Angle-Independent Cellulosic Photonic Crystals for Smart and Sustainable Colorimetric Sensing

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Cellulose nanocrystals, as well as hydroxypropyl cellulose, can form lyotropic liquid crystals, which can be processed into pigments or glitter products for sustainable coloration. Some stimuli-responsive polymers or nanoparticles are expected to form colorimetric sensors *via* co-assembly with these cellulosic photonic crystals. The co-assembly behavior of CNCs with polymers is determined by the hydrogen bonds and physical adsorption. Thus, adjusting the molecular chain structure, hydrophilicity, and electrostatic interaction of co-assembled polymers can lead to flexible and tunable colorimetric cellulosic sensors. Despite the advantages of cellulose-based amorphous photonic crystal (APC) pigments or glitters as sustainable and visually captivating sensors, there are still problems in efficient preparation and co-assembly conditions. This editorial will provide a brief discussion of the benefits, applications, and challenges of cellulose-based APCs.

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Cellulose-based Optics

Cellulose is a renewable resource, abundant in nature, with excellent biodegradability, low cost, as well as thermal and chemical stability. The nano-units of cellulose and some types of modified cellulose molecular chains have lyotropic liquid crystal behavior to self-assemble into periodic microstructures, exhibiting iridescent structural colors. The representative cellulosic nano-rods are the cellulose nanocrystals (CNCs) known as a type of tactoid. These are capable of self-assembling or co-assembling with other polymers or nanoparticles to form chiral nematic structures. The chiral nematic CNC films obtained via evaporation-induced self-assembly (EISA) are iridescent, with the structural color controlled by the pitch which is determined by the hydrogen bonding and electrostatic interactions between CNCs and co-assembling polymer chains or nanoparticles (Andrew *et al.* 2023). External stimuli such as stress, ionic strength, and humidity can affect the pitch of cellulose nanocrystals, causing changes in the corresponding reflective wavelengths. Meanwhile, the appearance is also affected by the viewing angle. With both the viewing angle and the external stimuli being the factors determining the reflective wavelength of the structural color, the visualization of external stimuli is highly dependent on precise measurement equipment and fixation of viewing angles, which limits the application in sensor, reflective display, optical encryption, and inkless printing. Therefore, weakening the influence of the viewing angle to ensure the dominant dependent of reflection wavelength on the pitch is crucial for the application of cellulose-based optics as pigments or sensing materials.

Amorphous photonic crystals (APCs) are photonic crystals having long-range random and short-range ordered microstructure, and this “defect state” structure enables APCs to have a photonic pseudoband gap that is independent of the viewing angle. Light is uniformly scattered in all directions, resulting in non-iridescent structural colors, which are more suitable as pigments and sensing materials for visual identification of external stimuli. There have been studies reported on the preparation of APCs using cellulose nanocrystals (CNCs) (Droguet *et al.* 2022; Parker *et al.* 2022). Generally, the self-assembled CNC film is heat-treated and then ground into particles with a fixed size range (typically below 25 μm) to prepare non-iridescent glitters. Unlike CNC, which forms a chiral nematic phase through the self-assembly of tactoids in the liquid crystal state, hydroxypropyl cellulose (HPC) is a cellulose derivative that forms a chiral nematic phase through the self-assembly of polymer chains at high concentrations in aqueous solutions (Ming *et al.* 2023). In the high-viscosity system, the self-assembly of HPC chain segments tends to form a local chiral nematic structure on the micron scale, while the material as a whole is isotropic. Therefore, the structural color of the resulting HPC optics is angle-independent. Both CNC and HPC can be used to fabricate angle-independent colorimetric sensors that can quantitatively measure external stimuli without coupling precise angle-fixing devices, which also allows for functional visual effects such as mechanochromism, ionochromism, and hygrochromism.

Applications of Cellulose-based APCs

The non-iridescent color and other excellent properties of APCs, including biocompatibility, sustainability, tunability, wide color range, fade resistance, heat stability, and chemical resistance, have opened up a myriad of innovative applications in diverse fields in environmental monitoring, biomedical sensing, food safety assessment, chemical processing, and gas detection. The inherent properties of cellulose-based APCs, such as flexibility, biocompatibility, and chemical/heat stability, make them suitable for integration into flexible electronics for continuous real-time monitoring and on-demand health diagnosis enabling the detection of various analytes such as proteins, enzymes, viruses, and bacteria. These sensors can be tailored to detect specific chemicals, gases, or volatile compounds for the measurement of temperature, humidity, gas composition, and air quality, which contributes to the research in climate analysis, indoor environmental control, and energy management.

Despite these advantages of cellulose-based APCs pigments, there are still some shortcomings when they are co-assembled with stimuli-responsive materials as sustainable and visually captivating sensors. The main problem lies in the complicated preparation process and high cost. For APCs prepared from CNC, thermal treatment is required before grinding the CNC self-assembled membrane to avoid damage to the ordered structure in the particles. This process will also greatly reduce the swelling and stimulus-response of APCs, causing excess stability against external stimuli or difficulty in incorporating stimuli-responsive materials. Therefore, how to regulate the interaction of CNC or HPC to effectively preserve the short-range ordered structure as well as maintain a certain stimulus responsiveness is the challenge of this research direction.

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