Symmetrically and Asymmetrically Functionalized Cellulose Nanocrystals

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Cellulose microfibrils contain reducing and non-reducing ends as a result of their biosynthetic pathway. Depending on the chemical treatment of the cellulose fibers, different cellulose polymorphs can be obtained. Mercerisation consists of treating native cellulose-I fibers with a concentrated NaOH solution, which transforms the parallel cellulose chains into an anti-parallel, thermodynamically more stable cellulose-II crystal structure. When the cellulose fibers are subjected to sulphuric acid hydrolysis, the amorphous regions of the fibers degrade and the crystalline parts remain, which are known as cellulose nanocrystals (CNCs) **(Figure 1)**.[1] The obtained CNCs are negatively charged and thus form stable aqueous dispersions. Different polymorphs yield their respective asymmetric cellulose I nanocrystals (CNC-I) and symmetric cellulose II nanocrystals (CNC-II).

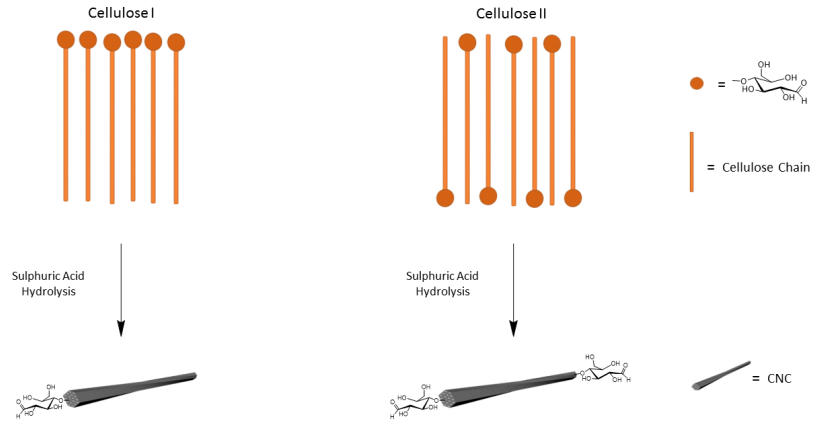

Figure 1: Hydrolysis of cellulose I and cellulose II towards asymmetric and symmetric cellulose nanocrystals.

Here, selective reducing end-group modification of these CNC polymorphs was investigated in order to attach atom transfer radical polymerization (ATRP) initiators with the ultimate objective of grafting hydrophilic polymers from the CNC reducing ends.[2] Quantification of the reducing end groups and subsequent reaction yields were obtained using colorimetric methods.[3] Finally, the self-assembly of asymmetric and symmetric CNC hybrids into lyotropic liquid crystal (LC) phases was investigated. The LC phases of such nanohybrids are expected to be more robust and easily controlled for templating chiral materials.

[1] Y. Habibi, L. Lucia, O. Rojas, Chemical Reviews, **2010,** 110 (6), 3479–3500
[2] J.O: Zoppe, A. Dupire, T. Lachat, P. Lemal, L. Rodriguez-Lorenzo, A. Petri-Fink, C. Weder, H. Klok. ACS Macro Letters, **2017,** 6, 892−897

[3] B. Risteen, G. Delepierre, M. Srinivasarao, C. Weder, P. Russo, E. Reichmanis, J. Zoppe, Small, **2018,** *accepted*