ESRF	Experiment title: **Stiffness recovery effects in biomimetic materials studied by in-situ WAXS combined with tensile tests**	**Experiment number**: SC-1491
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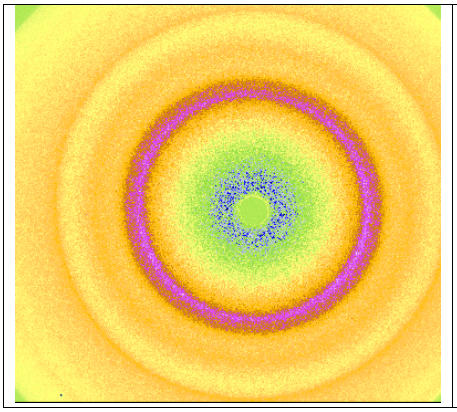
Report:

The outstanding mechanical performance of biological materials resides in their fibrous architecture and in specific *molecular mechanistic phenomena* operating between and/or in the fibres. It has been demonstrated that these phenomena are responsible for stiffness recovery effects in nacre and in bone [1,2] and also in wood [3-8]. This feature documents a tremendous variability of polymer interactions in biomaterials which has been implemented also in man-made biomimetic composites [9]. In-situ WAXS combined with mechanical tests provide a unique opportunity to evaluate structural changes in composite tissues at specific stages of tensile experiments. The hierarchical architecture of biological materials combined with compositional variability down to the *nm* level represents, however, a serious difficulty when studying bond recovery phenomena and viscoelastic effects. On the other hand, biomimetic materials with only few constituents provides an opportunity to controllably modify the mechanical behaviour by selecting a specific tissue architecture and well-defined fiber and matrix constituents.

The main goal of the present experiment was to study the mechanical behaviour of various cellulose-based materials using in-situ WAXS combined with mechanical tests and analyse the structural changes as a function of the external loading. A variety of materials based on bacterial cellulose and cellulose II were analysed. Additionally, some tests were performed on *Picea abies* [L.] Karst. tissues and on coir fibers. The users delivered their own computer-controlled tensile stage to the beamline which was used to strain the samples. The tensile stage was mounted on the "Huber tower" inserted in the goniometer. The straining of the samples was performed with different strain rates (0.001-0.3 mm/s) and the WAXS signal in transmission was collected using the present CCD. Thin foils with dimensions of 50 x 5 x 0.2 mm were tested. During the straining process, the stress responce of the tissue as well as the WAXS data were collected simultaneously. The main intentation was to understand the structure-property relationship in the relatively simple cellulose-based composites and compare it with the behaviour wood and coir fibres.

All materials exhibited significant changes in the orientation of the cellulose crystallites induced by the external strain. In Fig. 1. an example of the morhophology of Debye-Scherrer rings document structural

changes in the composite of cellulose II. The external force induced an irreversible reorientation of the cellulose fibers in the tissue.



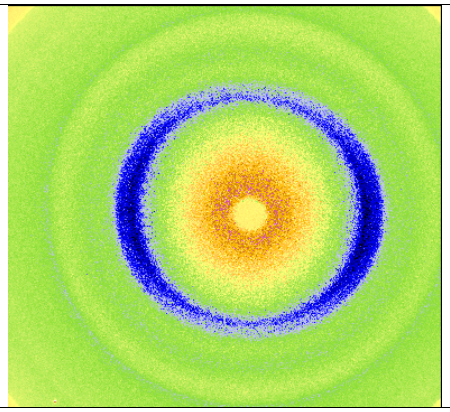


Figure 1 WAXS patterns collected during a tensile experiment on a composite of cellulose II. The frames right and left represent the structural information from the beginning and from the end of the tensile experiment, respectively.

The measurement data were used to evaluate the changes in the orientation of the cellulose fibrils as well as the elastic strain in the cellulose nanocrystals. The orientation of the cellulose fibrils was determined from FWHM of the intensity distribution along the Debye-Scherrer rings. The texture in the tissues was found to depend only on the actual magnitude of the external strain applied on the samples. In Figure 2, mechanical data and preliminary evaluated FWHM along Debye-Scherrer ring of celllulose 200 reflection are presented. The results indicate that the orientation of the fibers is a function of the external strain.

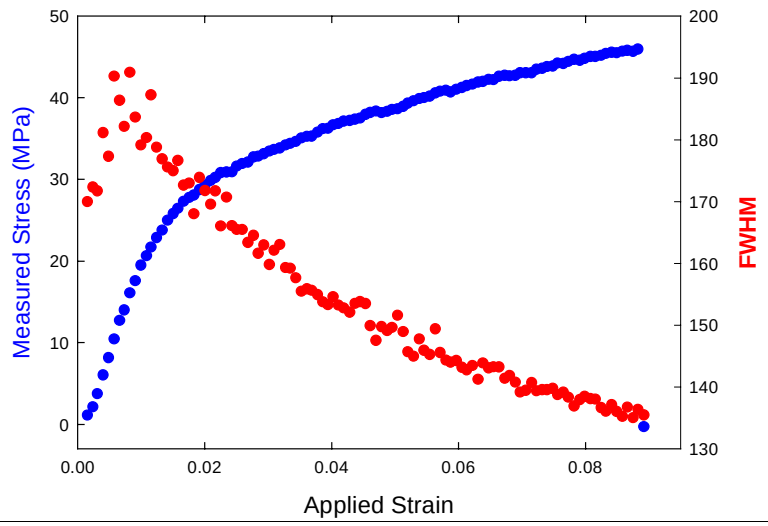


Figure 2 Mecahnical data (stress-strain curve) are correlated with the orientation of the cellulose fibrils in a cellulose II composite. The decrease of FWHM documents the reorientation of the cellulose in the tissues due to the external loading.

Similar effects were observed in other tissues. The reorientation of the fibrils in wood, bacterial cellulose, cellulose 2, cellulose composites and coir fibres is a function of the external strain.

In conclusion, the experiment was very successful and, after the thorough data evaluation, the users will present the results in referred journals.

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