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Multi-scale investigation of the effect of photocurable polyethylene glycol diacrylate on the self-assembly of cellulose nanocrystals (CNCs)

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Adding photocurable polymers to aqueous suspensions of cellulose nanocrystals (CNCs) preserves their structural organization after processing, as recently demonstrated for nanocomposites with gradient chiral nematic structure produced by membrane ultrafiltration and UV-curing [1]. Nevertheless,understanding the physiochemical interactions between the nonionic polymer (PEGDA) and the CNCs is necessary to rationalise their structuring under external stimuli. In this work, the effect of increasing amounts of PEGDA (0.7 kDa) on the organisation of CNCs has been investigated with a multi-scale approach, including small angle X-Ray and Light scattering. The results showed a peculiar non-monotonic trend: at low PEGDA/CNC mass ratios (RPC), the polymer adsorbed on the surface of CNCs, blurring their particle morphology and weakening their chiral strength (resulting in a looser cholesteric pitch). Further amounts (RPC=1-1.5) increased the thickness of CNCs, with higher volume fractions and higher chiral strength (smaller pitch). Finally, above RPC=2 (16 wt. %) the amount of PEGDA impacted also the properties of the medium. In fact, a lower dielectric constant increased the electrostatic repulsion between CNCs, while reducing further the pitch period. This complex trend was effectively captured by calculating the twist angle between CNCs, which could be used to describe the behaviour of CNCs suspensions in combination with other additives [2].

- [1] Mandin et al., Carb. Pol. 337 (2024), p. 122162.
- [2] Metilli et al., J. Colloid Interf. Sci. 658 (2025), p. 476-486

Abstract Title

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