Physical: Full-length

In situ SEM micro-indentation of single wood pulp fibres in transverse direction

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Abstract Fibre deformations such as kinks and micro-compressions are significant parameters in determining the quality of industrial pulps. Undoubtedly, very little information has been obtained so far on fibre deformation because it is very tedious to handle the specimens. In this study, a novel *in situ* scanning electron microscope (SEM) micro-indentation technique was adopted for the first time to study the deformation of single industrial pulp fibres in the transverse direction. A one-to-one correspondence between load drops in load–displacement curve and cell wall deformation was obtained by using the SEM video sequence recorded during micro-indentation. The cell wall deformation occurred by ‘elastic’ sinking-in and lateral bulging of the microfibrils. Finally, the critical load (stress) required to initiate a crack in the cell wall was measured for different unbleached pulp fibres.

Keywords single pulp fibre, *in situ* SEM micro-indentation, cell wall deformation

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Introduction

Wood pulp is obtained from wood either by mechanical, chemical or a combination of these treatments. In chemical pulping, lignin is degraded and dissolved, resulting in long and high strength pulp fibres [1]. In addition to lignin, the quantity of cellulose and hemicellulose is also slightly reduced on pulping. Compared with wood fibres, which are tubular in shape, pulp fibres are flattened in shape and thin-walled (P and S1 layers are peeled off) due to the pulping, bleaching and associated drying processes [2]. Pulp quality is generally determined by preparing hand sheets and then performing standard paper testing methods such as zero-span tensile tests and short-span compression tests [1,2]. Due to the non-uniform geometry and chemical composition of individual pulp fibres, only few studies have

been performed to understand the single pulp fibre deformation and mechanical properties [3–10].

In earlier studies, Sachs [5] reported the deformation behaviour of a pulp by subjecting the single kraft pulp fibre to longitudinal compression inside a scanning electron microscope (SEM). Dunford and Wild [6] studied the mechanical behaviour of single pulp fibres using cyclic transverse compression. *In situ* measurements performed on wood fibres basically focused on tensile testing under the light microscope [11] or inside the environmental SEM [12–14] and corresponding strain measurements. Recently, the hardness and elastic modulus of the S2 cell walls of wood fibres and pulp fibres across the fibre cross section (in the plane of its length) were routinely obtained via nano-indentation [15,16]. Micro-indentation has earlier been used to

Fig. 1. (a) *In situ* SEM micro-indentation of the pulp fibre in transverse direction. Flattened cross section of the pulp fibre and the shape of the lumen can be observed. (b) SEM image of the indenter tip having the flat-top surface. Inset picture corresponds to the schematic diagram of the flat-top conical micro-indenter showing conical angle and depth of penetration h_u .

study the compressive characteristics of paper sheets [17] and wood [18]. Neither micro- nor nano-indentation has been used to study the transverse properties of single wood or pulp fibres.

Bergander and Salmén [19] developed an *in situ* environmental SEM technique to study the transverse elastic modulus of the native wood fibre wall and pointed out the importance of single fibre properties in transverse direction. Hartler and Nyrén [10] studied the compression behaviour of pulp fibres in transverse direction and reported that sulfite fibres had a higher transverse modulus than kraft fibres and earlywood fibres a lower transverse modulus than latewood fibres, but their cell wall deformation studies were restricted to observations using light microscopy. Mohlin *et al.* reported that fully bleached commercial pulps are more deformed than unbleached pulps [3]. The deformations such as kinks, angular folds and twists which are prone to change the direction of the fibre axis are found to have a negative effect on tensile index, compression strength, *etc.* So it is important to study the ‘localized’ deformation behaviour of single pulp fibres in addition to the above studies.

In this study, a novel approach was undertaken to understand the ‘localized’ deformation and fracture behaviour of a single pulp fibre in transverse direction. This novel technique allows real-time correlation between the cell wall deformation and the response from the load cell in addition to advantages of the SEM over an optical microscope used in other studies. In the present paper, *in situ* SEM micro-

indentation adopted to pulp fibre is explained and preliminary results are reported.

Experiment

Softwood kraft pulp from Scots pine (*Pinus sylvestris*) of commercial quality was received as dry sheets from the Laboratory of Pulp and Paper Science and Graphic Arts, INP-Pagora, Grenoble (France). The pulp was unrefined and unbleached (Kappa number: 29). A few grams of pulp were suspended in water and stirred magnetically for 5 min, and a drop of pulp solution was placed on a glass slide and allowed to dry at room temperature. Ten long single pulp fibres (~1 mm length) were carefully extracted using micro-tweezers and fixed at both ends with epoxy droplets on 10 different conductive glass slides, which contained indium tin oxide as a top layer to minimize charging during SEM imaging. Then, each conductive glass slide was fixed to the SEM stub using double-sided carbon tape and sputter coated with a 6-nm-thick Au–Pd layer to avoid further charging. Micro-indentation was carried out at 0% moisture content of the pulp fibres in an environment of 10^{-6} mbar and 296 K inside a Zeiss DSM962 SEM. A detailed description of the *in situ* SEM micro-indentation set-up is available elsewhere [20]. As shown in Fig. 1b, a conical diamond indenter (cone angle of 43°) ending in a flat top (diameter of ~2 μm) was used in the present study. For all the tests, deformation was carried out at a constant displacement rate of 100 nm s^{-1} and corresponding load (P) vs displacement (h) curves were obtained.

Fig. 2. Load (P)–displacement (h) curve and corresponding SEM video sequence of the micro-indentation test. Arrows labelled in SEM micrographs represent crack initiation and crack propagation probably along the microfibril angle. Schematic diagram of the mechanism of the micro-indentation test is shown in inset (top left). The corresponding *in situ* SEM micro-indentation video is provided as supplementary data online.

Results and discussion

Figures 2 and 3 show the P – h curves obtained from *in situ* SEM micro-indentation of single pulp fibres. Insets are SEM micrographs revealing the real-time deformation and fracture sequence of the fibre. In all cases, initial non-linear behaviour of the P – h curve arises due to the non-uniform geometry of the fibres and results in proper contact with the glass substrate.

The deformation and failure sequence of the fibre shown in Fig. 2 is described as follows: Initial deformation was accommodated by sinking-in of the upper cell wall, followed by cracking, which corresponds to the kink or load drop in the P – h curve. The crack propagation occurs along the longitudinal axis of the fibre, while the initiation seems to be

dominated by stress concentrations at the surface. As the load increases, a second crack was initiated at ~10 mN. The two cracks propagated along the axis to a length of around 10 μm , probably parallel to the microfibril angle. Crack formation led to local collapse of the cell wall and the indenter punched through the upper cell wall. Continued indentation into the lumen and the lower cell wall yielded an increase in the steepness of the P – h curve due to substrate effects (schematic diagram in Fig. 2). The cracks formed on the upper cell wall, close on unloading, and a residual imprint of the conical punch can be seen in the pulp fibre. The corresponding *in situ* SEM micro-indentation video is provided as supplementary data online.

Fig. 3. Load (P)-displacement (h) plot of pulp fibres and corresponding SEM images of micro-indentation test. Note the cell wall 'sinking-in' and 'pile-up' behaviour (white arrow) for fibre-a and cross-sectional cell wall expansion (white arrow) for fibre-b. Indentation was stopped before cell wall fracture for fibre-a. Schematic diagram of the mechanism of the micro-indentation test is shown in inset (top centre).

Tougher responses were observed during the deformation of fibres shown in Fig. 3. While the typical initial 'sinking-in' of the upper cell wall was observed around the contact zone, significant crack formation was not observed. Nevertheless, kinks are observed in the P - h curves, which may correspond to micro-cracks in conjunction to lateral bulging. The extensive bulging of the cell wall (Fig. 3b) could be due to the high cellulose and hemicellulose content (lignin content: 4.4%) and partial fibrillation of the fibre wall caused by the pulping process [8]. The slope difference between fibre-a and fibre-b (Fig. 3) could be due to the differences in the fibre (cell) structure and fibre-indenter alignment. It is assumed that the cell wall structure of fibre-a is less affected by pulping processes because

of the intact cross section compared with the flattened cross section of fibre-b (Fig. 3).

It is understood that the fibre geometry will play a significant role when a fibre is subjected to lateral compression because fibre collapse precedes the cell wall compression [2,9]. But it is important to note that, in the present study, the load is applied locally, which leads to large stresses and correspondingly large strains. Hence, the P - h curves shown in Figs. 2 and 3 are mostly based on the indentation response of the cell walls and specific to the given geometry of the fibres and indenter. Based on these assumptions, the critical stress required to initiate a crack or deformation in the cell wall was calculated and presented in Table 1 for the three fibres discussed in this study.

Table 1. Critical stress of pulp fibre cell walls in transverse direction

Fibre shown in	Load P (mN)	Displacement h (μm)	Critical stress σ (MPa)
Fig. 2	9.2	5.4	48.3
Fig. 3a	11.2	5.3	63.7 ^a
Fig. 3b	13.9	8.8	38.5

^aIndentation was stopped prior to the fracture of the upper cell wall.

Failure sequence of single fibres subjected to longitudinal compression differs from the failure sequence found in this study. Loosening of the fibrils, piling-up of the fibrillar walls and eventual fibril fracture reported [5] during longitudinal compression were also observed in the present study. But, as shown in Figs. 2 and 3, SEM micrographs did not reveal any slip bands similar to those observed during longitudinal compression of single pulp fibres.

Conclusion

The deformation response of single pulp fibres was studied using *in situ* SEM, which revealed a correspondence between the cell wall failure mechanisms and the P - h curve response. The failure sequence of single fibre cell walls begins with sinking-in and lateral bulging of the microfibrils followed by cell wall fracture. It was found that a critical stress of ~50 MPa is required to initiate cracks in the cell wall, which propagate parallel to the microfibril angle. This technique can be used to understand the difference between unbleached, bleached and refined single fibres in terms of their 'localized' deformation and critical load-carrying capacity of the cell wall.

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