Fibre Development during Stone Grinding; Ultrastructural Characterization for Understanding Derived Properties

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ABSTRACT

Fundamental knowledge on fibre development at the cell wall ultrastructural level is still very limited for mechanical pulps, particularly for groundwood pulps. Therefore, a pilot scale stone groundwood (SGW) pulping study was performed on selected juvenile and mature Norway spruce wood with emphasis on ultrastructural aspects of fibre development and cell wall modification using scanning electron microscopy (SEM). SGW pulps and handsheets from juvenile wood (JW) exhibited improved physical properties such as light scattering, tensile- and tear strength and higher sheet density compared to mature wood (MW). SEM investigation indicated that an explanation for the differences is most likely related to the manner of fibre processing and development at the ultrastructural level. Results showed MW fibres to contain reduced long-fibre fractions and, more open fibres as a consequence of longitudinal fibre breakage and enhanced S1 fibrillation. In contrast, juvenile fibres showed a 14% increased long-fibre fraction and typical S2 fibrillation. JW fibre fibrillation showed features similar to that reported for thermomechanical pulp fibres where the native morphological fibre micro- and nanostructure regulate the manner of fibre fibrillation. Studies suggest that raw materials rich in juvenile wood (e.g. top logs) may be advantageous for the SGW process.

INTRODUCTION

Stone groundwood (SGW) pulping is an important mechanical pulping process particularly in Scandinavia and more recently is increasing in Southern European countries because of the lower energy consumption (cf to thermomechanical pulp (TMP)) and paper property trends towards smoother papers. However, it is well-known that the SGW process generates large amounts of fines with short fibres due to its very harsh mechanical action compared to TMP (1-3). The generally better strength properties of TMP is primarily due to the characteristic nature of the process and process conditions (4), where wood fibres are dismantled according to the native morphological architecture of their cell walls allowing for the generation of highly fibrillated (i.e. S2 fibrillation) fibres and fines of high quality (4). The SGW process produces pulps with poor strength properties in comparison to TMP processes that produce high quality furnish for a broad variety of tailor-made products (1, 6). Nevertheless SWG-based furnishes provide superior print surfaces for high quality supercalandered (SC) and light-weight coated (LWC) magazine papers (7).

Understanding fundamentals relating to fibre development is of prime importance for mechanical pulping as this is central to the mechanical pulp process in which almost all wood components are retained. Further, fibre cell wall ultrastructure plays a major role in determining most physical properties of final products. Intensive studies over many decades have provided better understanding of the process-property relationships of mechanical pulp-based furnishes (e.g. SGW pulps). However, fundamental knowledge on fibre development at the cell wall level for mechanical pulp fibres is still very limited particularly for SGW pulp fibres. Understanding both the fundamental native cell wall structure (i.e. topochemical and morphological) and changes that occur to its ultrastructure during processing will undoubtedly lead to a better use of these property developments and thereby provide possibilities for improvements in the process for products with specific qualities.

The present study therefore focused on micro/ultrastructural aspects of fibre development and cell wall modifications that occur during the SGW process of Norway spruce and their implications for the pulp properties developed.
EXPERIMENTAL

Wood Samples

Wood samples of Norway spruce (*Picea abies* (L.) Karst) collected from stands in Uppland (Mid-Sweden) were used in the study. Samples represented a wide range of wood properties with respect to basic density and juvenile wood content. Four different wood types (1, 5, 6, 7; Table 1) were selected for the grinding experiments and were characterized by measurement of diameter, number of annual rings, dry matter content according to SCAN-CM 39:94 and basic density according to Tappi T 258 os-76.

<table>
<thead>
<tr>
<th>Table 1. Properties of the four wood types used</th>
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<tr>
<td>Wood class (type)</td>
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<tr>
<td>Average ring width (mm)</td>
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<tr>
<td>Basic density (g cm⁻³)</td>
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<tr>
<td>Moisture content (%)</td>
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<td>Fibre length (mm)**</td>
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* Wood types 5 and 7 were processed but not utilized for further study since their pulp and handsheets properties were close to 1 and 6

** Fibre length measurement was conducted after delignification and maceration of representative wood pieces

SGW Pulping

Samples of juvenile- and mature wood (stored at -20 °C) were sawn into pieces 4.0 x 4.0 cm x length and ground using a laboratory scale grinder for SGW pulping at Åbo Akademi University. Wood pieces were ground with a grinding stone of type A605N7V21. Five different pulps corresponding to a CSF span of 50-200 ml (Table 2) were produced from each wood type. This was possible by controlling the feed of the wood pieces onto the grindstone from 0.7 - 1.8 mm s⁻¹. The target grinding effect corresponded to 1 MW h per ton groundwood pulp (range 0.5-1.3 MW h t⁻¹). The temperature of the shower water used was 70 °C and the peripheral speed of the stone was 30 m s⁻¹. Course material was first removed by passing the pulp through a Tampella vibration screen with 6 mm holes followed by a TAP screen with 0.15 mm slit openings. The reject was taken into consideration in SEC calculations.

Pulp analyses

Pulp properties analyzed included: pulp consistency using rapid drying of samples between an electrically heated plate—a common practice in industry-, CSF according to SCAN-C 21:65 and fibre dimensions and fractions using Kajaani FiberLab™ (Table 2).

Laboratory sheet analyses

Ten laboratory sheets of SGW pulp were made from each pulp batch using a recirculating water system with a target sheet grammage of 60 g m⁻² according to SCAN-CM 64:00. Analyzed properties were sheet grammage according to SCAN-P 6:75, thickness with L&W micrometer, density according to SCAN-P 7:96, ISO brightness, opacity, light scattering and light absorption with Elrepho 2000 Datacolor, tear index with L&W Tearing tester and tensile index with L&W Tensile strength tester.
Sample selection for scanning electron microscopy (SEM)

Pulps from the two most extreme wood types (i.e. Type 1- juvenile wood (JW) and Type 6- mature wood (MW)) with respect to juvenile wood content and basic density were chosen for microscopy. Samples for SEM were selected based on the relationship between specific energy consumption (SEC) and freeness (CSF) (Figure 1). Never-dried whole pulp samples from JW (CSF 56 and 152 ml) from Type 1 and MW (CSF 85 and 168 ml) from Type 6 (Figure 1; Table 2) were used for detailed observations using SEM.

![Graph showing SEC vs freeness for different wood types](image)

**Fig. 1.** SEC vs freeness of the SGW pulps from different MW and JW types used in the study. The two extreme samples from Type 1 (i.e. JW; JW1 and JW2) and Type 6 (i.e. MW; MW1 and MW2) were used for SEM studies.

Scanning electron microscopy

Pulp samples (each sample contained ca 400 wood fibres representing both long and fragmented fibres) were dehydrated with ethanol (20 mins, 20% steps 10 min. each) and then acetone (2:1, 1:1 ethanol: acetone and finally 100% acetone for 10 min. each) (8). Thereafter, samples were critical point dried (CPD) in an Agar E3000 CPD apparatus using liquid CO₂ as the substitution fluid, mounted on stubs using double-sided cellotape and coated with gold using an EMITECH K550X Sputter Coater. Observations were performed using a Philips XL 30 ESEM at 15 kV with images recorded digitally.

RESULTS AND DISCUSSION

Pulp properties

Previous TMP experiments have shown that wood rich in early- and juvenile wood have poorer tensile strength development than wood rich in mature wood (9-12) and JW is commonly known as a poor quality raw material for most pulp and paper products (13). In contrast, in this SGW trial the early- and juvenile rich wood (Type 1) gave the best electrical energy-tensile strength relationship (Figure 7a). The difference in CSF development between Types 1 and 6 was less evident indicating that the juvenile pulp had a better bonding capacity compared with the mature wood pulp (Figure 1).

Scanning electron microscopy observations

Ultrastructural studies of the SGW pulps provided insights on fundamental aspects of fibre development (i.e. mechanisms of fibre breakage, fibre cell wall fibrillation) that explained most of the pulp and paper properties developed. Results from SEM showed there were significant differences in fibre development at the cell wall level between the two wood types used, i.e. JW and MW.
Table 2. Physical pulp and handsheets characteristics of SGW JW and MW

<table>
<thead>
<tr>
<th>Pulp and handsheet property</th>
<th>Juvenile wood (JW) Type 1</th>
<th>Mature wood (MW) Type 6</th>
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<tr>
<td></td>
<td>1* 2 3 4 5</td>
<td>1 2 3 4 5</td>
</tr>
<tr>
<td>Specific energy consumption (MW h t(^{-1}))</td>
<td>1.29 0.99 0.86 0.77 0.66</td>
<td>0.98 0.78 0.66 0.59 0.53</td>
</tr>
<tr>
<td>Freeness (ml)</td>
<td>56** 83 114 126 152</td>
<td>85 110 135 150 168</td>
</tr>
<tr>
<td>Fibre length (mm)</td>
<td>0.54 0.42 0.43 0.4 0.39</td>
<td>0.37 0.39 0.4 0.39 0.35</td>
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<tr>
<td>Fractions, % by wt.</td>
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<tr>
<td>Long (0.5-7.6 mm)</td>
<td>29.8 29.6 30.6 28.6 28.0 (29.3)***</td>
<td>24.2 27.9 28.3 25.9 22.1 (25.7)</td>
</tr>
<tr>
<td>Middle (0.2-0.5 mm)</td>
<td>40.3 41.6 40.9 41.8 42.1 (41.3)</td>
<td>41.5 40.1 39.8 41.3 41.5 (40.9)</td>
</tr>
<tr>
<td>Fine (&lt; 0.2 mm)</td>
<td>30.0 28.5 28.5 29.5 29.9 (29.3)</td>
<td>34.3 32.1 32.0 32.8 36.4 (33.5)</td>
</tr>
<tr>
<td>Light scatt. coeff. (m(^2) kg(^{-1}))</td>
<td>87.7 83.7 80.3 79.9 79.4</td>
<td>74.6 73.2 66.7 66.9 63.0</td>
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* Numbers 1-5 represent pulps from 5 different freeness values ground from each wood type (i.e. Types 1 and 6).
** Extreme pulps selected for SEM studies in boxes
*** Numbers within brackets are average values of the 5 different pulp types.

Mechanism of SGW fibre breakage

SEM observations revealed that development of cracking and splitting of SGW pulp fibres leading to fibre breakage or cell wall fibrillation were initiated in the vicinity of pits (simple and bordered) as shown previously for TMP fibres (5). This indicates the presence of weak sites in the fibre cell wall that cracked/split as a response to the external forces applied during grinding, no matter the difference between the SGW and TMP processes (Figure 2a, b, arrows). However, unlike TMP fibres, most SGW pulp fibres showed (particularly MW fibres) cracking at weak sites in the fibre wall advanced further perpendicularly leading to transverse fibre breakage (Figure 2c-e). Both EW (Figure 2c, d) and latewood (LW; Figure 2e) showed the same mechanism of fibre breakage. Compared to TMP, the process conditions in SGW are rather rough and thus the viscoelastic properties of the cell wall materials are thought to be only poorly developed. SGW fibres showed brittle failure that resulted in total fibre breakage due to the attrition generated by the stone grits hitting the fibres perpendicular to their axis (3, 14). This mechanism of fibre breakage requires less energy than that required to deform fibres leading to fibre surface development (14, 15). Possibly, the temperature during stone grinding (i.e. shower water in the grinder was 70 °C) was insufficient for softening cell wall lignin (glass transition temperature of lignin is 115 °C) (16) and also there was no pre-conditioning of the fibres in order to develop the necessary viscoelastic properties of the cell wall materials. In contrast, pre-steaming of wood chips during TMP processing normally at ca 120 °C causes the fibres to develop viscoelastic properties (16) that not only allow fibres to deform during the subsequent refining stage, but also allow generation of fibrils by peeling.
Fig. 2a-e. SEM micrographs showing initiation of splits near pits (arrows; a, b, and arrowhead; c) of Norway spruce SGW pulp fibres. In contrast with TMP fibres, the majority of SGW pulp fibres showed transverse fibre breakage mechanism (c-e) due to perpendicular advancement of cracks close to pits of MW SGW pulp fibres (arrow, c). Both EW (c, d) and LW (e) showed the same mechanism of fibre breakage. Bars: a, b, d, e, 5 µm; c, 2 µm.

During the present SGW trial, the mechanism of transverse fibre breakage occurred most often close to the fibre tips resulting in fibre cleavage at both ends (arrows, Figure 3) contributing broken tip pieces to the fine fraction. This was apparent for both EW and LW as shown in Figure 6 (a, c for EW; b for LW). The remaining central part of the fibres (i.e. 0.5-0.2 mm) provided the large middle fraction of short fibres found in the SGW pulps (Table 2) and contributed to the poor strength properties in the final paper product, particularly tear strength. Fibre breakage at tips, indicative of brittle failure of SGW fibres compared to TMP, has been attributed to the increased pit density and pit size towards the fibre tips; the effective fibre modulus and strength being lowest near the fibre ends (17). In addition, the reduced cell wall thickness in the apical fibre regions (18) collectively contributes to enhanced fibre tip breakage during the attrition process. In contrast, fibre length is preserved more efficiently and provides the largest fraction of the long-fibre content of pulps during TMP processing. The TMP process creates favourable conditions for developing viscoelasticity within wood fibres (4) allowing cracks and splits to follow the microfibrillar organization of the cell wall (5) rather than causing cross-sectional fibre breakage as observed during the present SGW process.

SEM observations showed the MW SGW pulps to have more fibre breakage than JW pulps which also showed a ca 14% increased long-fibre fraction (Table 2). More broken fibre ends from MW fibres undoubtedly contributed to its increased fine fraction (ca 12%) compared to that of JW (Table 2). High paper strength properties, i.e. both tensile and tear strength (Figure 7a, b) recorded for the juvenile wood sample during the present study were partly explained by its greater long-fibre fraction. The high microfibril angle (MFA; 19°-54°) (19) and relatively high lignin content reported for Norway spruce JW cell walls (i.e. ca 29%) (19) may also have contributed by absorbing the energy during grinding thus resulting in reduced fibre breakage compared to the MW sample.
Fig. 3a-f. Norway spruce fibre breakage at their ends (arrows in a, c; EW, b; LW) during SGW pulping contributing broken fibre tips (d; from LW and e-f; from EW) to SGW fine fraction. Bars: a, 50 µm; b, 60 µm; c-f 10 µm.

In addition to transverse fibre breakage, present study also showed longitudinal fibre breakage of SGW pulp fibres (Figure 4) that was rarely observed previously with TMP fibres (5). The attrition process of SGW pulping with sharp grits generated “combing” effects upon the fibres causing them to break longitudinally resulting in the tearing of fibres, most often along the fibre axis (arrow, Figure 4a-b). More longitudinal fibre wall tearing was observed with MW SGW pulps while fewer fibres with torn fibre walls were apparent for JW pulps. Final paper products containing such types of furnishers (i.e. fragmented fibres without ends and fibres with only 50% wall remaining) rather than long intact fibres should contribute considerably to poor paper strength properties as was observed for the MW pulps (Figures 3 and 4). In contrast, during TMP processing fibre development is based primarily on fibre cell wall morphological architecture and microfibrillar organization (5). TMP fibres generally show typical S2 fibrillation providing good quality fines from the S2 secondary cell wall layer consisting of long and thin fibrils such as “semi-macro ribbons”, macrofibrils and microfibrils. In addition, the TMP process also generates higher ordered (i.e. broader) cell wall structures from the S2 layer such as “macro-sheets” and much broader “lamelliar sheets” (5) that could form carpet-like structures within paper webs (20, 21). These structures from S2 layer possessing a large surface area have high bonding capabilities and are thought to be excellent binders of particles within paper web. They provide strength and a much smoother and dense paper structure although they are known to have a negative effect on light scattering mainly due to their high relative bonded area (20, 21).

Fig. 4a-b. Longitudinal fibre breakage during stone grinding due to “combing” effects of grits onto the fibre cell wall. Bars: a, 50 µm; b, 10 µm.
Fibre cell wall fibrillation during SGW pulping

The most striking difference observed between SGW and TMP in respect of fibre development was the manner of fibre splitting and fibrillation of fibre cell wall layers. Most SGW pulp fibres (i.e. EW and LW) showed cell wall splitting/fracturing and subsequent development to occur randomly over the fibre cell wall even across the fibre microfibril angle (MFA) (Figure 5a, b). This phenomenon influenced the mechanism of surface fibre wall fibrillation resulting in multi-directional fibrillation that did not follow the MFA of the cell wall layers. It therefore negatively affected the degree of delamination and/or “peeling mechanism” particularly of the thick S2 layer. As a consequence, the generation of ribbons from the S2 cell wall layer as seen with TMP (5) was impeded and only smaller amounts of ribbon-like fibrils were produced in the SGW pulps; observations consistent with previous reports (22, 23). However, fibre crushing by the protruding stone grits of the grindstone perpendicular to the fibre axis (i.e. forces in the direction of S1 MFA) caused fibrillation of the S1 layer. Therefore most of the SGW fibres had a severely damaged and highly fibrillated (not as smooth as TMP fibres) fibre surface (Figure 5c, d) giving a misleading impression of S2 fibrillation (referred here as “pseudo-fibrillation”). The SGW process removed more material from the S1 layer contributing to the fines fraction as short, but broad “flake-like” fibrils. Therefore, the direction of forces applied to fibres, in addition to process conditions like pre-heating are important for determining the types of fines produced as well as fibre surface development (24). Nevertheless, some fibres were occasionally observed that fibrillated according to their cell wall architecture generating ribbons from the S2 layer due to “peeling”. Most often these fibrils were narrow and thread-like rather than sheet-like as seen with TMP (5). The distinct mechanism of SGW fibre development and subsequent contribution to the particle size and shape distribution is therefore thought to govern most of the SGW pulp properties.

![Fig. 5a-d. During SGW processing, fibre splitting/fracturing occurred in a random way over the cell wall across the fibre MFA (black lines in a, b). The characteristic rugged surfaces of SGW pulp fibres due to S1 fibrillation (i.e. “pseudo-fibrillation”; c, d) that may be mistaken as S2 fibrillation. Bars: a, b, c, 10 µm; d, 20 µm.](image)

It is common knowledge that SGW pulping produces high bulk and inferior strength properties particularly tensile strength (25). This was partly explained by present results where SGW produced furnishes containing fines with poor bonding potentials (i.e. fines and/or fibrils mainly from S1 layer together with broken fibre parts) in addition to repression of the important S2 fibrillation. The shorter fines from S1 layer (“flake-like”) and fibre fragments and narrow thread-like fibrils exhibit good light scattering (21, 26). Light scattering of SGW pulps is thus related to the manner of fibre breakage and fibrillation that occur during processing.

The very short fibre fragments of SGW pulps derived from the fibre ends effectively fill the gaps within the fibre network of the paper web during sheet formation and thereby contribute to improved surface smoothness (3, 27, 28). In addition, the shorter “flake-like” fibrils from the S1 layer also enhance the filling properties and thereby contribute further to the characteristic high surface smoothness of the SGW handsheets produced here and reported in previous studies (27).
Surface ultrastructure of SGW juvenile wood fibres and relation to final pulp fibre properties

There were considerable differences between the properties of handsheets made from SGW pulps of JW and MW. Greater light scattering with increasing SEC was observed with JW than with MW at a given freeness/SEC (Table 2). One reason for the greater light scattering of juvenile wood fibres was due to their thinner fibre cell walls compared to mature wood giving the JW pulps increased specific surface area (21). According to the detailed SEM analysis of fibres from the two pulps (Figure 6), JW fibres showed a high degree of S2 fibrillation exhibiting both delamination (i.e. lamellar sheets, Figure 6a-d) and cell wall peeling leading to the generation of ribbon-like (i.e. broad, narrow and thread like, Figure 6e-i) fibrils. A possible reason for the observed S2 fibrillation may be due to the slight development of viscoelastic properties of the cell wall of JW fibres. Development of viscoelasticity within the cell wall of JW fibres may reflect the lignin content of JW fibres that is slightly higher than MW fibres (29 vs 27%) (19). Thus in the present SGW study, the JW fibres behaved like TMP fibres where the development of fibrils followed the cell wall microfibrillar organization. Improved degree of S2 fibrillation in the JW pulps, especially the increased amount of ribbons and thread-like fibrils (Figures 6e-i), that have a pronounced positive effect on light scattering (21), enhanced the overall light scattering ability of the pulp giving rise to the large increment in light scattering coefficient of the JW pulps (Table 2).

Fig. 6a-i. SEM micrographs of JW SGW pulp fibres showing characteristic S2 fibrillation; a-d) Delamination of lamellae (curved arrows) producing sheet-like fibrils (arrows); e-i) Development of ribbon-like fibrils by peeling. Note the typical high S2 MFA of JW fibres (i.e. 20-40°; arrows in e, f, g and h) and narrow, thin and long fibrils produced (arrows in i). Figures e-h show the stripping of narrow fibrils of different sizes from the fibre surface. i; development of narrow fibrils (arrows) by peeling. Bars: a, c-h, 5 µm; b, 10 µm; i, 2 µm.

The unexpected observation of high strength properties (tensile and tear indices, Figures 7a, b) of handsheets made from JW compared with MW was attributed to the presence of increased amounts of high bonding lamellar sheets and broad ribbon-like fibrils (Fig. 7a-d) that were rarely observed in MW. In addition, the preserved increased fibre length at all freeness levels (i.e. from 56 to 152 CSF ml, Table 2) as well as the increased percentage of the long fibre fraction (Table 2) contributed to its high tear and the tensile strength further suggesting improved viscoleastic properties among JW fibres compared with MW fibres.
Figure 8 shows the tensile indices of JW and MW plotted against the corresponding handsheet densities. It is evident that both the density at a certain energy input (Figure 7a and 8) and resulting tensile strength are significantly higher in JW pulps. The observed “peeling off” mechanism of JW fibre cell walls leading to S2 fibrillation and removal of more cell wall materials during fibre development probably made the JW fibres more flexible even though the fibre cell wall is naturally stiffer than MW fibres due to the slightly higher cellulose MFA (i.e. 19°-54°, vs 4°-33° in MW) (19) in the S2 layer. In addition, the thinner cell walls of JW allowed the fibres to be more conformable. The improvement in fibre flexibility together with the greater bonding potential of the S2 fibrils in the fine fraction of JW pulps was also reflected by the higher density of the JW pulps that significantly increased their associated tensile strength properties compared to MW.
In summary, this work points to the importance of the morphological and hierarchical structures of fibre cell walls at the ultrastructural level even during SGW mechanical pulp processing. While certain parameters appear less important (e.g. MFA) during the SGW process, JW fibres behaved like mature wood fibres during TMP processing and produced better paper properties than SGW mature wood fibres. The improved quality of the fibre furnish of juvenile fibres over mature fibres in the SGW process may offer an alternative process for a more effective utilization of raw materials like top logs rich in juvenile wood.

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