Magnetic Alignment of Cellulose Nanocrystals in Suspension and in Hydrogels: Kinetics and Order Parameters

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Cellulose nanocrystals (CNCs) align in suspension when subjected to external forces, such as electric/magnetic fields and shear stress. The ability to orient CNCs is significant for a number of applications including the production of composite materials with enhanced mechanical performance, control of CNC optical properties in liquid and dried states, as model surfaces to study cellulose interactions, and as templates and scaffolds such as those used in tissue engineering. Above a critical concentration in suspension, CNCs self-assemble into a chiral nematic liquid crystal phase and it has been well documented that CNCs align perpendicular to a magnetic field. However, the cooperative nature of this assembly, the effect of magnetic field strength and the associated alignment kinetics have been largely uninvestigated.

This presentation will describe the use of small angle x-ray scattering (SAXS) to capture the real time ordering of CNCs in relatively weak magnetic fields (up to 1.2 T) in both aqueous suspension and in an injectable PEG-based hydrogel. This is the first experimental evidence of significant CNC alignment achieved in such a low magnetic field over short time periods. For CNC suspensions above the critical concentration, the time evolution of alignment was well described by a sigmoidal equation (i.e., cooperative ordering) with initial ordering related to the magnetic field strength. After 129 minutes at 1.2 T, nearly perfect anti-alignment (S=-0.499) of CNCs was achieved and a uniform chiral nematic texture across the entire sample was observed.[1]

In the injectable hydrogel matrix, the CNC concentration was orders of magnitude below the critical concentration but magnetic alignment was observed nonetheless. An order parameter of only S=-0.1 was achieved (almost instantly) and was "locked" into the quickly gelling system.[2] This demonstrates that alignment of very low concentrations of CNCs can be obtained if the surrounding media does not allow for fast relaxation, and indicates that the formation of magnetically ordered CNC materials is more readily accessible than previously thought.

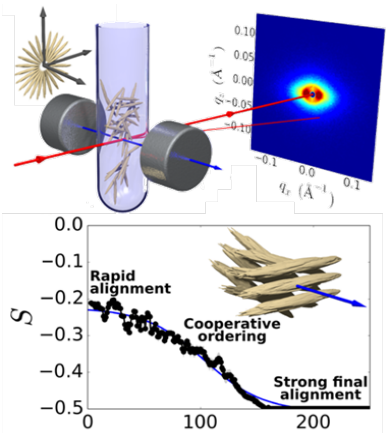

Figure 1. Experimental SAXS setup and evolution of order parameter over time showing cooperative ordering of a CNC chiral nematic suspension.

References:

[1] K. J. De France, K. G. Yager, T. Hoare, E. D. Cranston, *Langmuir* **2016**, *32*, 7564.

[2] K. J. De France, K. G. Yager, T. Hoare, E. D. Cranston, *Submitted to Advanced Functional Materials.*