ESRF	**Experiment title:** Influence of water on the mechanical properties of native cellulose: Microdiffraction and in situ stretching experiments on single flax fibres	**Experiment number**: SC-1439
Beamline: ID13	**Date of experiment**: from: 24.02.05 to: 28.02.05	**Date of report**: 01.09.05
Shifts: 9	**Local contact(s)**: Richard Davies	*Received at ESRF:*

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Report:

The semi-crystalline biopolymer *cellulose* consists of fibrous nanocrystals, the so-called *microfibrils*, embedded in a softer matrix of disordered cellulose [1]. Native cellulose fibres like flax have unique mechanical properties with a Young's modulus as high as the artificial high-performance fibre Kevlar. The understanding of the mechanical properties of cellulose fibres has to be based on their composite nature. Upon stretching, the cellulose microfibrils (i) *rotate* towards the longitudinal fibre axis so that their orientation distribution is narrowed [2] and (ii) are *stretched*, visible in a change of the lattice spacing in fibre direction [3]. These two mechanisms can be investigated with X-ray fibre diffraction.

Water drastically influences the mechanical properties of cellulose fibres since it penetrates the disordered matrix but not the crystalline microfibrils [4]. The Young's modulus is reduced as the matrix becomes less stiff. In the experiment reported here we investigated the different microstructural changes in dry and wet flax fibres, respectively.

We extracted single fibres with the help of an optical microscope and glued them into small plastic frames (Fig. 2). These films were afterwards mounted within a specially designed mechanical holder, which permits easy handling of the samples and ensures that there are no mechanical preloads before the measurement starts. Then the polymer films were cut apart using a soldering gun.

The sample holder loaded with the fibre was transferred into our humid stretching environment (HUSTEN) that allows the simultaneous acquisition of X-ray diffraction patterns and of force-elongation data within a humidity controlled atmosphere (Fig. 1).

To ensure a defined atmosphere around the sample the temperature of a water vat in the bottom of the chamber containing a saturated solution of NaCl was controlled with a standard controller (Lakeshore model 340). The resulting warm air with its high moisture content ascents to the sample where the relative humidity of the air was measured using a capacitive sensor.

The elongation of the sample was applied using standard linear guides with a ball screw of 1 mm pitch (THK) driven by a 2 phase stepping motor (MICOS, Pollux SMC) in high-resolution micro-step mode. The real

movement of the guide was monitored using an industry standard strain gauge (Heidenhain MT25) with a guarantied spatial resolution of 0.2 µm.
In order to measure the loading of the sample a force sensor was applied, which was a strain-gauge beam arrangement (Entran) with a maximal permitted force of 0.5 Newton. This sensor was powered by a 10V DC power supply (TTi) and read out with a data acquisition device (National Instruments PCI-6035E) floating in 0.1 Volt range at 16 bit resolution every few milliseconds.
To allow the X-rays to pass through the chamber there were two windows (Kapton) included. Behind the exit window the diffraction patterns were collected with a Photonic Science Gemstar CCD X-ray detector. To protect this detector against any damage due to the high intensity of the primary beam a lead beam stop of 200 µm diameter held with a glass capillary was placed in between the detector entrance and the sample chamber exit window. The detector was triggered with a 5V TTL signal generated by the PC controlling the experiment to synchronise the diffraction patterns with the rest of the measurements. This TTL signal was also shared to control the ID13 fast shutter.
To be able to vary the position at the sample that was illuminated by the X-ray beam the whole sample environment was placed on top of a xyz-translation table (MICOS) that was driven by the ID13 experimental control PC running spec©. The movements of this table were induced via TCP/IP connection between the ID13 PC in listener mode and the experiment controlling PC running a G code (bacon v0.5).
The whole experiment was observed with a CCD Camera (Lumera Lu205C) through a telecentric lens system (Zeiss) looking sidewise and via a heated mirror onto the sample itself.

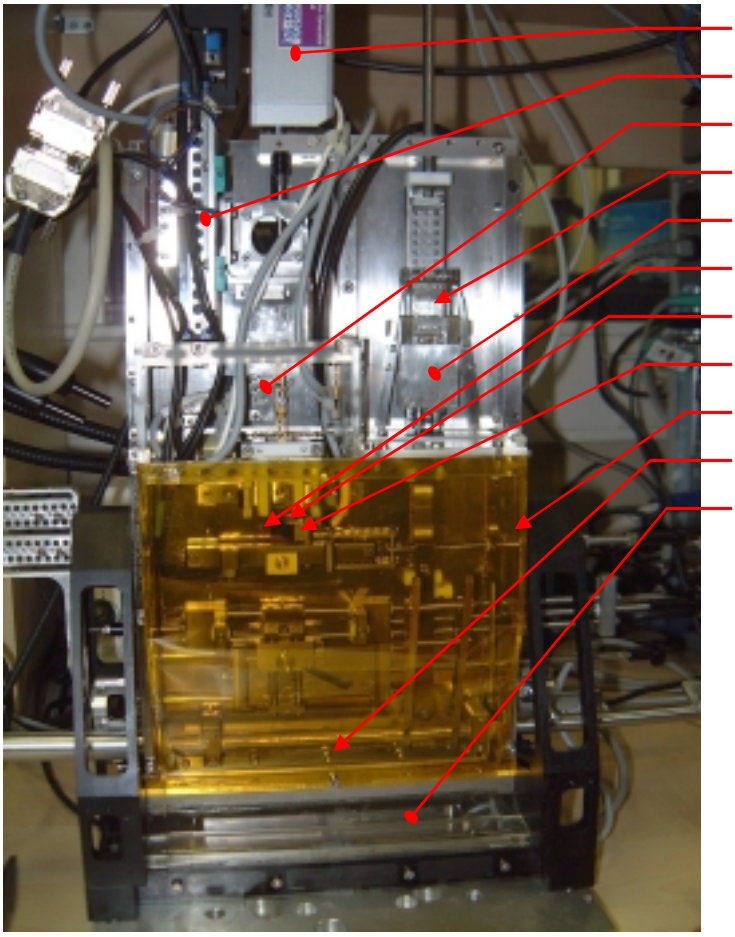

Fig. 1: Sample chamber HUSTEN viewed against the X-ray beam direction, showing the large Kapton exit window (yellow). For the experiment, the sample holder is moved into the beam position behind the entrance window.

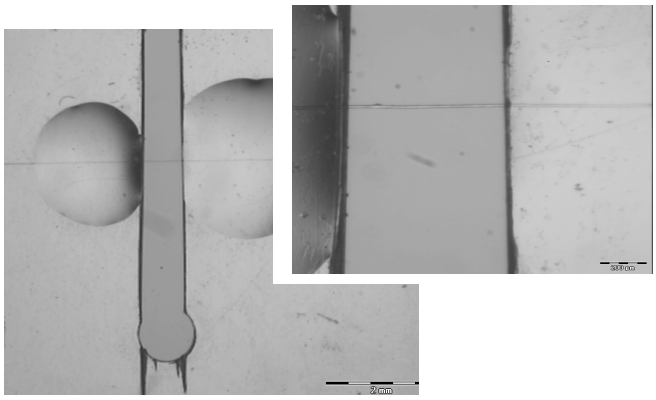

Fig. 2: Optical micrographs of a single flax fibre (20 µm in diameter) glued into a plastic frame. Sample free length is 660 µm.

In the stretching experiments, fibres were continuously stretched at a rate of about 0.36 %/min, i. e., faster than relaxation. Simultaneously, diffraction images were collected for 1 s every 2 s in horizontal sample scans with 21 positions every 10 µm over a scan range of 200 µm. Fig. 3 illustrates the importance of this fast stretching/scanning set-up. The single flax fibre was not completely straight at the start of the stretching experiment and thus moves laterally (by ca. 35 µm) when it is straightened. Like most fibres, also this one was slightly tilted with respect to the stretching direction, resulting in a further shift of about 10 µm. Eventually, the fibre broke and moved again.

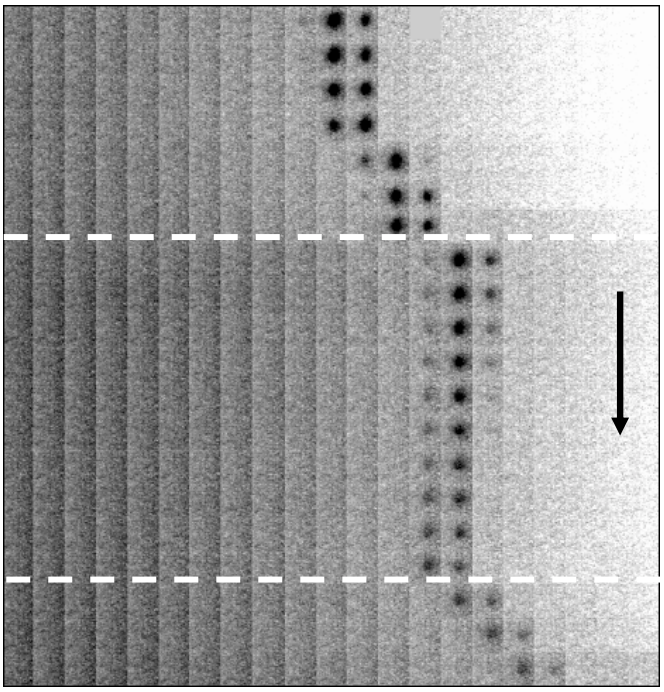

Fig. 3: Illustration of the fast scanning experiment during continuous stretching of a single flax fibre. Every single frame is a zoom into the region of the strongest cellulose reflection 200. Every line represents 21 CCD images with 1 s exposure time each and 1 s sleeping time between two images, i. e., 42 s in total. Scanning range was 200 µm for every row, corresponding to a step size of 10 µm.
The dashed lines separate the regions (from top to bottom) of initial straightening of the fibre, of the stretching process and of the rupture of the fibre.

Data analysis is still in progress. A first result is that the deformation of the crystalline cellulose microfibrils is much smaller than for dry fibres where the crystal strain in fibre direction amounts to about 25 % of the macroscopic fibre strain [3]. This would mean that the stress transfer in the wet (and therefore softer) matrix is less efficient. In contrast, the changes to the microfibril orientation distribution seem to be very similar in the wet and the dry case.

References

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