Wide and small angle X-ray scattering measurements of strong cellulose filaments: Fibril alignment during processing and in the dried material

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The mechanical properties of many biological materials, e.g. cellulose fibres from trees and spider silk, are determined by the nano-structure of the material[1, 2]. For the particular example of a wood fibre, properties comparable to glass fibres can be reached. In order to obtain this, the cellulose nano-fibrils (CNF) (near-crystalline bundles of cellulose fibres) that are the building block of the fibre has to be aligned with the fibre axis.

Recently, energy efficient disintegration of cellulose fibres into nano-fibrils has been made possible, making CNF a viable raw material for filaments, films and composites[3]. However, it has not been possible to reproduce the mechanical properties of the fibre originating from the tree.

We use a new approach to assemble CNF into filaments. The fibrils are aligned and assembled in a flow focusing device[4, 5] (see figure 1).

Small and wide angle X-ray scattering at the P03 beamline have been used to detect and measure fibril alignment (i) during processing and (ii) in the dry filament. Sample results are shown in figure 2. Figure 2a shows a visualisation of fibril alignment with polarised light. The micro focus SAXS

![Figure 1: Sketch of the flow focusing process and a photo of the flow cell. An CNF dispersion is ejected in the central flow \( Q_1 \) and is focused by a sheath flow \( Q_2 \). During focusing, the fibrils are aligned. Diffusion of ions from the sheath flow to the central thread can turn it into a gel, which can be ejected for further processing.](image-url)
images in figure 2b show that the nano-scale fibrils are aligned during focusing. In figure 2c, nano-focus WAXS images demonstrating alignment of the fibrils (or actually the crystals in the fibrils) are shown.

First of all, the SAXS measurements have proven that the alignment during processing actually occurs. The combination of SAXS during the process and WAXS of the dry filament have also made it possible to quantify the fibril alignment at both stages of processing. A considerable increase of alignment during drying (when the solids concentration of the filament increases from 0.3% to almost 100%) has been demonstrated. Fibril alignment measurements have also been performed in mixed CNF/carbon nano tube filaments with great success[6].

Last but not least, the fibril alignment in the dry filament was shown to be comparable to equally strong cellulose fibres extracted from wood. This is an important step that gives the hope of manufacturing high performance filaments from CNF. In order to achieve this, the alignment and assembly process needs to be fully understood. It is clear that SAXS and WAXS will be extremely valuable techniques during this work.

References