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In situ ESEM study of the deformation of elementary flax fibres

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The deformation behaviour of single elementary flax fibres was investigated in an ESEM, using a modified loop test. Plastic deformation starts on the compressive side of the loop, whereas fibre failure occurs on the tensile side of the loop. The primary and the secondary cell wall show a different deformation behaviour. The primary cell wall breaks in a brittle manner, whereas in the secondary cell wall, due to its fibrillar nature, a coarse crack grows, bridged by fibrils. The secondary cell wall was found to split relatively easily along the length direction, indicating that the lateral strength of the fibre is lower than its tensile strength, which also accounts for the lower compressive strength of the fibre compared to its tensile strength.

1. Introduction

Recently the environmental scanning electron microscope (ESEM) has proven to be a versatile instrument for the study of biological tissue under controlled humidity conditions [1]. An additional advantage of the ESEM is that, due to the presence of gas molecules in the chamber, samples do not need to be gold-coated, thereby offering the possibility to perform *in situ* mechanical measurements.

In this paper a study on the deformation behaviour of flax fibres is presented. Flax is an annual crop from which, since old times, the fibres have been used to make linen fabric. However, since the flax fibre is a relatively strong, stiff and light weight fibre, its use as a reinforcing agent in polymeric composites recently has drawn much attention [2].

A flax fibre has an intricate structure. The 1 m long so called technical bast fibres that are isolated from the flax plant consist of elementary fibres (see Fig. 1) with lengths generally between 2 and 5 cm, and diameters between 10 and 25 μm. The elementary fibres are glued together by a pectin interface. They are not circular but a polyhedron with 5 to 7 sides to improve the packing in the technical fibre. The elementary fibres are single plant cells, they consist of a primary cell wall, a secondary cell wall and a lumen, which is a small open channel in the centre of the cell. The lumen can be as small as 1.5% of the cross section [3]. The primary cell wall is relatively thin, about 0.2 μm [4] and consists of pectin, some lignin and (hemi)cellulose [5]. The secondary cell wall makes up most of the fibre diameter. It consists of oriented highly crystalline cellulose fibrils and amorphous hemicellulose. The secondary cell wall gives the fibre its high tensile strength. The crystalline cellulose fibrils in the secondary cell wall are oriented at an angle of $+10^\circ$ with the fibre axis [3, 6]. From mechanical studies on flax elementary fibres [7] it was found that the tensile strength of the elementary fibres is around 1500 MPa, and the compressive strength is approximately 1200 MPa. Furthermore it was observed that the fibres generally show deformed zones, presumably kink bands, due to the decortication process by which the fibres are isolated from the plants. Within the deformed areas, confocal scanning laser microscopy (CSLM) [7] revealed the presence of a fibrillated structure, with fibril diameter in the order of 0.1 μm. This structure was not observed on the fibre surface (the primary cell wall) and is therefore expected to appear in the secondary cell wall.

In the present study, the deformation of flax fibres in a modified loop test is investigated *in situ* in the ESEM. This test set-up allows the compressive and tensile failure of the fibres to be examined closely, without the need for coating. Since ESEM is a surface characterisation technique, obviously only the features appearing at the fibre surface can be examined. However, eventual failure of the primary cell wall also allows a view deeper into the secondary cell wall of the fibre.

2. Experimental

Flax (JS2-33-1995 from Cebeco, NL) was warm water retted and decorticated (braked and hackled) on a laboratory scale at ATO-DLO. Elementary fibres were isolated by hand from the technical bast fibres and glued with one end onto a small cardboard frame. At the free fibre end a single knot subsequently was made, leaving a loose loop of approximately 1.5 mm radius. The free fibre end was then glued onto the other end of the cardboard frame. The cardboard frame was mounted

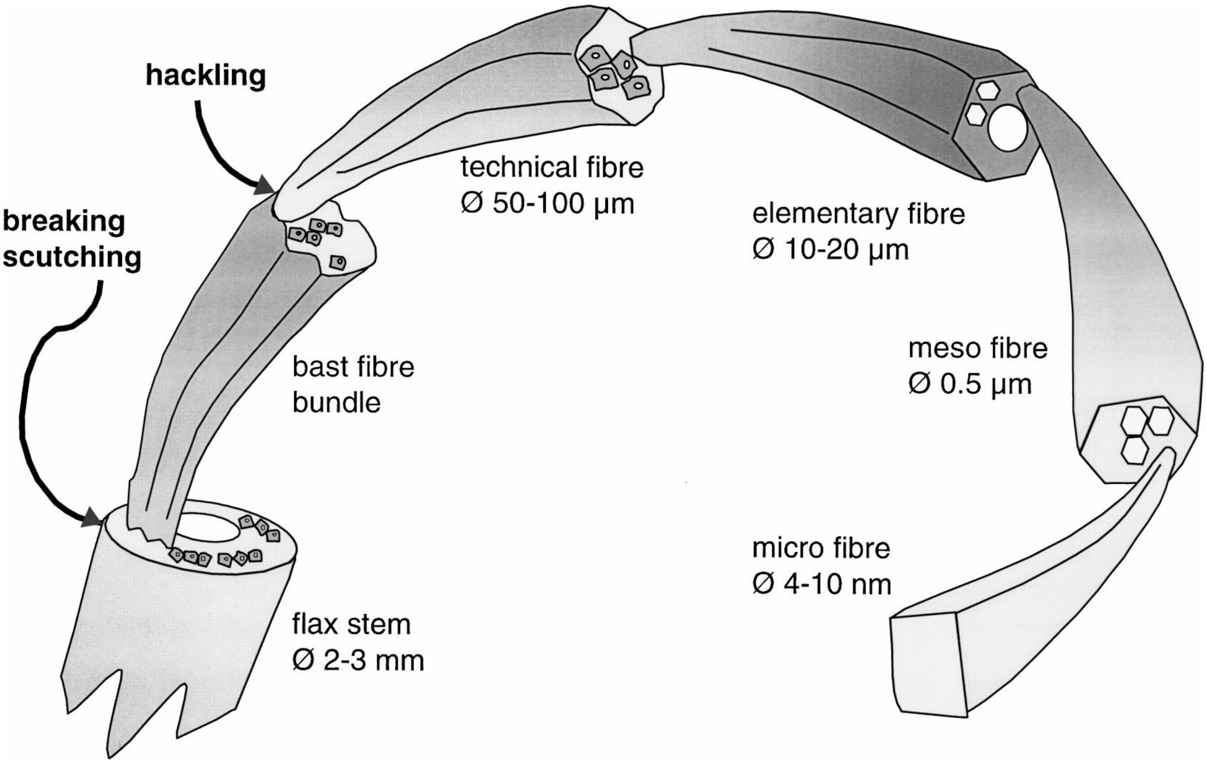


Figure 1 Composition and built of flax fibres.

on the tensile stage of the ESEM (Electroscan ESEM 2010), and the edges of the cardboard frame were cut, allowing the fibre to be strained freely. The microscope was pumped down to vacuum and flooded with water vapour up to 4 Torr pressure (this improves imaging), leaving the fibres relatively dry, indicative of their state in a composite material or for instance during extrusion compounding together with a thermoplastic polymer. The beam voltage was kept as low as 12 keV in order to avoid beam damage to the fibres. No evidence of beam damage was found at this level. The experiments were performed at room temperature.

3. Results and discussion

Straining of both fibre ends causes the loop to tighten slowly, thereby inducing compressive deformation on the inner side and tensile deformation on the outer side of the fibre. Since the loop is closed with a single knot, it is a nearly perfect circle, indicating that the stresses on the fibre along the loop are constant. The compressive stress on the inner side of the loop is approximately equal to the tensile stress on the outer side of the loop and can be written as:

$$\sigma_z = \left(\frac{E}{R}\right) z \tag{1}$$

with σ_z representing both the compressive and tensile stresses on the inner and outer side of the looped fibre, E the fibre modulus, which can be taken as 50 GPa [7], R the radius of the loop and z the radius of the fibre. A fibre of a radius of 20 μm reaches its compressive yield stress of 1200 MPa at a loop radius of approximately 0.4 mm, which is exactly within the accessible range of the experiment.

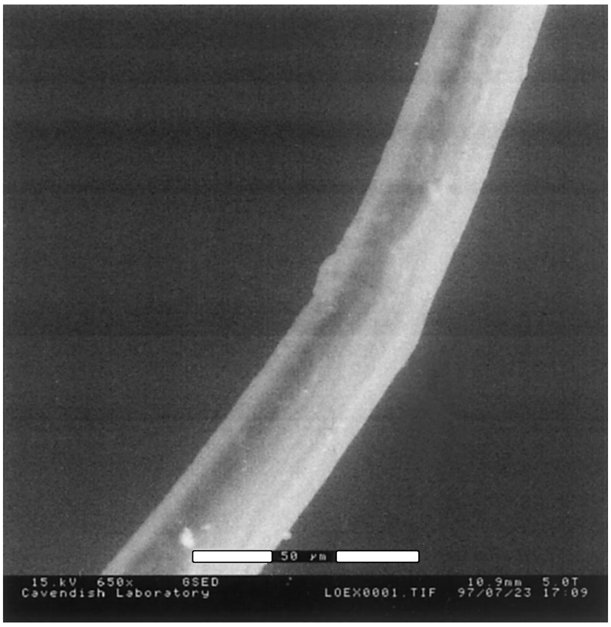


Figure 2 Elementary flax fibre which has just developed a kink band.

Fig. 2 shows a fibre which has just started to develop a kink band. From the curvature of the upper halve of the fibre, it can be estimated that the radius of this fibre loop is approximately 430 μm. This would correspond to a compressive strength of the fibre of approximately 1300 MPa, which is remarkably close to the value of 1200 MPa. Although this is of course a very rough estimate, it indicates that during the experiment in the ESEM the compressive behaviour of the fibre is likely to be comparable to its behaviour under experimental conditions outside the microscope.

Upon further straining the loop beyond the radius where the first kink band appears, the number of kink

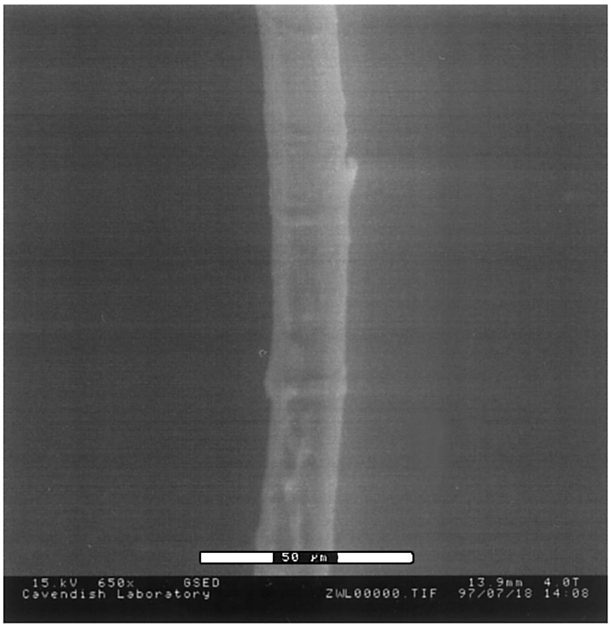


Figure 3 Elementary flax fibre in which a number of kink bands appear.

bands along the loop increases. The kink bands simultaneously become more pronounced (see Fig. 3), up till the point where they show a strong outward buckling of the primary cell wall. However, the primary cell wall never shows actual fracture on the compression side, the deformation is of a plastic, and irreversible, nature. The deformation possibly taking place in the secondary cell wall is of course not visible in the ESEM, since it will occur beneath the visible surface.

Fracture of the primary cell wall of the fibre is observed on the tensile side of a deforming kink band. Fig. 4a–f clearly show the initiation and development of fracture of an elementary flax fibre. Just before the primary cell wall of the fibre begins to tear, the curvature of the fibre on the tensile side is strongly increased, presumably due to failure in the secondary cell wall. A sharp crack then occurs perpendicular to the fibre long axis and the direction of the highest stress (Fig. 4a and b). In Fig. 4c clearly both the primary and the secondary cell wall can be seen. At the point where fracture has taken place, the primary cell wall appears to have separated from the secondary cell wall. Fig. 4d gives a view inside the fibre in the deformed secondary cell wall. A fibrillated structure in the secondary cell wall is clearly visible. The failure in the secondary cell wall bears little resemblance to the sharp crack which runs in the primary cell wall. The crack in the secondary cell wall is bridged by relatively thick fibrils. Further straining of the loop (Fig. 4e) causes the primary cell wall to tear further, and the fibrils bridging the crack in the secondary cell wall show plastic drawing up to failure. Fig. 4f is a micrograph taken just before the fibre failed. On the fracture surface some fibrils can be seen sticking out.

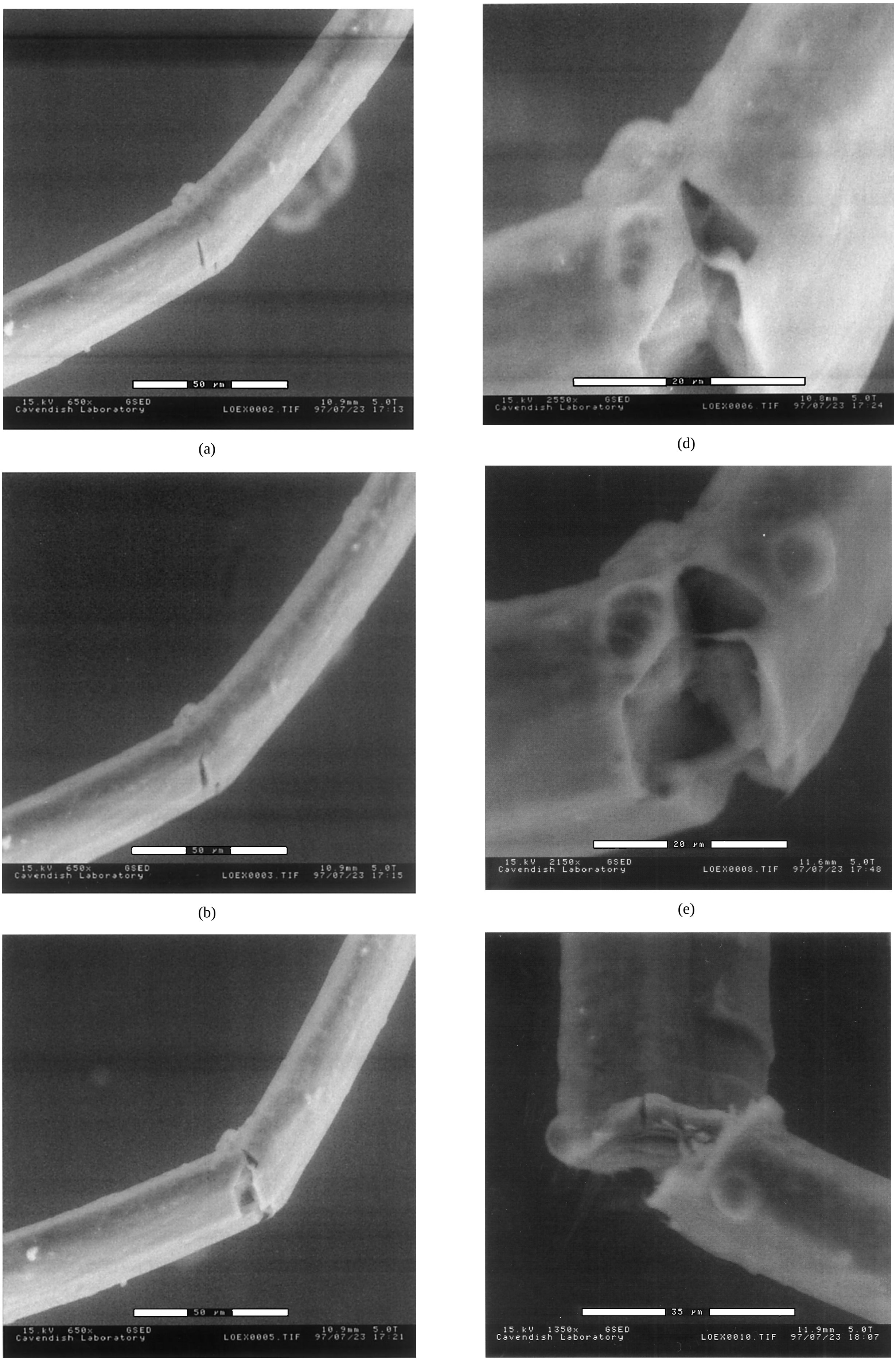
From this series of micrographs it is clear that the primary and the secondary cell wall show different mechanical behaviour, presumably as a consequence of the different chemical composition and morphology of both cell walls. The primary cell wall contains a large fraction of amorphous pectins and hemicelluloses and crosslinked lignins. It is therefore not surprising that the crack growth in the primary cell wall is of a rather brittle nature.

The secondary cell wall however consists of ca. 70% of crystalline cellulose. The fibrillar nature of the secondary cell wall has a large influence on its mechanical behaviour. Fibres with highly oriented crystalline structures, like carbon, aramid and PE fibres, are known to be sensitive to kink band formation under compressive loading. In view of the structure of the secondary cell wall, flax fibres are likely to fall in the same category. Williams *et al.* [8] have proposed a model for the bending failure of carbon fibres which seems also to be applicable for the case of flax fibres. They depict the fibre as similar to a steel cable, consisting of twisted steel wires. Upon bending such a cable, the wires easily come apart. It is likely that the holes in Fig. 4e are formed by a similar mechanism; the fibrillar structure of the secondary cell wall separates under bending or compression. There is of course interaction between the fibrils in the secondary cell wall: the amorphous hemicelluloses form the glue that keeps the fibrils together. However, the hemicelluloses will at some point fail due to the lateral stresses induced by the compressive strain and holes as seen in Fig. 4e will be formed. Since the fibrils themselves will not fail under compression, the resulting structure will be a crack bridged by coarse fibrils, as seen in Fig. 4e and similar to the structure found with a CSLM by Bos *et al.* [7].

As mentioned in the introduction, isolated flax fibres often already contain damaged zones due to bending of the fibres during the decortication process. These would obviously also be the spots where the first kink bands become visible during a compression or bending test. Also for the fibres studied here it can not be excluded that damage in the secondary cell wall already exists at the beginning of the test. Moreover, the fact that Fig. 4a indicates an increased curvature on the tensile side, which develops rather early during the deformation of the fibre, could indicate that the secondary cell wall structure of the fibre at this spot was already damaged before the test. Since no damage was seen in the primary cell wall, it can then be concluded that, similar to Williams' model for carbon [8], also in the flax fibres the primary cell wall could serve to keep the fibre together, even though the secondary cell wall might already be badly damaged.

It is unclear how deep the crack in Fig. 4 penetrates into the secondary cell wall. However, if this damage was already present before the test, it is not unlikely that the damage extends through the whole thickness of the fibre. It is therefore also quite possible that the secondary cell wall of finely hackled flax fibres, which have undergone a rather severe mechanical decortication and combing procedure, is actually full of damaged zones like these. A more careful way of isolating the fibres might then improve both the tensile and the compressive strength of the elementary flax fibre, which would be of interest for the potential use of these fibres in structural composite materials.

Another striking event is shown in Fig. 5a and b. This fibre has failed under tension as in the previous example. However, further tightening of the loop has at some

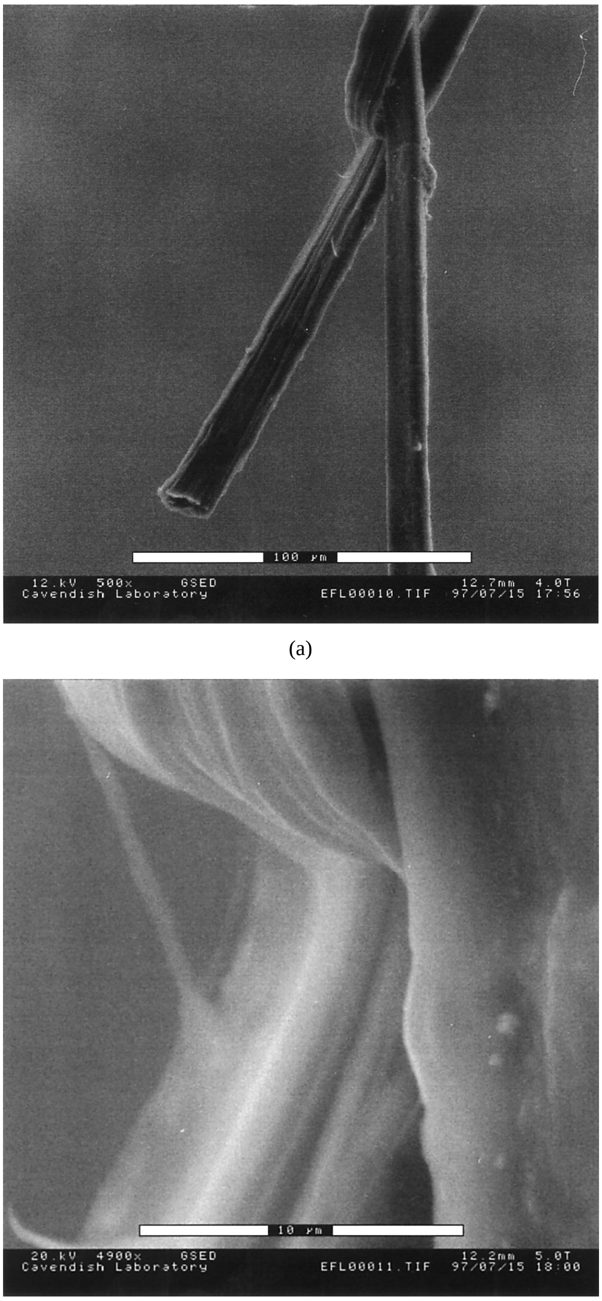


(a) (d)

(b) (e)

(c) (f)

Figure 4 Initiation and development of fracture in an elementary flax fibre. (a) Fracture initiates on the tensile side of the fibre. (b). Crack in the primary cell wall widens. (c) Primary and secondary cell wall have separated. (d) View into the deformed secondary cell wall. (e) Extended plastic deformation of the fibrils in the secondary cell wall. (f) The fibre just before complete failure.



(a)

(b)

Figure 5 Fibre separating in the secondary cell wall due to tightening of the loop. (a) Overview, the loop is visible in the upper right of the micrograph. (b) Magnification of the crack front.

point caused the crack to deviate along the direction of the fibre long axis, splitting the fibre in two halves. Clearly visible on the fracture surface now is the fibrillar structure of the secondary cell wall, with the fibrils at a slight positive angle to the fibre long axis. The apparent ease, at which part of the fibre is stripped off over a large length indicates again that the lateral strength of the fibre is lower than the tensile strength in the length direction. This is also indicated by the fact that the crack stays within the same layer; it does not cross the fibrils. The crack front in Fig. 5b shows that there is little tendency to crack bridging once the direction of crack growth has changed to the length direction of the fibre.

These results indicate that once a crack is running in the length direction of the fibre it will easily split the fibre over its entire length. In composites this could lead to a preferred direction of crack growth. Furthermore, in a composite with optimised fibre/matrix bonding, splitting of the fibre over its length might prove to be the dominant failure mechanism. Further composite strength optimisation can then only be reached by improving the strength of the hemicellulose bonding between the microfibrils in the secondary cell wall.

The fracture surfaces in Fig. 5 show that at this depth in the fibre all fibrils are oriented in the same direction, the angle is approximately 10° off the fibre long axis. In the literature there is still some confusion regarding the actual structure of the secondary cell wall. Herzog [3] has described the secondary cell wall as at least a two layered fibrillated structure with a change in fibril direction from $+10^\circ$ to -5° relatively close to the fibre surface. Mark [9] describes the cell wall as a three layered structure with winding angles of -30°, 6.5° and -30° for three fibril layers with thicknesses of respectively 1.2, 5 and 0.5 μm. Davies and Bruce [10] use the same fibre model in their calculation of the mechanical response of flax fibres. The micrographs shown here, however, indicate no change in fibril direction close to the surface and a main fibril direction of 10° down to a depth of approximately 4.5 μm into the secondary cell wall. No conclusions can be drawn with respect to any change in fibril direction deeper into the secondary cell wall.

4. Conclusions

The compressive behaviour of a lignocellulosic fibre like flax can very well be studied *in situ* in an Environmental SEM. The modified loop test, in which the loop is closed with a single knot, allows the study of compressive and tensile deformation on respectively the inner and outer side of the loop.

The difference in chemical composition and morphology of the two cell walls that form the flax fibre gives rise to differences in deformation behaviour. Failure of the primary cell wall is brittle and takes place on the tension rather than the compression side of the fibre loop. Failure of the secondary cell wall under compressive loading takes place in the lateral direction due to its highly oriented crystalline nature. Cracks bridged by coarse fibrils are formed in the secondary cell wall before the primary cell wall fails.

The strength of elementary flax fibres in the lateral direction is lower than in the fibre direction. For application of flax fibres in structural composites this is a fact which should be taken into account.

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