THE MULTI-SCALE ARCHITECTURE OF CELLULOSE HYDROGELS INVESTIGATED BY SMALL ANGLE SCATTERING TECHNIQUES

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Regardless of its source, native cellulose is characterised by a complex architecture composed of distinct structural features (i.e.cellulose nanocrystals, microfibrils and bundles or ribbons) which are hierarchically arranged. Although deconstruction methods based on sequential component extraction and/or hydrolytic treatments have been typically used to study the structure of cellulose in plant-based resources, they are of limited relevance due to the intrinsic structural alterations induced by these treatments. An alternative approach is the synthesis of highly pure cellulose hydrogels by means of bacterial fermentation. Small angle neutron and X-ray scattering (SANS and SAXS) techniques are an extremely powerful tool to characterise the native structure of highly hydrated cellulose hydrogels, covering the whole size range of interest and, unlike most characterisation methods, avoiding sample drying processes which affect the native cellulose structure. This work demonstrates the potential of a multi-technique approach based on the combination of SANS and SAXS with X-ray diffraction, spectroscopy and microscopy to elucidate the hierarchical structure of cellulose hydrogels. A multi-scale model based on core-shell cylindrical structures has been built and applied to the scattering data. This has revealed the multi-phase structure of the cellulose microfibrils and ribbons, as well as the essential role of water at the different structural levels. In addition, ultra-small angle neutron scattering (USANS) experiments are presented as a promising method to characterise the structure of cellulose in thelongitudinal direction, providing information on the microfibril length andribbon twisting periodicity.