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Page 1 to 10
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UTILIZATION OF LEFTOVER PULP FROM THE CARRAGEENAN INDUSTRY MADE FROM Eucheuma cottonii WITH THE ADDITION OF BAGASSE AS RAW MATERIAL FOR MAKING PAPER FOLDERS

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ABSTRACT

The carrageenan industry with Eucheuma cottonii seaweed as raw material produces 65-70% of pulp which still contains cellulose components so that it can be used as a diversified product for the paper industry as an alternative to tree fiber. The purpose of this study was to determine the best concentration of E. cottonii pulp added in making map paper made from E. cottonii pulp and bagasse and to determine the quality of map paper made from E. cottonii pulp and bagasse. Methods: The research method used was experimental laboratories using the research basis of Completely Randomized Design (CRD) with the treatment of different concentrations of E. cottonii pulp, namely 0%, 10%, 15%, and 20%. The test parameters carried out in this study were grammage, density, folding resistance, stiffness, water absorption, and SEM. Data obtained in the variance test($p \leq 5\%$), followed by Tukey test to see different treatments. Results: Based on the results of the study, there was an increase in the value of density, water absorption and a decrease in grammage, stiffness and folding resistance of paper due to the addition of E. cottonii pulp. Conclusion: The best treatment results, namely the addition of 10% E. cottonii pulp, resulted in

paper that can be utilized as folder/stopmap paper and fulfill Indonesian National Standard No. 0155:2010 on folder paper with a grammage value of 273.33 g/m2, density of 3.37 g/m3, folding resistance of 52.89 df, stiffness of 36.31 gf.cm, and water absorption of 27.84 g/m2.

KEYWORDS: Seaweed Pulp; Bagasse; Eucheuma cottonii; Paper.

1. INTRODUCTION

Papermaking materials still rely on fibers from woody plants, resulting in the exploitation of many forests. Alternative sources of materials are needed as a substitute for wood in order to minimize environmental damage. In an effort to find alternative raw materials to replace wood in papermaking, we are still fixated on resources on land.

Exploration carried out on marine organisms as an alternative material for wood substitutes can still be developed, one of which is Eucheuma cottonii seaweed . (Apriani & Zulfahmi, 2017)

E. cottonii is a type of red seaweed that is included in the Rhodophyta division. The derivative product that is widely produced *from E. cottonii* red algae is carrageenan which is widely known as a gelling, chewing and stabilizing agent in food. The abundant production of carrageenan is followed by the abundance of waste pulp from the production. Siswanto & Widowati (2018) stated that the remaining results of seaweed processing are in the form of solid waste that has not been widely utilized. The by-products of this type of seaweed processing plant reach 60%-70% of the total amount of raw materials. The characteristic results of the residual seaweed industry pulp contain 28.19% cellulose; 38.83% holocellulose; 10.63% hemicellulose; 8.27% ash; and 3.54% insoluble acid ash (Munifah & Irianto, 2018). Cellulose extract from E. cottonii seaweed can be used as a substitute material in papermaking which is usually made from wood pulp. Cellulose in E. cottonii seaweed is the basic material needed for pulp making, the higher the cellulose content, the better the quality of the paper (Ariefta et al., 2019). Furthermore, Alaydin et al., (2020) also stated that seaweed pulp contains sufficient cellulose so that it has the potential as a raw material for paper.

E. cottonii pulp has a type of short fiber, where in papermaking a combination of other materials is needed as a filler that functions to fill the gaps that exist between fiber bonds to be able to improve the print quality of the resulting paper. The results of research by Mandegani et al (2016) on papermaking made from seaweed pulp produce paper with characteristics that are easily brittle and easily torn because seaweed pulp is classified as short fibers which have weak bonding properties between fibers. Based on this research, it is concluded that a combination of materials is needed to strengthen fiber bonds.

Bagasse is the residue of processing or milling sugar cane stalks (Saccharum officinarum) in the sugar industry that has been taken by the juice and still contains fibrous material which is very useful as raw material for pulp making. Bagasse is usually just piled up around the factory and used as fuel. The processing of sugar cane around the community is usually only by selling sugar cane drinks and the bagasse will be thrown away because it is considered to have no economic value, increasing the amount of waste if not managed properly (Legiso & Kalsum, 2018). The main component of bagasse consists of about 43-52% fiber and has a fiber length of 1.7-2 mm with a diameter of 20 μm (Gustriani et al., 2013).

The chemical components present in bagasse consist of 37.65% cellulose, 27.97% pentosan, 22.09% lignin, 3.28% ash, potentially as an alternative material and can replace the role of wood in making paper pulp is bagasse (Allita et al., 2018). Bagasse has the potential as an alternative material and can replace the role of wood in papermaking because it has a high fiber content of 45%-55%. Fiber with high enough strength can be used as a reinforcing fiber material. Based on research conducted by Mardiah & Jannah (2016) on the utilization of bagasse into kraft paper with the organosolv method, namely a papermaking method that uses organic solvents to dissolve lignin and hemicellulose, it will produce a paper that is easy to shape and dense. Novia et al. (2022)

The research used a mixture of *E. cottonii* seaweed pulp and bagasse. The paper produced in this study can be utilized as map paper, which is paper that functions to store and protect the sheets of paper inside so that it is more neat and protected. Folder paper is a rectangular fold of paper or manila cardboard that is used to store archives. The types consist of ordinary folders which are often called folio stopmaps, laced stopmaps (portapel), clip folders (snelhechter), thick folders which are better known as ordner or brieforner (Hayati, 2020). Paper folder is paper that functions as a protective device in storing temporary documents such as invoices, offers, agreement letters and others (Putri & Trisninawati, 2020). The purpose of this study was to determine the best concentration between the mixture of *E. cottonii* pulp and bagasse added in making folder paper and determine its quality.

2. METHOD

2.1 Materials *E. cottonii* pulp was obtained from CV Karagen Indonesia, Semarang. Seaweed pulp is in powder form, white color. The other material is bagasse obtained from sugarcane juice beverage traders in the form of sugarcane residue after pressing the juice.

Chemicals used in the pulping process are NaOH and bleaching using H2O2, in addition to the adhesive material used in the form of PvAC and tapioca starch.

2.2 Place of implementation

The research process included the preparation of pulping materials and papermaking was carried out at the Fishery Products Technology Processing Laboratory, Diponegoro University. Testing of test parameters was carried out at the Integrated Laboratory of Diponegoro University and the Center for Pulp and Paper (BBPK) in Bandung, West Java.

2.3 Method used

The method used in this research was laboratory experiment with independent variables in the form of map paper products with variations in the treatment of the addition of E. cottonii pulp 0%, 10%, 15%, and 20%. The dependent variables were grammage, density, folding resistance, stiffness, and water absorption. This study used a variation of *E. cottonii* pulp concentration of 0%, 10%, 15%, and 20%. E. cottonii pulp was substituted with bagasse used in the formulation of papermaking materials.

Seaweed pulp samples were washed using running water until the pH was neutral, then dried and sieved. Bagasse was pulped by alkaline extraction, the alkaline solution used was NaOH with a concentration of 5%, the ratio of sugarcane waste to NaOH solvent was 1:15 (b/v). Pulping of

bagasse is done at 80o for 1 hour then washed with running water and filtered. The process continued with bleaching using a 12% concentration of H2O2 solution. Bagasse pulp then washed with running water and filtered.

Papermaking was done by mixing seaweed pulp, bagasse pulp, and adhesives. The concentrations of E. cottonii pulp used were 0%, 10%, 15%, and 20%. The mixed materials were then dissolved with water in a ratio of 1:15. The next process was printing using a paper printing screen and pressing using a sponge to reduce the water content and then transferred to a glass plate followed by drying in the sun for 1 day.

2.4 Experimental design

The experimental design in this study was using a completely randomized design (CRD) with the addition of different concentrations of E. cottonii seaweed pulp, namely concentrations of 0%, 10%, 15%, and 20% with three replicates in each treatment.

2.5 Data analysis

Some of the research data were compared with the BSN (National Standardization Agency) standard in SNI 0155:2010 on folder paper stating that the quality of folder paper was determined by the parameters of grammage, folding resistance, stiffness and water absorption. SEM data was analyzed qualitatively, while the test data was analyzed using SPSS version 16 which includes normality test, homogeneity test, and ANOVA test analysis.

3. RESULTS AND DISCUSSION

3.1 Grammage

The addition of E. cottonii pulp affected the increase in paper grammage when compared to the control treatment (Figure 1). The treatment of the addition of seaweed pulp with a concentration of 10% caused an increase in the paper grammage value of almost 2x of the control treatment. The increasing value was due to the addition of seaweed pulp which is able to fill the cavity in the bagasse fiber bond so as to minimize the occurrence of empty space between fiber bonds. The value of paper grammage is influenced by the type and amount of raw materials that will determine the thickness of the resulting paper. According to Putri et al (2022), grammage and thickness have a very close relation with each other. This is because the formulation of paper grammage can be obtained from the ratio of paper weight (g) to paper area (m2).

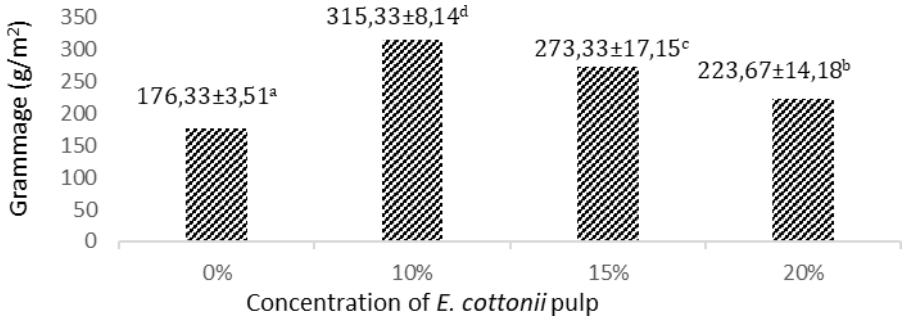


Figure 1. Grammage Value of *E. cottonii* and Bagasse Paper.

The grammage value of paper is influenced by the manual paper printing process. Pressing using a sponge to reduce the water content, if done too strongly, will result in short fiber particles that act as fillers of cavities between fiber bonds being released and dissolved with water. This causes the bond between fibers to weaken due to the formation of voids and results in decreased paper durability. In the paper industry, the printing process has been using machines, this is done for time efficiency and to minimize the difference in the paper produced.

3.2 Density

The results of the density test presented in Figure 2 show an increase in density value of almost three times in the 15% treatment when compared to the control treatment. This proves that the addition of E. cottonii pulp has an effect on increasing paper density.

The use of *E. cottonii* pulp and bagasse results in stronger bonds between fibers and minimizes empty space or voids in the paper bond structure. E. cottonii pulp, which is classified as short fiber, acts as a filler in the paper fiber bond structure so that it can increase the density value of the resulting paper. Damayanti et al (2022), examined short fibers that have filler properties so that they can fill empty spaces and increase the bonding power between the fibers that make up the paper.

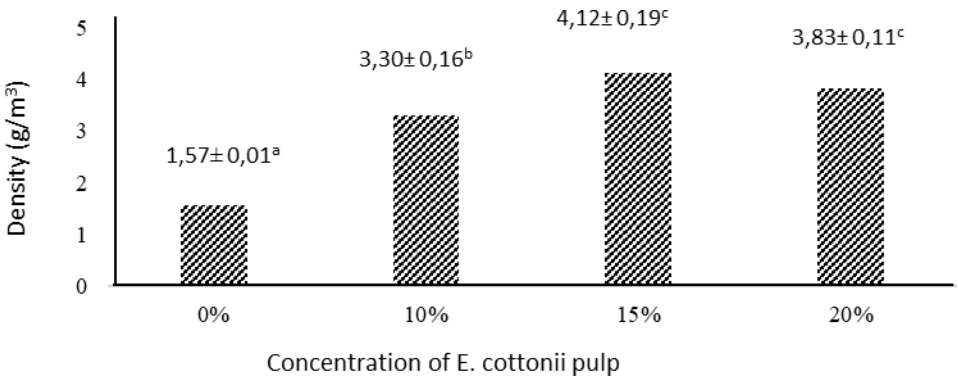


Figure 2. Density Value of *E. cottonii* and Bagasse Paper.

The combined use of long and short fibers in the papermaking process can increase the paper density value. The density value is influenced by the thickness of the paper, which is the distance between the top surface and the bottom surface of the paper. Thickness is affected by the pressing process in the paper printing process. A strong and long pressing process will make the printed recycled paper produce a good density so that the fibers are not damaged and the fiber bond becomes tighter (Putri et al, 2020).

3.3 Folding resistance

The results of the paper folding resistance test presented in Figure 3, show that the treatment of adding 20% *E. cottoni* pulp concentration decreased when compared to the other 3 treatments. The magnitude of the decrease was almost half of the control treatment. This is thought to be due to the fiber composition dominated by E.cottonii pulp which is classified as a short fiber type. The more *E.cottonii* pulp added causes more cellulose content so that the folding resistance weakens and causes the bond between fibers to be weak and easily broken. Longer fibers will provide greater physical strength, because the longer the fiber, the surface area of the contact area that binds between the fiber and the matrix becomes larger than the short fiber (Mengga et al, 2015).

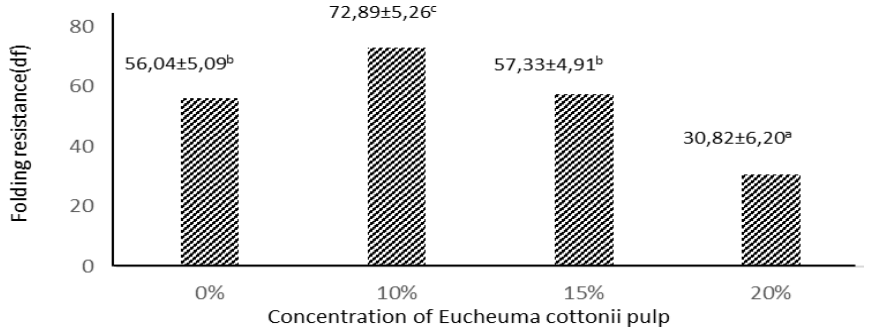


Figure 3. Folding Resistance Value *of E. cottonii* and Bagasse Paper.

Furthermore, Kathomdani (2018) found that folding resistance is the most sensitive indicator in the paper deterioration process. The folding resistance of paper is inversely proportional to the length of time the paper is stored. The longer the paper is stored, the more the folding resistance of the paper decreases. This decrease is due to the bond between fibers in the paper will be more fragile due to temperature and relative humidity.

The value of paper folding resistance is influenced by the value of grammage, density, fiber length, the use of adhesives and the use of alkaline materials in the pulping process. The alkalization process affects fiber breakage which serves to remove the lignin content in the fiber (Gian et al, 2017). The study states that the alkalization process aims to remove the lignin, hemicellulose, wax and oil content surrounding the fiber surface so that the pure fiber content can be separated.

3.4 Stiffness

The results of the stiffness test with the treatment of adding E. cottonii pulp with concentrations of 0%, 10%, 15% have fulfilled the value of the Indonesian National Standard (SNI No. 0155: 2010 on folder paper) of 35 gf.cm, while the 20% concentration has not fulfilled. The stiffness value of paper with the treatment of adding 20% E. cottonii concentration decreased up to 4x compared to the control treatment. This is because paper with the addition of 20% E. cottonii pulp is dominated by short fibers when compared to other treatments that have more long fiber content. Long fibers have better strength than short fibers. Research by Boimau (2020), shows that long fibers are able to distribute load and stress slowly from the stress center to all areas of the composite evenlyconcentration has not fulfilled. The stiffness value of paper with the treatment of adding 20% E.

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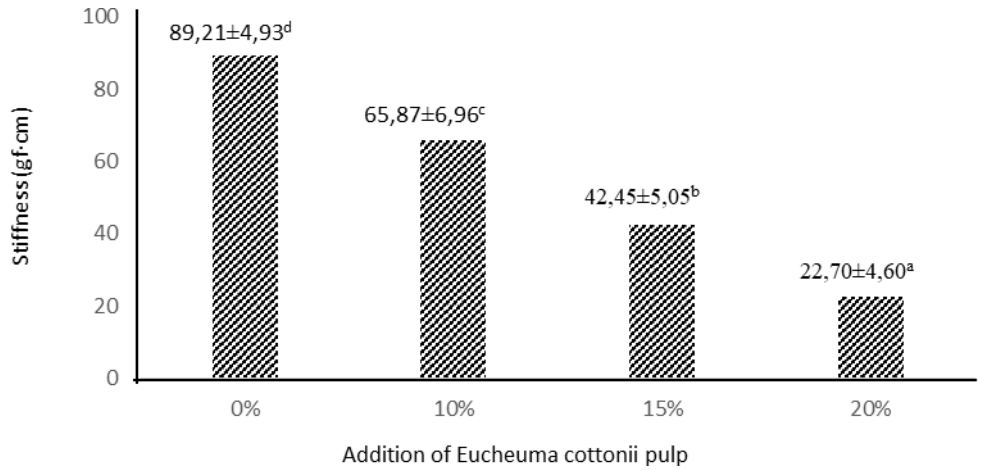


Figure 4. Stiffness Value of E. cottonii and Bagasse Paper.

Lignin content in the fiber makes the fiber stiff, therefore in papermaking the lignin content needs to be removed. The longer the fiber, the stronger the bonding power between fibers, which causes the paper to have a high stiffness value. Safrizal et al (2022) stated that paper stiffness is also influenced by the stiffness of the constituent fibers. The lignin content in the fiber has cellulose-binding properties that cause the fiber to become stiff. The lignin content in the fiber can be reduced by pulping using an alkaline NaOH solution. The higher the concentration of alkaline

solution used, the lower the lignin content in the fiber. The low lignin content causes the cellulose in the fiber material to be flexible. Longer fibers will provide greater tensile strength, because long fibers cause the surface area of the contact area that binds between the fiber and the matrix to be greater than that of short fibers (Mengga et al, 2015). Aminah (2020) further explained that paper that has a high stiffness value has better strength. Vice versa, paper that has a low level of stiffness or tends to be flexible, has a lower strength of the paper.

3.5 Water absorption

Based on the research data, the water absorption test produced increased as the amount of E. cottonii seaweed pulp increased. The value of water absorption in the 0% concentration treatment was the lowest value while the 20% concentration increased 4x compared to the control treatment. The results of the water absorption test showed that paper with treatment without the addition of E. cottonii pulp has not fulfilled the Indonesian National Standard (SNI No. 0155: 2010 on folder paper) of 25 g/m2. Paper treated with the addition of 10%, 15%, and 20% *E. cottonii* pulp has fulfilled the SNI standard limit.

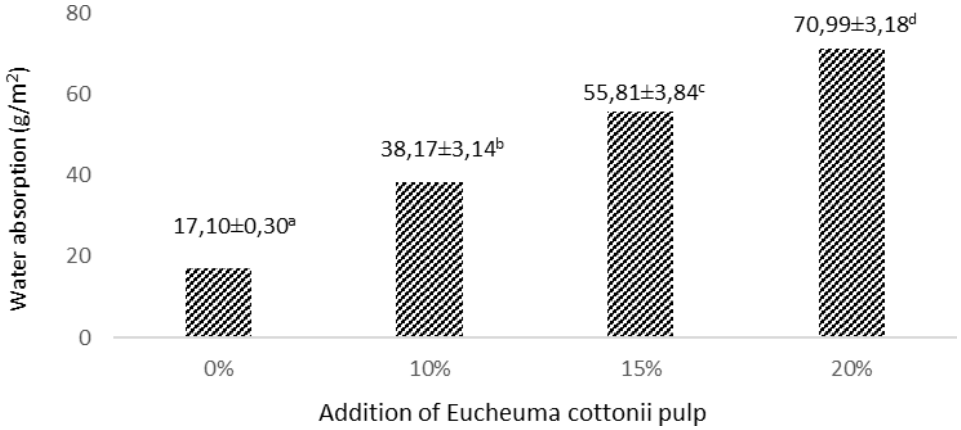


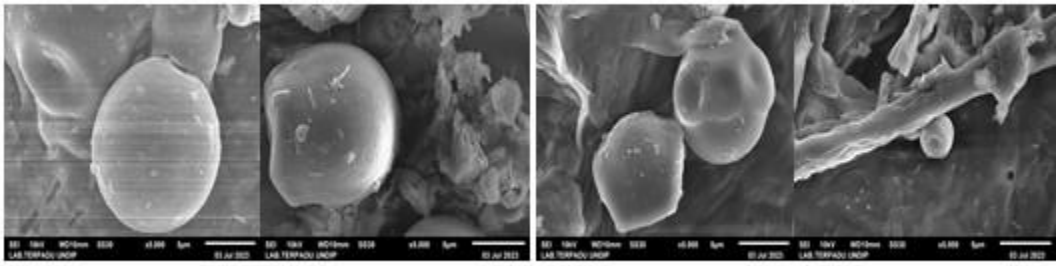
Figure 5. Water Absorption Value of E. cottonii and Bagasse Paper.

The water absorption of folder paper affects the durability of the paper. The low water absorption value is caused by the high composition of the constituent fibers, so that the paper's resistance to water is higher and produces paper with good quality. Haroen and Santosa (2017) found that the smaller the water absorption value, the higher the paper's resistance to liquid penetration. This is reinforced by Utami et al (2015), the fiber component of the paper determines the water absorption of the paper. The greater the ratio of mixed materials containing cellulose, the smaller the water absorption rate, which means the paper quality is very good.

The factor that affects water absorption is the length of the fibers that make up the paper, the shorter the fiber size, the wider the surface of the absorption area which results in higher absorption (Paskawati & Retnoningtyas, 2017). The level of fiber surface density also affects water absorption, the higher the density, the lower the water absorption rate. This is because the tighter bond between fibers will hinder water absorption (Sijabat et al., 2022).

Barbash & Yashchenk (2020) stated that the higher the fiber density, the lower the water absorption rate. The decreased value is because the tighter fibers will block the water absorption process. Paper composed of tenuous fiber bonds results in the formation of empty space, causing pores to appear, causing high water absorption which is influenced by capillarity forces.

SEM (*Scanning Electron Microscope*)



(a) (b) (c) (d)

Figure 6. SEM test results of paper made from E. cottonii pulp and bagasse (a) 0%, (b) 5%, (c) 10%, and (d) 15%.

Magnification 5000x

SEM allows obtaining morphological and concentrated images of the microstructure of the mixture of materials used as the constituent material of the test sample (Septiano et al., 2021). The results of SEM testing carried out with a magnification of 5000x captured an image of spherical particles that were thought to be E. cottonii pulp. This is due to the carrageenan extraction process using chemicals and the heating process causing changes in the structure of seaweed constituents, so that the microstructure of the resulting pulp is damaged into uneven shape changes.

Paper folder with treatment without the addition of *E. cottonii* pulp has voids between the bonds between the fibers. This is assumed to be because the paper consists of more bagasse, which has a long fiber size so that no filler can fill the voids between the bagasse fiber bonds. According to Ristianingsih et al (2018), paper from long-fiber pulp has high-strength properties, because the fibers bind each other strongly. However, the formation is lacking because between the bonds between the fibers there are small pores that cannot be filled by long fibers.

The paper consisting of bagasse with the treatment of adding *E. cottonii* pulp at a concentration of 10%, 15%, and 20%, the fiber bond was tighter due to the short fibers of E. cottonii pulp being able to fill the empty cavities in the fiber bond. The higher the concentration of E. cotton ii pulp added, the tighter the bond between the fibers that compose the paper. Setiawan et al (2015), showed that the higher the concentration of short fibers that act as cavity fillers, the smaller the pore size, this can occur because the higher the concentration level of short fibers, the higher the particle density which results in tighter fiber bonds.

4 CONCLUSIONS

The best treatment to produce paper characteristics that can be utilized as map paper was the treatment of 10% E. cottonii seaweed pulp concentration. The higher the concentration of *E. cottonii* pulp added, the effect on increasing water absorption and decreasing grammage, density, folding resistance and stiffness. The quality of paper produced in the treatment of 10% concentration of E. cottonii seaweed pulp has fulfilled SNI No. 0155: 2010 concerning map paper.

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