Alkaline Pulping of Red Meranti (Shorea selanica Blume)

Ganis Lukmandaru, Fajar Setiaji, and Fatimah Ayu Warahapsari

Abstract

The suitability for papermaking of red meranti (Shorea selanica Blume) wood for three alkaline pulping processes i.e. soda, soda-anthraquinone (AQ), and kraft was studied. The fiber morphology and chemical properties were also examined. Cooks were made for 20% (as Na2O) activate alkali. The resulting pulp and paper properties were investigated. The basic density of red meranti was 0.42 g/cm³ and can be classified to be of medium density. The fiber proportion (67.14%) and fiber length (1.07 mm) of red meranti in this study were within the range of tropical hardwoods. The derived values for Runkel ratio, slender ratio, and flexibility coefficient were 0.54, 54.93, and 0.62, respectively. Furthermore, total extractives, lignin, and cellulose contents as well as solubility in 1% NaOH were 5.17%, 31.05%, 45.20%, and 26.02%, respectively. The alkaline pulps showed low kappa number (16~22) and reject level (0.5~1.5%) with the best results for screened yield (47.41%) being achieved in soda pulping. With regard to strength and optical properties of the paper hand-sheets, soda-AQ pulping showed the highest value in burst index (2.36 kPa m²/g), tear index (8.47 ± 1.13 mNm²/g), and brightness (19.81%), whereas kraft pulping gave the best result in tensile index (28.39 N/m). The comparatively low values of yield and strength properties in kraft pulping might be due to overcooking in this experiment.

Keywords: red meranti, pulp yield, kappa number, strength properties, brightness.

Introduction

The escalation of paper consumption has prompted investigations into the potential of fast-growing species as raw material for the pulp and paper industry. Some dipterocarps species have been reported to have comparatively high mean annual diameter increment (1.16~1.30 cm/year) and considered to be high potential for rehabilitation of the logged-over area in a large scale (Adjers et al. 1995; Shono et al. 2007; Hassan et al. 2007). Shorea selanica Blume is a fast-growing tree which is grown commercially for the production of hardwood timber. Along with S. leprosula and S. ovalis from Kalimantan, Shorea selanica from Maluku are classified as 'red meranti' timber and as 'Critically Endangered' in the IUCN Red List of Threatened Species (Ashton 2011). Therefore, establishment of fast growing species plantations with high productivity such as the line planting of red meranti with intensive silviculture in tropical rain forest has been intensified (Na‘iem and Widiyatno 2012).

Kraft (sulphate) pulping is the dominant chemical pulping process used for the production of pulp fibres from various lignocellulosic materials. On the other hand, soda-anthraquinone pulping has environmental and economic advantages. Since no sulfur compounds are used, there is no such unpleasant smell generation. Furthermore, it increases the pulp yield and requires less cooking duration to obtain the same pulp quality and high yield values (Francis et al. 2006). Due to the effectivity, the anthraquinone addition may be applied to any raw – wood or non-wood – material (Biswas et al. 2011; Feria et al. 2012; Garcia et al. 2012; Gonzales et al. 2013).

In recent years, the attention has been focused on pulping of several hardwoods as alternatives to A. mangium in Indonesia (Lukmandaru et al. 2002; Yahya et al. 2010; Theo 2011). However, there is still little knowledge on pulping of new potential species. To the best of our knowledge, no study has been conducted to investigate the pulping of Shorea selanica. One paper reported the pulping potentials and occurrence of pith problems of other shorea species i.e. Shorea albida, Shorea richetia, and Shorea polita (Su et al. 1992) with varied results. Therefore, the present work aimed at evaluating the wood properties and the alkaline pulp produced from red meranti wood and comparing it with those from Acacia mangium wood. The kraft, soda and soda-anthraquinone (AQ) pulping of the wood were conducted.

Materials and Methods

Sampling

Wood samples (Fig. 1) were obtained from a single tree (20 years) grown at the campus yard of Faculty of Forestry, Universitas Gadjah Mada, Jogyakarta. Stem disks (diameter of 20~30 cm, heartwood proportion ca. 27%) were cut from the base, center, and top parts at a certain height. The wood samples from each tree height were chipped (3 cm × 2 cm × 2-3 mm) manually, mixed, and air-dried.
Fibre Morphology

Cross sections (12 µm thick) were microtomed from wood block (American Optical Corp., New York, USA) and stained with a 0.1% solution of safranin (WAKO Pure Chemical Industries, Osaka, Japan), and mounted in glass slides. The cross-sectional images were captured under a light microscope (Olympus BX 51; Olympus Corporation; Japan) with a digital camera (Olympus DP 70; Olympus Corporation; Japan) and converted to digital format. Proportion of wood cell types, which are vessel, fibre, ray parenchyma and axial parenchyma, were measured in percentage. Fibre morphology, which comprised fibre diameter, fibre lumen diameter, and fibre wall thickness according to IAWA (1989), was also measured. Lengths of wood fibres were measured with image-analysis software (Image pro Plus). The cell length of 100 fibers, macerated from small sticks with Franklin’s solution, was measured by a digitizer (Olympus DP 70; Olympus Corporation; Japan) coupled to a light microscope (Olympus BX 51; Olympus Corporation; Japan). The cell morphologies of 100 randomly selected fibers were measured by image-processing software (Image pro Plus) according to IAWA (1989).

The derived fibre properties are defined as follows (Yahya et al. 2010 and the literatures cited therein):

- Runkel ratio = double fibre wall thickness / fibre lumen diameter
- Slenderness ratio = fibre length / fibre diameter
- Flexibility coefficient = fibre lumen diameter/ fibre diameter

Basic Density

The basic density was determined as the ratio of oven-dry weight to green volume as determined by the water displacement method.

Chemical Analysis

Extractives content was determined by extraction sequences with ethanol/toluene (1/2, v/v) (ASTM D1107 - 96) and hot water (ASTM D1110 - 84). The acid-insoluble Kason lignin (SNI 0492:2008), holocellulose (Wise’s chlorite acid method, Browning 1967), and α-cellulose (NaOH extraction, Rowell et al. 2005) were determined in extractive-free wood. Hemicellulose content was determined by subtracting holocellulose from the cellulose contents. Ash content and solubility values in 1% NaOH were determined according to SNI 14-1031-1989 and ASTM D 1109 – 84 (2001), respectively.

Alkaline Pulping of Wood

Alkaline pulping of red meranti wood (250 g, oven-dried chip) was performed using a digester. The following kraft, soda, and soda-AQ cooking conditions were used: maximum cooking temperature of 170 °C, time required to reach the targeted maximum temperature of 60 min, cooking time at maximum temperature of 120 min, and liquor-to-wood ratio of 4 to 1. The active alkali was 20% as NaOH on oven-dry wood in all cases. Sulfidity of 25% was used in the kraft pulping, whereas the AQ percentage was set 0.1% based on the oven-dried chip in the case of soda-AQ pulping. A total of two replications were then made for each treatment and the average reading was taken. Subsequently, the pulps were washed, screened and processed using fibre sorter equipped with a 100 mesh slot screen. Pulp yields and rejects were determined based on the oven-dry weight of wood chips initially charged to the digester. Pulps then were evaluated for kappa number (SNI ISO 302, 2014). Pulpability factor was calculated by dividing screened pulp yield by the kappa number (Little et al. 2003).

Beating and Physic-mechanical Properties of Pulp

Pulp samples were withdrawn on the Niagara beater after beating for 200~300 ml Canadian Standard Freeness (CSF) testing (SNI ISO 5267-2-2010) and hand-sheet formation (SNI ISO 5269-1-2012). Hand-sheets, each weighing 80 g/m², were made in a laboratory-scale sheet (diameter of 15.9 cm). Hand-sheets were conditioned (23 ± 2 °C and 50 ± 2% relative humidity) and tested for tear (SNI 14-0436-1989), tensile (SNI 14-0437-1989-A), burst (SNI ISO 2758-2011), brightness (SNI ISO 2470-1-2014), and opacity (ISO 2471, 2008).

Results and Discussion

Physical, Morphological, and Chemical Properties

The pulp and paper properties of wood species depend considerably upon their basic properties. Therefore, physical properties (Table 1) and the chemical composition (Table 2) of red meranti were determined to better understand the pulp and paper properties. The basic density of wood affects the pulp production. Pulpwod with a basic density greater than 0.60 g/cm³ is not recommended (Little et al. 2003). Lower wood density certainly gives less pulp production per digester. Table 1 shows data on the properties of red meranti and compares the data with those of A. mangium (7 years,
The basic density of red meranti was 0.42 g/cm³, which was slightly lower than that of *A. mangium* (Table 1), and the wood can be classified as medium density wood. It is assumed that the basic density of this sample was good for kraft pulping.

As observed under light microscope (Figs. 2), several cell types could be distinguished (fibres, rays, parenchyma cells, and vessel elements). Based on the measurement of various cell areas, the fibres of red meranti composed 67.14% of its total cells. The fibres of red meranti are long and cylindrical. The higher proportion of fibre and lower proportion ray (7.14%) and parenchyma (7.85%) tissues compared to *A. mangium* will give an advantage to produce higher yield. Fibre proportion and tear factor or folding endurance were positively correlated (Ona et al. 2001). However, the higher vessel proportion technically potentially causes vessel picking in paper manufacturing.

Longer fibre length, higher flexibility coefficient and/or lower wall-to-lumen ratio in wood are important aspects in pulping and papermaking (Xu et al. 2006). The fibre length of red meranti in this study was 1.07 mm, which was in the range of fibre length of most tropical hardwoods (Fengel and Wegener 1984). The fibre diameter was within medium range (21.16 μm). The fibre length, fibre wall thickness, and fibre diameter of red meranti were more than those of *A. mangium*. A positive correlation was found between fibre length and burst strength (Ona et al. 2001), tear strength (Shmulsky and Jones 2011) and folding endurance (Ona et al. 2001). Thick-walled fibres produce paper with low burst and tensile strength (Shmulsky and Jones 2011).

A direct correlation exists between fibre morphology and paper properties (Ververis et al. 2004). The slenderness and Runkel ratio values of red meranti were higher but its flexibility coefficient was lower than *A. mangium*’s. Raw materials with low Runkel ratio are preferred for paper making (Ohshima et al. 2005). It indicates the ability to collapse easily and form good fibre-to-fibre bonding. There is a positive correlation between slenderness ratio and folding endurance (Ona et al. 2001). Less flexible fibers do not produce large contact areas for fiber-to-fiber bonding. Its high flexibility is expected to have a positive effect on tensile and bursting strengths as well as on folding endurance (Xu et al. 2006). The results of the derived values in Table 1 indicate that red meranti sample fall within desirable ratios for paper making.

Table 1. The basic density, fibre morphological characteristics, and derived values of red meranti as compared to *A. mangium*

<table>
<thead>
<tr>
<th>Physical properties</th>
<th>Shorea selanica</th>
<th>Acacia mangium*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Basic density (g/cm³)</td>
<td>0.42</td>
<td>0.46</td>
</tr>
<tr>
<td>Cell Proportion</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fibre (%)</td>
<td>67.14</td>
<td>62.46</td>
</tr>
<tr>
<td>Ray (%)</td>
<td>7.14</td>
<td>9.77</td>
</tr>
<tr>
<td>Parenchyma (%)</td>
<td>7.85</td>
<td>15.66</td>
</tr>
<tr>
<td>Vessel (%)</td>
<td>17.85</td>
<td>12.11</td>
</tr>
<tr>
<td>Fibre Dimension</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fibre length (μm)</td>
<td>1070 ± 5.44</td>
<td>982</td>
</tr>
<tr>
<td>Fibre lumen diameter (μm)</td>
<td>13.20 ± 3.88</td>
<td>14.29</td>
</tr>
<tr>
<td>Fibre wall thickness (μm)</td>
<td>3.78 ± 0.45</td>
<td>2.55</td>
</tr>
<tr>
<td>Fibre diameter (μm)</td>
<td>21.16 ± 3.24</td>
<td>19.39</td>
</tr>
<tr>
<td>Derived values</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Runkel ratio</td>
<td>0.54 ± 0.12</td>
<td>0.37</td>
</tr>
<tr>
<td>Slenderness ratio</td>
<td>54.93 ± 8.55</td>
<td>51.29</td>
</tr>
<tr>
<td>Flexibility coefficient</td>
<td>0.62 ± 0.10</td>
<td>0.73</td>
</tr>
</tbody>
</table>

Remark : * Yahya et al. (2010), 7 year-old trees.
The lignin and cellulose contents in red meranti were almost similar to those in A. mangium (Table 1). The ethanol-toluene extract of this species was lower than that of A. mangium. Low extractive contents in wood are desirable for pulping, bleaching, and paper making. Total extractive content (the sum of ethanol-toluene extractive content and hot-water solubility) was about 5.17%. However, red meranti also had high level of solubility in 1% NaOH (26.02%). High 1% alkali solubility may be attributed to higher amount low molecular weight polysaccharides. The comparatively high 1% alkali solubility value may result in lower pulp yield from red meranti in the present investigation. The hemicellulose in red meranti was lower than that in A. mangium (Table 1). The lower hemicellulose in red meranti in this investigation may be the basis of higher pulp yield. The ash content (0.7%) was much lower than that of usual tropical species (1-3%) (Khristova et al. 1997).

Table 2. The chemical composition of red meranti as compared to A. mangium

<table>
<thead>
<tr>
<th>Chemical properties</th>
<th>Shorea selanica</th>
<th>Acacia mangium</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ethanol-toluene extractive content (%)</td>
<td>3.74</td>
<td>5.38</td>
</tr>
<tr>
<td>Hot-water solubility (%)</td>
<td>1.97</td>
<td>-</td>
</tr>
<tr>
<td>Holocellulose (%)</td>
<td>69.83</td>
<td>80.43</td>
</tr>
<tr>
<td>α-Cellulose (%)</td>
<td>45.20</td>
<td>45.71</td>
</tr>
<tr>
<td>Hemicellulose (%)</td>
<td>24.65</td>
<td>34.72</td>
</tr>
<tr>
<td>Lignin (%)</td>
<td>31.05</td>
<td>31.30</td>
</tr>
<tr>
<td>Ash (%)</td>
<td>0.73</td>
<td>-</td>
</tr>
<tr>
<td>Solubility in 1% NaOH (%)</td>
<td>26.02</td>
<td>-</td>
</tr>
</tbody>
</table>

Remark: * Yahya et al. (2010), 7 year-old trees

Pulping and Pulp Properties

Red meranti was pulped in soda, soda-AQ, and kraft pulping processes (Table 3). In all processes, the kappa number of pulp obtained was low (15.11-17.72). In 2 h of cooking, pulp yield obtained was 39.94%-47.41%. The highest screened yield (47.41%) and moderate kappa number (20.52) were obtained in soda pulping process, which were better than those of A. mangium. In the similar cooking condition, soda-AQ pulp had lower screened yield and higher kappa number than soda pulp. However, it is clearly seen that the kraft process showed its superiority for delignification over soda and soda-AQ processes with kappa number being 16.17 and reject being 0.56% but low in screened yield (39.94%).

Chemical additives can be used in pulping to reduce reactions of polysaccharides or increase reactivity of lignin. AQ additionally accelerates the delignification rate of pulping (Dimmel et al. 2003). Red meranti showed similar kappa number (about 20–22) in the soda and soda-AQ pulping processes, but the yield was higher in soda pulping process. In an earlier report, delignification was accelerated by AQ addition in soda liquor (Parthasarathy et al. 1995). Similar trend was also observed in A. nilotica pulping (Khristova and Karar 1999). It means that addition of AQ in soda liquor during red meranti pulping did not increased selectivity of pulping. Thus, another approach for different concentrations should be attempted.

The higher values of pulpability factor (greater than 2.34) is recommended with the ideal basic density between 0.46 and 0.52 g/cm³ (Gardner et al. 2001). Kraft process showed pulpability factor greater than 2.34, which indicate good pulpwod quality without having to do multiple cooks and interpolate to the desired 20 Kappa number. The values of red meranti were higher than those of A. mangium (2.41).
In this investigation, kraft pulp yield of red meranti was lower than that of A. mangium. Lower pulp yield in the present investigation may also be caused by high active alkali (20%) or sulfidity (25%) used in this experiment. The chips may be overcooked because of the use of more severe cooking conditions. A high 1% alkali solubility value, may be another reason of lower pulp yield.

Beatability of pulps is significant variable for the energy consumption of mills and generally depends on the chemical composition of pulps. The highest initial beatability degree was observed in soda-AQ samples (686 ml CSF), which indicates more energy consumption. It indicates higher lignin content as indicated by higher kappa number (21.92) and lower hemicellulose content.

Table 3. The pulp properties of red meranti alkaline pulp obtained at 20% active alkali

<table>
<thead>
<tr>
<th>Pulp Properties</th>
<th>Soda</th>
<th>Soda-AQ</th>
<th>Kraft</th>
<th>Acacia mangium-kraft a</th>
</tr>
</thead>
<tbody>
<tr>
<td>Screened yield (%)</td>
<td>47.41 ± 1.90</td>
<td>45.75 ± 3.44</td>
<td>39.94 ± 0.21</td>
<td>45.02</td>
</tr>
<tr>
<td>Reject (%)</td>
<td>1.51 ± 0.90</td>
<td>1.50 ± 1.17</td>
<td>0.56 ± 0.08</td>
<td>3.54</td>
</tr>
<tr>
<td>Kappa number</td>
<td>20.52 ± 0.79</td>
<td>21.92 ± 2.44</td>
<td>16.17 ± 0.44</td>
<td>18.61</td>
</tr>
<tr>
<td>Pulpability factor</td>
<td>2.31</td>
<td>2.08</td>
<td>2.47</td>
<td>2.41</td>
</tr>
<tr>
<td>Initial beatability degree (ml CSF)</td>
<td>630.0 ± 14.1</td>
<td>686.6 ± 11.5</td>
<td>637.0 ± 77.8</td>
<td>-</td>
</tr>
</tbody>
</table>


In order to examine the paper properties of red meranti, standard paper hand-sheets were produced from their pulps. The results of physical paper properties obtained at different alkaline processes are shown in Table 4. For the purpose of comparison, properties of hand-sheets from pulps of mangium pulp-woods obtained through kraft process (Haroen and Dimyati 2006) are also included in the Table. In an earlier study, the burst index produced from kraft-AQ gave lower values compared to those of soda-AQ in orange tree wood (Gonzales et al. 2013). Kraft pulp of Acacia auriculiformis showed slightly better tensile index than the soda pulp did and showed lower burst index than soda and soda-AQ pulps did (Jahan et al. 2009). In addition, kraft pulp had higher tear index than soda and soda-AQ pulp at higher beating degree. In this experiment, kraft pulping process showed slightly better tensile index compared to soda and soda-AQ processes. Furthermore, strength properties (burst and tear indices) and brightness level were improved with AQ addition. Kraft pulp showed higher tear index level than the soda pulp. Another paper demonstrated that the soda pulp of non-wood bagasse has better tensile and burst strength than the soda-AQ pulp owing to lower degree of delignification and higher pulp viscosity (Hedjazi et al. 2008). In kenaf pulping, kraft and kraft-AQ pulping processes produced slightly lower or comparable quality pulp than soda-AQ pulping process (Ang et al. 2010). Those varied patterns might be due to the different nature of raw materials and pulping conditions.

Brightness level has linear negative correlation with kappa number (Gulsoy and Tufek 2013). The comparatively high brightness value of soda-AQ pulp might be because residual lignin containing highly colored chromophoric groups were more intensively reduced (Serkov and Alen 2004). Although almost in similar levels, the highest opacity (99.66%) was obtained in soda pulping process. The high opacity values indicate the high the light scattering coefficient of the pulps which increases the fiber-air interface number. This behaviour is probably due to differences between pulps at physical and chemical levels (Gonzales et al. 2013).

Compared to kraft A. mangium pulp, the red meranti kraft pulp showed considerably lower tensile and burst indices. Technically, tensile and burst indices depend on bonding ability of fibers. It indicates that red meranti paper does not produce large contact areas for fiber-to-fiber bonding or has less flexible fibres. Based on fibre properties, lower burst and tensile indices of red meranti pulp could be explained by lower fiber flexibility or bonding ability (Xu et al. 2006) and higher Runkel ratio (Ohshima et al. 2005) of red meranti fibers (Table 2). Another explanation could be that severe cooking kraft conditions in this experiment caused polysaccharides degradation. More moderate cooking kraft conditions should be performed to improve the sheet physical properties in the future work. The tear index of red meranti was almost similar to that of A. mangium. It could be attributed to longer, (Shmulsky and Jones 2011), thicker fibres (Scott et al. 1995) and also its higher slenderness ratio (Ona et al. 2001) (Table 2). Brightness of red meranti pulp gave lower values compared to that of A. mangium. It indicates more residual lignin containing highly colored chromophoric groups on fiber surfaces (Serkov and Alen 2004). Based on national standard (SNI) for leaf (hardwood) bleached kraft, only tear index and opacity of red meranti pulp met the requirements (Table 4). This suggests that red meranti kraft pulp could be a potential reinforcement component in products based on mechanical pulp, such as newsprint. Furthermore, it will be necessary also to evaluate the pulp properties of S. selonica wood at the harvest age (5-6 years) from industrial plantation forests.
Table 4. The paper properties of red meranti alkaline pulp obtained at 20% active alkali

<table>
<thead>
<tr>
<th>Physical Properties</th>
<th>Soda</th>
<th>Soda-AQ</th>
<th>Kraft</th>
<th>Acacia mangium -kraft</th>
<th>SNI</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tensile index (Nm/g)</td>
<td>27.47 ± 2.10</td>
<td>27.83 ± 1.19</td>
<td>28.39 ± 0.28</td>
<td>78.75</td>
<td>45</td>
</tr>
<tr>
<td>Burst index (KPa m²/g)</td>
<td>2.18 ± 0.04</td>
<td>2.36 ± 0.13</td>
<td>1.98 ± 0.34</td>
<td>6.58</td>
<td>2.5</td>
</tr>
<tr>
<td>Tear index (mNm/g)</td>
<td>4.79 ± 0.75</td>
<td>8.47 ± 1.13</td>
<td>6.82 ± 0.47</td>
<td>6.94</td>
<td>5.5</td>
</tr>
<tr>
<td>Brightness (%)</td>
<td>17.89 ± 0.48</td>
<td>19.81 ± 0.99</td>
<td>17.25 ± 0.84</td>
<td>46.98</td>
<td>-</td>
</tr>
<tr>
<td>Opacity (%)</td>
<td>99.66 ± 0.19</td>
<td>98.20 ± 1.16</td>
<td>99.38 ± 0.10</td>
<td>-</td>
<td>80-90</td>
</tr>
</tbody>
</table>

Remark : ¹ Haroen and Dimyati (2006), ² Indonesia National Standard (SNI 6107, 2009)

Conclusions

The physical, morphological, and chemical properties of red meranti were evaluated in terms of its suitability for papermaking. The basic density, fibre proportion, and fibre length of red meranti in this study were within the range of those of tropical hardwoods. The lignin content in red meranti (and α-cellulose) was almost similar to that in Acacia mangium. The high level of total extractive content and high solubility in 1% NaOH of red meranti would potentially reduce the pulp yield. Soda, soda-AQ, and kraft pulping processes were studied. The highest screened yield was obtained in soda pulping. Kraft pulping showed the lowest kappa number and reject. Acceptable pulp yields were obtained at cooking in soda and soda-AQ pulping processes. Compared to soda and soda-AQ pulping processes, kraft pulping process gave slightly better tensile index whereas soda-AQ pulping process produced the highest value in tear index and burst index. Kraft pulp of red meranti showed considerably lower tensile and burst indices than the kraft pulp of A. mangium, which was probably due to higher concentration of chemicals during the cooking. However, soda-AQ pulp of red meranti produced better tear index compared to kraft pulp of A. mangium.

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