ESRF	**Experiment title:** Inelastic behaviour of single plant fibres under tension: in-situ tensile tests with microdiffraction on flax, ramie and single wood cells	**Experiment number**: SC-910
Beamline: ID13	**Date of experiment**: from: 26.02.02 to: 02.03.02	**Date of report**: 02.03.03
Shifts: 12	**Local contact(s)**: Stephan V. Roth	*Received at ESRF:*

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Report:

This first report on experiment SC-910 refers to the *in situ* stretching experiment on single native cellulose fibres (flax). Analysis of the second part of the experiment (single wood cells) is still in progress.

Native cellulose fibres are biological composite materials. The biopolymer cellulose is semicrystalline with nanocrystals (so-called microfibrils) embedded in a softer matrix of disordered cellulose [1]. In the case of flax, all cellulose molecules are aligned along the longitudinal fibre axis [2], giving the fibre its high Young's modulus and strength. Cellulose microfibrils in flax have a typical diameter of 40 Å and a length of about 0.1 µm; their orientation distribution has a width of only a few degrees [3].

Upon stretching of a cellulose fibre, two processes can be expected: (1) Rotation of the cellulose microfibrils towards the fibre axis, leading to a narrowing of their orientation distribution; (2) straining of the microfibrils, visible in a shift of the lattice parameter in fibre direction, *c*. The rotation had already been observed previously in a static *in situ* stretching experiment using microdiffraction at the ESRF Microfocus Beamline ID13 [4]. The experiment was termed static since after increasing the strain, the fibre was allowed to relax until a stable stress value was measured. The X-ray diffraction diagram was then taken in the relaxed state.

In nature, relaxation is generally not possible while tensile load is applied. Thus, ideally the strength and the structural properties of a fibre under tension should be measured in a dynamic way, i.e., with a constant stretching rate ε/t that has to be high enough so that relaxation effects can be neglected. In close collaboration with the ESRF (Jens Meyer, Manfred Burghammer) we developed a SPEC macro that allowed fibres to be stretched in the ID13 piezo stretching device with a given rate. With the new Photonics Science image intensified CCD detector on ID13 it was possible to acquire a microdiffraction pattern (beam size 10 µm) from a single flax fibre (typical diameter 20 µm) in 2 seconds (Fig. 1 a). The readout time was just 300 ms, and the strain rate amounted to ε/t = 4 %/min. Since 4 % is about the typical maximum strain of flax fibres, a stress-strain curve of the fibre with about 26 points is obtained in one minute (Fig. 2, triangles). The Young's modulus (slope) is practically constant, in particular there is no bend in the curve at higher strain indicating plastic or viscoelastic behaviour.

Both rotation and strain as discussed above are already visible in the raw data (compare Fig. 1a and b, taken at 0 % and 2 % strain, respectively): (1) The azimuthal width of the reflections (most prominently of the equatorial 200 reflection) is reduced considerably; (2) the position of the meridional 004 reflection shifts inwards. It is interesting to note that the process of reorientation (1) is already completed at about 1.8 % strain (diamonds in Fig.2). The strain of the microfibrils was calculated from the position of the 004 cellulose reflection. In a first approach we assumed isotropic stress distribution in the composite material. In order to draw the microscopic stress-strain curve of the microfibrils (circles in Fig. 2) the respective stress is taken from the macroscopic (fibre) stress-strain curve at the fibre strain where the diffraction diagram was taken at (see dashed line in Fig. 2). The Young's modulus of the crystals is about a factor of five higher than that of the composite material, the fibre.

The aim of this study is to develop a consistent model for the mechanical properties of cellulose fibres. The assumption of isotropic stress distribution, particularly in a dynamic stretching experiment, is certainly an over-simplification. Furthermore, the interplay between the processes (1) and (2) have to be taken into account. An important parameter will be the mechanical properties of the soft matrix of disordered cellulose.

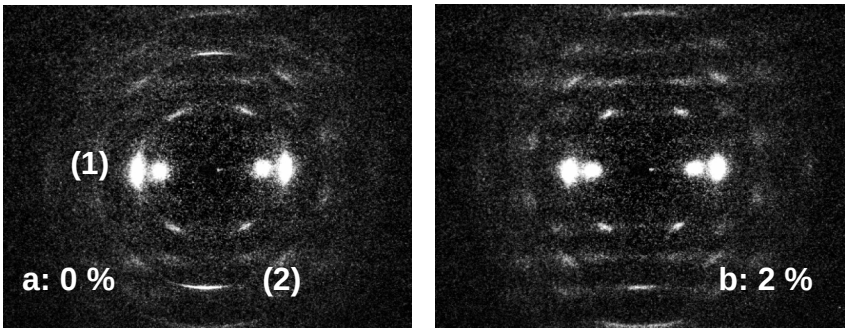

Figure 1: Microdiffraction fibre diagrams of a single flax cellulose fibres at 0 % and 2 % strain (a and b, respectively). Two effects are observed upon stretching *in situ*: (1) Reduction of the azimuthal width of the Bragg reflections; (2) inward shift of the position of the meridional 004 reflection. Thus, the cellulose microfibrils rotate (1) and are stretched (2).

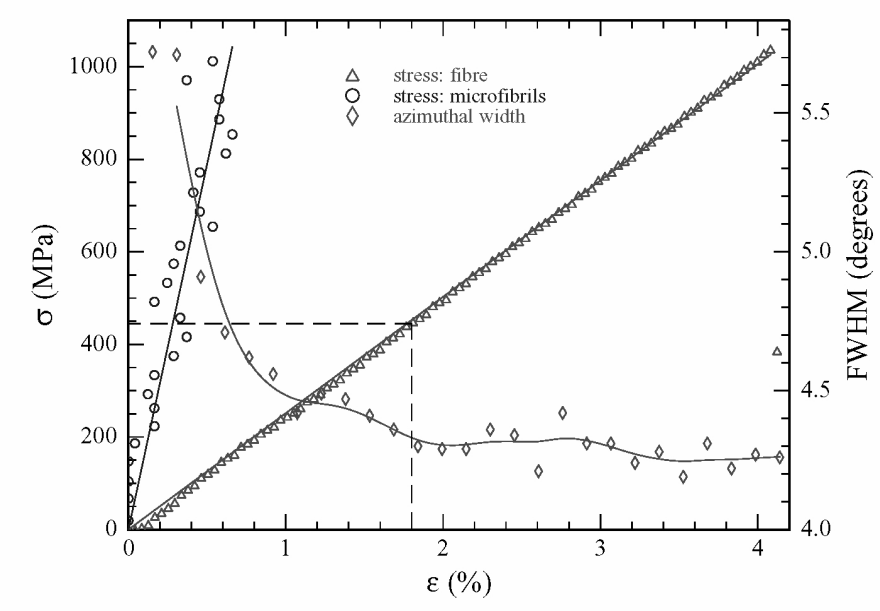

Figure 2: X-ray microdiffraction results of a dynamic *in situ* stretching experiment on a single flax fibre. The macroscopic stress-strain curve of the fibre (triangles) has an almost constant slope. The internal strain of the cellulose microfibrils is calculated from the shift of meridional Bragg reflections. Under the assumption of isotropic stress distribution (dashed line, see text) the microscopic stress-strain curve of the microfibrils can be calculated (circles). Rotation of the microfibrils (diamonds) is already finished at about 1.8 % fibre strain.

References

[1] A. C. O'Sullivan. Cellulose: the structure slowly unravels. *Cellulose* **4**, 173-207 (1997).

[2] M. Müller, C. Czihak, H. Schober, Y. Nishiyama, C. Riekel. All disordered regions of native cellulose show common low-frequency dynamics. *Macromolecules* **33**, 1834-1840 (2000).

[3] M. Müller, C. Czihak, G. Vogl, P. Fratzl, H. Schober, C. Riekel. Direct observation of microfibril arrangement in a single native cellulose fibre by microbeam small-angle X-ray scattering. *Macromolecules* **31**, 3953-3957 (1998).

[4] *ESRF Experimental Report* on experiment **SC-568** (on ID13)