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Fundamental understanding of pulp property development under different thermomechanical pulp refining conditions as observed by a new Simons' staining method and SEM observation of the ultrastructure of fibre surfaces

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Abstract

The morphological and chemical characteristics of cell walls govern the response of wood fibre to mechanical pulping processes and thereby influence the energy efficiency of the process and determine most pulp and paper properties. A study has been carried out at the microstructural/ultrastructural level of fibre cell walls by means of a newly developed Simons' staining (SS) method and scanning electron microscopy to characterize thermomechanical pulps (TMPs) produced under different refining conditions. The SS method allows assessment and quantification of pulp fibre development during the process in terms of cell wall delamination/internal fibrillation (D/IF) under different process conditions, and the degree of D/IF can be statistically evaluated for different TMP types. In focus was never-dried Norway spruce TMP from primary stage double-disc refining running in a full-scale mill, where specific refining energy was varied at different refining pressure levels. Improved energy efficiency was gained at the same tensile index level when applying high pressure (temperature). Under conditions of high pressure and refining energy, a significant enhancement of the degree of D/IF of pulp fibres was observed. The surface ultrastructure of these fibres exhibited an exposed S2 layer with long ribbon-type fibrillation compared to pulps produced with lower pressure and energy input. A given TMP type can be classified in the categories of high-severity and low-severity changes and quasi-untreated concerning the degree of D/IF of its fibres. The relative proportions of these are important for the development of pulp properties such as tensile strength. The presence of higher amounts of fibre fractions in the categories high D/IF and low D/IF will improve the tensile index of a TMP.

Keywords: cell wall ultrastructure; delamination/internal fibrillation (D/IF); energy efficiency; fibre development; Norway spruce; SEM; S1 layer; S2 layer; Simons' stain (SS); temperature; thermomechanical pulp (TMP).

Introduction

During the last 5 years, the energy prices for mechanical pulping (MP) have risen considerably and the industry has tried to reduce the energy consumption of the process. Diminishing energy for refining, however, leads to products with lower quality. Therefore, research is focusing on reducing energy consumption while maintaining or improving pulp quality. As a step towards this goal, in 2008 Holmen Paper AB invested in a new thermomechanical wood refining process at the Braviken mill. Single-stage high-consistency refining is one of the key steps in this innovative energy-saving process, which has been shown to have a strong influence on fibre development (Muhić et al. 2010). The final pulp quality is influenced by the first fibre treatment (Höglund and Wilhelmsson 1993; Härkönen et al. 2003).

In the course of optimizing the refining conditions, increased temperature was beneficial to reduce the specific energy consumption (SEC) in double-disc (DD) refining and simultaneously keep the same pulp quality (e.g., tensile index) (Kure and Dahlqvist 1998; Muhić et al. 2010). Any change in pressure inside the refiner will also influence the temperature (Nilsson 1987). High temperature contributes to softening of fibres, making them easier to refine, and provides for a broader range of treatment possibilities; for example, fibres can be treated with higher intensity and higher SEC without fibre shortening (Salmén 1984; Höglund et al. 1995). Temperature is also one of the most important variables that influence initial fibre separation and fibrillation efficiency, and these properties have a great effect on TMP quality (Salmén et al. 1983; Höglund et al. 1995; Fernando and Daniel 2008). Development of fibre fractions, preservation of long fibre fraction, shive content, and average fibre length also correlate positively with temperature. In softened fibre material, the proportions of the long fibres and fines will change, depending on how the fibres are treated (Nurminen 1999; Omholt and Miles 2008). Often, the develop-

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ment and quality of fines follow fibre development (Rundlöf et al. 2000). The shive content is lowered with increased temperature owing to easier defibration, i.e., improved fibre–fibre separation (Irvine 1985; Sabourin et al. 2003; Fernando and Daniel 2008).

The term intensity explains the energy transfer to mass unit of fibres per impact; it is a function of cumulative forces applied on the fibre during its residence time in the refiner. Higher power increases the refining intensity (Miles 1991; Huhtanen et al. 2009). The plate gap is an important parameter, which influences the fibre residence time in the refiner. The residence time affects overall fibre development via the refining intensity and can be controlled by the differential pressure over the plate gap (Härkönen et al. 2003; Huhtanen et al. 2009). Fibre length reduction caused by high-intensity refining can be mitigated by temperature increases (Kure et al. 2000; Vuorio and Bergqvist 2001; Huhtanen et al. 2009). However, increased temperature may lead to brightness reduction (Sabourin and Wiseman 2002). The plate gap (intensity) affects the brightness, as low plate gap increases the specific light scattering coefficient at a given SEC (Stationwala et al. 1991; Muhić et al. 2010). A smaller gap (effectuated by pressure increase) elevates the tensile index and specific light scattering at a similar SEC level (Muhić et al. 2010).

Pulps are normally characterized by laboratory tests according to standards, such as Canadian standard freeness (CSF) or the tensile index, or by routine measurements of fibre size distributions with FiberMaster, KajaaniMAP, PulpEye, etc. Various in-depth morphological characterizations by means of light and electron microscopy – sometimes in conjunction with staining – have also gained much interest in the last decade (Reme et al. 2002; Fernando 2007; Huang et al. 2008; Daniel et al. 2009). Such methods give information at the fibre wall level and provide profound understanding of the fibre behaviour during processing, as structural changes (both internal and external) are to a great extent responsible for the pulp and paper properties produced (Claudio-da-Silva 1983; Clark 1985; Tchepel et al. 2004; Kang et al. 2006).

Delamination/internal fibrillation (D/IF) is an example of structural changes that take place inside the fibre cell wall, which improve flexibility (Stone and Scallan 1965). Flexibility is a key factor as it governs most physical and optical properties of pulp and paper, including paper formation (Abitz and Luner 1989; Mohlin 1989; Paavilainen 1993; Petit-Conil et al. 1994). In addition, the surface ultrastructure of pulp fibre walls (e.g., type of external fibrillation) contributes significantly to both the physical (e.g., tensile strength, breaking length, bonding potential) and optical properties (Mohlin 1989; Kang et al. 2006; Fernando 2007). Currently, however, there is a gap in our knowledge on the behaviour of wood fibres at the cell wall level, mainly due to the unavailability of suitable fibre characterization techniques.

Recently, Fernando and Daniel (2010) introduced a microscopic method that can be used to measure and evaluate statistically the degree of fibre wall D/IF of different mechanical pulps by means of Simons' stain (SS). SS is a two-colour dye consisting of a low- and high-molecular weight polymeric mixture of direct blue (DB) and direct orange (DO) dyes. The stain was previously successful in visualizing changes in pulp fibre fibrillation and mechanical damage of beaten fibres (Simons 1950; Koljonen and Heikkurinen 1995).

The major objective of the present study was to explain the influence of process conditions on fibre characteristics. Thermomechanical pulps (TMPs) produced under different refining conditions were characterized with regard to the microstructure and ultrastructure of fibre cell walls by means of the new SS method and scanning electron microscopy (SEM). The influence of structural changes in the fibre wall on some pulp and paper properties will be discussed. The main goal is, however, to gain further insights into fibre development at the wall level that should provide clues to the question of how pulp production can be combined with energy efficiency and high quality.

Materials and methods

The refining system

Full-scale trials were carried out at one of the TMP lines of the Holmen Paper Braviken mill, with Norway spruce [*Picea abies* (L.) Karst.] as the raw material. The primary refining system of counter rotating RGP68DD (pressurized double disc, Metso; DD1) was in focus. The system had an atmospheric preheater (retention time of around 10 min) installed before the refiner. Chips were transported from the preheater to the pressurized refiners via production and plug screws. Process operating values and pulp quality targets are presented in Table 1.

Sampling method and refiner control

The refining temperature was controlled by pressure adjustments (feed and housing). There was no difference between feed and housing pressure. The refining curve at 4.4 barg (bar gauge) was the reference. Consistency was changed to control the refiner stability. Other process parameters were kept constant (Table 2).

The relevant parameters were determined based on point average measurements made during the sampling period, e.g., dilution water flow, pressure, motor load, and production rate (t h^{-1}) (needed for

Table 1 Standard operating values for the double-disc refiner (DD1) at Holmen Paper Braviken.

Parameters	Target value
SEC (kW h adt^{-1})	1650
Consistency (%)	36
Production (adt h^{-1})	11
CSF (ml)	120
Tensile index (N m g^{-1})	43
Fibre length (mm)	1.0
Tear index (mN m^2 g^{-1})	7
Specific light scattering (m^2 kg^{-1})	52
Opacity	93
Shives (sum g^{-1})	350

SEC, specific energy consumption; CSF, Canadian standard freeness.

Table 2 Process data from the trials.

Parameters	Trial 1	Trial 2
Inlet/housing pressure and temperature of saturated steam	4.4 barg, 155°C 6.4 barg, 167°C 6.9 barg, 170°C	4.4 barg, 155°C 6.4 barg, 167°C 7.0 barg, 170°C
Plate gap temperature (°C)[a]	166–171 172–178 176–179	167–171 173–177 176–179
Consistency (%)	36.9±2.6	35.6±3.4
Motor load (MW)	14.4–19.8 16.8–20.8 17.2–20.5	14.3–21.6 14.9–21.0 14.7–19.8
Production (adt h^{-1})	10.6±0.2	11.4±0.3
Plate life (h)	178	140

[a]TDC-measured plate gap temperature, called temperature in the text.

Table 3 Standard pulp and handsheet measurements used.

Measurement	Method
Pulp consistency	ISO 4119:1995
Hot disintegration	SS-EN 2471
Sheet basis weight	SS-EN ISO 536
Canadian standard freeness	ISO 5267-2
Rapid Köthen[a]	ISO 5269-2
Specific light scattering and light absorption coefficient	SS-ISO 9416
Opacity	SS-EN 2471
Tear and tensile index	SS-EN ISO 5270
Paper conditioning ratio	SS-EN 20187
Long fibre and shive content[b]	–

[a]Handsheet making method; [b]results of fibre length and shive content in the pulp were gained by Eurocon.
PulpEye measurements. References are available at Braviken paper mill laboratory.

calculations of specific energy consumption). The SEC calculations were based on air-dried pulp (adt; air-dried tonne). Pulp samples were collected from the blow line behind the refiner under stable refiner conditions.

Analysis of pulp and paper properties

Handsheets were produced by Rapid Köthen (PTI GmbH, Vorchdorf, Austria). All pulp and handsheet properties were analysed according to standard SCAN and ISO measurement methods (Table 3). All measurements and analyses were carried out at the Braviken Paper Mill laboratory.

TMP samples and Simons' staining

Pulp fibre wall D/IF induced owing to different refining conditions was evaluated and assessed by a new SS method (Fernando and Daniel 2010). Seven never-dried samples were selected from a larger collection of TMPs produced at Holmen Paper Braviken (Figure 1) based on the criteria pressure level, SEC, and tensile index (these are marked with circles in Figure 1). TMP type 4.4 (1510) (first number represents the pressure level in barg, and the number within parentheses represents SEC level) was used as reference pulp. Approximately 1 g of TMPs was stained by the SS method and immediately examined using light microscopy (LM) as described by Fernando and Daniel (2010).

Statistical analysis on stained pulp fibres

Following SS, data on five subfibre populations (S-FPs) existing in each sample (Figure 2) were obtained with LM and statistically analysed using SAS software [SAS/STAT, version 9.1 for Windows (XP-Pro platform), SAS Institute, Cary, NC, USA] (Fernando and Daniel 2010). An ordinal logistic regression (OLR) test was performed during data analysis.

Scanning electron microscopy

Some 2 g of wet samples from each TMPs was dehydrated separately, as described by Fernando and Daniel (2008). Samples were then dried in an Agar E3000 critical point dryer (Agar Scientific Ltd, Stansted, UK) with carbon dioxide as the drying agent, and subsequently coated with gold using an Emitech K550X sputter device (Quorum Technologies Ltd, Ashford, Kent, UK). Observations were made in a Philips XL 30 ESEM (FEI Company, Eind-

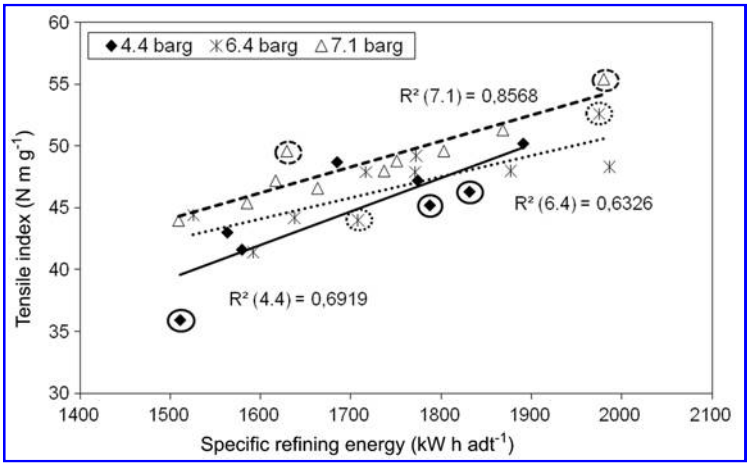

Figure 1 Tensile index development at three different pressure conditions. Fibre characteristics were investigated on pulps marked with a circle.

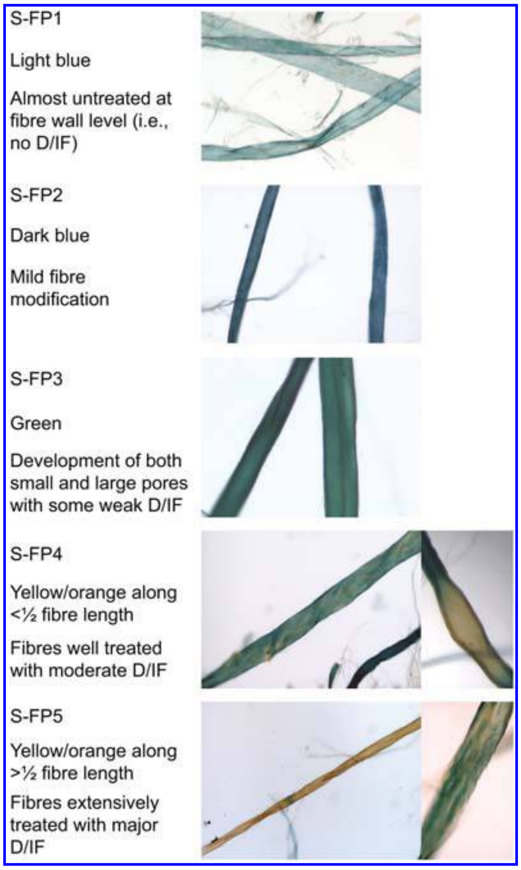

Figure 2 Summary of subfibre populations (S-FPs) present in a thermomechanical pulp type identified using Simons' stain, and the different levels in degree of fibre wall delamination/internal fibrillation (D/IF) they reflect.

hoven, Netherlands), operated at 10 kV with images recorded digitally.

Results and discussion

Tensile indices of the trials are presented in Figure 1. An increase in pressure (and temperature) in the refiner gave higher tensile indices at a given SEC level. Highest energy efficiency was noted at lowest SEC and highest pressure. This tensile index increment is due to a lower plate gap (Figure 3a) and fibre softening was caused by increased temperature.

Figure 3(b) illustrates how fibre length changes with increasing SEC. When the SEC and pressure were increasing, the plate gap was decreased, giving a higher intensity and thereby a more severe fibre treatment. Fibre shortening was observed if the intensity became too high, which was shown for pulps produced above 1750 kW h adt^{-1} during high-pressure conditions. This contradicts the hypothesis of fibre length being preserved with higher temperature (Kure et al. 2000; Vuorio and Bergqvist 2001).

CSF was also affected by the pressure increase. At a given SEC level, lowest CSF was gained at highest pressure (Figure 3c). The specific light scattering coefficient was strongly affected by the pressure rise (Figure 3d) and the s-value increased with increasing SEC (at a given pressure) and with increasing pressure (at a given SEC).

As outlined in Materials and methods, refiner stability was controlled by changing consistency. Hence, consistency changes occurred for RGP68DD refiners with inner (DO-52B036-037) and outer (DN72N816-817) segments at consistencies of 32–40%. Earlier studies describe the essential influence of consistency on pulp quality (Miles and May 1990; Berg et al. 2003). In the present study, the blow-line consistency varied by ±3%. It was assumed that this small variation would not affect the interpretation of the results.

TMP fibre characterization and assessing fibre development at the cellular level by SS

The SS method reveals information on fibre development/fibre treatment (D/IF) induced by refining and treatment conditions. It also allows quantitative and statistically relevant assessments of TMPs in respect of the degree of fibre wall D/IF. The D/IF data are collected from stained single fibres mostly in the range of the fibre fraction, and the fibres have varying patterns of colours between blue and yellow/orange depending on the degree of wall D/IF (Fernando and Daniel 2010).

According to Figure 4, diverse fibres with varying levels of fibre wall D/IF are present. The heterogeneity in a population of native wood fibres is due to the morphological origin of the fibres, e.g., as earlywood, latewood, reaction wood, etc. Thus, the fibres have very different properties, including cell wall thickness and stiffness, to mention just two. The natural heterogeneity and the stochastic nature of the TMP processes are the reason why the TMP fibres (which have undergone considerable changes in both the chemical and morphological structure of the native fibre wall) and the development of their paper-making properties are so different (Claudio-da-Silva 1983; Karnis 1994; Fernando 2007; Daniel et al. 2009). For example, the tendency for swelling and shrinking is heavily influenced by the parameter D/IF of the fibre wall. The flexibility of pulp fibres, which leads to improved collapsibility of the pulps, is also enhanced by the D/IF. Fibre collapsibility/conformability influences the strength properties, light scattering, and the runnability in paper machines (Abitz and Luner 1989; Mohlin 1989; Paavilainen 1993). Hence, D/IF of pulp fibre walls is important for most physical and optical properties of pulp and paper.

After SS, localized patches and their distribution of intracellular D/IF over pulp fibre walls could be visualized (Figure 5). A great variety can be observed: (1) a few patches are visible, so that the total area coverage is less than half the fibre length (Figure 5a–d and S-FP4 in Figure 2), and (2) many patches are observable, so that more than half the fibre length is covered (Figure 5e,f and S-FP5 in Figure 2). The distribution patterns are also different: (1) only the fibre end(s) (Figure 5g), (2) small patches (Figure 5a–d), or (3) one or few large continuous patches along the fibre (Figure 5h) can be visualized by staining. The quantitative and qualitative characteristics of D/IF contribute to the degree of an

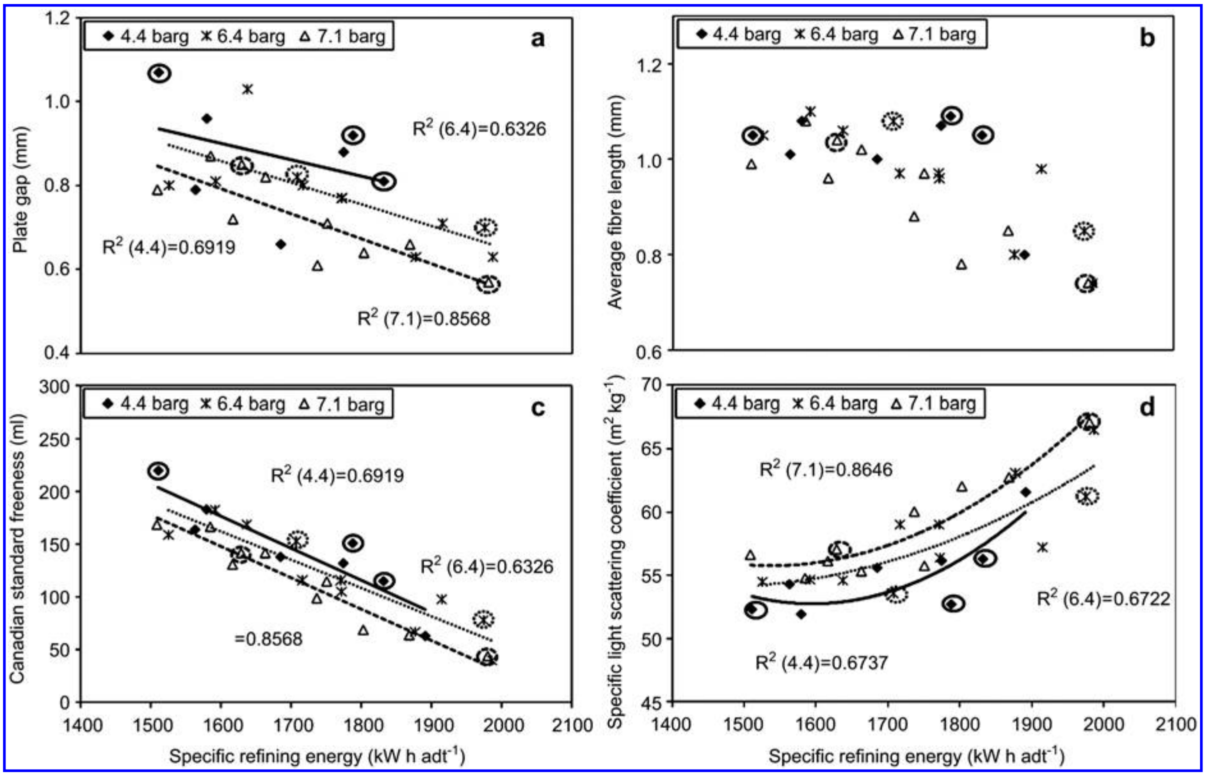

Figure 3 Relationship between specific energy consumption and pulp and paper properties produced in different thermomechanical pulp (TMP) types: (a) the plate gap is decreased with increasing pressure in the refiner; the smallest plate gap is achieved at highest pressure; (b) average fibre length is reduced with the introduction of high pressure; fibre cutting is probably occurring for pressure conditions above 1750 kW h adt^{-1}; (c) the Canadian standard freeness (CSF) value after the double-disc chip refiner is lowest for the highest pressure; (d) the highest s-value is attained with the highest pressure; the s-value has an increasing trend when applying more specific refining energy.

individual fibre's flexibilities, which sum up to the overall flexibility and subsequent collapsibility of a pulp as a whole.

A general overview of the results of SS is presented in Figure 6. Here, the five subfibre populations (S-FP1 to S-FP5, presented in Figure 2) present in a TMP are classified into three major contrasting groups, namely no D/IF, low D/IF, and high D/IF (the three groups represent non-treated or the least treated, moderately treated, and the most severely treated fibre populations, respectively, in a TMP) by combining data of S-FP1 and S-FP2 ''light blue'' and ''dark blue'' to form the group ''no D/IF'', and S-FP4 and S-FP5 to form the group ''high D/IF''. The S-FP3 fraction was classified as a low-severity treatment group (i.e., ''low D/IF'').

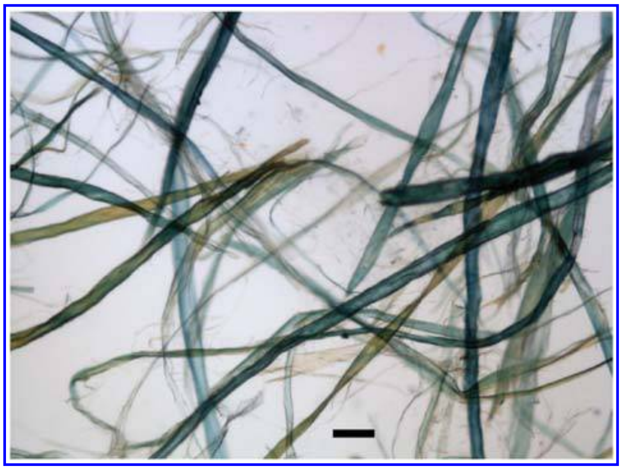

Figure 4 Following Simons' staining, thermomechanical pulp (TMP) fibres stained in colours between blue and yellow, reflecting different levels in severity of fibre wall delamination/internal fibrillation (D/IF) induced by the refining conditions. Bar: 100 µm.

The amount of quasi-untreated fibres is significantly reduced at increased SEC and increased refining intensity (in the sense of increasing pressure) (Figure 6). This means that fibres are well treated at their cell wall level by increasing the two refining conditions with an increasing trend in the two groups low D/IF and high D/IF. However, the two groups are present in varying percentages among different TMPs. For example, TMP type 7.1 (1980) (highest pressure of 7.1 barg and SEC 1980 kW h adt^{-1}) had the highest fraction of high D/IF fibres (i.e., ca. 40%, Figure 6) with the lowest fraction of non-treated stiff fibres (i.e., ca. 38%, Figure 6). This pulp thus contained well-developed TMP fibres. In contrast, a combination of lowest pressure and minimum energy input resulted in a pulp, i.e., type 4.4 (1510), dominated by quasi-untreated stiff fibres (ca. 66%) and the lowest proportion of well-treated fibres (i.e., ''high D/IF''; ca. 11%, Figure

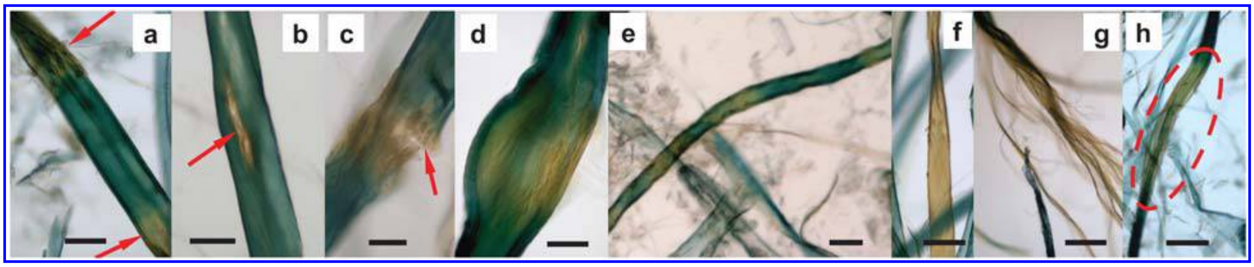

Figure 5 Information on the internal structural changes, i.e., delamination/internal fibrillation (D/IF), of pulp fibre walls could be extracted and transformed into quantitative data using the Simons' staining (SS) method: (a) two localized patches (arrows) of yellow (localized D/IF) staining over of an earlywood fibre; (b, c) SS stained only the area of fibre wall damage yellow (arrows); (d) only the swollen fibre wall area due to localized D/IF is stained yellow with SS; (e) latewood fibre showing several small patches of localized D/IF along the fibre length as visualized from presence of yellow patches; (f) whole fibre stained yellow due to severe D/IF along the fibre; (g) fibrillated fibre ends stained yellow; (h) an extended patch of D/IF along relatively large area of a fibre (red circle of broken line). Bars: a,d, 20 μm; b,e, 50 μm; c, 30 μm; f,h, 100 μm; g, 150 μm.

6). Pulp 4.4 (1510) was therefore considered the least developed TMP type produced during this trial.

Statistical evaluation of degree of D/IF of different TMPs

Statistical analysis (OLR) was performed on the degree of D/IF of pulp fibres in focus (Table 4). There was a highly significant difference in the degree of D/IF of fibre walls between the TMPs ($P<0.0001$). As to the effects of two refining conditions, both the SEC and the refining pressure were statistically significant, with the SEC being highly significant ($P=0.0038$). Accordingly, the influence of SEC on D/IF is most pronounced, as shown previously (Abitz and Luner 1989; Heikkurinen et al. 1991), while refining pressure also has a significant impact on enhancing fibre wall D/IF. The effect of levels of each refining condition on the degree of fibre wall D/IF is also presented in Table 4. Although SEC has the largest effect at the cell wall level, the effect is different at various energy levels. The latter affected fibre wall D/IF significantly, particularly when increased to levels between 1500 and 1800 kW h adt^{-1} at 4.4 pressure [$P<0.0003$ for 4.4 (1510) vs. 4.4 (1830)]. However, a small energy increase either at low [$P=0.2909$ for 4.4 (1790) vs. 4.4 (1830)] or at higher pressures [$P=0.0684$ for 7.1 (1630) vs. 7.1 (1980)] did not have a significant effect. Similarly, refining pressure also significantly enhanced the D/IF when increased from low to higher levels, e.g., from 4.4 to 6.4 ($P=0.0400$). However, no significant change in D/IF occurred when changing from 6.4 to 7.1 barg at an energy level ca. 1650 kW h adt^{-1} [$P=0.5768$ for 6.4 (1710) vs 7.1 (1630)]. The results thus indicate that significant modifications in the pulp fibre cell walls concerning D/IF can be made with the higher pressure of 6.4 barg, but a further increase to 7.1 barg

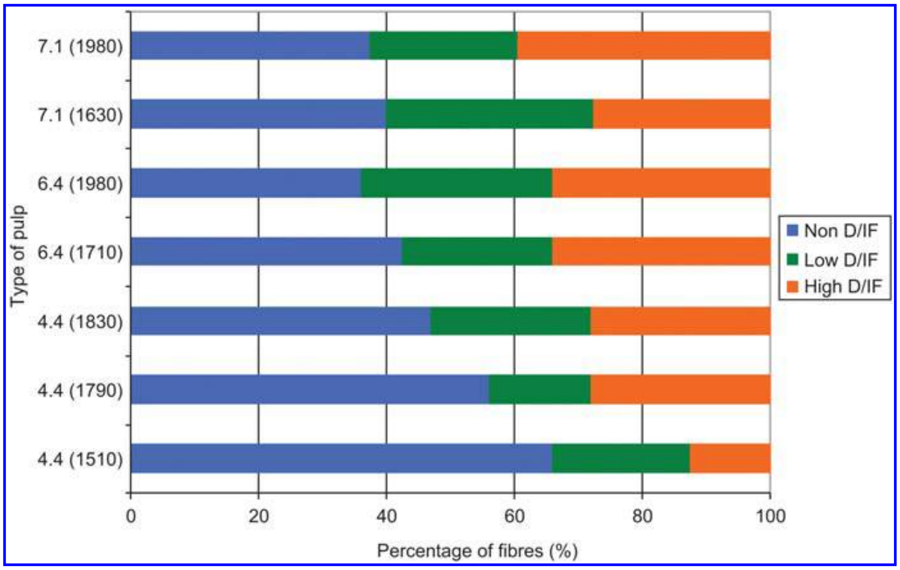

Figure 6 Percentages of pulp fibres in the three major groups of no D/IF, low D/IF, and high D/IF that were revealed with the Simons' staining (SS) method and represented three levels in degree of fibre wall delamination/internal fibrillation (D/IF).

Table 4 Logistic regression statistics for type 3 analysis of ordinal logistic regression test for significant differences (a) among the pulps, (b) in the energy input and refining pressure, and (c) between different pulps at either the same energy or same pressure on the degree of fibre wall delamination/internal fibrillation (D/IF).[a]

Source	df	χ^2	Pr>χ^2
Pulps (a)	6	45.98	<0.0001
Energy (b)	4	15.48	0.0038
Pressure (b)	2	6.38	0.0329
4.4 (1510) vs 4.4 (1830) (c)	1	13.01	0.0003
4.4 (1790) vs 4.4 (1830) (c)	1	1.12	0.2909
7.1 (1630) vs 7.1 (1980) (c)	1	3.32	0.0684
4.4 (1790) vs 6.4 (1710) (c)	1	4.22	0.0400
4.4 (1830) vs 7.1 (1980) (c)	1	5.42	0.0199
6.4 (1710) vs 7.1 (1630) (c)	1	0.31	0.5768

[a] In the sample designation, e.g., 4.4 (1510), the first number is pressure in barg and the specific energy consumption (SEC; kW h adt^{-1}) is in parentheses.

at higher energy levels (e.g., 1600 kW h adt^{-1}) does not trigger any further significant D/IF in the pulp fibre walls.

Surface ultrastructure of TMPs as revealed by SEM observations

There was a clear difference between two extreme pulps, i.e., 4.4 (1510) (Figure 7a) and 7.1 (1980) (Figure 7d). While the majority of pulp fibres of the TMP 4.4 (1510) exhibited the S1 layer as their outer layer (Figure 7a) with flake-like S1 fibrils protruding from the fibres (Figure 8a, b), almost all of the fibres from the pulp 7.1 (1980) showed an S2 layer (Figure 7d) with ribbon-type fibrillation on the outer surfaces (Figure 8c). This implies that deep layers in the secondary S2 cell wall could not be removed under mild processing conditions and, as a consequence, important S2 external fibrillation did not take place. Higher input of pressure and energy resulted in more flexible fibres with thinner walls, as reflected by exposed S2 layers and a fine fraction that exhibited the presence of higher amounts of bunches of long ribbon fibrils from the S2 layer (Figure 8d). It is also apparent from SEM observations that more fibres were shortened during high-pressure refining (Figure 8e). This probably contributes to a higher yield of middle fraction, correlating with results obtained on fibre length distribution (Figure 3b). Also here, more fibres were found with severely fibrillated ends within the TMP 7.1 (1980) (Figure 8f,g). This indicates the presence of a harsh mechanical action at a 7.1 barg pressure level, presumably on fibres that are weakened/susceptible as a result of a concomitant high temperature.

Other pulp types showed very similar surfaces where some fibres had their S1 exposed while others exhibited the S2 as the outer layer. Therefore, they are not presented here, with the exception of pulp 4.4 (1790) (Figure 7b). Nevertheless, in fibres in pulp 6.4 (1710) the S2 was most often the outer surface (Figure 7c). Sometimes, ripped-off macrofibrils were visible from the S2 layer along its microfibril angle (MFA) (Figure 8h). In other words, most of the ribbons from S2 were completely peeled-off from the fibre surface to be included in the fine fraction. On the other hand, fibres with fibrillated surfaces in pulp 4.4 (1790) have a rugged appear-

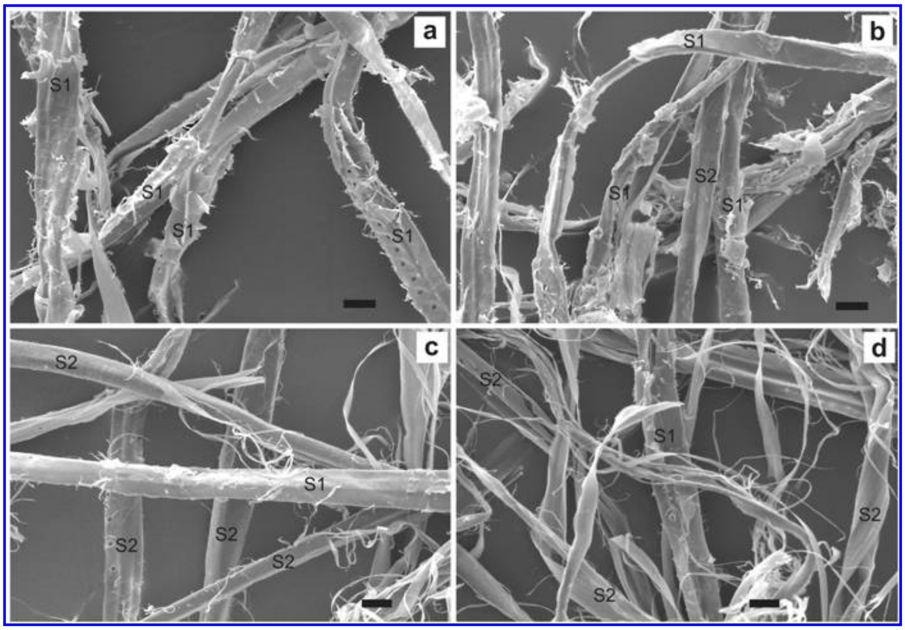

Figure 7 SEM micrographs of double-disc refined thermomechanical pulp (TMP) fibres showing their surface morphological ultrastructural characteristics induced owing to different refining conditions: (a) fibres of TMP type 4.4 (1510) exhibiting a secondary S1 layer of the fibre wall as their outer surface layer; (b) while most fibres of 4.4 (1790) showed S1 surface layers, some showed an S2 secondary layer; (c) surface ultrastructure of pulp 6.4 (1710) showing S2 as surface layer sometimes with clear outer surfaces presumably due to complete peeling of the S2 fibrils; (d) the highest energy and pressure conditions produced pulp 7.1 (1980) possessing an S2 layer as the fibre outer layer with ribbon-type fibrillation from the inner S2 secondary layer. Bars: a,b,c,d, 30 μm.

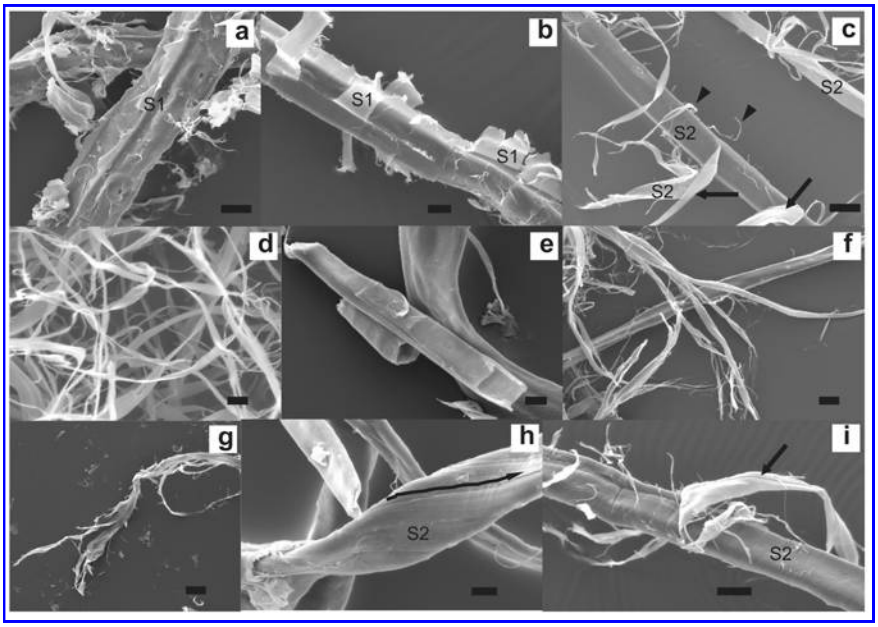

Figure 8 Close-up views of SEM micrographs for different thermomechanical pulp (TMP) fibres: (a, b) rugged appearance of 4.4 (1510) pulp fibres due to fibrillated surface layer of S1 generating short flake-like fibrils; (c) TMP 7.1 (1980) showing extensive ribbon-type fibrillation [broad sheet-like (arrows) and thin thread-like (arrowheads)] from its outer surface layer of inner S2 secondary wall; (d) fines from TMP 7.1 (1980) showing bunches of long ribbon-type fibrils peeled from the fibre S2 surface; (e) severe fibre cutting observed with pulp 7.1 (1980); (f, g) fibre ends of pulp 7.1 (1980) were severely fibrillated by the extreme refining conditions; (h) severe peeling of S2 from fibres of 6.4 (1710) leaving marks of ripped-off macro/microfibrils along the S2 microfibril angle (arrow); note better collapsing of these fibres; (i) broader sheet-like S2 fibrillation from a fibre from TMP 7.1 (1980); note that part of S2 lamellae is protruding out of fibre surface (arrow). Bars: a,i, 20.0 μm; b, 1.0 μm; c,f,g, 30.0 μm; d,e,h, 10.0 μm.

ance due to ''flake-like'' S1 fibrillation (Figures 7b and 8a,b).

Relation between fibre wall D/IF and pulp properties

For simplicity, the three major groups of no D/IF, low D/IF, and high D/IF, representing the three major subpopulations of fibres within a TMP type (as described above), were focused on for these analyses (Figure 9). There is a clear correlation in so far as TMP fibres possessing high D/IF exhibited a strong positive correlation with the tensile index ($r^2=0.75$, Figure 9), whereas quasi-untreated (i.e., no D/IF). TMP fibres have a similar correlation coefficient ($r^2=0.79$), but the correlation is negative. TMP fibres with moderate D/IF (low D/IF) have a positive trend but the strength is low. Accordingly, the strength of a TMP is improved by increasing the proportion of the two groups low and high D/IF, which contain a sufficient quantity of desirable flexible fibres.

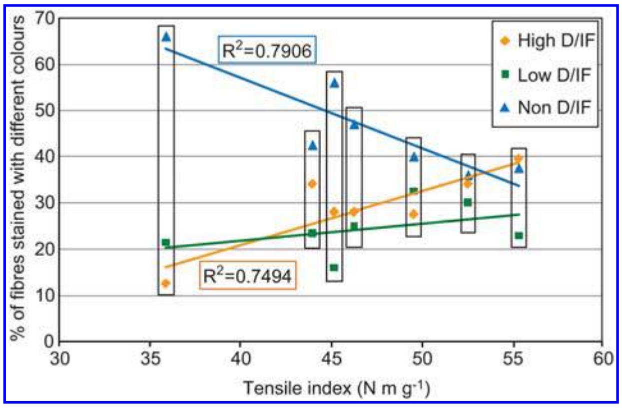

Figure 9 Relationship of the percentage of fibres in the three major groups of subfibre populations in each thermomechanical pulp (TMP) to the tensile index reflecting the influence of the degree of fibre wall delamination/internal fibrillation (D/IF) to the development of strength.

Development of pulp strength properties

By combining information gained from the SS method revealing fibre development of a TMP at the cell wall level and surface ultrastructural studies, a more comprehensive understanding of the fundamentals relating to the development of pulp strength properties is attained. As the influence of fines was discussed exhaustively in previous studies (e.g., Rundlöf et al. 2000), this aspect is not considered here. However, it is assumed that their influence in this case is proportional to the amount of fines as per Rundlöf et al. (2000), who conclude that the fibre fraction properties dominate the properties of the whole pulp. The results shown in this study give clues to why the fibre fraction properties created in the chip refining stage are so important.

Despite higher fibre length reduction in pulp type 7.1 (1980) resulting from extreme refining conditions, the tensile strength property of the pulp was the highest of all pulps tested. This is due to the high portion of treated and thus flexible fibres, which tend to collapse well, as revealed by SS (ca. 62% of the whole fibre population disposed of 65% ‘‘high D/IF’’ fibres, Figure 6). Flexibility and collapsibility of the pulp, which account for enhanced fibre-fibre bonding, are well known to be key factors in the development of pulp and paper properties such as tensile strength, apparent density, and light scattering (Emerton 1957; Stone and Scallan 1965; Abitz and Luner 1989; Heikkurinen et al. 1991; Paavilainen 1993).

Pulp 7.1 (1980) showed an exposed S2 layer and greater S2 fibrillation with long ribbon-like fibrils (Figure 8c, d and i), and the cellulose concentration on its surface was enriched. These characteristics are advantageous for a good bonding ability of the pulp. Ribbon-type fibrils acquire higher bonding potentials, particularly the broad and long fibrils (Braaten 1998). The highest tensile strength of the pulp can thus be explained based on the cell wall characteristics (i.e., internal and external) of their fibres.

In contrast, pulp 4.4 (1510) exhibited the lowest tensile index despite the fact that it retained its higher fibre length (Figure 3b); the moiety of quasi-untreated stiff fibres (i.e., no D/IF fraction ca. 66%) was high, which impaired the collapsibility/conformability of the pulp. In addition, ultrastructural characteristics of their fibre walls (i.e., the presence of lignin-rich S1 at the outer surface layer with flake-like short S1 fibrils) are responsible for obstructing the development of their strength properties. The bonding ability of flake-like fibrils is inferior to that of ribbon-like fibrils from S2. The presence of outer wall layers such as S1 is also a sign of poor fibre wall thinning, which leads to low flexibility and collapsibility.

The development of varying tensile strength properties in the rest of the TMPs can also be explained with fibre flexibility caused by D/IF and cell wall ultrastructural characteristics. For example, in pulp type 6.4 (1710) the proportion of treated fibres was slightly above 50% of the whole pulp fibre population (the moieties with low and high D/IF were 52%, Figure 6), which most likely contributed to its mediocre tensile index of 44 N m g^{-1}, being between the two extreme pulps (Figure 1). Morphological features of the pulp fibre walls with S2 exposed with clear surfaces indicated complete peeling and release of S2 fibrils into the fine fraction leading to high tensile strength properties. The example of pulps 4.4 (1830) vs. 7.1 (1630) with similar morphological properties indicates that higher pressure with less SEC could be suited for the production of fibres with similar ultrastructural characteristics to the pulps produced with higher SEC. The information relating to improved and efficient fibre development at the cell wall level during high-pressure refining therefore provides important clues on energy-efficient TMP processes that produce pulps with even better fibre properties. However, it should be noted that thicker cell walls, and a varied fibre population of earlywood and latewood, also affect pulp strength properties (Daniel et al. 2009).

Conclusions

Lower energy consumption is required to a given tensile index at elevated pressures/temperatures in chip refining. Similar fibre surface ultrastructure characteristics were gained by pulps with high pressure at lower SEC and by pulps with low pressure and at high SEC. The SS method is suited for statistical evaluation of the severity of fibre degradation, i.e., degree of D/IF of different TMP pulps. High temperature in combination with high SEC resulted in significantly elevated D/IF of pulp fibres. The surface ultrastructure in these cases had S2 layers with long ribbon-type fibrillation. The amount of fines is important for the handsheet properties of TMP, while fibre properties can be changed by adjusting the chip refining conditions as shown here. It is suggested that the relative proportions in the categories no D/IF, low D/IF, and high D/IF, representing three levels in the degree of D/IF of pulp fibres in a given TMP, are of prime importance for the development of pulp properties such as tensile strength. If the amount of low- and high-severity fibres is elevated, better tensile indices of the pulps can be expected.

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