

Mechanical properties of magnetically aligned cellulose nanocrystals/polymer nanocomposites

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Cellulose is the most abundant macromolecule on Earth and has gained significant attention in recent years due to its sustainable properties. Cellulose nanocrystals (CNCs), obtained through the acid treatment of natural cellulose (Fig. 1), are being explored as reinforcing fibers for nanocomposites due to their high aspect ratio, crystallinity, and stiffness. The orientation of CNCs within a matrix can create anisotropy in nanocomposites, paving the way for new applications. Earlier studies have demonstrated that when exposed to magnetic fields, the hard magnetization axis of CNCs aligns with the *c*-axis (fiber axis) of cellulose, causing the long axis of the CNCs to align perpendicularly to the applied magnetic field [1]. In this study, we prepared anisotropic CNC nanocomposite films by dispersing CNCs in aqueous polyvinyl alcohol (PVA) and cellulose acetate (CA) / acetone solutions, and then casting them in a static magnetic field. We investigated the relationship between the strength of the magnetic field and the orientation of the CNCs, and we evaluated the mechanical properties of the resulting nanocomposites.

Cellulose was treated with sulfuric acid to prepare a CNC suspension. Polyvinyl alcohol (PVA) was added to the suspension, and then CNC/PVA dispersions were prepared. The carboxyl groups were introduced onto the surface of the CNC and subsequently replaced with tetrabutylammonium groups through an ion exchange [2]. Using this surface-modified CNC, a CNC/cellulose acetate (CA) acetone suspension was prepared. These CNC/polymer suspensions were cast at 35°C under a horizontal magnetic field of 0–7 T to create nanocomposite films. The structure of the nanocomposites was evaluated using X-ray diffraction, and their mechanical properties were assessed through tensile testing.

The X-ray diffractogram of the CNC/PVA nanocomposites indicated that the degree of orientation increased with the strength of the applied magnetic field. Additionally, a linear relationship was observed between the half-width of the azimuthal angle plot of the diffraction peaks and the inverse of the magnetic field strength. The findings also revealed that the cellulose fiber axis was aligned perpendicular to the applied magnetic field. A similar orientation was observed in the CNC/CA nanocomposites, and the magnetic alignment of the CNCs was successfully achieved even in organic solvents. Mechanical tests demonstrated that the addition of CNCs to the nanocomposites improved the elastic modulus, even at random orientations (0 T), thereby confirming the reinforcing effect of the CNCs. Moreover, it was determined that the elastic modulus further increased as the degree of orientation intensified with rising magnetic field strength.

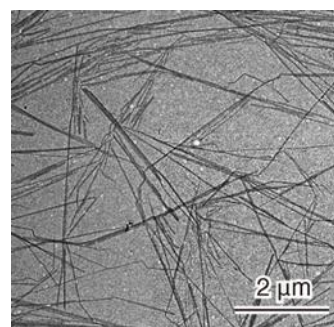


Fig. 1 TEM image of CNC

[1] M. Wada et al., *Cellulose* **28**, 6757–6765 (2021)

[2] M. Shimizu et al., *Biomacromolecules*, **15** (5), 1904–1909 (2014)