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## Hybrid Chiral Plasmonic Films via an Electrochemical Approach

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We report green, straightforward, one-pot and scalable approaches to produce chiral cellulose nanocrystal (CNC) films electrophoretically, and chiral plasmonic CNC-gold nanoparticle films electrochemically in the absence of reducing agents. The 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.

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-translucent 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

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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.

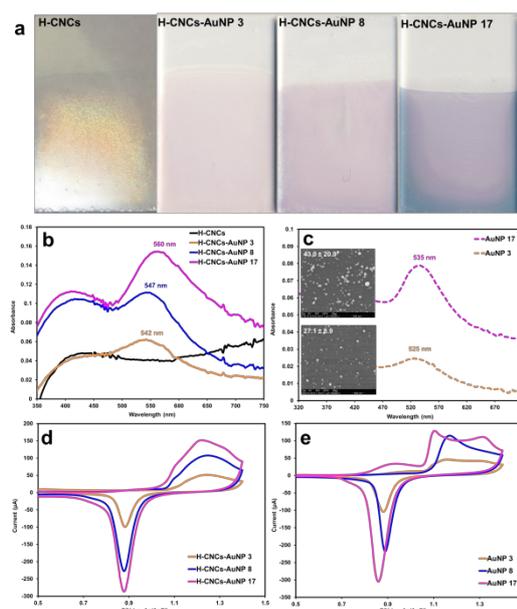


Figure 1. Macro images, optical and electrochemical properties of the electrodeposited AuNP and H-CNCs-AuNP films. a, Pictures of the electrodeposited H-CNCs-AuNP films with varying gold concentration. b and c, UV-vis absorption spectra of the electrodeposited H-CNCs-AuNP films and gold nanoparticles, respectively, with varying gold concentration. The inset images show the corresponding SEM indicating the sizes of electrodeposited AuNP. d and e, Cyclic voltammograms recorded in a 0.1 N H<sub>2</sub>SO<sub>4</sub> solution at 50 mVs<sup>-1</sup> scan rate for 10 scans of the electrodeposited H-CNCs-AuNP films and AuNP, respectively, with varying gold concentration.

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