrapping paper is the paper made of banana peel waste within the additive cinnamon of 2%, containing pH of 6.95, the water content of 7.14%, grammage of 347.6 gram/m² and the brightness level of 24.68%.

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