Effect of Cooking Liquor Ratio on Lignin Reduction in Pulping Process from Cogongrass and Lemongrassleaves using Soda Process

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Keywords: Alpha cellulose, lignin, cogongrass, lemongrass leaves, soda process

Abstract: Pulp is the raw material for paper making. The quality of pulp is determined by the content of cellulose and the remaining lignin contained in the pulp. Alpha cellulose is a parameter used to determine the amount of cellulose contained in the pulp. The alpha cellulose content in the pulp is affected by the concentration of the cooking liquor. The level of lignin in the pulp shows the residual lignin left from incomplete hydrolysis. This study aims to reduce the amount of lignin in the pulping process from cogongrass and lemongrass leaves using soda process. The liquor that is used for cooking process is sodium hydroxide (NaOH) and sodium carbonate (Na_2CO_3) with a cooking time of 120 minutes, at a temperature of 110 ºC, with ratios of cooking liquor and raw materials variation such as 1:1, 2:1, 3:1, and 4:1. The results of this investigation shows thatthe best cooking conditionwas obtained in the ratio of cooking liquor and raw material 4:1 with alphacellulose content 83.11%, 56.31% ofyield, 0.78% ofash content, and Kappa number which is a method to determine the amount of lignin remaining in pulp of 13.17, while the highest yield of 78.33% was obtained in the ratio of cooking solution 2: 1. In spite of that, almost all produced pulp in this study has pulp quality in accordance with SNI 7274 and the Center of Pulp (alpha cellulose content > 40%, ash content <3%, and Kappa number 13.5).

1 INTRODUCTION

The need for pulp in Indonesia has increasedover the years as the demand for paper keep increasing. The government, through the Industrial Research and Development Body (BPPI), encourages increasing the production of pulp and paper from non-wood raw materials because Indonesia has a large potential of non-wood materials.

Head of the Center of Pulp and Paper, Andono Sugiharto, said that Indonesia currently ranks 6th as the world's paper producing country and 9th at world pulp producer (Pikiran Rakyat, 2017). So far, most of the raw materials for producing pulp were made of wood, this can be seen from the total production of 4.55 million tons of pulp in 2013, only 18,680 tons or 0.0004% that is the pulp made of bamboo plants.

Other types of non-wood plants that are easily obtained and can be used as raw material for pulping include hemp, cotton, agricultural waste such as bagasse, pineapple skin, rice straw, lemongrass and various types of grasses and reeds (Yandha and Sherren 2014) . The use of non-wood raw materials has advantages, such as the potential is quite high, easily made into pulp, easily milled, and easily bleached with environmentally friendly materials. This study uses cogongrass and lemongrass leaves as raw material, mainly because cogongrass and lemongrass leaves possess high cellulose content. Besides, cogongrass also has become undesired plants which is grown in the midst of other productive plants so that they are always eradicated using cogongrass poisonwhich can also damage the environment.

The use of reeds which are difficult to eradicate and lemongrass leaves as agricultural waste that has not yet been used as raw material for pulp contribute to become a good solutions in environmental management. One of the factors that influence the pulping process is the concentration of cooking liquor, namely the right ratio between the cooking liquor and the raw material. In this study, the ratio between the cooking solution and the raw material were 1: 1, 2: 1, 3: 1 and 4: 1 with 1: 1 ratio of cogongrass and lemongrass leaves to obtained good quality pulp.

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2 LITERATURE REVIEW

2.1 Cogongrass

Raw materials used in pulp making usually are woods, but non-wood raw materials can also be used. One of them is cogongrass. *Imperata cylindrica* is also known as japgrass, bladygrass, speargrass, reeds and weeds. This plant is extremely easy to grow and develop, and is widely distributed in tropical and subtropical regions. Until now, this plant can be found in more than 73 countries.

Because of the short growth cycles, abundant, unsuitable to become animal feed and the lack of commercial application of this grass, this plant can be proposed as an alternative fiber in the pulp and paper industry to reduce or replace the use of pure pulp widely (Kassim et al, 2015).

Imperata has an alpha cellulose content of 40.22%, hemicellulose (pentosan) 18.40% and lignin 31.29% (Sutya et al, 2015). The cellulose content of more than 40% (Kartikasari et al, 2015), making this plant has a huge potential as raw material for pulp making.

2.2 Lemongrass

Lemongrass is a plant that has its own commodity which results in many plants being used as agricultural crops. Lemongrass which is used or sold in the form of lemongrass stems, causing lemongrass leaves become waste. Lemongrass leaves contain quite highcellulose content, so that it can be used as an alternative material for pulp making.

2.3 Cellulose and Lignin

2.3.1 Cellulose

Cellulose is mostly found in cell walls and woody parts of plants. Cellulose has a role that determines the character of fiber and allows its use in paper making. In pulping, fibers are expected to have high cellulose content. The properties of materials containing cellulose are related to the degree of polymerization of cellulose molecules. Reduced molecular weight below a certain level will cause reduced strength. Cellulose fibers show a number of properties that meet paper-making needs. The best balance of papermaking properties occurs when most lignins are excluded from fibers. Fiber toughness is mainly determined by the raw material and the process used in pulping (Surest and Dody, 2010).

2.3.2 Lignin

Lignin is the third macromolecule found in biomass, functions as a binder between fibers. Lignin can be removed from cell wall material which is insoluble with chlorine dioxide.

The molecular structure of lignin is very different when compared to polysaccharides, because it consists of an aromatic system composed of propane phenyl units. The properties of lignin are insoluble in water and strong mineral acids, dissolve in organic solvents and dilute alkaline solutions. The lignin attached to the pulp product decreases the strength of the paper and causes the paper to turn yellow.

The pulp will have good physical properties or strength if it contains just a little lignin. This is because lignin is water-repellent and stiff, making it difficult for the grinding process. The lignin content for wood raw materials is 20-35%, while for non-wood materials is lower (Surest and Dody, 2010).

2.4 Pulp Making Process

2.4.1 Sulfate Process (Kraft)

In the Kraft process, sodium hydroxide and sodium sulfide are used in wood pulp. This process is widely used in the pulp and paper industry. In the Kraft process, about half of the wood is dissolved in chemicals and will form black liquor. This liquid is cleaned from pulp by washing and being incorporated into the Kraft renewal system where pulp inorganic chemicals are recycled and reused, whereas in dissolved organic matter can be used to produce steam and energy (Tran and Vakkileinnen, 2011).

2.4.2 Sulfite Process

The sulfite process uses a cooking liquor in the form of an acid which is a combination of acids mixed with magnesium bisulfate. In this process, the cooking liquor enters the wood and decomposes the lignin which when cooked is converted into water-soluble compound that can be washed (Sappi, 2003).

2.4.3 Soda Process

The soda process involves heating the fiber in a pressurized reactor at a temperature of 100-170 °C using 13-16% sodium hydroxide which is the cooking liquor. The ratio of cooking liquor to dry fiber is 5:1. In this process, lignin is separated from cellulose and suspended in a liquid phase. In liquid phase which is black liquor is separated from solid phase containing cellulose due to lignin. This solid phase containing cellulose is called pulp. The resulting pulp is then processed in the manufacture of paper, boards, composite materials, polymers and others. This black liquor contains lignin and sodium hydroxide (soda) which are usually further processed to be recycled and reused in the process (Doherty and Thomas, 2006).

2.5 Factors Affecting Pulp Making

2.5.1 Cooking Liquor Concentration

The higher the concentration of cooking liquor, the greater the amount of cooking liquor that reacts with lignin, but the use of excess cooking liquor is not good because it will cause the cellulose to degrade, so that the cooking solution ratio must be in accordance with the portion (proportional).

2.5.2 Ratio of Cooking Liquor to Material

The ratio of cooking liquor to raw material must be sufficient so that the lignin fragments are perfect in the degradation process and can dissolve completely in the cooking liquor. Ratios that are too small can cause redeposition of lignin so that it can increase the Kappa number (the quality of the pulp decreases).

2.5.3 Cooking Temperature

Cooking temperature is related to the reaction rate which can increase the rate of delignification (removal of lignin), but at temperatures above 180 ^oC causes degradation of cellulose which at this temperature lignin has been dissolved. Then the optimum temperature ranges from 80-140 ^oC [10].

2.5.4 Cooking Time

The longer the cooking time will cause the lignin hydrolysis reaction to increase. However, cooking time that is too long will cause cellulose to hydrolyze, thus reducing the quality of the pulp. For cooking before 1 hour the pulp has not been formed and for 5 hours of cooking the cellulose are degraded. The optimum time in the delignification process is around 60-120 minutes with a constant lignin content in that time span (Putra, 2012).

3 RESEARCH METHODOLOGY

3.1 Materials and Equipments

The raw material in the form of cogongrass and lemongrass leaves with a ratio of 1: 1.
Cooking solution of 12.5% (NaOH and Na_2CO_3) with a ratio of 85% NaOH and 15% Na_2CO_3 and the remaining is water (87.5%) (TAPPI, 1999).

3.2 Equipments

Set of Equipments

The set of equipmentsused in the research (Figure 1)

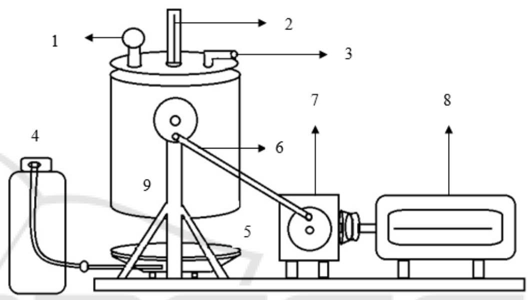

Figure 1: Series of Equipments in Pulp Making

Caption:

1. Barometer 7. MotorReducer
2. Thermometer 8. Driving motor
3. Safety valve 9. *Digester*
4. Gas holder
5. Fire heater
6. Driving connector with digester

3.3 Analysis

The analysis was carried out based on SNI 2009 and TAPPI 1999 namely:
Analysis of Water Content of Raw Materials
Analysis ofAsh Content
Analysis of alpha, beta and gamma cellulose content
Analysis of Kappa Numbers
Analysis of Yield

3.4 Main Procedure

The raw material was weighed as much as 200 grams and tested for water content and the calculation of the need for cooking liquor. The raw material and cooking liquor were put into the digester and the operating conditions were set at constant temperature and time at 120 minutes and a temperature of 110 ºC. After the specified operating conditions were reached, the pulp was removed from the digester and washed. The washing process was carried out with hot water and followed by cold water until the washing water is clean. The pulp obtained is analyzed for ash, cellulose, Kappa number and yield.

4 RESULTS AND DISCUSSION

4.1 Effect of the Ratio of Cooking Solution on Alpha, Beta and Gamma Cellulose Content of Pulp

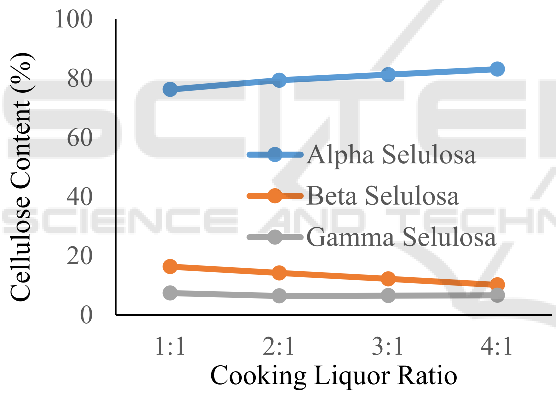

Figure 2: Cellulose Content (Alpha, Beta and Gamma) to the Cooking Liquor Ratios at a Temperature of 110 ºC and Cooking Time of 2 Hours

From Figure 2 above, it can be observed that alpha cellulose content has increased with increasing cooking liquor ratio. In the ratio of cooking liquor to raw material of 1: 1, 2: 1, 3: 1, and 4: 1 the alpha cellulose content had an increase of 76.22%, 79.32%, 81.32% and 83.11% respectively. The levels of beta cellulose decreased by 16.34%, 14.21%, 12.22% and 10.21%, respectively, and for gamma cellulose content was constant at 7.44%, 6.47%, 6.57%, and 6.68%.
An increase in the ratio of the cooking solution increased the amount of lignin dissolved, thereby increasing the cellulose percentage. In this case is an increase in alpha cellulose, increasing the amount of alpha cellulose will make the pulp quality better, according to SNI 7274 and the Center of Pulp (SNI, 2008), good pulp quality standards with a minimum amount of alpha cellulose 40%. In this research, on all cooking liquor ratios produce higher amounts of cellulose than 40%.

4.2 Effect of the Ratio of Cooking Liquid to Ash Content of Pulp

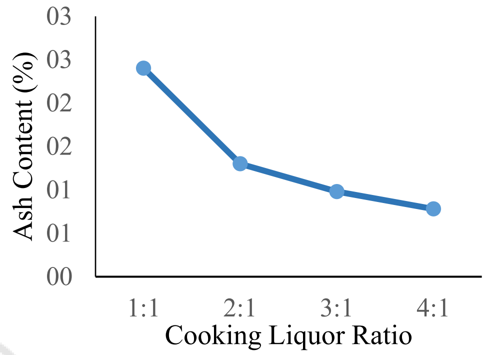

Figure 3: Ash Content of Pulp to the Ratio of Cooking Liquor at Temperature of 110 ºC and Cooking Time of 2 Hours

From Figure 3 it is shown that the higher the ratio of cooking liquor to raw materials, the ash content will decrease. At the ratio of 1: 1, 2:1, 3:1, and 4:1, the ash content was 2.4%, 1.3%, 0.98% and 0.78% respectively. The decrease in ash content is caused by the dissolution of organic substances by the cooking liquor. The more the cooking liquor is used, the more organic matter can be dissolved.

According to SNI 7274 and the Center of Pulp (SNI, 2008), a good pulp quality standard has 3% maximum amount of ash.In this research, the pulp was obtained with ash content at various variations lower than 3%. This shows that the pulp produced were in accordance to the standard and has good quality.

4.3 Effect of the Ratio of Cooking Liquor on Kappa Numbers

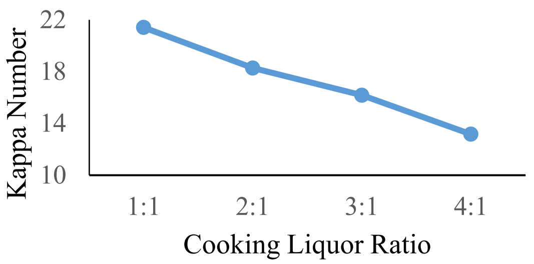

Figure 4: Pulp Kappa Number on the Ratio of Cooking Liquor at 110 °C and 2 hours Cooking time

From Figure 4 above it can be observed that the Kappa number has decreased along with the increase in the cooking liquor ratio. The Kappa number is the required number of milliliters of potassium permanganate ($KMnO_4$) required or absorbed by 1 gram of dry pulp under standard conditions [11]. The purpose of Kappa number analysis is to determine the effectiveness of the delignification process, where the Kappa number shows the remaining lignin content of the pulp.

At the ratio of 1:1, 2:1, 3:1, and 4:1, the Kappa number is 21.41, 18.27, 16.17, and 13.17 respectively. An increase in the ratio of the cooking liquor increased the amount of lignin dissolved. The greater the dissolved lignin, the lower the Kappa number, which shows the better quality of the pulp with a low lignin content.

4.4 Effect of the Ratio of Cooking Liquor to Pulp Yield

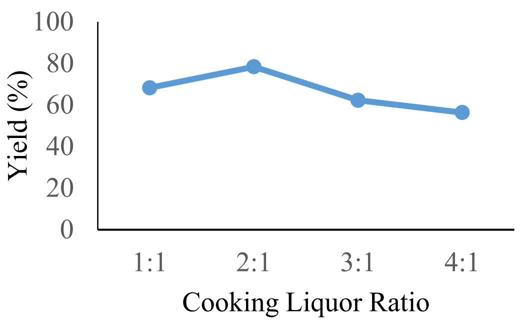

Figure 5: Yield of Pulp on Ratio of Cooking Liquorat 110 °C and Cooking Time of 2 hours

From Figure 5 above shows that at the ratio of 1:1, 2:1, 3:1, and 4:1,the yield obtained were 68.23%, 78.33%, 62.21% and 56,31% respectively. Yield has increased from a ratio of 1: 1, to a ratio of 2: 1, this increase is because at the ratio of 1:1 less cooking liquor are used so that the pulping process was less effective, and it caused a decrease in the yield. In the ratio of 2:1 to 3:1 the yield decreased, and also for the ratio of 4:1, which were caused by the dissolution of cellulose (degraded cellulose) in the cooking process.

5 CONCLUSION

1. The lowest ash content is at a ratio of 4:1 of 0.78%.
2. The highest alpha cellulose content isat a ratio of 4: 1 of 83.11%.
3. The lowest Kappa number is at a ratio of 4:1 of 13.17.
4. The highest pulp yield at a ratio of 2:1 is 78.33%.
5. In applications that require high levels of alpha cellulose, and lignin as low as possible, it can be done with a ratio of 4:1 cooking liquor, but if the high yield is required, the comparison of cooking solution 2: 1 can be used.

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