## Polymer Nanocomposites with Cellulose Nanocrystals made by Coprecipitation

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The reinforcement of industrially produced polymers by way of incorporating micro- or nanosized fillers is of great interest in material science. Cellulose nanocrystals (CNCs) are rigid, strong, and toxicologically benign nanofibers and have attracted significant interest in this context, due to their facile extraction from renewable bio-sources [1,2]. The commercial production of CNCs has recently been started and CNCs have great potential applications such in as paper reinforcement, manufacturing, composite or rheology modifier. However, one main challenge of using CNCs as reinforcing filler in polymer nanocomposites is to achieve a high level of dispersion in the polymer matrix using established processing techniques [3]. For many industrially produced polymers the direct melt-mixing with CNCs is not satisfying, as it results in composites with large-scale CNC aggregates and limited mechanical properties. For some of these polymers, a homogeneous dispersion of CNCs in the polymer matrix can be achieved by solution casting from organic solvents, but important commodity polymers (e.g. polyolefins such as polyethylene and polypropylene) cannot be solution-cast with CNCs due to their large polarity difference. We report here on two different approaches towards scalable processing of polymer/CNC nanocomposites. In a first approach, we functionalized CNCs with a 2ureido-4[1H]pyrimidinone (UPy) motif that can serve as a "universal" compatibilizer for CNCs [4]. The motif allows the dispersion of CNCs in both polar (DMF, DMSO) and non-polar (toluene, THF) solvent and thus permits their homogenous dispersion in a great variety of polymers such as polyethylene or natural rubber via solution casting. As an alternative to solution casting, which features disadvantages associated with the drying process, we explored a co-precipitation method, in which polymer/CNC mixtures in a good common solvent were precipitated into water. This process was designed to kinetically trap well-dispersed polymer/CNC mixtures and serves to remove the

organic solvent. Indeed, polymer/CNC mixtures thus precipitated form solid beads (Figure 1a), which could be dried and melt-processed using conventional techniques such as injection molding (Figure 1b). Using this process, we prepared nanocomposites of polyurethane with up to 30% of CNCs. A comparison of the mechanical properties of these materials, established by dynamic mechanical analysis and tensile testing, with those of reference materials made by solution casting, revealed virtually identical characteristics.

## References

[1] Eichhorn S. et al., J. Mater. Sci., 45 (2010) 1.

[2] Endes, C.; Camarero-Espinosa, S.; Mueller, S.; Foster, E.J.; Petri-Fink, A.; Rothen-Rutishauser, B.;

Weder, C.; Clift, M. J. D., Journal of Nanobiotechnology, 14 (2016) 78.

[3] Mariano, M.; Kissi, N. K.; Dufresne, A., J. Polym.Sci. Part B Polym. Phys., 52 (2014) 791.

[4] Natterodt, J. C.; Sapkota, J.; Foster, E. J.; Weder,C. Biomacromolecules, 18 (2017) 517.

## Figures



**Figure 1.** (A) Optical image of polyurethane/CNC beads (10% w/w CNCs) after precipitation from DMSO into water. (B) Photograph of a bar of this nanocomposite made by injection-molding.