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Review

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Fabrication strategies and microscale sensing functionalities of mechanochromic colloidal photonic crystals for underwater applications

Feng-Lian Qi[1], Qun Li[3], Cai-Feng Ding[1], Jia-Bin Wu[2]

[1]Key Laboratory of Optic-electric Sensing and Analytical Chemistry for Life Science, Ministry of Education; College of Chemistry and Molecular Engineering, Qingdao University of Science and Technology, Qingdao 266042, Shandong, China.
[2]Department of Chemistry, Tsinghua University, Beijing 100084, China.
[3]CAS Key Laboratory for Nanosystem and Hierarchy Fabrication, CAS Center for Excellence in Nanoscience, National Center for Nanoscience and Technology, Beijing 100190, China.

Correspondence to: Dr. Jia-Bin Wu, Department of Chemistry, Tsinghua University, No.1, Chengfu Road, Haidian District, Beijing 100084, China, E-mail: jiabin7295@163.com

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Abstract

Mechanochromic colloidal photonic crystals (PCs), which typically integrate a self-assembled PC array with a highly elastic medium, exhibit the ability to reversibly respond to external mechanical stimuli by altering the periodicity of PC structures. Nowadays, leveraging visible indications and optical signals for mechanical forces, mechanochromic colloidal PCs have been widely used in reflecting body motion, health monitoring, and communications in daily life. However, despite their extensive applications on land, it is vital to explore the potential of mechanochromic sensing applications underwater, where message transmission mainly relies on body gestures and motions. This review comprehensively examines recent advancements in mechanochromic colloidal PCs and their underwater applications. The first part introduces the response mechanism of mechanochromic colloidal PCs, emphasizing the main principles that facilitate sensing on the microscale. The second part describes the fabrication strategies for constructing these PCs, demonstrating various approaches to establish optical sensors with specific functionalities. The final section discusses the confronted challenges and summarizes the potential opportunities in developing mechanochromic colloidal PCs for underwater sensing applications.

Keywords: Mechanochromic, colloidal photonic crystal, underwater sensor, flexible elastomer, strain sensor

INTRODUCTION

Photonic crystals (PCs) represent a type of functional material characterized by a periodic arrangement of the refractive index. In recent years, the creation of readable structural colors and the emergence of optical signals through the interaction with light have propelled PCs into the forefront of research for developing smart displays[1,2] and sensors[3,4]. Inspired by natural PCs, artificial PCs have undergone rapid development. Currently, the preparation of artificial PCs primarily employs two approaches. The first is the "top-down" method, derived from traditional lithography technology, which establishes periodic structures through lithography or micro-printing techniques[5]. The PC structures designed by this method are advantageous in few defects, making them applicable in a broad spectrum of optical fields, including optical fibers, optical computers, and chips. Another common method is named the "bottom-up" method, typically involving the organized stacking of colloids to create periodic structures[6]. These constructed PCs, namely colloidal PCs, have gained prominence due to their effective cost, ease of functionality, and simple large-scale production. Serving as long-range ordered micro-superstructures formed through the colloidal self-assembly of colloids with monodispersed nanoparticles as building blocks, colloidal PCs allow the tunable self-assembly of nanoparticles driven by interactive forces[7,8]. Consequently, they play indispensable roles in the advancement of diverse fields[9-13].

By using colloidal PCs as optical templates, it is noted that incorporating responsive media into colloidal PCs enhances their applicability in sensing areas, such as spanning display[14], temperature[15-17], humidity, electromagnetic fields[18-21], and various target molecules related to human health[22-26] and public safety[27], *etc*. Notably, the ability to visually reflect gestures and body motions through the mechanical deformations of flexibly mechanochromic sensors[28-31] distinguishes mechanochromic colloidal PCs as an efficient approach of visually conveying body messages[32]. Therefore, mechanochromic colloidal PCs stand out as a promising sensor for achieving health diagnosis and communication through body motions.

Compared with sensing on land[33-37], underwater sensing has recently attracted much attention due to the growing number of individuals participating in underwater explorations and activities[38-40]. Thus, there is an increasing demand to advance underwater sensing capabilities, crucial for ensuring public safety and communication in water-filling environments[41,42]. Challenges persist in extending sensors to underwater areas, stemming from the conflict between sensing reliance on electronic energy and the potential occurrence of current leakage in water-filled environments. To address the drawback of current leakage, water-resistant conductive gels have been developed underwater with electrical signals[43]. However, electrical sensors still provide indirect readout ways of recognizing gestures and body motions under water, offering delayed feedback on messages, and posing potential challenges in extreme conditions, such as underwater rescues. Notably, mechanochromic colloidal PCs serve as optical sensors with visually tunable structural color under varied stress conditions, circumventing the issues associated with electrical leakage and providing instantly readable symbols. This characteristic not only prevents undesirable side effects but also ensures prompt feedback, especially crucial in critical situations such as underwater rescues.

In this review, we provide a summary of the latest research focusing on the strategies employed in establishing mechanochromic colloidal PCs and their developments in underwater sensing. Different from previously published reviews related to mechanochromic sensors that concentrate on the development of fabrication methods[11] or diverse sensing applications[9], here we focus on bridging the relationship between the mechanochromic colloidal PCs and the promising underwater sensing directions and exploring the crucial factors that could promote the rapid development in this direction. First, we will introduce the response mechanism of mechanochromic colloidal PCs, elucidating their capacity for modulating optical signals. Second, we will provide an in-depth exploration of the designed strategies for mechanochromic

colloidal PCs, categorizing them based on the types of responsive materials employed. Third, we will highlight the recent developments in the application of mechanochromic colloidal PCs for underwater sensing [Scheme 1]. Finally, we will address the challenges encountered and explore potential opportunities in the realm of mechanochromic colloidal PCs and their underwater sensing applications.

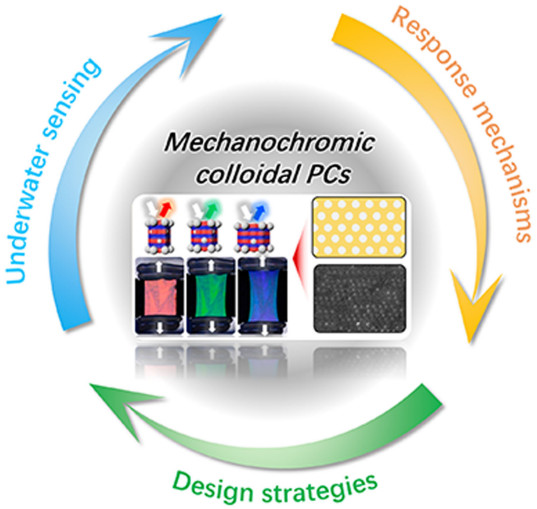
RESPONSE MECHANISM OF MECHANOCHROMIC COLLOIDAL PCS

Microstructure classification of colloidal PCs

Colloidal PCs are ordered arrays self-assembled by mono-dispersed nanoparticles at sub-micro scales[44-46]. Based on the periodicity of colloidal PCs in spatial directions, they can be divided into colloidal one- (1D), two- (2D) and three-dimensional (3D) PCs. Figure 1A-C illustrates three typical examples of PCs that exist in nature based on the repeated number of periodicities on the refractive index. For instance, the wings of the *Morpho Menelaus* butterfly displayed bright blue in normal light owing to their stacked layer microstructures, which could be regarded as 1D-PC due to the periodicity on the normal direction of the layer plane[47]. As to the natural 2D-PC, the *Mosquito C. pipiens* is representative since the spherical-like units assembled to ordered configuration on one plane, which thus could be found two periodicities on this plane[48]. For the natural 3D-PC, the building blocks in the body of weevil *L. augustus* prefer to form stacked microstructure on the whole spatial range, which thus formed three periodicities[49]. Inspired by the assembled configurations from natural PCs, Figure 1D-F illustrates three representative ways for the construction of artificial colloidal PCs with the assembled unit cells considered as spherical nanoparticles.

Colloidal 1D-PCs

As illustrated in Figure 1D, the representative single chain-like superstructure, where the periodic structure exists only in one direction, can be defined as colloidal 1D-PC. Due to its fast preparation speed and easy modulating optical signal, the magnetic field-induced self-assembly approach has become prevalent in the design of 1D-PCs in recent years. For instance, Ge *et al.* pioneered the construction of magnetically responsive colloidal 1D-PCs with single chain-like structures in aqueous solutions by controlling the spherical-like Fe_3O_4[50] and Fe_3O_4@SiO_2[51] colloidal nanoclusters at low concentrations, and achieved the structural color change through the tuning of external parallel magnetic fields. Moreover, the incorporation of anisotropic building blocks provides a new concept for constructing 1D chiral PCs. For instance, Li *et al.*[52] tried the magnetic Fe@SiO_2 cubes as the assembled units to construct a new type of 1D-PCs with edge-to-edge assembled chains, exhibiting optical signals that display intense dependence on observation angles. Moreover, using magnetoplasmonic Ag@Fe_3O_4 nanoparticles as building blocks, Jeong *et al.*[53] constructed a helical superstructure that could achieve the dynamic modulation of the chiroptical property under dynamic magnetic fields. Except for magnetic force, the spontaneous assembly of cellulose nanocrystals (CNCs) also enriched the establishment of 1D-PC with cholesteric ordering[54]. Serving as the most abundant biopolymer that is accessible from nature, the bulk cellulose is easy to get CNCs with diameters of ~5-15 nm and lengths of ~100-300 nm by the sulfuric acid-catalyzed hydrolysis[55]. It is noteworthy that CNCs are capable of organizing into liquid crystals in water with chiral nematic configurations, which further form photonic film with layered structures where the CNCs are oriented orderly in each layer and form a helicoidal chiral nematic order with a repeated pitch. Interestingly, Lv *et al.*[56] constructed biomimetic chiral 1D-PCs through the orientational assembly of inorganic nanowires by Langmuir-Schaefer deposition approach, which opened a brand-new way for the establishment of artificial chiral PCs with different optical outputs under circularly polymerized light. Further, with the integration of semiconductor quantum dots with nanowire-based chiral 1D-PC, they also developed a novel chiral semiconductor nanostructures for the modulation of circularly polarized luminescence with high intensities, which thus showed the great promise for designing high-performance circularly polarized luminescence-based devices[57].



Scheme 1. Overview of the topics covered in this review.

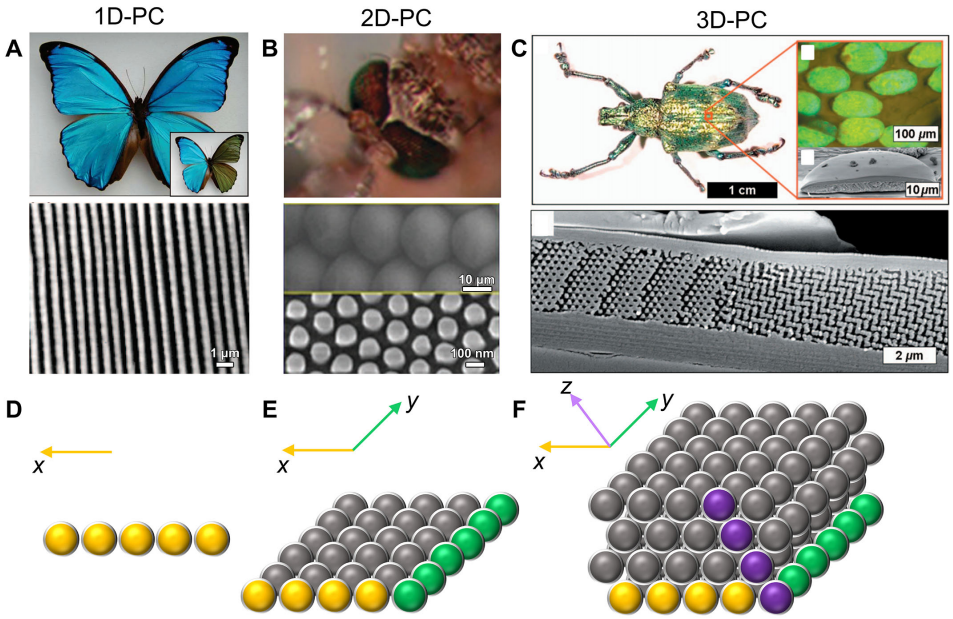


Figure 1. Typical examples of natural PCs with different periodicity: (A) natural 1D-PC formed on the wings of *Morpho Menelaus* butterfly[47]; (B) natural 2D-PC formed on the *Mosquito C. pipiens* moth[48]; (C) natural 3D-PC formed on the body on weevil *L. augustus*[49]; (D-F) diagram illustrating the construction of corresponding colloidal PCs as the building blocks are spheres. PCs: Photonic crystals; 2D: Two-dimensional; 3D: Three-dimensional.

Colloidal 2D-PCs

When the nanoscale building blocks assemble into single layer-like superstructures with periodic repeating units in two different directions, they are considered as colloidal 2D-PCs. Normally, the construction of colloidal 2D-PCs involves prevalent approaches such as Langmuir-Blodgett assembly and tip-diversion assembly. The Langmuir-Blodgett self-assembly method refers to dispersing hydrophobic colloidal spheres at the water/air interface, forming closely packed single-layer PCs by the physical compression of the interface volume[58]. Moreover, leveraging the surface tension difference between the dispersive solvent and

water, Zhang *et al.* originally used the tip-diversion assembly method to uniformly spread spherical nanoparticles onto the water-air surface. This approach facilitates the spontaneous assembly of nanoparticles, constructing densely packed 2D-PC arrays[59]. Besides, when the 2D colloidal PC array integrates with responsive hydrogels, a 2D-PC hydrogel sensor is further established, showcasing both discernible color changes and quantifiable alterations on the Debye diffraction ring under the illumination of monochromic laser. This illustrates the potential applications of 2D-PC hydrogels in the sensing field[60-62].

Colloidal 3D-PCs

It is noteworthy that spheres dispersed in aqueous solutions tend to assemble into a multilayer structure extending throughout the whole space. This structure is typically periodic in three spatial directions, normally adopting face-centered cubic configurations, resulting in close-packed 3D colloidal PCs after the solvent evaporates completely. Therefore, various approaches, such as gravitational sedimentation[63,64], vertical deposition[65,66] and co-solvent evaporation[67,68], are employed to establish colloidal 3D-PCs with intense optical signals and vivid structural colors owing to the extended order of superstructures. Besides, by using electrostatic interactions among charged nanoparticles, the 3D colloidal PCs with the body-centered-cubic lattice, namely non-close-packed 3D-PCs, are stably formed in aqueous solutions[69,70]. Typically, as compared with closed-packed 3D-PCs, non-close-packed 3D-PCs allow higher tolerance to the size polydispersity of nanoparticles because of the existing voids among them, and a lower particle concentration is required for the superstructure construction[71].

Microstructure-performance relationships

Optical mechanisms in mechanochromic PCs

This section elaborates on the optical response mechanisms in mechanochromic colloidal PCs, emphasizing the impact of microstructure changes on optical properties. Generally, colloidal PC arrays consisting of spheres just provide the skeleton with brilliant optical signals. However, to obtain satisfied mechanically responsive performances in colloidal PC arrays, it is essential to introduce responsive materials that exhibit both flexibility and robustness into the interstices among assembled building blocks. When white light illuminates colloidal 3D-PCs at certain incidence angles, the visible light within the range is blocked and diffracted to the same side, which induces the formation of visible structural color on colloidal PCs. Basically, Bragg's equation that defines the relationship between the diffracted wavelength (λ) and the corresponding parameters in colloidal PCs is given as follows[72]:

$$2nd\cos\theta = m\lambda \tag{1}$$

where n is the effective refractive index, d represents the lattice spacing, θ is the angle between the incidence and the normal direction of the crystal plane, m is the diffraction order, and λ is the wavelength of the reflected light. Notably, there also exists a relationship in close-packed colloidal 3D-PCs between the lattice spacing (d) and the interparticle/interpore spacing (D)[73]:

$$d = \sqrt{\frac{2}{3}}D \tag{2}$$

which demonstrates that the lattice spacing is positively correlated with interparticle spacing. This indicates that the precise tuning of the interparticle spacing can control the structural color of colloidal PCs.

Mechanochromic responses in colloidal PCs

Precise tuning of lattice spacing

Colloidal 3D-PCs, when illuminated with white light at specific incidence angles, exhibit visible structural colors due to the blocking and diffraction of light within a certain range. This section delves into the effect of external mechanical forces on the microstructures of colloidal PCs, which thus offers the opportunity to reveal the relationship between the micro-scalar lattice spacing (d) change and the optical readouts of colloidal PCs. As shown in Figure 2, three possible micro-changes that occur in the ordered structures of PCs are discussed when mechanical forces are exerted. Normally, with the enlarging on the two sides of PC films or the pressure from the top side, the spacing between two planes along the vertical direction would directly decrease, thus leading to the periodic decrease of colloidal PCs. Second, while the embedded templates are soft or removed, it is foreseeable that the lattice spacing would decrease, accompanied by changes in effective refractive index. Third, based on the second type of PC nanomaterials, as the medium produces an irregular deformation even under uniform external stimuli, the ordered PC structure would change to a disordered state.

Mechanical forces and micro-changes

In principle, based on Equation (1), the microchanges on colloidal PCs are closely related to the observed optical signals. Variations in lattice spacing on mechanochromic PCs lead to discernible color changes, emphasizing the role of this parameter in the overall performance of colorimetric sensors. As illustrated in Figure 2, when mechanical forces are applied to mechanochromic colloidal PCs, three possible microchanges may occur in the ordered structures, each resulting in corresponding optical performances that depend on the compositions of PCs. Typically, three types of occasions involving hard spheres, soft building blocks or micropores, and shape-memory media are detailed as follows.

In the first scenario, where the building blocks consist of hard spheres, only the lattice spacing (d) decreases with the exerted stretching forces applied along two sides or the pressures from the top. This micro-change on PCs leads to the blueshift of optical reflection signals based on Equation (1), which thus could inversely reflect the strength of forces according to the change of structural colors as observed from the naked eye[42]. Besides, for mechanochromic PCs consisting of soft building blocks or micropores, distinctive fluctuations occur in both the morphology of units and the distance between unit cells. It discloses that forces exerted on PCs alter both the parameters of effective refractive index and interplanar spacing. It is noted that the effect of interplanar spacing change on the wavelength of reflected light exceeds that of effective refractive index change, thus predominantly inducing a blueshift in the reflected light as well. The last response mechanism, which typically occurs in PCs after the introduction of shape memory polymers (SMPs), involves the reversible switch between ordered and disordered states with the removal or addition of external stimuli. SMPs can memorize one or more temporary distorted states and can recover to their original states or some intermediate morphologies under external stimuli[72]. Meanwhile, this transition is concurrent with the reversible disappearance or emergence of structural color, which thus could be employed as the indicator for suggesting dangerous or extreme conditions in our daily lives[45].

DESIGN STRATEGIES OF MECHANOCHROMIC PHOTONIC CRYSTALS

Building block-based PC film

Directly designing PCs to sense mechanical forces involves enhancing the robustness of building blocks, necessitating the formation of intact films during the assembly. CNCs, which are renewable nanomaterials with soft structures, could assemble into integral PC films with readable structural color post-solvent

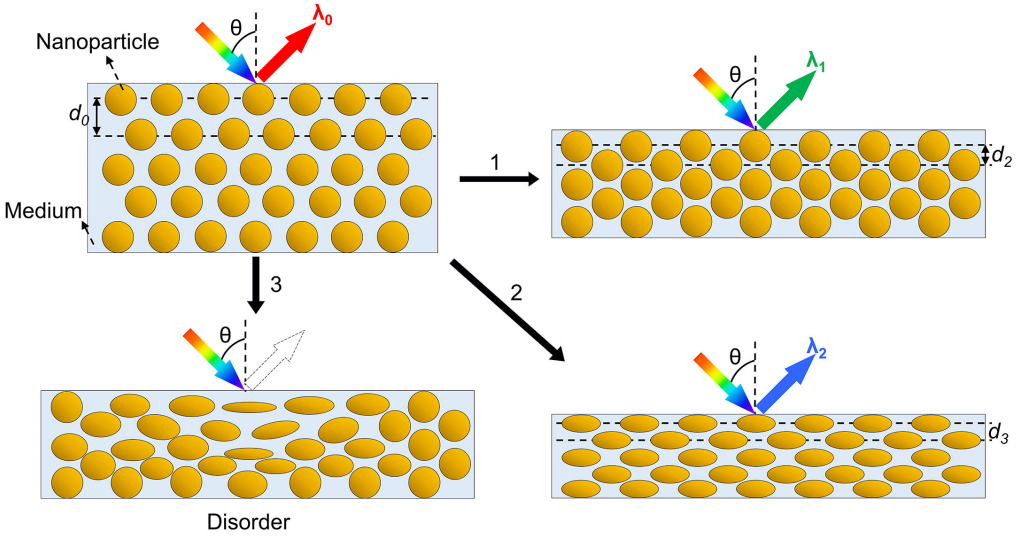


Figure 2. Three typical response mechanisms of mechanochromic colloidal PCs (3D configurations) under external forces based on various compositions: 1-hard building blocks; 2-soft building blocks; 3-shape-memory media. PCs: Photonic crystals; 3D: Three-dimensional.

evaporation[74]. However, these CNC-based PC films face limitations in pressure loading owing to the inherent lack of elasticity in CNCs. To address this, glucose, which usually serves as a plasticizer to prevent film from cracking, is introduced into CNC precursors, resulting in the construction of chiral photonic cellulose films with a strain-at-break up to 40.8%, exceeding current reports on chiral photonic cellulose films[75]. Remarkably, this crack-free film exhibits multi-stimuli-responsive characteristics, selectively reflecting circularly polarized light in response to both mechanical forces and chemicals.

PC gel

Hydrogels stand out among various gel types as promising candidates for decorating PC arrays to diverse forms, such as film[76], fiber[77] or microfluidic microneedles[78,79] due to their easy fixation processes under UV light. For instance, as shown in Figure 3, Jia *et al.* developed a mechanochromic photonic hydrogel by incorporating Fe_3O_4@C nanoparticles into an acrylamide (AM) hydrogel under a parallel magnetic field[80]. This hydrogel showed a compression detection range from 0-4.3 to 0-130.6 kPa by controlling monomer concentrations, demonstrating potential applications in displays and sensing. Moreover, a two-step filling strategy was also introduced for constructing non-close-packed 3D-PCs after etching the polymer core of nanoparticles and further filling the responsive medium, achieving ultra-sensitivity to temperature and compressional strain owing to the high refractive contrast of ZnS nanospheres (n = 1.91) and polymers (n ~ 1.3-1.5)[81].

In underwater applications, the self-adhesion of flexible sensors to smooth substrates, such as skins and diving suits, is crucial. To address this, a smart wearable visual sensor, integrating colloidal PC arrays with self-adhesive hydrogels, serves as mechanochromic indicators for monitoring underwater deformations[42]. This biocompatible PC sensor achieves excellent self-adhesion to human skin, reaching ~27 kPa, using 2-acrylamido-2-phenylacetic acid as the functional monomer in hydrogels. Besides, for mechanochromic sensors, high pressure and strain endurance capability are essential. Introducing a double-network structure to hydrogel systems enhances their endurance on mechanical forces. For instance, Liu *et al.* developed a mechanochromic photonic sensor for visual stress monitoring in flexible medical instruments[82]. In this work, the robust hydrogel, designed with a poly(acrylamide-co-N-acryloyl phenylalanine)/polyacrylamide

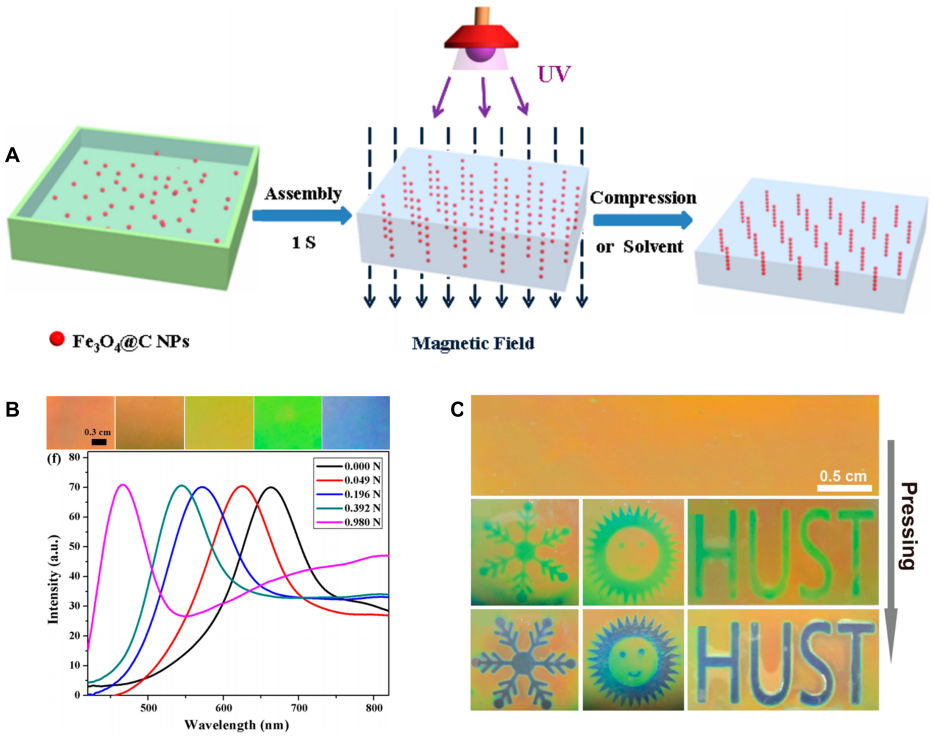


Figure 3. Mechanochromic photonic hydrogel with magnetically responsive 1D-PCs. (A) Scheme of the preparation of the photonic hydrogels assembled by Fe_3O_4@C nanoparticles; (B) photographs and related reflection spectra of the photonic hydrogel under different compressions; (C) photographs of the stamp patterns as pressed by a PDMS pattern. Reproduced with the permission of Ref.[80] Copyright 2015, ACS Publications. PCs: Photonic crystals; 1D: One-dimensional; PDMS: Polymerized deep eutectic solvent.

double network, exhibits a sensitive color shift covering the visible range with stress changing from 0 to 85 kPa, simultaneously reflecting a shift from 658 nm to 467 nm. Leveraging its mechanical-resistant property, a double-network PC hydrogel sensor, combining polyacrylamide and poly-methacryloxyethyl trimethyl ammonium chloride, enhances the reusability of PC papers in ion detection test[83].

Beyond hydrogels, organic gels offer advantages in constructing magnetochromic PCs. For instance, Hu *et al.* employed nonpolar and polymerizable di(ethylene glycol) ethyl ether acrylate (DEGEEA) to infiltrate ordered silica nanoparticles, resulting in a mechanochromic non-close-packed 3D-PCs with a broad tuning range of the photonic bandgap[84]. The main driving force for simultaneous assembly was strong electrostatic repulsion among silica nanoparticles, maintaining equivalent and untouchable distance in the DEGEEA precursor. This significantly enhanced optical performances, leading to a distinct change in effective refractive index under the same stress.

PC elastomer

Elastomers show promise as ideal materials for establishing mechanochromic PCs due to their robust flexibility and toughness. Unlike gels, elastomers, such as rubber, are not reliant on solvents for stress sensing, overcoming common solvent evaporation problems. Rubber, known for its excellent flexibility, corrosion resistance, shock absorption, and sound insulation, finds extensive use in the automotive and chemical industries. Integrating rubber or rubber-like materials into colloidal PCs can enhance mechanochromic PC sensors with these advantageous properties. As illustrated in Figure 4, an artificial skin combining a 3D-PC array with a rubber matrix demonstrates reversible mechanochromic performance, suitable for mechanical sensors and color displays[85]. The 3D-PC array, driven by interparticle repulsion

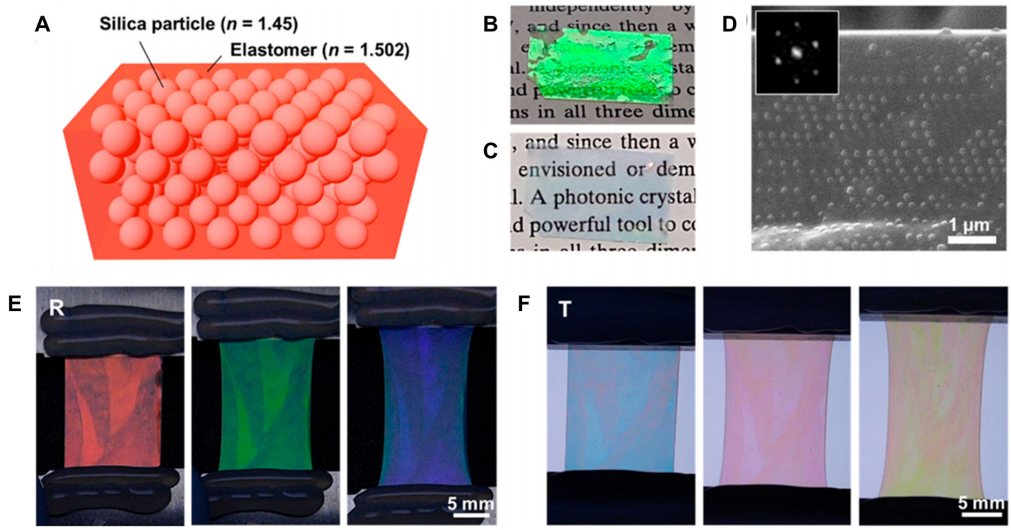


Figure 4. Mechanochromic photonic film containing a non-close-packed 3D-PC array. (A) Scheme of the photonic film consisting of silica particle as building block and elastomer as medium; Photographs of photonic film taken under (B) reflection condition and (C) transmission condition; (D) Cross-sectional Scanning electron microscope (SEM) image and related fast Fourier transform image; A photonic film under three different strains recorded in (E) reflection mode and (F) transmission mode. Reproduced with the permission of Ref.[85] Copyright 2017, ACS Publications. 3D: Three-dimensional; PCs: Photonic crystals.

from solvation layers on silica particles' surfaces, adopts a non-close-packed configuration, resulting in broader color changes compared to close-packed PCs. Combining chiral PCs with rubber-like materials, as illustrated in Figure 5, leads to reflection signal discrepancies under unpolarized and circular polarized light. Hussain *et al.* developed a photonic cholesteric liquid-crystal elastomer (CLCE) film with highly stretchable properties, achieved through cross-linking of thiol terminal groups and the formation of disulfide connections (-S-S-) in mesogenic oligomers[86]. Figure 5 showcases the dynamic exchange reaction between the -S-S- bond and preheating, allowing various hidden patterns in the CLCE film to reappear during stretching. This capability suggests potential applications in data encryption for anti-counterfeiting purposes.

Polydimethylsiloxane (PDMS), a widely used hydrophobic elastomer known for its superior structural integrity and transparency, is crucial for constructing the responsive medium in mechanochromic PC films[87]. PDMS films can be designed to possess ordered configurations with brilliant structural colors. For instance, Li *et al.* developed a mechanochromic device exhibiting distinct thickness-dependent wrinkling behavior for programmable optical responses[88]. As shown in Figure 6, the periodic wrinkles formed on PDMS films after oxygen plasma treatment, inducing lively patterns on thin PDMS films (below 1 mm) under stretching, changing the incident plane, or observing angle. Besides, integrating a colloidal PC array with a PDMS medium is effective for constructing efficient mechanochromic sensors. By encapsulating the PC/poly (ethylene glycol) phenyl ether acrylate (PEGPEA) polymer in PDMS films, a soft mechanochromic device with a multilayer architecture is designed for monitoring mechanical stress cycles in real situations[89].

Shape memory PCs (SMPCs) are intelligent composites merging the characteristics of PCs with SMPs. These materials visually sense external stimuli through temporary nanoscale shape deformations[72]. SMPCs facilitate the design of photonic sensors with colorful and colorless behaviors, achieved by the reversible switch between ordered and disordered states. Incorporating macroporous PC superstructures into the sensors allows SMPCs to exhibit distinct responses to temperature owing to the temporarily 'frozen' features

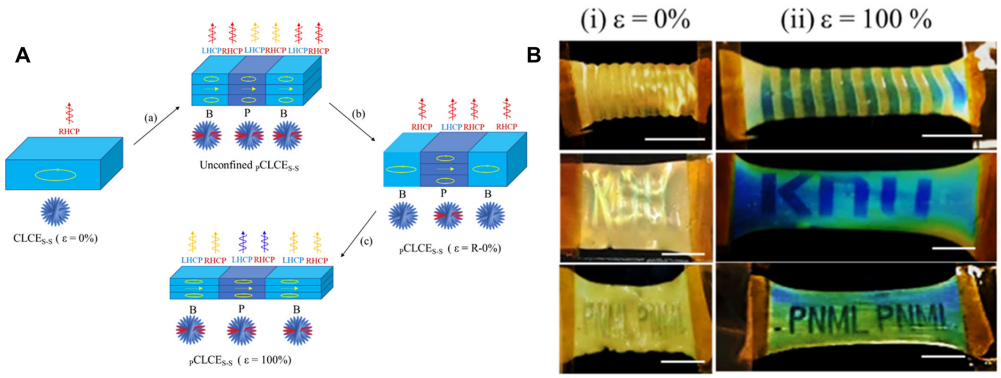


Figure 5. (A) Scheme illustrating the handedness change of the CLCE photonic film. (B) Photographs of CLCE photonic films under unpolarized light with lines, "KNU", and "PNML" letters (i) before and (ii) after stretching at ε = 100%. Reproduced with the permission of Ref.[86] Copyright 2021, ACS publications. CLCE: Cholesteric liquid-crystal elastomer.

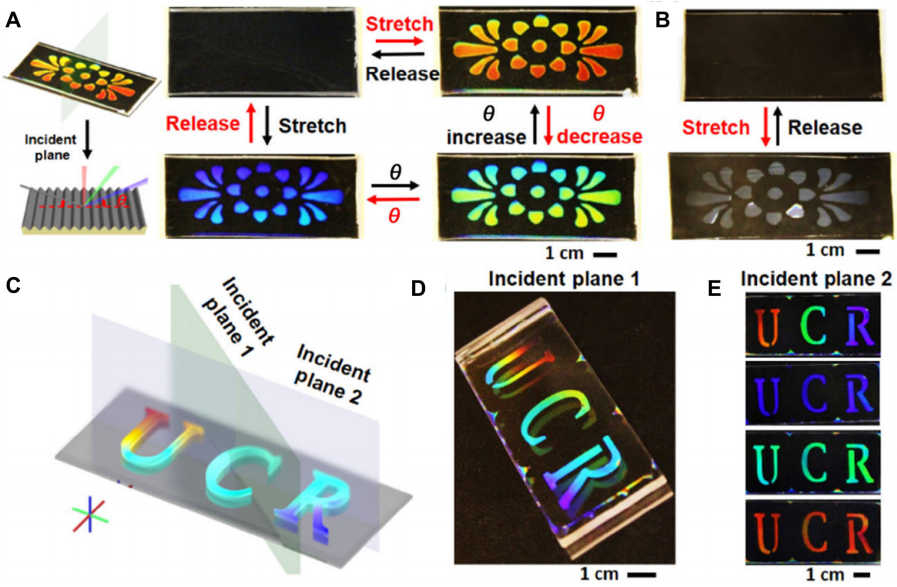


Figure 6. (A) Fabrication processes and (B-E) signal outputs of mechanochromic films based on the periodic wrinkles of PDMS for data encryption. Reproduced with the permission of Ref.[88] Copyright 2020, Springer Nature. PDMS: Polymerized deep eutectic solvent.

of SMPs at low temperatures. For instance, a SMPC microporous film with reconfiguration/rewriting capabilities was constructed using a polyurethane-based shape memory copolymer as the sensing material[90]. This sensor displayed all-room-temperature shape memory cycles, showcasing color control under programmable temperature regulations. Multifunctional SMPC sensors are also designed for sensing complicated conditions with integrated devices. Liu *et al.* developed biomimetic soft actuators based on liquid crystal elastomers (LCEs) for thermo- and mechanochromic camouflage and self-healing[91]. Integrating multi-stimuli-responsive tetraacrylsuccinonitrile (TASN) chromophore into LCEs, the TASN-LEC soft actuators exhibited reversible shape switching and color changing behavior in response to heat and mechanical compression simultaneously. Additionally, a facile hot-pressing approach and post-photocuring technique were developed for large-scale fabrication of SMPCs with both mechanochromic and thermochromic features[92].

PC-based textile material

Textiles, as a common element in daily attire, are considered ideal for developing wearable smart devices, making them potential representatives in the future intelligent textile industry[93-95]. Leveraging the inherent flexibility and toughness of fabrics, textile-based strain sensors have gained prominence for visually monitoring local deformations with easy wearability[96,97]. Combining commercially accessible textiles with colloidal PC arrays can lead to the design of mechanochromic textile sensors. For instance, Zhao *et al.* developed a mechanochromic electronic textile (MET) sensor for visual and electronic monitoring of body activities[98]. This MET sensor was designed by coating supramolecular photonic elastomers onto conductive polyester textiles with hierarchical fibers [Figure 7]. Notably, this integrated textile sensor exhibited obvious structural color, robust toughness up to 35.6 kJ m^{-3}, mechanical resilience, rapid optical and electrical response (~0.30 s), and fast recovery speed. These features highlight the potential of textile-based strain sensors for wearable devices.

Mechanochromic structures with high elasticity and stability can be incorporated into fiber shapes for intelligent textile construction. For instance, Zhang *et al.* utilized a continuous dip-coating method to combine photonic microspheres with elastic polymer fibers, creating a mechanochromic fiber that allows rapid and reversible monitoring of external forces visible to the naked eye[99]. This approach presents a novel avenue for developing smart wearable textiles. Another example for the design of magnetochromic PC fiber was designed by coating elastic rubber, involving a non-close-packed 3D-PC array, onto a spandex yarn[100]. By adjusting the size and volume fraction of silica particles in the PC array, efficient tuning of the original color of PC fiber was achieved. During stretching, the PC fiber exhibited a broad color change from red to blue, with a corresponding reflectance shift of 127 nm. This demonstrated the reversible and distinct optical changes in PC fibers, highlighting their potential applications in the field of mechanochromic PC textiles. Except for coating approaches, entire PC fibers have been woven into mechanochromic textiles with superior performances. For instance, Sun *et al.* developed a novel mechanochromic fiber by depositing a PC array onto a continuous sheet of aligned-carbon-nanotubes, followed by embedding PDMS on the fiber surface[101]. Benefiting from the excellent flexibility of PDMS, this elastic PC fiber maintained high sensitivity and stability even after 1000 deformation tests.

Multifunctional composite materials

Incorporating multifunctional materials into a system allows the creation of multi-modal photonic structures with diverse responsive abilities[102]. Conductive gel, known for its outstanding electrical capacity, is widely considered in structural color materials for designing flexible sensors with dual electrical and optical signal responses to external forces[98,103]. For instance, the integration of polymerized deep eutectic solvent (PDES) with a 3D polystyrene colloidal array resulted in a dual-mode photonic-ionic skin (PI-skin) with remarkable adhesion for human motion monitoring[41]. The strong adhesion, mechanical strength, and self-healing features are attributed to the multiple hydrogen bond network of PDES. Another way to introduce electronic signals into PC films is by incorporating ionic conductivity through the infiltration of ionic liquids into the gel, which is crucial for cultivating underwater stability. For instance, Li *et al.* reported a mechanochromic chiral nematic nanostructured film by penetrating fluorine-rich ionic liquids (FILs) into a CNC film[104]. Besides, Ma *et al.* also designed an ionic conductive cholesteric LCE (iCLCE) through the *in-situ* Michael addition and photopolymerization of cholesteric LCE precursors on ionic liquid-based polymeric networks[105]. Additionally, by directly coating a flexible conductive layer onto mechanochromic photonic fibers, a fiber-shaped dual-mode strain sensor is established with a strain range of 0%-80% and sensitivity of as high as 1.90[106].

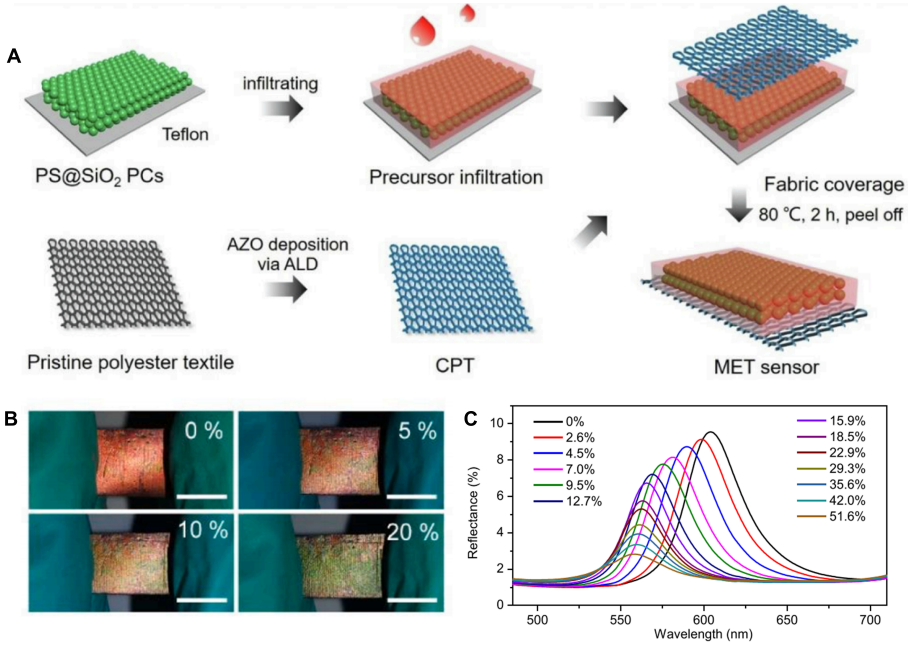


Figure 7. Mechanochromic electronic textile (MET) sensor constructed by coating supramolecular photonic elastomers onto conductive polyester textiles. (A) diagram of the fabrication of MET sensor; (B) photographs and (C) reflection spectra of MET sensors under varied stretching forces. Reproduced with the permission of Ref.[98]. Copyright 2021, Elsevier.

By combining hydrogels with elastomers, multifunctional mechanochromic PC films can be developed for integrated wearable devices. For instance, Zhang *et al.* designed a rational mechanochromic optical/electrical skin comprising an ionic electrode integrated with an elastomer layer, a chromotropic layer embedded with PC arrays, and a conductive MXene layer involving carbon nanotubes [Figure 8][107]. This mimic skin demonstrated excellent feedback to strain and pressure, providing both readable optical signals and ultrasensitive electrical signals simultaneously, thanks to the integration of PCs with conductive and flexible responsive materials. Embedding functional PC films into robust elastomers is an efficient way for developing mechanochromic PC sensors[108]. This method prevents solvent evaporation from photonic hydrogels, expanding the application environment of sensors to extreme conditions. Additionally, introducing flexible elastomers enhances the toughness and pressure-resistant capability of photonic sensors. Liang *et al.* developed a self-healing sandwich structure with a hydrophilic photonic hydrogel embedded into a modified PDMS layer[109], serving as a cuticle to prevent water evaporation. Similarly, Yang *et al.* developed a more deformable composite film consisting of poly(ethylene glycol) methacrylate and PDMS film[110]. This mechanochromic photonic gel displayed improved sensitivity to weak external forces, with a color change covering the entire visible range, attributed to the introduction of ethylene glycol into the photonic gel before particle assembly. Moreover, drawing inspiration from human skin, an electronic skin capable of sensing strain, temperature, and pressure was constructed by combining ionic hydrogel, PDMS film, and carbon nanotubes/PDMS electrodes into the PC array[111].

Through the combination of stimuli-responsive liquid crystal network bilayers involving azobenzene units with a colorless inverse opal film consisting of flexible polymers, a multifunctional photonic film responsive to temperature, light, and mechanical forces has been successfully designed[112]. Besides, as shown in Figure 9, Cui *et al.* developed a rewritable photonic paper capable of mechanical writing, electrically erase, and programmable optical modulation of structural colors[113]. In detail, reconfiguring the periodicity parameters, such as helical axes, allowed the switch between two high-contrast optical states for writing and

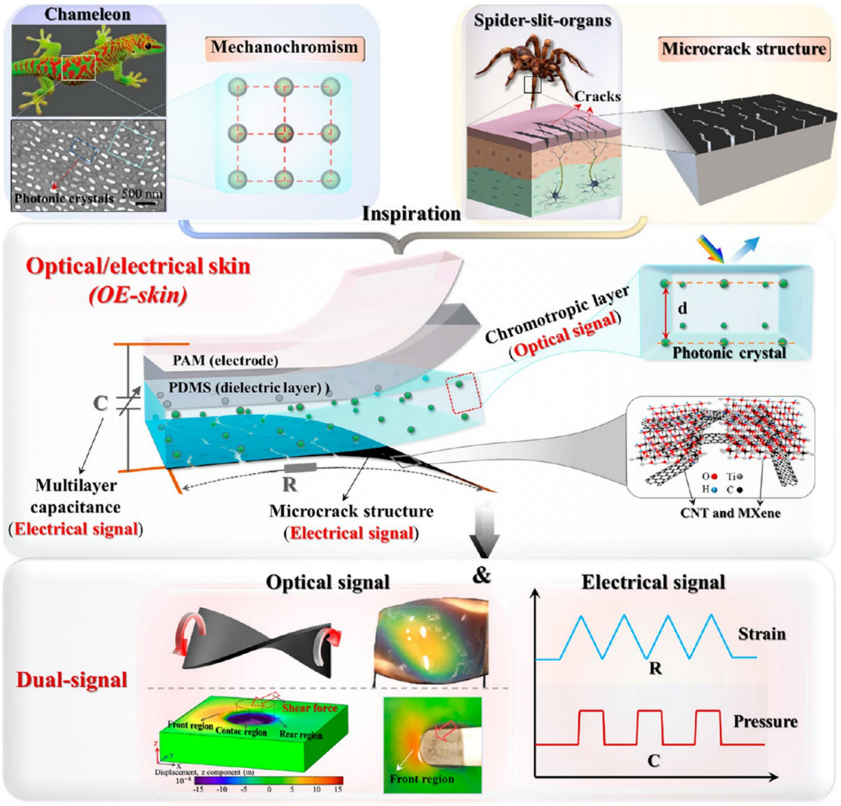


Figure 8. Construction of mechanochromic optical/electrical skin. Reproduced with the permission of Ref.[107] Copyright 2023, ACS publications.

erasing by pressure and electricity, respectively. Besides, the introduction of light-driven cholesteric liquid crystals (CLCs) offers the opportunity to design patterns with diverse colors, demonstrating multiple tuning ways for the pitch lengths of CLCs.

APPLICATIONS FOR UNDERWATER SENSING

For decades, research has predominantly focused on sensing applications on land owing to the urgent need for analyzing body gestures, which has implications for health monitoring, public safety, messaging, interactions, *etc.*. Additionally, there is a growing demand for flexible and visually readable photonic sensors in extreme conditions where non-optical signals are challenging to collect and detect, such as underwater environments in rivers and oceans. The development of mechanochromic PC sensors enables instantaneous and visible color changes in response to external mechanical forces, facilitating the transmission and interpretation of information by gestures or dynamic body motions.

Underwater body motion monitoring

In addition to flexible performance, self-adhesion plays a crucial role in constructing mechanochromic PC sensors for underwater sensing, ensuring stable attachment to wet substrates. As shown in Figure 10A and B, a self-adhesive PC hydrogel, comprising a polymethylmethacrylate PC array and biocompatible elastomer, was developed to monitor joint motion activity in water. This sensor achieved visual monitoring by structural color changes and electrical reflection peak shift[42]. Notably, it exhibited excellent self-adhesion (~27 kPa to human skin), impressive stretchability (~1050%), and wide color variability, showcasing its effectiveness in underwater motion monitoring. Furthermore, the incorporation of PDES

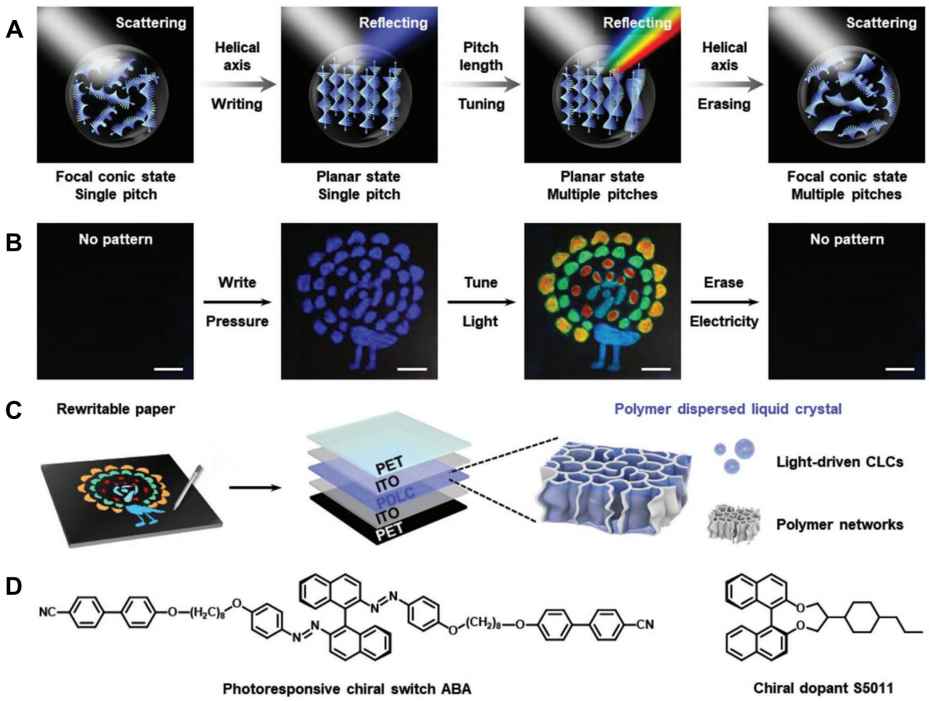


Figure 9. Construction and tuning of rewritable photonic paper. (A) Scheme of the working principle; (B) Photographs of the photonic paper showing the writing, tuning, and erasing process; (C) Scheme of the microstructure; (D) Chemical structures of photoresponsive chiral switch ABA and chiral dopant S5011. Reproduced with the permission of Ref.[113] Copyright 2022, Wiley-VCH GmbH.

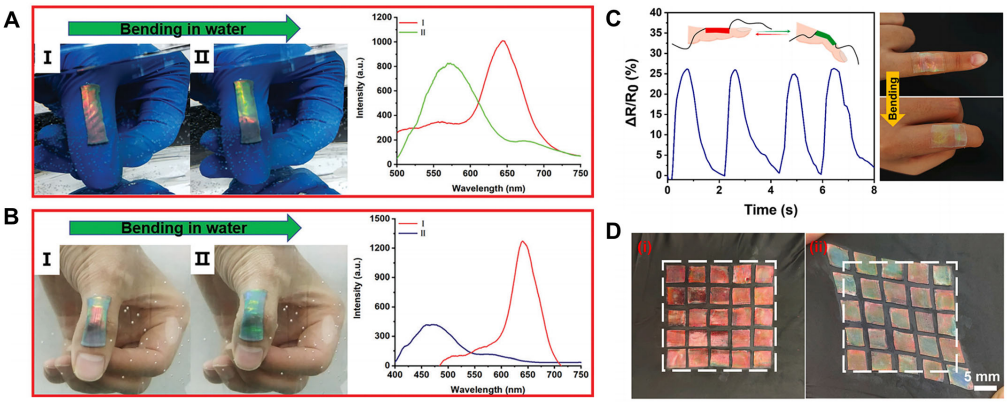


Figure 10. (A and B) Underwater wearing application of the PC film. Reproduced with the permission of Ref.[42] Copyright 2022, Royal Society of Chemistry; (C and D) PI-skin sensor for monitoring the joints' motions. Reproduced with the permission of Ref.[41] Copyright 2023, Elsevier. PC: Photonic crystals.

into the colloidal PC array led to the creation of a bifunctional mechanochromic and electromechanical PI-skin. With superior adhesion (~1.44 MPa on glass) owing to the multiple hydrogen bond network of PDES[41], this PI-skin simultaneously produced optical and electrical signals, along with self-healing capability [Figure 10C].

The 5 × 5 array construction facilitated efficient visual monitoring of strain distribution under nonuniform strain [Figure 10D]. Inspired by these achievements in both biocompatibility and self-adhesiveness, the potential of mechanochromic PC films for underwater activities is poised for enhancement.

Underwater communication

Transforming external changes into encrypted signals and converting them into texts provides an efficient approach for encrypting and transmitting information. Recognizing complex body gestures in this manner holds significant value, particularly for exploring the application of mechanochromic colloidal PC sensors underwater, where direct communication is challenging. Although underwater encrypted information transmission using PC sensors is in its early stages, some work has highlighted its advantages. For instance, the integration of CNC-based photonic film with FILs led to the construction of a FIL-CNC nanostructured film with excellent optical/electrical dual signals for underwater sensing and encrypted information transmission[104]. Hydrophobic FILs offered robust thermal and chemical stability during gel volume changes. As shown in Figure 11A, the FIL-CNC nanostructured film presented distinct color changes, shifting from red to blue as the film was stretched from 0% to 100% underwater, demonstrating maintained flexibility and readable optical signals. Besides, as shown in Figure 11B-M, the underwater relative resistance changes of the film are dependent on the bending degree of fingers, enabling the transmission of complex information underwater through Morse code. Using simple gestures, efficient information related to "HELLO", "YES", "HELP" and "SOS" was successfully transmitted via Morse code underwater [Figure 11F-J]. Moreover, this bifunctional PC sensor could recognize hands touching the water surface or users, the number of hands, and standing postures of the human body [Figure 11J-M], reflecting the initial but valuable potential of mechanochromic PC sensors in underwater communications.

CONCLUSION AND PERSPECTIVES

Mechanochromic colloidal PCs exhibit tremendous potential for underwater sensing applications, offering advantages such as energy independence and resistance to current leakage. This review has demonstrated the response mechanisms of typical mechanochromic colloidal PCs, emphasizing detailed designs on responsive media, including building blocks, hydrogels, organic gels, conductive gels, textiles, and multifunctional materials. The comprehensive overview of recent developments in mechanochromic colloidal PCs encompasses their initial applications in monitoring underwater body motion and encrypted information transmission, highlighting their significant potential in the underwater sensing field.

Despite the exciting accomplishments in underwater PC sensors, several urgent challenges need to be addressed. First, enhancing the integration of mechanochromic colloidal PCs with skins in water-filled environments demands focused efforts. It is noteworthy that more studies related to the biocompatibility of PC materials, involving toxicity, allergy, excitability, *etc.*, should also be considered owing to their direct contact with skin. While the design strategies for mechanochromic colloidal PCs have rapidly advanced, their limited application underwater is hindered by reliance on gels as main responsive media. The inevitable solvent evaporation from gels, crucial for maintaining volume change, restricts the application of mechanochromic PC sensors in extreme conditions. To overcome this drawback, the introduction of hydrophobic media that are not reliant on solvents for stress sensing, such as elastomers, would be an ideal approach for extending the use of sensors in particular environments. Additionally, there is ample room for exploring the use of mechanochromic PCs underwater, especially in developing self-adhesive or textile-based PC sensors owing to the wearable need in aqueous conditions. Second, improvements in the speed of underwater communication through mechanochromic PC sensors are essential. Although these sensors could provide instant and readable color changes, message transformation and feedback remain time-wasting, potentially leading to issues in critical situations such as underwater diagnosis and rescues. Notably, integrating responsive media with radio transmission materials holds potential for establishing a wireless information platform, which would be a new direction for achieving fast and accurate readout of optical signals and feedback simultaneously.

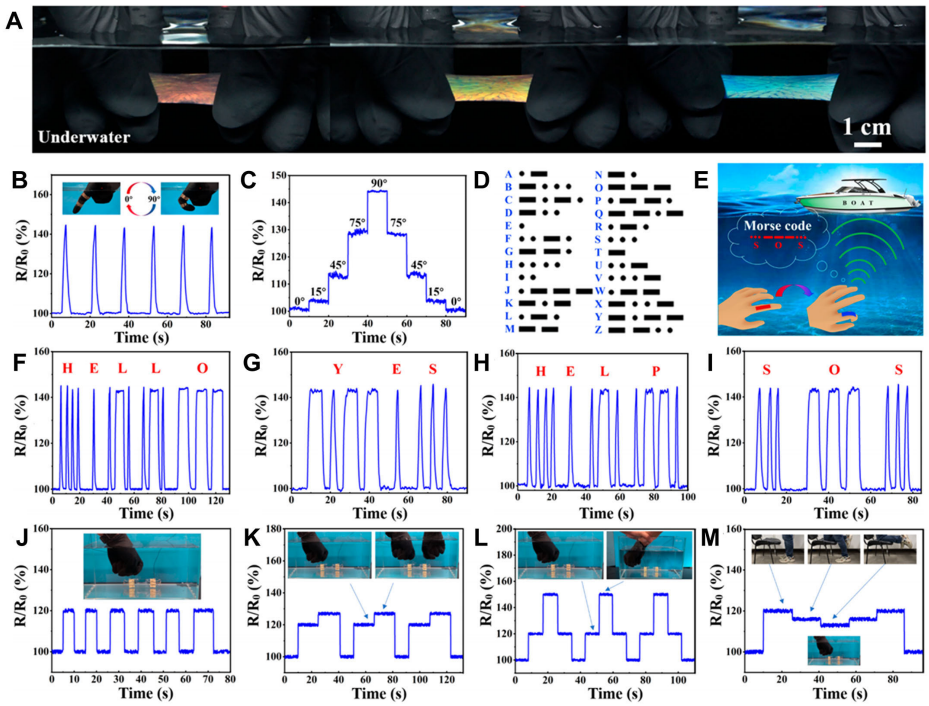


Figure 11. Underwater encrypted information transmission of the FIL-CLC nanostructured photonic film. (A) Underwater photographs of the photonic film under different strains; (B-M) Electrical signal monitoring of the photonic film adhered to a glove for gesture recognition. Reproduced with the permission of Ref.[104] Copyright 2023, ACS Publications. FIL: Fluorine-rich ionic liquids; CLCs: Cholesteric liquid crystals.

After addressing the issues on message transformation speed, four promising applications for mechanochromic PC sensors in underwater environments are envisioned. First, the development of an underwater pressure PC sensor capable of visually and precisely monitoring pressure fluctuations in water-filled environments, which thus is applicable for divers and underwater operators to make a rapid and relatively accurate judgment on the degree or direction of confronted current danger. Second, achieving underwater optical labeling using mechanochromic PC sensors enables the labeling and monitoring of underwater entities or alterations in the surroundings. This envisioned application is valuable for underwater engineers to rapidly locate the same underwater environments or places for multiple operations. Third, exploring the potential of mechanochromic PCs for underwater energy harvesting, utilizing their optical properties for light energy collection and conversion, which might be useful for providing lifesaving lighting or power generation functions for the entities or submarines that are working on underwater activities in an emergency. Finally, leveraging the mechanical characteristics of mechanochromic PCs for direct underwater imaging enables precise image acquisition in challenging water quality conditions or varying depths. The envisioned detecting method might provide a convenient and efficient way for collecting underwater information for related underwater monitoring departments. This review focuses on the latest progress in mechanochromic colloidal PCs and their applications in underwater sensing, suggesting that next-generation underwater sensors based on mechanochromic colloidal PCs will greatly enhance human exploration in underwater environments.

DECLARATIONS

Authors' contributions

Drafted the manuscript: Qi, F. L.

Revised the manuscript: Li, Q.; Wu, J. B.
Coordinated the writing and finalized the manuscript: Ding, C. F.; Wu, J. B.

Availability of data and materials
Not applicable.

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Conflicts of interest
All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate
Not applicable.

Consent for publication
Not applicable.

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