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**Thermomechanical pulping of novel Brazilian Eucalyptus hybrids**

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Thermomechanical pulping of novel Brazilian *Eucalyptus* hybrids

**Abstract:** The importance of *Eucalyptus* wood as a sustainable resource is well established. Mechanical pulp production is an energy-intensive process, and methods for decreasing energy demand are needed. In the present article, the structure of *Eucalyptus* hybrids and the produced pulps from the hybrids were assessed in terms of energy consumption, technical properties, and fiber morphology. The defibration and fiber development were investigated by traditional thermomechanical pulping in laboratory scale. There was no clear difference in the extent of defibration and fibrillation among the hybrids, with the exception of one hybrid (U2xGL1). Guaiacyl lignin contents of more than 7.2% had a minor influence on energy consumption, and this finding is different from the results of previous studies on chemimechanical pulp- ing processes. However, a low amount of guaiacyl lignin (6.1% based on wood) decreased the energy demand. This assessment is of major importance concerning the energy-saving defibration during thermomechanical pulping production.

**Keywords:** defibration, energy consumption, *Eucalyptus*, lignin structure, pulp properties, TMP

**Introduction**

*Eucalyptus* wood has become one of the most important fiber sources for the forest industries all over the world due to its fast growth in certain regions and its fairly good wood quality for a variety of applications. *Eucalyptus* has especially good properties for the production of chemical pulps (Magaton et al. 2009). Moreover, *Eucalyptus* fibers have a high potential as a reinforcement in biocomposites (Tonoli et al. 2009, 2010a). For example, *Eucalyptus* fibers produced by the mechanical processes, which have high lignin content, improve the long-term performance of the fiber-cement composites (Tonoli et al. 2010b; Muguet et al. 2012). The refining of thermomechanical pulping (TMP) can be thought to involve two main phases: (1) defibration or fiber separation, where the wood matrix is softened, and the separation of fiber bunches and single fibers begins, and (2) fiber development, where the wood fibers start to be disintegrated with the main effects, such as delamination and internal fibrillation (Fernando and Daniel 2008; Fernando et al. 2011, 2012).

There are several data available concerning *Eucalyptus* mechanical pulp production (Cort and Bohn 1991; Area et al. 1995, 1998, 2007; Xu et al. 1996; Ognar and Xu 1998; Xu and Sabourin 1999; Jones and Richardson 2000, 2001; Browne et al. 2001; Xu 2002; Area and Kruzolek 2008; Muguet et al. 2012). However, the high amount of energy demand during mechanical pulping is a key limiting factor for the process feasibility (Browne et al. 2001). Therefore, understanding the defibration as well as the factors affecting it is necessary. Chips' chemical pretreatment and the application of enzymes have been shown already to decrease energy consumption during refining (Cort and Bohn 1991; Area et al. 1995; Xu et al. 1996; Xu and Sabourin 1999; Xu 2002; Hart et al. 2009a,b; Masarin et al. 2009; Lecourt et al. 2010; Li et al. 2011). In addition, energy demand can be decreased by reducing raw materials variability (Dundar et al. 2009) or by changing the spout angle of wood chipper (Hellström et al. 2011). In TMP production, however, other factors have been shown to decrease the energy demand, such as decreasing discharge consistency from the primary refiner, on a double-stage refiner (Alami et al. 1997), increasing refining temperature/pressure (Muhic et al. 2010), and compressing chips before refining (Gorski et al. 2010). Earlywood and latewood fractions of wood also have been...
shown to affect TMP process energy demand (Lanouette et al. 2010).

The studies regarding the understanding of the main variables affecting the refining energy demand, such as the chemical and morphologic structure of wood, are scarce. Such studies may have important implications because the results may open new possibilities for hardwood TMP production.

Lignins in hardwoods consist mainly of guaiacyl units (i.e., G units, with one methoxy group in the aromatic ring) and syringyl units (i.e., S units, with two methoxy groups) (Sarkanen and Ludwig 1971; Boerjan et al. 2003). In our previous studies (Muguet et al. 2012), high G unit contents of Eucalyptus hybrids increased the specific energy consumption (SEC) during alkaline peroxide mechanical pulping (APMP). However, it would be also important to understand the behavior of Eucalyptus wood in nonchemical systems. TMP has been shown to be feasible in industrial scale for softwoods and in case of some hardwoods, such as aspen wood. A deeper understanding of the factors of SEC of hardwoods might contribute to more production of TMP from hardwoods. New results may also serve as guidelines for the traditional and genetic breeding of trees, which could lead to hybrids with less energy demand during TMP production.

In this work, mainly traditional TMP processes were in focus. The objective was the evaluation of the behavior of a new generation of Eucalyptus hybrids during the process. Relations are sought for between the structural components of wood, focusing mainly on lignin, and the SEC development of fibers and their properties during TMP production.

Material and methods

Four different hybrids of Brazilian-grown Eucalyptus trees were in focus, which are from the Brazilian Network of Eucalyptus Genome Research — Genolyptus (a nationwide network of laboratories and forestry companies devoted to an integrated molecular breeding approach) (Grattapaglia 2004). The wood samples are coded based on the crossings: E. urophylla ×E. globulus (U2xGL1), [E. dunnii ×E. grandis] ×E. globulus (DGxGL2), E. grandis ×[E. urophylla ×E. globulus] (G1xUGL), and [E. dunnii ×E. grandis] ×E. urophylla (DGxU2). These hybrids were chosen from 18 other hybrids of the same Genolyptus project due to their similarities in chemical and physical characteristics. Such properties ensure that differences in total carbohydrate and total lignin content, density, and some morphologic features (e.g., cell wall thickness) would have a minimum impact on the SEC variations. However, the guaiacyl lignin (G lignin) content differed notably (in the range of 6.1–8.3%) based on wood; Table 1) among the hybrids. The syringyl-to-guaiacyl (S/G) ratio was determined by the nitrobenzene oxidation method (Lin and Dence 1992) and the G lignin content was calculated taking into account the S/G values and the total lignin content from the wood samples.

Wood density was evaluated according to the TAPPI standard T258 om-06. The carbohydrate composition was analyzed by high-performance anion exchange chromatography with pulsed amperometric detection after acid hydrolysis following the procedure described by Wallis et al. (1996). Klasson lignin and acid soluble lignin were measured according to Gomide and Demuner (1986) and Goldschmid (1971), respectively. Lignin content was defined as the sum of Klasson and acid soluble lignin as described by Dence (1992). S/G ratio was evaluated according to Lin and Dence (1992). Total uronic acids and acetyl groups were evaluated according to Scott (1979) and Solar et al. (1987), respectively.

The TMP process was carried out in two steps: (1) The wood chips were impregnated with deionized water in a rotating digester. To ensure complete saturation of the chips, 10 bar of pure oxygen was inserted in the digester, for 5 h, at 4.1 kg m⁻³ liquid-to-wood ratio and 25°C. Then, the chips were removed from the digester and rested for at least 48 h at room temperature and atmospheric pressure in sealed containers. (2) The mechanical defibration was carried out on 100 g o.d. chips samples in a wing defibrator consisting of four static blades; the refining gap between blades and inner refiner wall was 1.0 mm. Other parameters are as follows: approximately 750 rpm; at approximately 37% consistency; 130°C; and refining times of 5, 10, and 15 min. Before the refining experiments, the chips were preheated inside the refiner for 5 min, and the condensate was released before the refining process started. Pulps were screened with a slot screener of 0.17 mm and tested for Canadian Standard Freeness (ISO 5267-2:2001). Handsheets were prepared (ISO 5269:1:2005) and tested for grammage (ISO 536:1995), density (ISO 534:1988), tear strength (ISO 1974:1990, Elmendorf method), tensile index (SCAN-P38), and opacity and brightness (ISO 2470:1999). Fiber morphologic analyses were carried out with FiberLab analyzer (Metso Automation, Vantaa, Finland).

Table 1  Chemical and physical characterization of the Eucalyptus hybrids investigated.

<table>
<thead>
<tr>
<th>Composition, properties</th>
<th>Eucalyptus hybrids</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>U2xGL1</td>
</tr>
<tr>
<td>Carbohydrates (%)</td>
<td></td>
</tr>
<tr>
<td>Glucans</td>
<td>46.8</td>
</tr>
<tr>
<td>Xylans</td>
<td>13.7</td>
</tr>
<tr>
<td>Others</td>
<td>2.3</td>
</tr>
<tr>
<td>Lignin (%)</td>
<td></td>
</tr>
<tr>
<td>Klasson</td>
<td>24.3</td>
</tr>
<tr>
<td>Acid soluble</td>
<td>5.8</td>
</tr>
<tr>
<td>Total</td>
<td>30.1</td>
</tr>
<tr>
<td>S/G ratios</td>
<td>4.0</td>
</tr>
<tr>
<td>Guaiacyl units (%)</td>
<td>6.1</td>
</tr>
<tr>
<td>Acetyl groups (%)</td>
<td>2.7</td>
</tr>
<tr>
<td>Uronic acids (%)</td>
<td>4.1</td>
</tr>
<tr>
<td>Density (kg m⁻³)</td>
<td>506</td>
</tr>
</tbody>
</table>

All percentages are based on wood. For the explanation of the abbreviations, see Materials and methods.
Results and discussion

TMP refinings

Because the wood composition is similar among the samples (Table 1), the refining behavior of almost all hybrids is also similar; an exception is the U2xGL1 hybrid (Figure 1a).

It is worth noting that the refiner used in this study, based on blades, consumes more energy than pilot-scale or industrial disc refiners (Xu and Sabourin 1999; Jones and Richardson 2000, 2001). It was assumed that, at least as an approximation, the total applied energy with different refining systems will be proportional to the energy that manifests itself as changes to the fibers (Muguet et al. 2012). Table 2 shows the summary of the refining results. The content of rejects after 0.17 mm screening is in line with TMP production data from other wood types (Lönnberg 2009). Obviously, the amount of fines approximately follows the trend of rejects content.

The lignin glass transition temperature has a direct influence on the defibration. At temperatures below the lignin glass transition temperature, the cell wall can be considered as consisting of largely tangentially oriented lamellae of two zones, each of which has both hard and soft components. This means that the microfibrils consist of crystalline cellulose (stiff and “hard” segments) and the paracrystalline cellulose (as “soft” segments), which is surrounded by a matrix, and also consist of the soft and hard components, namely from the “soft” hemicelluloses and the “hard” lignin. Each zone is incompletely softened, and the overall fracture modulus of the wood remains high (Irvine 1985). That is the case of the TMP process, where the lignin as the main component of the compound middle lamella (CML) remains hard, whereas the rest of the cell wall is more softened. In this case, the defibration takes place mostly in the segment between the primary wall and the S1 layer of the secondary fiber wall. However, in a chemimechanical process, such as chemithermomechanical pulp and APMP processes, where wood is treated with chemicals before defibration, the lignin properties are altered in such a way that the softening temperature is lowered. This leads to improved defibration, which takes place mostly on the middle lamella or CML (Franzén 1986). Our previous studies (Muguet et al. 2012) showed a high positive correlation between the G lignin content and the SEC to reach a given freeness level. This observation was interpreted as that the G lignins of

<table>
<thead>
<tr>
<th>Eucalyptus hybrids</th>
<th>Refining time (min)</th>
<th>SEC (MWh o.d.⁻¹)</th>
<th>Freeness, CSF (ml)</th>
<th>Rejects (%)</th>
<th>Fines (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U2xGL1</td>
<td>5</td>
<td>2.59</td>
<td>445</td>
<td>3.4</td>
<td>21.8</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>4.39</td>
<td>320</td>
<td>2.5</td>
<td>22.2</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>5.87</td>
<td>200</td>
<td>2.5</td>
<td>22.0</td>
</tr>
<tr>
<td>DGxGL2</td>
<td>5</td>
<td>2.65</td>
<td>525</td>
<td>4.8</td>
<td>19.9</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>4.37</td>
<td>370</td>
<td>3.6</td>
<td>15.3</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>5.80</td>
<td>310</td>
<td>3.4</td>
<td>15.3</td>
</tr>
<tr>
<td>G1xUGL</td>
<td>5</td>
<td>2.62</td>
<td>525</td>
<td>2.2</td>
<td>14.0</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>4.39</td>
<td>375</td>
<td>2.2</td>
<td>14.6</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>5.83</td>
<td>295</td>
<td>1.2</td>
<td>12.5</td>
</tr>
<tr>
<td>DGxU2</td>
<td>5</td>
<td>2.62</td>
<td>525</td>
<td>3.4</td>
<td>22.1</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>4.42</td>
<td>370</td>
<td>3.3</td>
<td>20.9</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>5.85</td>
<td>300</td>
<td>2.8</td>
<td>20.6</td>
</tr>
</tbody>
</table>

For the explanation of the abbreviations, see Materials and methods. CSF, Canadian Standard Freeness.

Figure 1 Relation of the SEC to (a) freeness (Canadian Standard Freeness) and (b) guaiacyl units at the freeness level of 450 ml in Eucalyptus hybrids.

For the explanation of the abbreviations, see Materials and methods.
hardwoods are mainly located in the CML of fibers and in the wall of vessels (Musha and Goring 1975). The validity of this was also shown for Eucalyptus wood (Watanabe et al. 2004). The content of G lignin varies notably among the Eucalyptus wood hybrids listed in Table 1. Figure 1b shows the relationship between G unit content and SEC to reach a 450 ml freeness. In the case of the U2xGL1 hybrid, the value was obtained by extrapolation (Figure 1a). The energy demand in three cases of the four is very similar probably because of the low amount of G units in the fibers’ secondary wall of hardwoods (Musha and Goring 1975; Watanabe et al. 2004).

However, the SEC for the U2xGL1 hybrid was substantially lower compared with the other samples to reach a given freeness (450 ml). This can be explained by the low G content in the wood of this hybrid. G lignin moieties are known to increase the glass transition temperature of lignins probably because they increase the cross-linking density of lignins (Olsson and Säkkinen 1997).

Pulp physical, mechanical, and optical properties

Ideally, paper sheets from TMP should have high opacity, brightness, bulk, smoothness, as well as suitable pore structure at low grammage, without the need of excessive reinforcement (Lönnberg 2009). Table 3 shows the summary of the handsheets properties. The differences in the brightness values among the hybrids reflect the natural differences in the color of the wood material. Expectedly, sheet density tends to increase with longer refining times as the more flexible fibers increase the relative bonding area. External fibrillation also induces sheet density increment by intensifying interfiber bonding.

Tensile index gives indications of how paper web will behave on the manufacturing process and can be highly affected by bonding strength between fibers, stiffness, and fiber length. In general, the tensile index values were low for all four hybrids, but are in line with previous results, at the same density level (Xu and Sabourin 1999; Muguet et al. 2012). Particularly, DGxGL2 and DGxU2 had similar fiber length, fiber width, and fibrillation index (Table 4). However, the fines content of DGxGL2 is slightly lower than of DGxU2, which could indicate that the fines are acting like spacers, reducing bonding ability and thus lowering both tensile and tear index values.

Tear index is mainly influenced by fiber strength and fiber length (Kärenlampi 1996). Differences can be seen for tear index, being greater for U2xGL1 hybrid. Some of the fiber morphologic characteristics, such as fibrillation index, have a great influence on tear index, which will be discussed on the next section. In addition, tear index is better interpretable when correlated with tensile index, because the former is related to intrinsic fiber strength and the latter is more connected to fiber bonding strength (Figure 2a). The great correlation demonstrates that woods with less favorable properties could be improved by more intense refining.

Table 4  Summary of morphologic properties of pulp fibers derived from all four Eucalyptus hybrids.

<table>
<thead>
<tr>
<th>Hybrids</th>
<th>Refining time (min)</th>
<th>Curl (%)</th>
<th>Fibrillation (%)</th>
<th>Length (mm)</th>
<th>Width (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U2xGL1</td>
<td>5</td>
<td>6.7</td>
<td>2.60</td>
<td>0.47</td>
<td>15.86</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>6.6</td>
<td>3.26</td>
<td>0.46</td>
<td>15.42</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>6.5</td>
<td>3.52</td>
<td>0.46</td>
<td>15.42</td>
</tr>
<tr>
<td>DGxGL2</td>
<td>5</td>
<td>6.0</td>
<td>2.11</td>
<td>0.48</td>
<td>17.37</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>6.5</td>
<td>2.60</td>
<td>0.50</td>
<td>17.20</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>5.6</td>
<td>2.90</td>
<td>0.51</td>
<td>16.45</td>
</tr>
<tr>
<td>G1xUGL</td>
<td>5</td>
<td>7.4</td>
<td>2.06</td>
<td>0.57</td>
<td>16.83</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>6.7</td>
<td>2.61</td>
<td>0.60</td>
<td>16.70</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>7.1</td>
<td>2.72</td>
<td>0.58</td>
<td>16.25</td>
</tr>
<tr>
<td>DGxU2</td>
<td>5</td>
<td>5.2</td>
<td>1.80</td>
<td>0.47</td>
<td>17.00</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>5.7</td>
<td>2.31</td>
<td>0.46</td>
<td>16.52</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>5.0</td>
<td>2.39</td>
<td>0.47</td>
<td>16.37</td>
</tr>
</tbody>
</table>
match their counterparts on more intense refining. An interesting feature is that the hybrid DGxU2 was the one with lowest values in all technical properties assessed, which could also be seen for APMP processes (Muguet et al. 2012).

As discussed previously, the G units did not have great influence on the defibration, with the exception of the U2xGL1 hybrid, which showed better technical properties in comparison with the other hybrids. However, the fiber development across the refining seems to have also an effect. This is a hint to the important role of morphologic properties in terms of the fiber development and the resulting papemaking properties.

### Fiber morphology

Fiber morphologic characteristics, such as microfibril angle (Page et al. 1977; Salmén and De Ruvo 1985), coarseness, fiber length (El-Hosseiny and Anderson 1999), curl index (Mohlin and Alfredsson 1990; Fellers et al. 2001), have been shown to be of extreme importance for pulp and paper properties. Table 4 shows the summary of these morphologic features. Prinsen et al. (2012) evaluated wood fibers’ properties from both G1xUGL and DGxU2 hybrids. Fiber width results are in line with the morphologic characteristics from the wood fibers (higher fraction of 12–23 μm), which is an indication that the presence of fiber bundles on the final pulp is minimal. Because wood density values of all hybrids are similar, the conclusions regarding fiber width are also valid for the U2xGL1 and DGxGL2 hybrids.

There is no strong correlation between the morphologic properties on the one hand and the physical and mechanical properties on the other, with the exception of the fibrillation index. External fibrillation has been already shown to have an effect on tensile strength (Kang and Paulapuro 2006); however, in other studies (Hartman 1984; Kang et al. 2006), such trend was not clear. In this study, fibrillation index has a strong effect on the physical, mechanical, and optical properties of the handsheets prepared from the TMP pulps (Figure 2c).

External fibrillation enhances interfiber bonding, enhancing the compatibility of the fiber network (density), which reflects directly on mechanical properties (e.g., tensile and tear indices). The intensity of fibrillation induces the formation of different types of fines, which affects directly the light scattering coefficient. In addition, opacity has an inverse correlation with curl index ($R^2=0.623$). Mohlin and Alfredsson (1990) showed that the fibrillation index has an effect on light scattering.
Seemingly, curled fibers of the handsheets tend to increase light absorption across the sheet Z-profile, decreasing the light transmission.

Conclusions

TMP of *Eucalyptus* is challenging, especially regarding the high energy demand of the process. G lignin contents of more than 7.2% (based on wood) had a minor influence on energy consumption, but the SEC was lower beginning with G lignin content of approximately 6.1% (based on wood), which was the case in the hybrid U2xGL1. However, it is not possible to give an exact threshold value, which is relevant for SEC lowering. The macrocharacteristics of the investigated *Eucalyptus* hybrid samples, such as density and total carbohydrate contents, and their influence on the SEC is complex. Moreover, no experiments could be carried out with *Eucalyptus* samples with G lignin content between 6.1% and 7.2%. Therefore, the results must be interpreted with care. More in-depth studies are needed to understand the complexity of influence of glass transition temperature of lignins and the hemicelluloses on the defibration mechanism and the SEC of TMP.

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