Hierarchically Structured Grating Films Constructed via Confined Self-Assembly of Cellulose Nanocrystal Colloidal Particles

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Colors generated by continuous and periodic micro/nanoscale structures play an important role in nature. Inspired by those structures, various micro/nano optical devices with diverse morphologies have been developed and made a difference across multiple fields. Metal/semiconductor colloidal particles, polymers, and organic small fluorescent molecules can all serve as the fundamental building blocks for structural optical devices. [1−3] Depending on different optical mechanisms, the resulting colors can be attributed to thin-film interference, diffraction, or scattering. [4] Such optical devices are widely used for waveguides, displays, photoelectric conversion, and optical sensing. [5] In this work, a hierarchic transparent cellulose nanocrystals (CNCs) film that integrates both linear structures and cholesteric structures is formed through evaporation-induced self-assembly (EISA) of renewable CNCs colloidal particles. [6] The asymmetric structural morphology was studied along with the asymmetric optical properties under polarized plane-wave illumination. A wrinkled surface was introduced as the template for utilizing its spatial confinement effect. CNCs colloidal particles undergo EISA within this confined one-dimensional wrinkling surface. During the helical self-assembly of CNCs, interfacial bending occurs in accordance with the template scale, resulting in the successful fabrication of diffraction grating structures with various wavelengths and heights up to approximately ten times the width of a single CNC. The resulting linear gratings exhibit unidirectional reflective grating properties. A clear resolution of ±3 diffraction orders can be observed with a first-order diffraction efficiency of up to 9%. The cut-off wavelength range covers from the visible region (508 nm) to the infrared region (1539 nm). Furthermore, due to a kinetic arrest effect during EISA, the fabricated films remain the left-handed helical layered structure even after interfacial bending and spatially confined drying. The stacked layered structure extends from flat regions into the linear grating regions. These films produced single-channel left-handed optical activity, and their circular dichroism (CD) signal intensity and reflection spectral position can be tuned by adjusting the helical structure of CNCs. Within this framework, additional composite structures (with or without diffraction grating features) can be patterned on the film to produce optical anti-counterfeiting elements that are both visually observable and specifically detectable. This work provides a scalable and cost-effective platform for fabricating optical components using natural renewable colloids and lays a foundation for the development of optical signal technologies based on green and sustainable resources.

Keywords:
Cellulose Nanocrystals, Self-Assembly, Gratings, Wrinkle Template

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