Chapter 14

ThermoChroma-smart Colorimetric Sensors as Cold Chain Indicators

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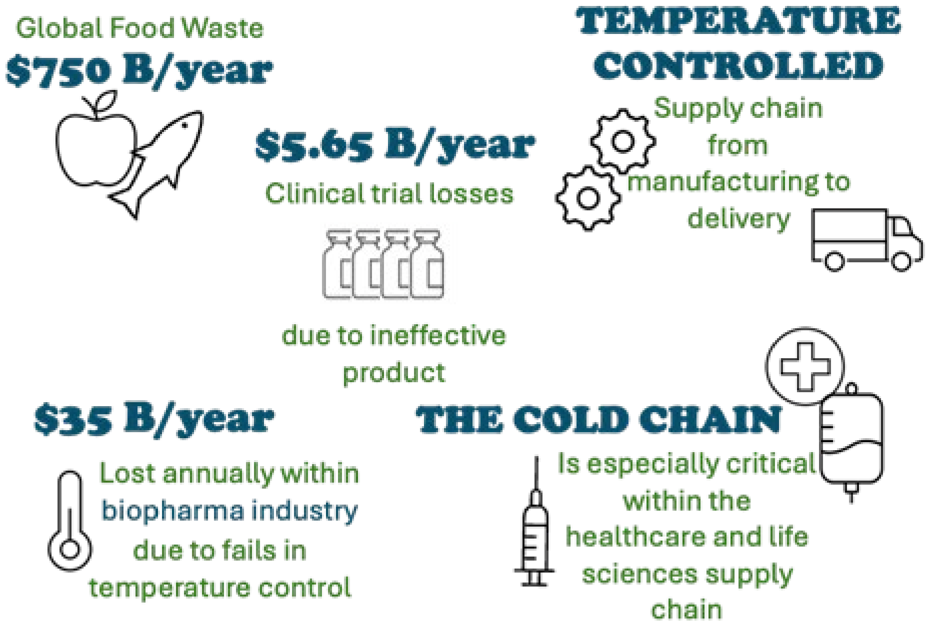
Abstract

Cold chain failures of pharmaceutics result in substantial economic and clinical losses. Conventional monitoring systems, like selected batch analysis, in-situ electronic sensors, and time-temperature indicators, provide reliable data but remain costly, infrastructure-dependent, and impractical for per-item implementation. This paper introduces a photonic colorimetric sensor design that provides a simple, visual readout of cold chain integrity. The photonic system exploits the thermoresponsive cholesteric phase of cellulose derivatives, which undergoes distinct and reversible colour shifts from 0°C to room temperature. Preliminary prototypes demonstrate integration into packaging formats, with scalability through solution processing and printing routes and colour response is just within minutes. With a scope to investigate temperatures below 0°C, this approach offers a sustainable, low-cost alternative to electronic monitoring systems and establishes photonic architectures derived from natural resources as promising candidates for cold chain monitoring.

Keywords: *Colorimetric Sensors, Sustainable Development, Structural Colour, Cellulose photonics, Cold chain detection*



Infographic illustrating key industries dependent on cold chain integrity

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Introduction

Temperature-controlled logistics, commonly referred to as cold chains, are essential for preserving the efficacy and safety of temperature-sensitive products across diverse sectors, including, life sciences and medical devices as well as perishable and frozen food and industrial chemicals, and reagents, as shown in Figure **1** below[1].

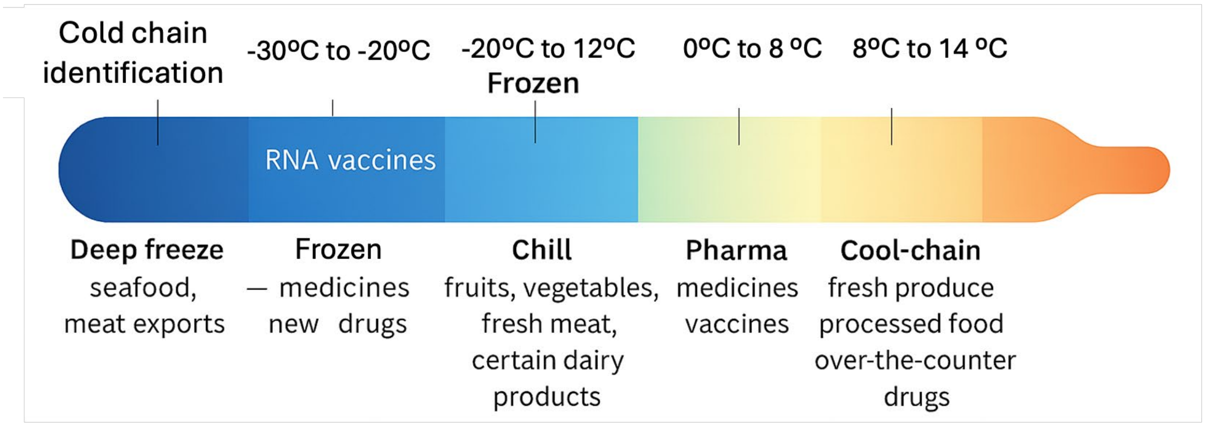

Figure 1: Schematic illustration of critical cold storage zones, highlighting temperature ranges required for the safe handling of temperature-sensitive goods across the cold chain.

One of the most important criteria in terms of sustainability is the shelf-life of a pharmaceutical modality. Temperature variations in transit, warehouse storage management conditions and delays as well as the breakdowns that might constitute moderate inconveniences in a typical supply chain can have devastating impacts for pharmaceutical cold chains crucial for the development of the new drug modalities such as gene and cell therapy relevant biological components, m-RNA drugs, and complex biochemicals. A single anomaly in the cold chain can render entire batches of products unusable, contributing to the alarming statistics mentioned above. The pharmaceutical industry faces staggering losses, with failures in the cold chain alone costing an estimated 29 billion GBP annually[2]. To mitigate these risks and reduce the environmental footprint of the chemical processes involved in pharmaceuticals, there is an urgent need for innovative tools that offer simple and cost-effective monitoring of the cold chain for individual components. Such intervention is expected to make a great impact by reducing wastage and decreasing the use of raw materials.

Existing cold chain monitoring technologies ranging from electronic data loggers and radio frequency identification (RFID)-enabled sensors to time-temperature integrators provide accurate tracking but are limited by cost, infrastructure requirements, and restricted suitability for dose-level deployment. These constraints leave a technological gap for simple, scalable indicators that can be integrated into individual packaging and interpreted without specialist equipment.

Monitoring visual colour changes is a widely used strategy in biological signalling systems. This allows direct interpretable signalling without supplemental tools. To achieve such monitoring capacity,

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[1] Tempk, "What Is the Cold Chain? 2025 Temperature-controlled Guide," November 14, 2025, https://www.tempcontrolpack.com/knowledge/what-is-the-cold-chain-2025-temperaturecontrolled-guide/

[2] Stefan Lutzmayer, Elina Osoianu, and Tom Rosenfield. Tip of the Iceberg: Economic and Environmental Impact of the Vaccine Cold Chain. IQVIA White Paper. 2025.

pigment-based indicators have been explored, but they often suffer from poor stability, leaching, and limited adaptability across different temperature regimes. By contrast, structurally coloured photonic materials provide robust optical responses arising from periodic nanoscale order, and their spectral properties can be engineered to respond to environmental stimuli.

This paper introduces the design of a photonic colorimetric sensor utilising the cholesteric liquid crystalline phase of cellulose with thermoresponsive block copolymers. Unlike pigments, structural colour arises from the periodic organisation of matter at the nanoscale, producing vivid and highly stable optical responses[3] (4). Hydroxypropyl cellulose (HPC), in particular, forms cholesteric liquid crystalline phases that display strong colour changes in response to temperature and humidity[4]-[5][6][7] (5-8). When combined with block copolymer (BCP) hydrogels, the resulting hybrid material enables tuning of the thermal response, improving sensitivity in narrow temperature ranges and enhancing the processability of cellulose into scalable formats. This system presents tuneable colour changes in the critical 0°C to 8°C range and up to room temperature relevant to pharmaceutical cold chains. The preliminary devices demonstrate rapid, visible, and reversible (or lockable) colour changes and compatibility with scalable processing methods such as thin-film deposition and printing. These results establish polysaccharide-based photonic systems as a promising platform for sustainable and low-cost cold chain monitoring. The hybrid HPC-BCP system responds rapidly, within minutes, and can be engineered for reversible or irreversible behaviour depending on the application. Importantly, the system operates without breaching the package seal, thereby ensuring integrity during storage and transport[8]

Results and Discussion

Inspired by the colour changing biological creatures such as beetles or cuttlefish skin, our photonic sensing design accounts for producing a multi-layered colorimetric smart indicator directly monitoring the cold chain. We coined this design as ThermoChroma, which is a photonic sensor (producing colour through light interference rather than pigments) that translates temperature fluctuations in the pharmaceutical cold chain into a clear colour signal visible to the naked eye. Such response stems from materials intrinsic temperature sensitivity. Unlike pigment-based indicators, which function like litmus

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[3] Ahu Gümrah Dumanli and Thierry Savin, "Recent Advances in the Biomimicry of Structural Colours," *Chemical Society Reviews* 45, no. 24 (2016): 6698–724, https://doi.org/10.1039/c6cs00129g

[4] Tadeusz Balcerowski et al., "Hierarchical Organization of Structurally Colored Cholesteric Phases of Cellulose via 3D Printing," *Small* 19, no. 8 (2022): e2205506, https://doi.org/10.1002/smll.202205506

[5] Hongning Ren, Tadeusz Balcerowski, and Ahu Gümrah Dumanli, "Achieving a Full Color Palette With Thickness, Temperature, and Humidity in Cholesteric Hydroxypropyl Cellulose," *Frontiers in Photonics* 4 (2023), https://doi.org/10.3389/fphot.2023.1134807

[6] Gen Kamita et al., "Biocompatible and Sustainable Optical Strain Sensors for Large-Area Applications," *Advanced Optical Materials* 4, no. 12 (2016): 1950–54, https://doi.org/10.1002/adom.201600451

[7] Gen Kamita, Silvia Vignolini, and Ahu Gümrah Dumanli, "Edible Cellulose-based Colorimetric Timer," *Nanoscale Horizons* 8, no. 7 (2023): 887–91, https://doi.org/10.1039/d3nh00006k

[8] Dumanli and Savin, "Recent Advances in the Biomimicry of Structural Colours," January 1, 2016.

paper and often lack stability, ThermoChroma relies on structural colour generated by periodic nanoscale order, delivering robust, tuneable, and sustainable performance.

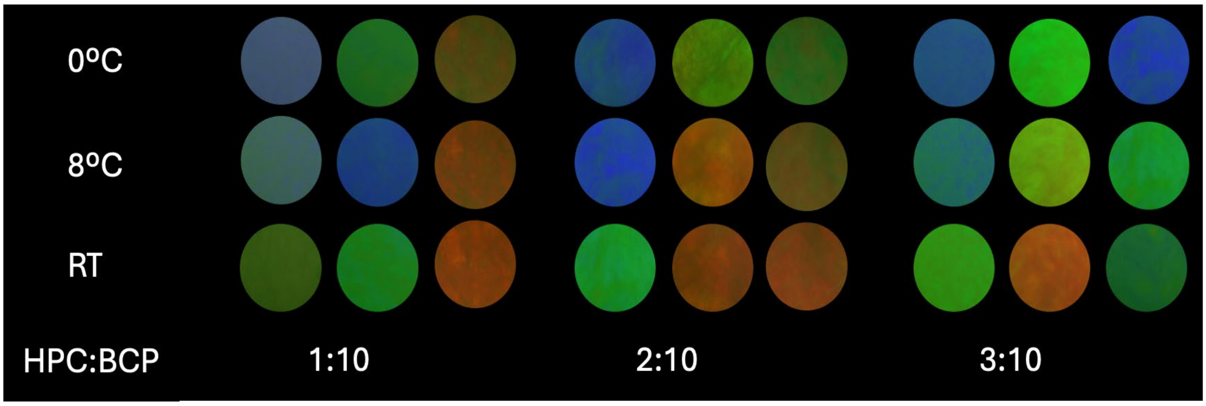

Figure 2: Digital camera images of the ThermoChroma's HCP/BCP sensors, and their colour response to cold simulation test where the temperature was varied between room temperature (RT) and 0°C, showing a distinct and reversible shift in their macroscopic appearance between 0°C and RT.

At the core of the device is HPC, a renewable polysaccharide that naturally forms cholesteric liquid crystalline phases. These phases produce vivid, angle-dependent colours that shift in response to external stimuli, in this case temperature. To enhance the sensitivity and tunability of this platform, we incorporated a block copolymer (BCP) hydrogel that modulates the thermal response. Specifically, temperature responsive block copolymerBCP, poly(glycerol monomethacrylate)-*b*-poly(2-hydroxypropyl methacrylate) (PGMA-*b*-PHPMA) was used in this work to acts as a responsive scaffold, adjusting the phase transition properties of the HPC system and amplifying subtle temperature changes into sharper optical shifts[9] (9). Figure **2** demonstrates HPC-based sensors with distinct and reversible transition in colour between 0°C and room temperature, marking the critical threshold for pharmaceutical cold chain integrity (6).

Photonic HPC-BCP constructs in the film form were prepared by casting HPC-BCP solutions with varying concentrations of HPC in water from 55% to 64% with a HPC to BCP ratio 10:1, 10:2, and 10:3. The film thickness of the colorimetric indicators were adjusted to 0.5 mm to 1 mm. The films were sealed in semipermeable membranes and the HPC-BCP composites were kept as hydrogels to allow dynamic colour responses. The HPC-BCP solutions were self-assembled to form a mesophase with cholesteric ordering (Figure **3**). The reflection wavelength of the cholesteric hybrid system can be described by the modified Bragg equation[10],[11],[12]:

$$\lambda = n\, p\, cos\, cos\, \theta \qquad \text{(Equation 1)}$$

and

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[9] Qi Yue et al., "Multifunctional Self-Assembled Block Copolymer/Iron Oxide Nanocomposite Hydrogels Formed from Wormlike Micelles," *ACS Applied Materials & Interfaces* 16, no. 16 (2024): 21197–209, https://doi.org/10.1021/acsami.4c03007

[10] Kamita, Vignolini, and Dumanli, "Edible Cellulose-Based Colorimetric Timer," January 1, 2023.

[11] Hl. De Vries, "Rotatory Power and Other Optical Properties of Certain Liquid Crystals," *Acta Crystallographica* 4, no. 3 (1951): 219–26, https://doi.org/10.1107/s0365110x51000751

[12] Ahu Gümrah Dumanli et al., "Digital Color in Cellulose Nanocrystal Films," *ACS Applied Materials & Interfaces* 6, no. 15 (July 9, 2014): 12302–6, https://doi.org/10.1021/am501995e

$$n_{av} = n_{HPC-BCP}\, \phi_{HPC-BCP} + n_{Water}\phi_{Water} \qquad \text{(Equation 2)}$$

where n is the average refractive index, and p is the cholesteric pitch (where the consecutive pseudo layers undergo a 360° rotation of the cholesteric structure), φ is the volume fraction and θ is the angle of incidence of light with respect to the normal. The refractive index of the hybrid system is consideredas $n_{HPC\text{-}BCP} = 1.5$.

To demonstrate potential integration into pharmaceutical contexts, our proof-of-concept prototype set up for the ThermoChroma films were sealed to emulate packaging and subjected to controlled cold-chain cycling between 0°C and 20°C. Distinct, visible colour transitions were observed under ambient lighting, readily captured with a smartphone camera (Figure **2**). This preliminary validation highlights the applicability of ThermoChroma sensors in real-world logistics environments. Microstructural analysis of the films using cross sectional imaging with scanning electron microscope (SEM) revealed that near perfect helicoidal ordering characteristic of cholesteric photonic systems (Figure 3a), closely resembling the layered cuticle of scarab beetles (Figure **3**b). This ordered architecture underpins the distinct spectral shift observed in reflectance measurements, with λ_{max} moving from orange (~600 nm) towards greenish blue (~450 nm) across the 0°C to 20°C range (Figure **3**c). The response can be engineered to be reversible, resetting with temperature, or irreversible, locking into a distinct colour once a threshold is breached. The latter provides a powerful tool for quality assurance: a single glance can confirm whether a medicine has been compromised.

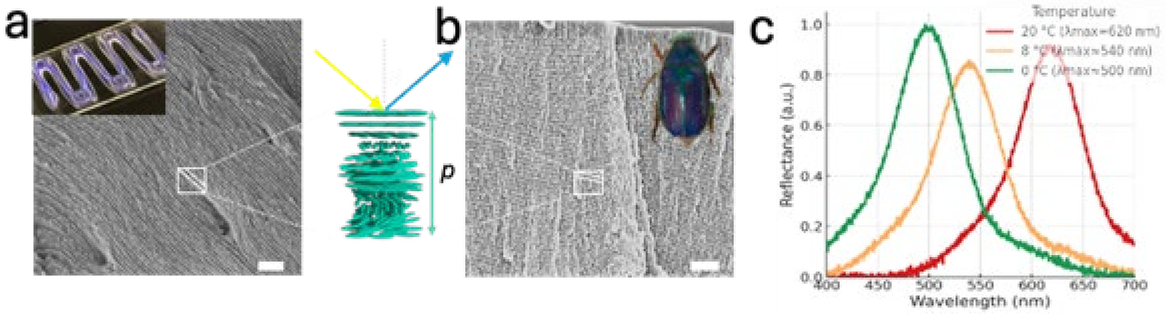

Figure 3: SEM micrographs of the a. HPC/BCP films reveal the cholesteric pitch (p) and periodic ordering responsible for structural colour and b. The cholesteric structure found in Anoplognathus smaragdinus beetle cuticle.Scale bars 1 micron. c. The reflectance analysis of the ThermoChroma sensor's colour response, showing a distinct and reversible shift in hue between 0°C and room temperature.

Beyond single-point readouts, the ThermoChroma platforms can be patterned into films, codes, or tags, using scalable methods such as 3D printing or roll-to-roll coating. This opens opportunities for integration into pharmaceutical packaging, food logistics, and even QR-like codes for digital verification. The technology provides not just a functional sensor but a pathway towards sustainable, low-cost, and item-level monitoring of cold chains, with the potential to substantially reduce waste and carbon footprints across supply networks.

Conclusion

ThermoChroma prototypes address key limitations in current cold chain monitoring by combining the cholesteric liquid crystalline HPC hybrids with self-assembled block copolymer BCP hydrogels to provide visible signal of temperature changes. The existing prototypes demonstrate optical responses across the critical 0–8 °C range and up to room temperature, directly relevant to pharmaceutical storage.

The devices exhibit fast, distinct, and reversible colour changes that can also be engineered into irreversible “lock-in” responses once a threshold is crossed, ensuring permanent visual confirmation of cold-chain breaches. Beyond simple visible readouts, ThermoChroma also enables the creation of advanced security features, such as 3D-printed codes or patterned indicators, adding a new layer of traceability and protection against tampering or counterfeiting. Importantly, the platform can be extended to lower-temperature regimes, including the −20 °C storage required for next-generation vaccines, complex pharmaceuticals, and mRNA therapeutics.

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