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Cellulose Nanocrystals: Towards New Transformations in Functional Polymers and Optoelectronic Materials

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Cellulose nanocrystals (CNCs) are renewable nanomaterials, derived from lignocellulosic biomass and are typically extracted from bleached chemical wood pulps by sulfuric acid hydrolysis to yield a colloidal suspension of spindle-shaped nanocrystals on the order of 5–20 nm in diameter and 100–300 nm in length. Slow evaporation of aqueous CNC suspensions leads to solid semi-transparent and iridescent films that retain the chiral nematic (cholesteric) order of the liquid-crystalline phase above a critical CNC concentration. Owing to their optical and self-assembling properties, CNCs are attracting extensive interest in advanced functional materials including organic semi-conducting materials, templating photonic materials, and a host matrix for a wide range of nonchiral guests, such as luminescent nanoparticles and plasmonic nanoparticles particularly gold nanoparticles (AuNP), which possess unique physical properties including surface plasmon resonance (SPR) resulting from the collective electron charge oscillation after resonant excitation by incident photons. When incorporated into the CNC matrix, gold nanoparticles induce a chiroptical plasmonic response. Such sustainable, chiral plasmonic materials are of particular interest and may extend the potential applications to catalysis, biosensors, surface-enhanced Raman scattering (SERS), optical nanocircuits, energy conversion, and biomedical applications.

We have developed a straightforward, one-pot and scalable approaches to produce chiral cellulose nanocrystal (CNC) films electrophoretically, and chiral plasmonic CNC-gold nanoparticle films electrochemically

electrodeposited films exhibit tunable chiroptical plasmonic characteristics by a simple adjustment of the medium. The simplicity of the method and the sustainability of CNCs can potentially expand their application to other metals and semiconducting nanoparticles for the development of new advanced bio-sourced optoelectronics and functional materials for sensing and detection.

Moreover, a bottom-up approach has been developed for synthesizing flexible, organic, semi-conducting nanocomposite films based on CNCs and polyaniline (PANI) through aqueous emulsion polymerization. Dodecylbenzene sulfonic acid (DBSA) was used as surfactant and dopant for the emulsion. CNCs and DBSA micelle concentrations, their structural organization and alignment with the monomer in the emulsion have a significant effect on the physical and mechanical properties of the resulting nanocomposite films with electrical conductivity reaching as high as $5.29 \times 10^{-1} \text{ S.cm}^{-1}$ which falls in the electrical conductivity range of Germanium ($2.24 \times 10^{-2} \text{ S.cm}^{-1}$) and Silicon ($0.43 \times 10^{-5} \text{ S.cm}^{-1}$). The CNCs-PANI-DBSA nanocomposite films show a maximum tensile stress and strain values of 22 MPa and 0.89 %, respectively and are significantly stronger and more flexible films than those obtained with, for instance, graphene/polyaniline composite paper or graphene paper, where it has been reported in the literature that the tensile strengths were 12.6 and 8.8 MPa, and maximum strains, 0.11 and 0.08 %, respectively.

Our work shows that the mechanical and conductive properties can be tailored to suit the desired end-use application. This development provides promising sustainable organic materials for use in electronic and opto-electronic applications. Our in-depth investigation helps provide strong insights into the structure-property interrelationships of the sustainable, organic semi-conducting films.

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