Characterization of fiber development in high- and low-consistency refining of primary mechanical pulp

Abstract: Primary refined softwood was subjected to high-consistency (HC) or low-consistency (LC) secondary refining, and the nature of the development of the internal and external fiber microstructure and ultrastructure has been compared. The primary refining of mixed softwood as a raw material was performed in pilot scale by the advanced thermomechanical pulp process. The study was aiming at the comparative characterization of LC and HC pulps at the fiber level when produced with similar and well-characterized handsheet properties. The formerly described Simons' staining method was applied. A significant degree of fiber wall delamination/internal fibrillation (D/IF) was observed during both LC and HC refining. Both the energy input and the refining consistency had a significant impact on elevating the degree of fiber wall D/IF. The statistical evaluation of internal fiber development indicated that the fiber populations in LC- and HC-refined pulps had a similar degree of fiber wall D/IF despite having a large difference in refining energy input (420 kW h odt⁻¹), confirming that D/IF was promoted more energy-efficiently in LC than in HC refining. The characteristic of the external fiber development from HC and LC refining was very different. Secondary LC refining promoted fiber surfaces with ribbons of thin hairlike threads arising from the inner secondary S2 layer that occasionally developed along the whole fiber length. Broad sheet- and lamella-type external fibrillation from the S2 was typical for HC refining, and these characteristics were rarely observed in the LC pulps. The mechanisms for LC and HC fiber development are proposed. The cell wall characteristics (internal and external) of the pulp fibers appear to govern most of the physical and optical properties in handsheets.

Keywords: ATMP, delamination/internal fibrillation (D/IF), fiber characterization, fiber development, surface ultrastructure of fibers, HC refining, LC refining, SEM, Simons’ staining

Introduction

Mechanical pulping is expected to produce pulp of suitable quality that meets the customer requirements at the lowest possible energy expenditure. The advanced thermomechanical pulp (ATMP) process gives rise to pulps with similar properties to those produced by a traditional TMP process, but the energy input is significantly lower (Gorski et al. 2011a, 2012c; Johansson et al. 2011). The low-consistency (LC) refining of primary ATMP further reduces the total energy requirement: ~300 kW h odt⁻¹ less energy is needed to achieve the target pulp and paper properties by secondary LC refining compared with the secondary high-consistency (HC) refining of primary ATMP (Gorski et al., 2012a).

The cross-sectional dimensions of fibers decrease with HC refining (Kure 1999) but remain unchanged or even increase with LC refining perhaps due to internal delamination and swelling (Gorski et al., 2012a). The distribution of fiber fractions from the two processes also differs considerably. During LC refining, the long fiber fraction (R30) is lowered and the middle fraction (P30/R100) is elevated (Gorski et al. 2012b). LC-refined fibers are more flexible but have a smaller external surface area compared with HC-refined fibers (Gorski et al., 2012a). The interpretation is that LC refining creates less external fibrillation (EF) and more internal delamination compared with HC refining, although the characteristic of the fibrillation has not been studied. The mechanisms leading to these observations are unknown.

The quality of the final paper product correlates strongly with the development of fiber properties such as the amount of split fibers and decreased cross-sectional
dimensions (Reme et al. 1998; Kure 1999; Gorski and Hill 2012), fiber flexibility (Corson 1989; Fernando et al. 2011, 2012; Gorski et al. 2011b,c, 2012a), fiber wall thickness (Braaten 2000; Ferluc et al. 2010), fiber bonding (Skowronski 1990; Reme et al. 1998), collapsibility of fibers (Reme et al. 1998; Kure 1999), development of the long fiber fraction (Corson et al. 2003), and EF/internal fibrillation (EF/IF) (Fernando et al. 2011, 2012; Gorski et al., 2012a). EF/IF can presumably be tailored by the parameters of the refining process (Miles and Karnis 1991; Kang and Paulapuro 2006; Fernando et al. 2011). It is known that IF (Claudio-da-Silva 1983; Abitz and Luner 1989; Mohlin 1989; Paavilainen 1993; Fernando et al. 2011, 2012) and EF (Mohlin 1989; Kang and Paulapuro 2006; Fernando 2007) determine, to a large extent, both the strength and the optical properties of paper.

The focus of this study was to investigate the mechanisms of fiber development in secondary HC and LC refining of primary ATMP. Therefore, the characterization of fiber development was performed, aiming to improve the understanding of the interrelation between HC- and LC-refined fiber profiles and the optical and strength properties of paper sheets. The intention was to contribute to a better understanding of the fundamental mechanisms governing this fiber property development in LC- and HC-refined pulps at the microstructural and ultrastructural levels of cell walls. Most of all, the influence of refining processes on the structural changes to the fiber walls and thereby the final paper quality should be better understood.

**Materials and methods**

**Raw materials and refining**

The mixed softwood chips were obtained from a Canadian TMP mill. Composition: ~80% lodgepole pine and 20% Sitka spruce and western balsam fir from the interior of British Columbia. The ATMP process (Hill et al. 2010; Johansson et al. 2011) for HC refining was conducted at the Andritz pilot plant (Springfield, OH, USA). The chips were preheated and defibrated by means of a mechanical pretreatment consisting of a modular screw device (Impressaﬁner; Andritz, 40 kW h odt⁻¹ energy input) and a Fiberizer (Andritz; 220 kW h odt⁻¹ energy input). The fiberized material was first stage refined under conditions of RTS (low retention time, high temperature, and high refiner speed) according to Sabourin et al. (1997), with an energy input of 530 kW h odt⁻¹; 3.1% bisulfite was added through the dilution water.

A part of the first-stage pulp was further refined by an atmospheric HC double-disc refiner with four different energy inputs (610–1000 kW h odt⁻¹). LC refining of the first-stage ATMP was conducted in two optimized stages at the pilot plant in the Pulp and Paper Centre, The University of British Columbia (Vancouver, British Columbia, Canada). The pulp was disintegrated in 60°C water for 4 h before refining at LC (3–3.4%) using a 16-inch LC pilot refiner equipped with a 16-inch overhang segments and driven by a 110 kW variable frequency motor. The refining conditions were varied by changing the gap of the refiner while the throughput was kept constant. This gave specific energy inputs of 70–180 kW h odt⁻¹ in the first LC stage and 70–240 kW h odt⁻¹ in the second LC stage. The FineBar (Aikawa Fiber Technologies) segments were used (bar edge length 5.59 mm, groove depth 4.8 mm, groove width 2.4 mm, bar width 1 mm, and bar angle 15°). The pulps and handsheets formed from the pulps (Table 1) were subjected to the Technical Association of the Pulp and Paper Industry standards testing as described previously (Gorski et al. 2012b).

**Pulps for fiber characterization**

Five pulps were chosen for the characterization of fiber properties (Table 1) based on specific energy consumption (SEC) and the properties of handsheets made from the refined pulps for representing extreme levels and/or similar properties (Figure 1; cf. Table 1). This is due to the fact that paper with similar physical properties can be produced from LC-refined pulp with significantly lower energy input compared with HC-refined pulp. Therefore, in this study, an emphasis was given to the comparisons of pulps with a similar level of handsheet properties.

**Simons’ staining of pulp fibers**

The development of fiber wall delamination/IF (D/IF) was evaluated according to Fernando and Daniel’s (2010) method of Simons’ staining (SS). Staining was performed on ~1 g of never-dried pulp from each of the five samples and then immediately examined and analyzed by light microscopy as described in the quoted literature.

<table>
<thead>
<tr>
<th>Code</th>
<th>Position (stage)</th>
<th>SEC (kW h odt⁻¹)</th>
<th><strong>Basic properties</strong></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>TI (N m g⁻¹)</td>
</tr>
<tr>
<td>HC1</td>
<td>First HC stage</td>
<td>800</td>
<td>21.6</td>
</tr>
<tr>
<td>HC2a</td>
<td>Second HC stage</td>
<td>1410</td>
<td>37.8</td>
</tr>
<tr>
<td>HC2b</td>
<td>Second HC stage</td>
<td>1570</td>
<td>43.6</td>
</tr>
<tr>
<td>LC2</td>
<td>Second LC stage</td>
<td>980</td>
<td>29.7</td>
</tr>
<tr>
<td>LC3</td>
<td>Third LC stage</td>
<td>1150</td>
<td>39.4</td>
</tr>
</tbody>
</table>

TI, tensile index; CSF, Canadian standard freeness; LS, light scattering coefficient.
Data collection and statistical analysis

Five subfiber populations (SFPs; see Fernando and Daniel 2010) that represent the different categories of fibers possessing varying degrees of D/IF in a given pulp were identified following SS. Each category reflects varying levels of internal fiber development in a given pulp. The SFPs from stained samples were obtained manually by light microscopy (200 fibers from each pulp were evaluated) and analyzed as described (Fernando and Daniel 2010). Raw data were first summarized graphically for the ease and direct observation of D/IF development in the cell walls. Thereafter, the data were evaluated and statistically analysed with SAS software (SAS/STAT version 9.3 for Windows XP-Pro platform; SAS Institute, Cary, NC, USA). Ordinal logistic regression tests were performed for the assessment of the significance of the differences concerning the degree of D/IF of the pulp fiber cell walls.

Scanning electron microscopy

The characterization of surface ultrastructure and EF of pulp fibers was performed by scanning electron microscopy (SEM). Approximately 2 g of wet samples were dehydrated separately in a series of increasing ethanol and then acetone concentrations (Fernando and Daniel 2008). The samples were then dried by means of an Agar E3000 critical point dryer (Agar Scientific, Stansted, UK) with CO₂ as a drying agent and coated with gold with an Emitech K550X sputter device (Quorum Technologies Ltd, Ashford, Kent, UK). SEM instrument: Philips XL 30 ESEM (FEI Company, Eindhoven, Netherlands) operated at 10 kV.

Results and discussion

The handsheets with a tensile index of 40 Nm g⁻¹ and a light scattering of 59 m² kg⁻¹ were produced from both HC- and LC-refined ATMP. The HC refining required ~1450 kW h odt⁻¹ SEC, whereas ~300 kW h odt⁻¹ less SEC was consumed in LC refining (HC2a vs. LC3; Figure 1). The apparent density of the handsheets developed similarly for both HC and LC refining compared at equal tensile index level, whereas the tear index, length weighted average fiber length, stretch, and tensile energy absorption (TEA) were lower for the LC-refined pulp (Table 1). LC refining significantly reduced the size of R30 fiber fraction and considerably increased the middle fraction (Gorski et al. 2012a,b).

Internal fiber development

The response of individual fibers to SS differed; fibers stained in varying color intensities from blue to yellow/orange with increasing severity of the fiber wall D/IF. The observation is in line with that of previous works (Blanchette et al. 1992; Fernando and Daniel 2010). This indicates a diverse distribution of the individual fiber development within a given pulp. This can be expected because the heterogeneous wood fibers are treated differently depending on SEC, temperature, and intensity, and these parameters change both the chemical and the morphologic structures of the native fiber wall as illustrated in the literature (Claudio-da-Silva 1983; Fernando 2007; Daniel et al. 2009). For example, an increase in SEC or refining intensity enhances the proportion of treated fibers with high degree of D/IF, thereby improving the overall internal fiber development of a pulp (Fernando et al. 2012).

An overview of the results from the SS study is shown in Figure 2 and outlines an initial assessment of fiber development. The five SFPs were simplified into three major groups: “non-D/IF”, “low D/IF”, and “high D/IF”, as described in the method for ease of understanding (Figure 2a). As expected, increasing the SEC significantly reduced the percentage of untreated fiber population resulting in enhanced fiber development (Figure 2a, red line). The difference was especially pronounced when the initial primary-stage pulp (HC1) was compared with any of the secondary pulps irrespective of refining consistency (e.g., HC1 vs. LC2). The primary pulp with the lowest refining energy input (HC1; 800 kW h odt⁻¹) consisted of ~63% untreated stiff fibers and only 16% fibers with a high degree of D/IF. In contrast, pulp HC2b with the highest refining energy input (HC1; 1570 kW h odt⁻¹) was the most developed at the fiber wall level. It was dominated by a treated fiber population (~63%) untreated stiff fibers and only 16% fibers with a high degree of D/IF. In contrast, pulp HC2b with the highest SEC (1570 kW h odt⁻¹) was the most developed at the fiber wall level. It was dominated by a treated fiber population (~65%), with the majority of fibers from the high D/IF group (~38%) that represents the most flexible fiber fraction. The preliminary analysis also indicated that pulps LC3 and HC2b appeared to have more or less similar fiber populations concerning the degree of D/IF (61% and 65%).
promotes D/IF of the cell wall more efficiently than HC refining. This implies that, within the LC system, more energy was transferred effectively onto fibers (i.e., interaction effect of SEC and LC system) as discussed below.

**Statistical analysis for the degree of D/IF**

Detailed statistical analyses by means of the ordinal logistic regression test (Table 2) shows (a) the overall significance in the degree of D/IF among all pulps, (b) the effect of the two refining conditions (i.e., the refining energy input and the refining consistency), and (c) the impact of the different refining conditions on morphologic changes (i.e., D/IF) in the different pulps by analyzing two at a time.

The greatest difference was found in the degree of wall D/IF among the five pulps at 0.01% significance level (P < 0.0001; Table 2). This resulted from both increasing SEC, which is in line with previous results (Fernando et al. 2011, 2012), and changing the refining conditions (HC/LC). The statistical evidence for this was provided when analyzing the two variables separately. The results indicated that both the energy input (P < 0.0001) and the LC/HC refining conditions (P < 0.0001) have highly significant influence on enhancing fiber wall D/IF (i.e., making stiff wood fibers flexible).

The statistical analysis performed pairwise is also informative and provides further evidence on the novel information regarding the internal fiber development mechanisms during the two processes. There is a significant difference in the development of fiber wall D/IF between primary HC and secondary LC pulps (HC1 vs. LC2; P = 0.0472). The difference in SEC between the

| Table 2 Logistic regression statistics for type 3 analysis of ordinal logistic regression test for significant differences between HC- and LC-refined pulps on the degree of fiber wall D/IF. |
|---|---|---|---|
| Source | DF | $\chi^2$ | Pr>\(\chi^2\) |
| Pulp$^a$ | 4 | 53.18 | <0.0001 |
| Energy$^b$ | 1 | 41.39 | <0.0001 |
| HC/LC system$^b$ | 1 | 15.66 | <0.0001 |
| HC1 vs. LC2$^c$ | 1 | 3.94 | 0.0472 |
| LC2 vs. LC3$^c$ | 1 | 8.43 | 0.0037 |
| LC3 vs. HC2a$^d$ | 1 | 0.04 | 0.8461 |
| HC2a vs. HC2b$^e$ | 1 | 2.36 | 0.1241 |
| LC3 vs. HC2b$^e$ | 1 | 1.50 | 0.2213 |

$^a$Overall significance.

$^b$Two refining conditions (energy and HC/LC system effect).

$^c$Comparing two pulps for differences in overall degree of D/IF of their fibers.
two pulps was 180 kW h odt\(^{-1}\). The internal fiber development between the two LC pulps is also significantly different with an increase in SEC of 170 kW h odt\(^{-1}\) (LC2 vs. LC3; \(P = 0.0037\)). However, the degree of D/IF developed between the two HC pulps was not significantly different, although the SEC difference of the two pulps was similar to that of the two LC pulps (HC2a vs. HC2b; \(P = 0.1241\)). The results thus indicate an interaction effect of energy and LC system that accelerates internal fiber development during LC refining. This is reflected in LC3 pulps that contained a greater percentage of treated fibers compared with LC2 (Figure 2a). An evidence for the interaction effect is also given by the raw data itself (Figure 2b) as discussed above. However, it should be noted that the interaction effect could not be statistically estimated because all the possible combinations of energy and consistency were not available in the data. For example, the combination high energy input and LC was missing due to the practical limitations of the refining system.

Furthermore, statistical analysis indicated that LC refining with low SEC produced pulps that was similar to high-energy HC pulps regarding the degree of internal fiber development (LC3 vs. HC2b; \(P = 0.2213\)). This provides a statistical evidence for the preliminary analysis and confirmed the improved energy efficiency of LC refining.

**Characterization of the fiber surface ultrastructure by SEM**

There were differences in the fiber surfaces especially between the primary HC1 pulp (Figure 3a) and the secondary HC2b pulp produced with high energy input (Figure 3b). The majority of fibers in HC1 (SEC of 800 kW h odt\(^{-1}\)) had the S1 secondary wall as their outer surface layer (Figures 3a and 4a). It was most often fibrillated into typical flake-like fibrils (Figure 4a, arrows), sometimes with remaining parts of the compound middle lamella (CML; Figures 3a and 4a). In addition, most of the HC1 fibers retained their native geometrical shape of wood fibers, which is indicative of intrinsic stiffness. On the contrary, almost all the fibers in the HC2b pulp (SEC of 1560 kW h odt\(^{-1}\)) displayed an exposed cellulose-rich S2 layer with a typical ribbon-type fibrillation and the fibers exhibited greater collapsibility/conformability (Figure 3b). The collapse of the fibers was most likely attributable to their thinner walls, which resulted from the removal of the major part of fiber wall materials as shown in Figure 3b.

Most of the fibers from the LC2 and LC3 pulps also exhibited an exposed S2 layer, often with typical ribbon-like fibrils projecting from the fiber surface (Figure 3c and d). Both LC-refined pulps appeared morphologically

---

**Figure 3** SEM micrographs of HC- and LC-refined TMP fibers with their surface morphologic characteristics (see Table 1). (a) HC1 fibers with the secondary S1 layer of the cell wall as the outer layer. The retention of the native geometrical shape of the fibers indicates inherent fiber stiffness. (b) Almost all the HC2b fibers possess the S2 as the outer layer with ribbon-type fibrillation or clear surfaces presumably due to complete peeling of fibrils.Collapsed never-dried fibers lost their square-like form. (c) Most fibers of LC2 show S1 as the outer layer. (d) The majority of LC3 fibers have the S2 layer exposed most often with hairlike fibrils protruding from the surface. Bars, 40 μm (a–d).
similar, although the LC2 with lower energy input had more fibers covered with outer S1 wall material, indicating an inferior peeling effect compared with the LC3 pulp from higher SEC.

A detailed SEM observation revealed important information related to external fiber development mechanisms during HC and LC refining. Severe fiber splitting was already initiated during the ATMP refining process in the first-stage pulp (Figure 4b, arrows). This is consistent with earlier observations where a split fiber index of ~0.30 was determined for first-stage pulp (Gorski et al., 2012a). Although this implied a strong refining action on the fibers already in the first stage, an extensive peeling of the fiber wall material into the inner secondary wall layers was observed only at high SEC.

Strong dissimilarities in fiber surface morphology were found between HC and LC pulps. HC refining with high SEC generated various types of characteristic S2 fibrillation (Figure 4c; e.g., ribbon-type fibrils) ranging from thin thread-like fibrils (Figure 4d, arrows) to broad sheet-like in the inner secondary wall layers was observed only at high SEC.

Strong dissimilarities in fiber surface morphology were found between HC and LC pulps. HC refining with high SEC generated various types of characteristic S2 fibrillation (Figure 4c; e.g., ribbon-type fibrils) ranging from thin thread-like fibrils (Figure 4d, arrows) to broad sheet-like
fibrils (Figure 4e and f) and lamellar sheets (Figure 4g and h) peeling from the fiber surfaces. Particularly, the broad sheet-like fibrils and lamellar sheets were common on fibers in HC2b, although they were also observed occasionally in the HC2a pulp with lower energy input.

LC refining promoted a mechanical interaction that led to a distinct fiber development mechanism, where fiber surfaces most often showed thin hairlike threads, sometimes along the whole fiber length (Figure 4i, l, and m). Broad sheet-like and lamellae types of EF were rarely observed in the LC pulps and the major hairlike surface wall structures were thread-like fibrils (i.e., similar to single macrofibrils; Figure 4j and k, arrowheads) or very thin ribbon-like fibrils originating from the S2 layer (Figure 4j and k, arrows). In mechanical pulping refining, microcracks are known to develop along naturally occurring weak zones in the lamellae of already exposed S2 layer (i.e., between single macrofibrils or aggregates of various sizes of macrofibrils within lamellae; Fernando and Daniel 2004). In this trial, LC refining enhanced the development of microcracks predominantly between smaller aggregates of macrofibrils (Figure 5a). The resulting ribbons of thin threads were thereby easily peeled off from the surface by the action of shearing forces inside the refiner (Figure 5b and c). The actual events taking place in the fiber outer wall during LC refining are illustrated in Figure 5.

A possible cause for the differences in the fiber surface ultrastructure may be the different refining environments in HC and LC refiners, which could influence the softening and surface properties of refined fibers. For example, Kerekes (2011) describes the difference in the coefficient of friction measured in HC (0.5–0.8) and LC (0.1–0.15) refiners. The motor effect and typical energy inputs in HC and LC refiners are also reported to follow the same relationship and are approximately 5–10 times less in the case of LC. The lower friction inside the LC refiners suggests a different type of mechanical interaction, which may explain the distinct fibrillation type observed.

**Hypothesis on the mechanisms of HC and LC refining**

Based on the results, it can be hypothesized that shear forces may dominate in HC refining and influence the observed fibrillation types (e.g., broader sheets/lamellae fibrils), which have a greater relative bonded area (RBA). In the case of LC refining, on the contrary, compressive forces are dominant. They are generated from fiber-to-fiber interaction by the hitting of less concentrated fibers/fiber bundles against each other, which is less probable in HC refining. The refiner bars play also a more prominent role (plate-to-fiber action) for cyclic compression in LC refiners. Therefore, it is most probable that these mechanical interactions (impact on fibers) in LC refiners impose high internal stresses/strains on fibers that should trigger...
D. Fernando et al.: Fiber development in refining efficiently and generate more microcracks in the wall leading to hairlike fibrillation. The previous studies on the action of shear and compressive forces conclude that the former contributes predominantly to EF via peeling mechanisms, whereas IF is the primary effect of cyclical repetitive compression (Hartman 1984; Kang and Paulapuro 2006; Kerekes and Senger 2006). Accordingly, the hypothesis proposed on refining mechanisms with different and selective refining actions between HC and LC refiners is the principle reason for the different fibrillation mechanisms and fiber responses observed.

Several studies report on the importance of plate clearance in terms of fiber properties (Härkönen et al. 2003; Murton and Duffy 2005; Kerekes and Senger 2006; Mohlin 2006; Eriksen and Hammar 2007). Compressive forces are known to dominate when the plate gap is small, and shearing forces are prominent when the gap is large. Härkönen et al. (2003) showed how consistency controls fiber residence time and plate clearance. The smallest values for the latter two parameters are attained with the lowest consistency. The plate clearance in LC refiners is very small compared with HC refiners, where it is several times larger (Eriksen and Hammar 2007). In addition, a very intensive turbulent motion of the pulps and the extension of the turbulence toward the outer radius of the refiner plate are obtained when the pulp consistency is lowest (Härkönen et al. 2003). These conditions most likely prevailed in the LC refiners of the present trial (at ~3% consistency), which promoted strong fiber-to-fiber as well as fiber-to-bar impacts (i.e., compressive forces). In contrast, at higher consistencies, repetitive compression was less influential during HC refining. However, more fundamental work is needed to clarify the mechanisms proposed for HC and LC refining.

Characteristic of fiber development and final product quality

The relationship of the three major D/IF groups to the tensile strength is established in earlier work (Fernando et al. 2011). The present study also showed very similar relationships (Figure 6a). A very strong positive correlation ($r^2=0.96$) to tensile index of handsheets was exhibited by pulp fibers with high D/IF. Untreated non-D/IF fibers had an inverse correlation with the same strength ($r^2=0.99$). Fibers with low D/IF showed a positive trend with lower strength gain. Furthermore, the three groups of fibers were correlated to the density of the handsheets with exactly the same strength and direction as they did with tensile strength (Figure 6b). The results thus emphasize the direct influence of the relative proportions of the two groups of high and low D/IF, which contain desirable flexible fibers, in a given pulp for the development of strength and density.

Accordingly, the significant differences in the degree of D/IF among the five pulps are the fundamental bases for varying tensile strengths, freenesses, and densities of the pulps developed (Table 1). The elevated D/IF in LC3 and HC2b pulps made them highly flexible and thereby improved the collapsibility and conformability of fibers. This accounts for the enhanced fiber-to-fiber bonding and better densification of handsheets (Stone and Scallan 1965; Abitz and Luner 1989; Heikkurinen et al. 1991; Paavilainen 1993).

In addition, the surface ultrastructure of each pulp also contributed substantially to most of the physical and optical properties of their handsheets. For example, the exposed S2 layer with enhanced EF (long ribbon-like cellulose fibrils), particularly the broad sheet-like and lamellae fibrils, exhibited by pulp HC2b were key characters for good bonding ability of the pulp (Kang and Paulapuro 2006).

![Figure 6](image-url) Linear correlation of the three major groups of SFPs in a pulp to the tensile index (a) and apparent density (b) of their handsheets. The non D/IF and high D/IF groups had very strong but opposite correlation. The figure illustrates the influence of D/IF on the properties of the sheets produces from the fibers.
2006; Li et al. 2010; Fernando et al. 2012). Ribbon-type fibrils acquire higher bonding potential, particularly the broad and long fibrils possessing greater RBA (Braaten 2000) and promote sheet density, tensile index, and Scott bond strength (Kang and Paulapuro 2006). The higher tensile index and density of HC2b and LC3 pulps were thus explained by enhanced fiber development (i.e., internal and external cell wall characteristics) of their fibers. In contrast, the inferior EF reflected by the lignin-rich S1 surface layer with flake-like short S1 fibrils of poor bonding potential is partly responsible for the lowest density and strength properties in HC1 and LC2 (Table 1). A considerably larger proportion of their untreated stiff fibers (63% for HC1 and 54% for LC2; Figure 2a) also collectively contributed to the observed lower quality of the pulp.

Light scattering was on the same level for LC- and HC-refined pulps compared at equal tensile indices (HC2b vs. LC3; Table 1). Gorski et al. (2012a) observed lower EF for LC pulps than for HC pulps. However, this study demonstrates that LC refining has a different mechanism of EF, which promoted hairlike long ribbons (Braaten 2000) describes RBA of external fibrils as the determining factor for both strength and optical properties of a paper sheet and the important role of thread-like and narrow ribbon-shaped fibrils for higher light scattering properties. Consequently, the morphologic characteristic of EF found in LC3 pulps was presumably the principle reason for its equally high light scattering ability. According to Braaten (1997, 2000), the RBA of the hairlike narrow long ribbons of LC3 fibers is very low and thus significantly influences the light scattering coefficient of the pulp.

A significantly lower stretch and TEA of handsheets was shown by the LC compared with the HC-refined pulp (Table 1). This means that the fiber network of sheets from the LC pulps was not able to stretch as much as the network of HC-refined fibers. This may be caused by several factors. LC-refined fibers are straighter and shorter compared with HC-refined fibers (Gorski et al., 2012a), which would lead to less stretch in the individual LC fibers, when a handsheet is subjected to tension. The morphologic characteristic of EF, which was deficient in carpet forming broad sheets and lamellae (which are excellent binders between fibers; Braaten 1997, 2000) compared with that of HC fibers, may have also collectively contributed to the inferior stretchability of the LC fiber network. A higher degree of D/IF normally causes higher flexibility and stretchability of individual fibers (Fernando et al. 2012), which did not seem to compensate here for the higher straightness of the LC-refined fibers. A possible explanation is that the highly flexible LC fibers formed bonds with a larger bonded area, which could stretch out the fiber segments between the bonds leading to less network stretchability.

Conclusions

HC and LC refining of primary ATMP along with the increasing SEC significantly elevated the degree of fiber wall D/IF. The internal fiber development during secondary HC and LC refining was similar at a similar level of handsheet tensile index. However, the refining energy input for the LC refining was 420 kW h odt⁻¹ less. LC promoted the fiber wall D/IF more energy-efficiently than HC refining. The external fiber development during HC and LC refining was very different as revealed by the ultrastructural characterization of the pulp fiber surfaces. LC refining is suggested to induce a distinct action that promotes fiber surfaces rich in thin hairlike threads of S2 ribbons, sometimes along the whole fiber length in addition to efficient D/IF development. In the course of HC refining, broad sheet and lamellar types of EF of the S2 layer occur, leading to high bonding potential of the fibers. These characteristics were rarely observed in LC-refined pulps. The different mechanisms of fiber development at the cell wall level were explored in HC and LC refining that governs most of the physical and optical properties of the pulps investigated.

Acknowledgements: Our work was carried out within the framework of the “Branschforskningsprogram för skogs- och trädindustrin” and “Process and product developments through unique knowledge of wood fiber ultrastructure” (2007–03230). The program is financed by VINNOVA, six forest-based industries (Eka Chemicals, Holmen, SCA, Smurfit Kappa Kraftliner, StoraEnso and Södra Cell), and involves collaborative work among SLU, Innventia, KTH, and MSU. The authors would like to express their gratitude to the staff of The University of British Columbia Pulp and Paper Centre and the Andritz pilot plant in Springfield, OH, especially Dr. Antti Luukkonen. Dr. Jens Heymer (Aikawa Fiber Technologies) and Prof. James Olson (The University of British Columbia Pulp and Paper Centre) are acknowledged for their advice and support during the LC refining trials. Jan Hill (QualTech AB) is acknowledged for valuable comments on the article. Dmitri Gorski would also like to acknowledge the financial support of Gålöstiftelsen, Hans Werthén Fonden, and Jansons Legat who all contributed to his postdoctoral at The University of British Columbia.

Received August 13, 2012; accepted February 11, 2013; previously published online xx
References


