

Hydrogen Bond-enabled Crack-free Colloidal Photonic Crystal towards Passive Daytime Cooling

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Electronic Supplementary Information

Abstract Colloidal photonic crystals (CPCs) as self-assembled microstructured materials have emerged as promising platforms for structural coloration and passive daytime cooling (PDC). However, their practical application is severely hindered by cracking propensity, and the intractable trade-off between mechanical robustness and optical performance in large-area fabrication. Herein, we report a hydrogen bond-assisted self-assembly strategy to construct waterborne polyurethane (PU)-enhanced CPC coatings that synergistically integrate crack-free scalability, vivid angle-dependent structural color, excellent mechanical stability, and efficient PDC performance. Systematic optimization reveals that incorporating 3 wt% PU establishes a homogeneous hydrogen-bonding network between PU chains and carboxyl groups on colloidal particle surfaces. This hydrogen-bonding network effectively dissipates capillary stress during evaporation self-assembly to eliminate cracking while preserving the periodic lattice structure. The resultant PU-enhanced CPC (CPC/PU) coatings exhibit full visible-spectrum structural color tunability with retained iridescence, and achieve Class 2 adhesion. Mechanical evaluations confirm the CPC/PU coatings withstand 100 fraction cycles, 2-min water flushing, 50 times bending cycles, and 6-day UV aging without structural failure and color fading. Optically, the CPC/PU coatings deliver 63% solar reflectance and 86% infrared emissivity within the atmospheric transparency window (8–13 μm), enabling an 18.9 °C reduction in surface temperature compared to bare cement under 1000 W/m² simulated solar irradiance. Moreover, a maximum sub-ambient cooling of 6.6 °C under natural sunlight (average $I_{\text{solar}}=538 \text{ W/m}^2$) in outdoor field tests is achieved. The strategy offers a facile, sustainable route to multifunctional CPC coatings for decorative thermal management and building energy conservation.

Keywords Colloidal photonic crystal; Hydrogen bond; Crack-free; Passive daytime cooling

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INTRODUCTION

Microstructured materials have attracted increasing interest in materials science, physics, and chemistry owing to their capability to manipulate light, mass, and heat at the nanoscale level.^[1,2] Colloidal photonic crystals (CPCs) with periodic three-dimensional arrangements, derived from the self-assembly of monodisperse nanoparticles, exhibit a pronounced photonic bandgap.^[3,4] This characteristic enables vivid structural colors through selective Bragg diffraction rather than molecular absorption by dyes or pigments, facilitating innovative advancements in structural color textiles,^[5] high-resolution patterning,^[6] and biomimetic intelligent materials.^[7] Beyond their well-es-

tablished optical functionality, recent advances have highlighted CPCs as promising platforms for passive daytime cooling (PDC). Based on engineering the lattice constant and refractive index contrast, CPCs can deliver selective solar reflection via Bragg diffraction while preserving strong infrared (IR) emissivity within the atmospheric transparency window (8–13 μm), thereby enabling subambient cooling without energy input.^[8–10] Recent progress in this field includes biomimetic microstructured photonic-crystal coolers,^[11] nested bilayer ZnS@SiO₂ photonic architectures with robust full-color and daytime cooling performance,^[12] and inverse-opal scaffolds that combine structural coloration with mechanically resilient radiative-cooling interfaces.^[13] However, most high-performance designs rely on lithographic patterning or inherently brittle multilayer assemblies, which pose challenges for large-area applications.

Despite remarkable progress has been attained, conventional CPC films suffer from susceptibility to cracking and poor large-area uniformity, which severely limits their practical application. Generally, these issues primarily originate

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from the excessive tensile stress induced when the shrinkage rate of the colloidal lattice exceeds the solvent evaporation rate during evaporation self-assembly.^[14] To address this issue, diverse strategies have been developed, including surfactant addition, colloidal surface modification, and external field assistance.^[15,16] Gu *et al.*^[17] utilized low-adhesive super-hydrophobic substrates to assemble crack-free and highly crystalline CPCs, where hydrophobic repulsion effectively released interfacial stress. Xie *et al.*^[18] demonstrated that the in situ grafting of functionalized carbon dots (CDs) onto PS@P(MMA-AA) particles created an interfacial hydrogen-bonding network, effectively suppressing assembly stress and enabling scalable fabrication of large-area, non-cracking films with bright structural colors. Polymer-assisted assembly is a viable strategy for this purpose. Flexible polymer matrices can serve as binders to bridge inter-particle contacts, suppress crack propagation, and improve substrate adhesion without significantly disrupting the photonic bandgap.^[5,19] Hu *et al.*^[20] prepared crack-free CPC coatings on pretreated leather using a colloidal particle/polymer binder hybrid system. The pre-wetting strategy mitigated evaporative capillary stress by promoting inward fluid flow, whereas polyacrylate nanobinders provided mechanical reinforcement to eliminate cracks. However, achieving large-scale crack-free CPC coatings on complex substrates while maintaining both high structural color saturation and mechanical robustness remains a significant challenge.

Herein, we design a polyurethane (PU)-enhanced CPC (CPC/PU) coating that synergistically integrates a large-scale crack-free morphology, angle-dependent structural coloration, desirable mechanical robustness, and PDC function-

ality. The key advances of this work include: (1) we demonstrate a hydrogen bond-assisted self-assembly strategy that promotes the efficient and ordered packing of colloidal particles, enabling the scalable fabrication of crack-free CPC films; (2) the combination of vivid structural color, superior mechanical stability, and strong substrate adhesion endows the CPCs with considerable potential for advanced coating applications (Schemes 1a and 1b); (3) crucially, the CPC/PU coating retains its characteristic photonic stopband and high mid-IR emissivity, ensuring effective PDC performance (Scheme 1c). Thus, this study establishes a foundation for next-generation photonic materials and provides a facile and sustainable route towards multifunctional photonic coatings for decorative thermal management surfaces.

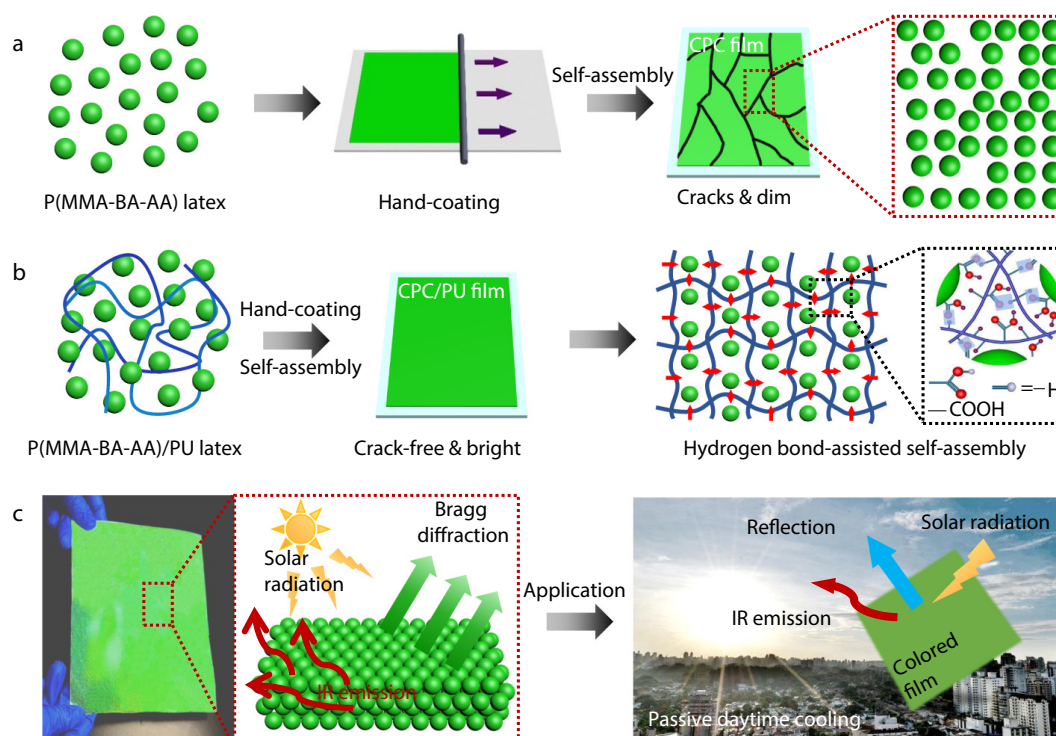
EXPERIMENTAL

Chemicals and Materials

Methyl methacrylate (MMA), butyl acrylate (BA), and acrylic acid (AA) were purified by repeated alkaline washing before use. The nonionic surfactant CO897, anionic surfactant sodium dodecyl sulfate (SDS), sodium bicarbonate (NaHCO_3), and potassium persulfate (KPS) were used for the synthesis of monodisperse colloidal particles. PU was also introduced as a reinforcing additive. All the chemicals were purchased from commercial suppliers. Ultrapure water with a resistivity of $18 \text{ M}\Omega\cdot\text{cm}$ was used in all the experiments.

Synthesis of P(MMA-BA-AA) Monodisperse Colloidal Latex

Monodisperse poly(methyl methacrylate-co-butyl acrylate-co-



Scheme 1 (a) Schematic illustration of the construction of the traditional P(MMA-BA-AA) CPCs, which suffers from cracking and dim structural color; (b) Mechanism of the hydrogen bond-assisted self-assembly in the P(MMA-BA-AA)/PU CPC. The resultant CPC is crack-free and bright; (c) Photograph showing the large-scale crack-free CPC film, demonstrating its potential application in PDC.

acrylic acid) (P(MMA-BA-AA)) latexes were synthesized via modified semi-continuous emulsion polymerization.^[21] Typically, to obtain latex with a particle size of 212 nm and a polydispersity index (PDI) of 0.020, 0.70 g of CO897, 0.08 g of SDS, and 0.65 g of NaHCO₃ were completely dissolved in 130 g of ultrapure water to form the base solution, which was then transferred into a four-necked round-bottom flask under nitrogen atmosphere with constant mechanical stirring. The system was then heated to 80 °C and maintained at this temperature for 30 min. Subsequently, a pre-emulsion consisting of 9 g of BA, 30 g of MMA, 4 g of AA, 0.30 g of CO897, 0.09 g of SDS, and 7.5 g of H₂O was added dropwise. Simultaneously, an initiator solution (0.55 g of KPS dissolved in 15 g of ultrapure water) was injected at a rate of 15 mL/h to initiate the polymerization. The reaction was allowed to proceed for an additional 4 h. Finally, the resulting P(MMA-BA-AA) monodisperse colloidal latex was filtered and dialyzed to remove any residual impurities.

Preparation of P(MMA-BA-AA)/PU Latex

Monodisperse P(MMA-BA-AA) colloidal latex (30 wt%, 5.0 g) was dispersed in a clean, dry 25 mL beaker. Subsequently, 0.05 g of PU was added dropwise under gentle stirring to ensure preliminary mixing and to prevent local concentration gradients. The mixture was ultrasonicated at 40 kHz for 30 min to ensure a homogeneous dispersion. Finally, the latex was allowed to stand for 30 min to facilitate dissipation of residual air bubbles and ensure system stabilization.

Construction of CPC Film

A facile hand coating method was employed to fabricate CPC films. A wire-wound rod with a defined groove depth was used to uniformly spread the latex onto a substrate, where the wet film thickness was precisely determined by the groove depth of the rod. For standard CPC preparation, 30 µm-thick poly(ethylene terephthalate) (PET) substrates were first treated with oxygen plasma for 250 s to enhance surface hydrophilicity. Subsequently, the P(MMA-BA-AA)/PU latex was selected as the coating ink and uniformly blade-coated onto PET substrates using a wire-wound rod (CCT-90), corresponding to a wet-film thickness of 90 µm. Crucially, solvent evaporation and colloidal self-assembly were conducted under controlled ambient conditions (25±2 °C and 45%±5% relative humidity). Under these conditions, the solvent evaporated gradually, enabling the colloidal particles to self-assemble into CPC films with vivid structural colors. This protocol was readily scalable, as demonstrated by the successful fabrication of large-area coatings on extended substrates.

Furthermore, for the patterned CPC fabrication, a patterned template was placed in contact with the substrate. The P(MMA-BA-AA)/PU latex was then coated onto the patterned substrate, where the colloidal particles underwent confined self-assembly within predefined regions, yielding bright CPC patterns.

Cooling Performance Measurement

The cooling performance of the CPC/PU films was evaluated through both indoor and outdoor field tests. Indoor evaluations were conducted using a custom-built apparatus consisting of an aluminum foil-wrapped PS foam insulation box and solar simulator (CEL-PE300L-3A) equipped with a xenon lamp. Samples were placed inside an insulated box and sealed with trans-

parent polyethylene (PE) film. The setup was exposed to a simulated solar irradiance of 1000 W/m² calibrated using an optical power meter (CEL-NP2000). Real-time temperature data were recorded continuously throughout the test using an IR thermal imager (Fluke Ti400). Similarly, outdoor measurements were performed on the rooftop of the State Key Laboratory at Nanjing Tech University (32°06'N, 118°40'E) on August 5, 2023.

Cooling Power Calculation

Theoretically, the cooling power of the CPC/PU coating was calculated using the following formulas:^[22–24]

$$P_{\text{cool}}(T_s) = P_{\text{rad}}(T_s) - P_{\text{sun}} - P_{\text{atm}}(T_{\text{atm}}) - P_{\text{cond+conv}} \quad (1)$$

$$P_{\text{rad}}(T_s) = \varepsilon_{\text{LWIR}} \sigma T_s^4 \quad (2)$$

$$P_{\text{sun}} = \int_{0.3}^{2.5} I_{\text{AM1.5}}(\lambda) A(\lambda) d\lambda \quad (3)$$

$$P_{\text{atm}}(T_{\text{atm}}) = \varepsilon_{\text{LWIR}} \varepsilon_{\text{atm}} \sigma T_{\text{atm}}^4 \quad (4)$$

$$P_{\text{cond+conv}} = q(T_{\text{atm}} - T_s) \quad (5)$$

where $P_{\text{cool}}(T_s)$ is the cooling power of the CPC/PU coating. $P_{\text{rad}}(T_s)$ represents the thermal radiation emitted by the CPC/PU coating, P_{sun} is the absorbed solar irradiance, $P_{\text{atm}}(T_{\text{atm}})$ denotes the atmospheric long-wavelength radiation power received by the CPC/PU coating, and $P_{\text{cond+conv}}$ is the nonradiative power lost via thermal convection and conduction. Meanwhile, $\sigma = 5.67 \times 10^{-8} \text{ W}/(\text{m}^2 \cdot \text{K}^4)$ is a Stefan-Boltzmann constant, A is the average solar absorptivity of the CPC/PU coating, $\varepsilon_{\text{LWIR}}$ is average emissivity of the CPC/PU coating within the atmospheric transparency window, and $I_{\text{AM1.5}}$ is the standard AM 1.5 global solar irradiance spectrum. ε_{atm} is the atmospheric emissivity. T_{atm} is the ambient temperature, T_s is the surface temperature of the CPC/PU coating, and q is non-radiative heat coefficient.

Characterization

The hydrodynamic size distribution and PDI of the colloidal particles were measured using dynamic light scattering (DLS, Malvern Zetasizer 3000). The morphology and microstructure of the CPCs were characterized by scanning electron microscopy (SEM, Hitachi S-4800). Fourier transform infrared (FTIR) spectra of the CPC films were recorded using a microscope (Nicolet iN10) to analyze the functional group intensities. Reflection spectra of the CPC films were collected using a fiber-optic spectrometer (Ocean Optics USB4000). Macroscopic optical images of the films were captured using a stereo-optical microscope (SHUNYU SZM45). Additionally, the surface wettability of the CPC films was evaluated using a contact angle measurement system (Kruss DSA100). The CIE 1931 chromaticity coordinates were collected using an ultraviolet-visible (UV-Vis) spectrometer (PerkinElmer Lambda 900). Reflectivity spectra were measured in the 0.3–2.5 µm range using an ultraviolet-visible-near-infrared (UV-Vis-NIR) spectrophotometer with an integrating sphere (Shimadzu UV-3600i Plus). An FTIR spectrometer (Nicolet iS50) with an integrating sphere was used to measure IR emissivity in the 6–15 µm range.

RESULTS AND DISCUSSION

Preparation and Characterization of the P(MMA-BA-AA)/PU CPC

Traditional CPCs commonly suffer from excessive tensile stress

induced by liquid menisci at the air-liquid interface during solvent evaporation, inevitably leading to cracking. To address this, we introduced into P(MMA-BA-AA) monodisperse colloidal latex as a film-forming agent. The abundant hydrogen-bonding sites within the PU network enhance particle-particle and particle-substrate interactions, thereby suppressing crack formation (Fig. S1 in the electronic supplementary information, ESI).^[25] Crucially, the incorporation of an appropriate amount of PU (1 wt%–3 wt%) did not significantly alter the particle size distribu-

tion and monodispersity of the colloidal latex. However, increasing the PU concentration to 5 wt% resulted in a broader size distribution and reduced uniformity owing to severe agglomeration, which is undesirable for high-quality CPC assembly (Fig. S2 in ESI).

Based on this, P(MMA-BA-AA)/PU CPC films with PU contents ranging from 0 wt%–5 wt% were fabricated. As shown in Fig. 1(a), the pristine CPC film (0 wt% PU) was predominantly covered with dense and interconnected cracks.

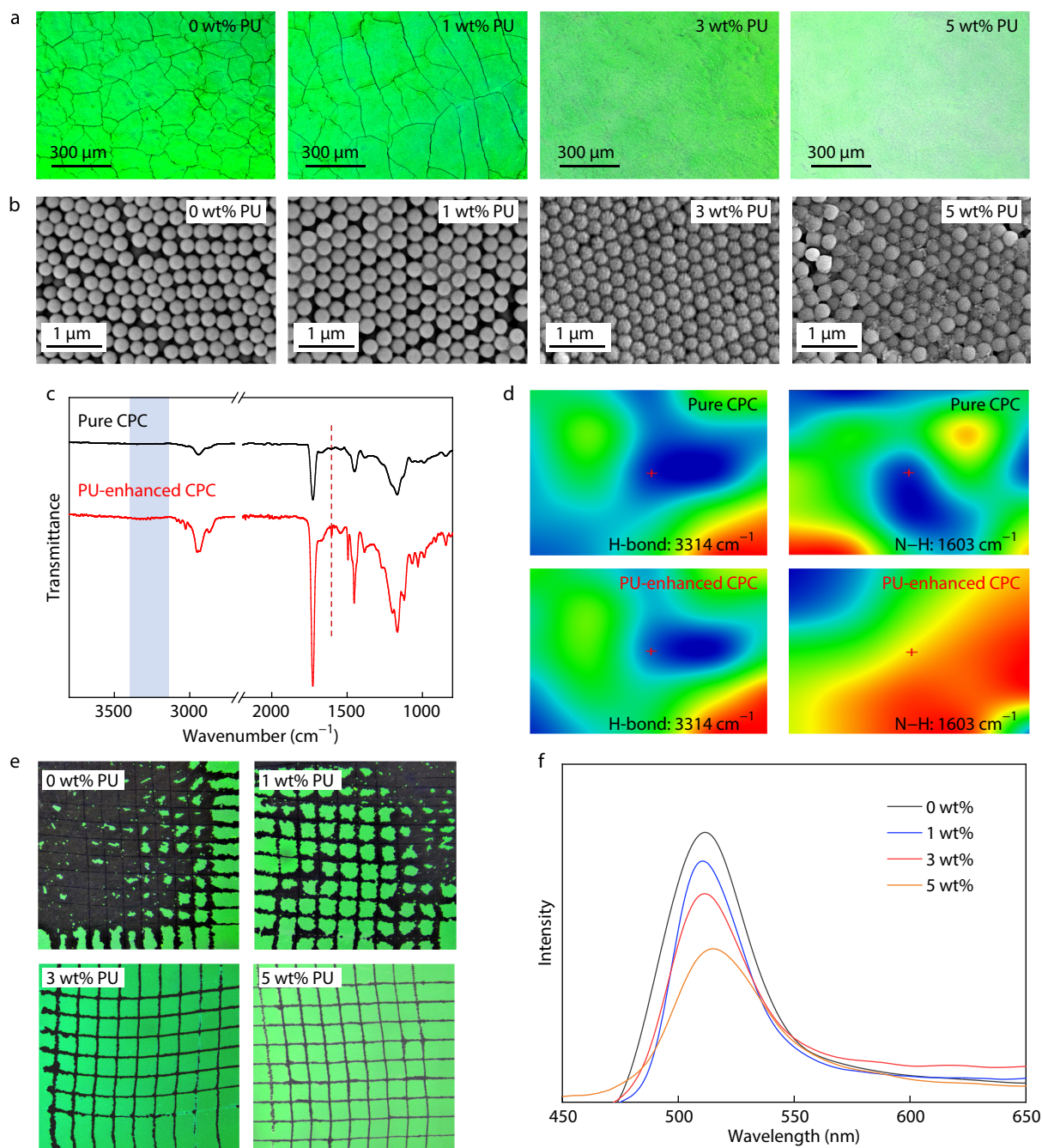


Fig. 1 (a) Optical photographs and (b) SEM images of CPC films with varying PU contents (0 wt%, 1 wt%, 3 wt%, and 5 wt%); (c) FTIR spectra of pristine P(MMA-BA-AA) CPC and P(MMA-BA-AA)/PU CPC; (d) Micro-IR images corresponding to characteristic peaks at 1603 and 3314 cm^{-1} , confirming the formation of hydrogen bond network; (e) Adhesion tests and (f) reflection spectra of CPC films with different PU content (0 wt%, 1 wt%, 3 wt%, and 5 wt%).

However, 1 wt% PU slightly mitigated this issue, and a critical transition occurred at 3 wt% PU, yielding a nearly crack-free film with a uniform green structural color. Further increasing the PU content to 5 wt% maintained the crack-free morphology but resulted in a visually duller color with reduced saturation. The SEM micrographs (Fig. 1b) further reveal the microstructural evolution governing these macroscopic changes. The pristine CPC sample (0 wt% PU) exhibited extensive microcracks and point defects. Distinct interparticle gaps were observed. Upon the introduction of 1 wt% PU, the particle arrangement became more regular, accompanied by an enhanced long-range order. Meanwhile, PU was incorporated, rendering the interparticle interfaces less distinct compared to the pristine sample. Notably, the 3 wt% CPC/PU sample exhibited a perfect crack-free face-centered cubic (FCC) lattice structure with nearly invisible interparticle interfaces. Conversely, excessive polymer filling at 5 wt% PU induced disintegration of the ordered lattice into disordered stacking and severe agglomeration, which disrupted the periodic arrangement essential for photonic performance.

To elucidate the crack-suppression mechanism, FTIR spectroscopy was employed to characterize the hydrogen-bonding interactions. As shown in Fig. S3 (in ESI), the spectrum of PU exhibited characteristic bands at $3300\text{--}3500\text{ cm}^{-1}$ (N—H stretching) and 1700 cm^{-1} (C=O stretching), confirming the presence of potential hydrogen-bonding sites. In addition, Fig. 1(c) presents the FTIR spectra of the control sample (pure P(MMA-BA-AA)) and P(MMA-BA-AA)/PU. The presence of a series of characteristic bands at $1200\text{--}1700\text{ cm}^{-1}$ in the control sample indicated the successful synthesis of P(MMA-BA-AA) colloidal particles. Upon incorporation of PU, the band profile in the $3100\text{--}3400\text{ cm}^{-1}$ region exhibited a noticeable broadening and shift relative to that of pristine P(MMA-BA-AA) CPC. This evolution was attributed to the formation of hydrogen bonds between the C=O/N—H groups of the PU molecular chains and the —COOH groups on the surface of the P(St-BA-AA) colloidal particles. Micro-IR imaging was employed to visualize the distribution of this interaction (Fig. 1d). The hydrogen-bonding map (3314 cm^{-1}) showed a weak blue signal for the pristine CPC film, whereas the CPC/PU film exhibited a uniform green distribution, indicating a homogeneous hydrogen-bond network. Similarly, the PU-specific N—H bending map (1603 cm^{-1}) revealed distinct signals in the CPC/PU film, verifying the uniform distribution of PU.^[26,27] Furthermore, to directly verify the formation of hydrogen bonds in the CPC/PU system, variable-temperature FTIR measurements were conducted on the CPC/PU film (Fig. S4 in ESI). The stretching vibration frequency located at $3400\text{--}3100\text{ cm}^{-1}$ decreased in intensity and shifted to higher wavenumbers upon heating to $75\text{ }^{\circ}\text{C}$ but largely recovered upon cooling, demonstrating the thermally reversible breakage and reformation of hydrogen bonds.^[28] These results directly verified the formation of hydrogen bonds. Thus, the abundant hydrogen bond network between the colloidal particles and PU matrix effectively dissipates internal capillary stress at both the macroscopic and microscopic scales, thereby accounting for the crack-free morphology of the CPC film. As expected, the incorporation of PU modulated the surface wettability of CPC films (Fig. S5 in ESI). This effect is also governed by the

hydrophilic groups (e.g., carboxyl, polyester, and polyether) in PU, which enhance hydrogen bonding with water molecules. Increasing the PU content from 0 wt% to 5 wt% increased the surface free energy of the CPC/PU film, causing the water contact angle to drop monotonically from 64° to 27° . This transition from weak to strong hydrophilicity further highlights the pivotal role of hydrogen bonding in governing the structural evolution of the CPC films.

The reinforced hydrogen-bonding network significantly improved the structural stability of the CPC/PU films. As shown in Fig. 1(e), the adhesion performance was evaluated using a cross-cut tape test (ISO 2409).^[29] The pristine CPC film (0 wt% PU) exhibited severe delamination under cutting stress and corresponded to a Class 5 rating (worst adhesion) owing to weak van der Waals forces. Incorporating 1 wt% PU improved the rating to Class 3, whereas 3 wt% PU achieved Class 2 with only slight peeling at intersections. Remarkably, the 5 wt% PU film demonstrated excellent adhesion (Class 1), showing almost no detachment and intact grid lines. This progression confirms that the dense PU network effectively strengthens interparticle connections, thereby boosting the mechanical adhesion integrity and scratch resistance of the CPC coatings. From the perspective of optical properties, the PU content strongly influences the structural color of the CPC films. As shown in Fig. 1(f), as the PU content increased from 0 wt% to 5 wt%, the reflectance peak intensity gradually decreased. Specifically, the peak intensity at 510 nm decreased from about 80% for the pristine CPC film to about 45% for the 5 wt% CPC/PU sample. A slight red shift in the reflection peak from 510 nm to 515 nm was observed. These results can be attributed to two factors. First, excess PU ($>3\text{ wt}\%$) reduced the overall refractive index contrast between the colloidal particles and the matrix. Another reason is that PU aggregation disrupts the ordered colloidal packing.^[30,31] Correspondingly, the CIE chromaticity diagram (Fig. S6 in ESI) shows that, while all samples remained within the green region, the coordinates shifted towards the center with increasing PU content, indicating a reduction in color saturation. Macroscopically, this manifested as a whitish and duller structural color. Taken together, these results demonstrate that while PU incorporation effectively addresses the inherent cracking issue of pristine CPC films via hydrogen bonding, it introduces trade-offs in the optical performance, including reduced reflectance, peak shifting, and saturation loss. Therefore, an optimal PU loading of 3 wt% was identified, which balances the robust mechanical integrity with the preserved optical quality.

Hydrogen Bond-assisted Self-assembly Mechanism Study

The second set of experiments focused on the assembly behavior and underlying mechanisms of CPCs. In the CPC/PU film, the abundant hydrogen-bonding sites within the PU network significantly strengthened the interfacial interactions between the colloidal particles, facilitating their assembly into a densely packed and ordered lattice structure (Fig. 2a).^[26,32] As evidenced by the SEM images in Fig. 2(b), a well-defined FCC lattice is clearly observed, and the regular hexagonal arrangement is well preserved throughout the entire structure. Conversely, the control sample (pure CPC film) lacked this driving force, resulting in

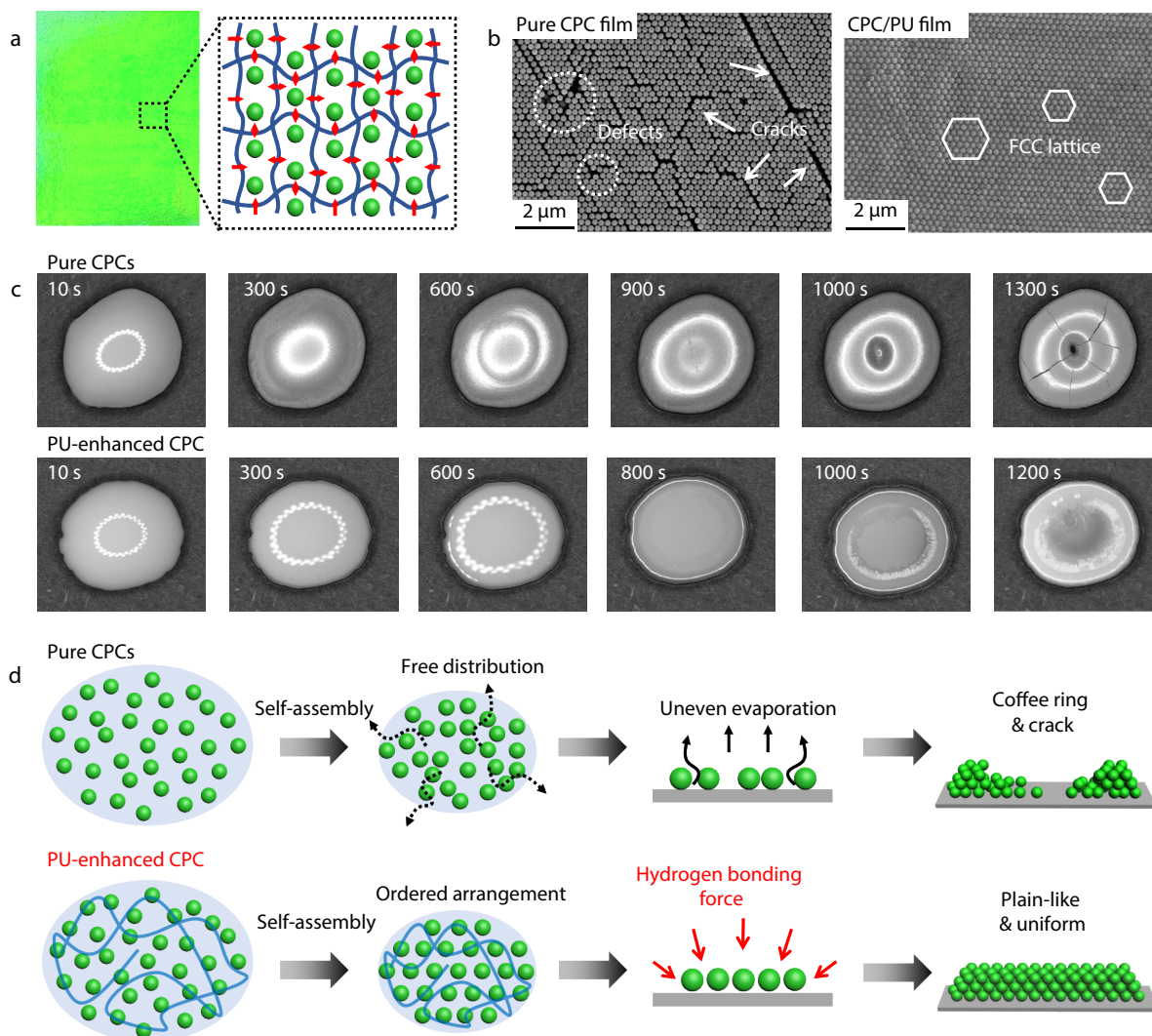


Fig. 2 (a) Schematic illustration of the hydrogen bond-assisted self-assembly for constructing the crack-free CPC film; (b) SEM images comparing the micro-structure of pure P(MMA-BA-AA) CPC and P(MMA-BA-AA)/PU CPC; (c) Time-dependent tracking of the self-assembly during solvent evaporation for both pure and CPC/PU systems; (d) Illustration of self-assembly mechanism of traditional CPC and hydrogen bond-assisted self-assembly of the CPC/PU.

a structure with cracks and defects. To further elucidate how PU modulates the assembly process, the self-assembly dynamics of the pure CPC and CPC/PU systems were monitored using a stereomicroscope. Fig. 2(c) illustrates this distinct drying behavior. In the pure CPC system, a prominent bright ring appeared at the droplet edge in the early stage, where capillary flow drove the particles to the periphery. Subsequent solvent evaporation induced massive capillary stress, causing the central particles to aggregate and contract and forming a thick edge deposit. Ultimately, internal stress accumulation leads to significant cracking, resulting in a non-uniform “coffee ring” morphology. In contrast, the CPC/PU systems exhibit markedly different behaviors. The droplets maintained a uniform distribution throughout the early stages. During the intermediate stage, the droplet retains its structural integrity without noticeable contraction or phase separation. In the late stage, flat, uniform, and crack-free deposition were achieved.

Based on these results, a possible mechanism for the assembly of CPC/PU is presented in Fig. 2(d). Pristine P(MMA-

BA-AA) colloidal particles lack sufficient interfacial interactions to dissipate the massive capillary stresses generated during solvent evaporation, leading to severe cracking and structural failure upon assembly.^[33,34] The incorporation of PU yields a robust hydrogen-bonding network that bridges colloidal particles. This network functions as a dynamic binder, effectively dissipating internal stress at both macroscopic and microscopic levels, thereby eliminating defects and enhancing the surface structure.^[26,35] Furthermore, PU mitigates the coffee ring effect by increasing system viscosity and modulating solvent evaporation, thus promoting uniform particle deposition.^[36] The synergistic effect of this hydrogen-bonding network and coffee-ring suppression endows CPC films with excellent optical and mechanical stability, providing a feasible strategy for developing robust CPCs.

Construction of the Robust CPC/PU Coating

Taking the application into consideration, full-spectrum structural color tunability is crucial for advancing high-performance

CPCs. By precisely tuning the size of colloidal particles, the structural color can be continuously modulated across the entire visible spectrum. As shown in Fig. S7 (in ESI), a series of P(MMA-BA-AA)/PU colloidal latices with varying particle sizes was synthesized. DLS measurements revealed that all these latex samples possessed narrow size distributions ($PDI < 0.04$) and excellent monodispersity, qualifying them as ideal building blocks for high-quality CPCs.^[37] Consequently, as shown in Fig. 3(a), the uniform colloidal particles assembled into highly ordered and periodic close-packed lattice structures, which endowed the CPC/PU film with high reflectance at specific wavelengths, yielding vivid blue (212 nm), green (246 nm), yellow (259 nm), orange (286 nm), and red (309 nm) structural colors. The reflection spectra of the five CPC films were measured to evaluate their optical performance (Fig. 3b). All the samples exhibited sharp reflection peaks, indicating excellent optical characteristics. As the particle size increased from 212 nm to 309 nm, the peak position experienced a continuous red shift from 463 nm (blue) to 636 nm (red), which is in good agreement with Bragg's diffraction law.^[38] These results confirm that the structural color

can be precisely tuned across the entire visible range by simply adjusting the particle size, enabling continuous color modulation from blue to red. The most intuitive optical characteristic of CPCs is the angular dependence of the structural color, known as the "iridescence effect," which is an inherent property dictated by Bragg's diffraction law.^[39] Experimental results confirmed that the introduction of PU did not alter this physical property. As shown in Fig. 3(c), as the observation angle increases from 0° (normal incidence) to 90° (grazing incidence), the structural color of the PU/CPC film sequentially shifts from red to orange, yellow, yellow-green, and green. Correspondingly, the reflection spectra revealed that the peak position shifted from approximately 613 nm at 0° to approximately 544 nm at 90° , exhibiting a distinct blue shift, consistent with the theoretical predictions. This behavior demonstrates that the CPC microstructure retains precise control over light interference and diffraction, as PU merely acts as a matrix filler without disrupting lattice periodicity. Thus, the PU/CPC film successfully preserved the iridescence effect.

The scalability of the PU/CPC system was validated by fabri-

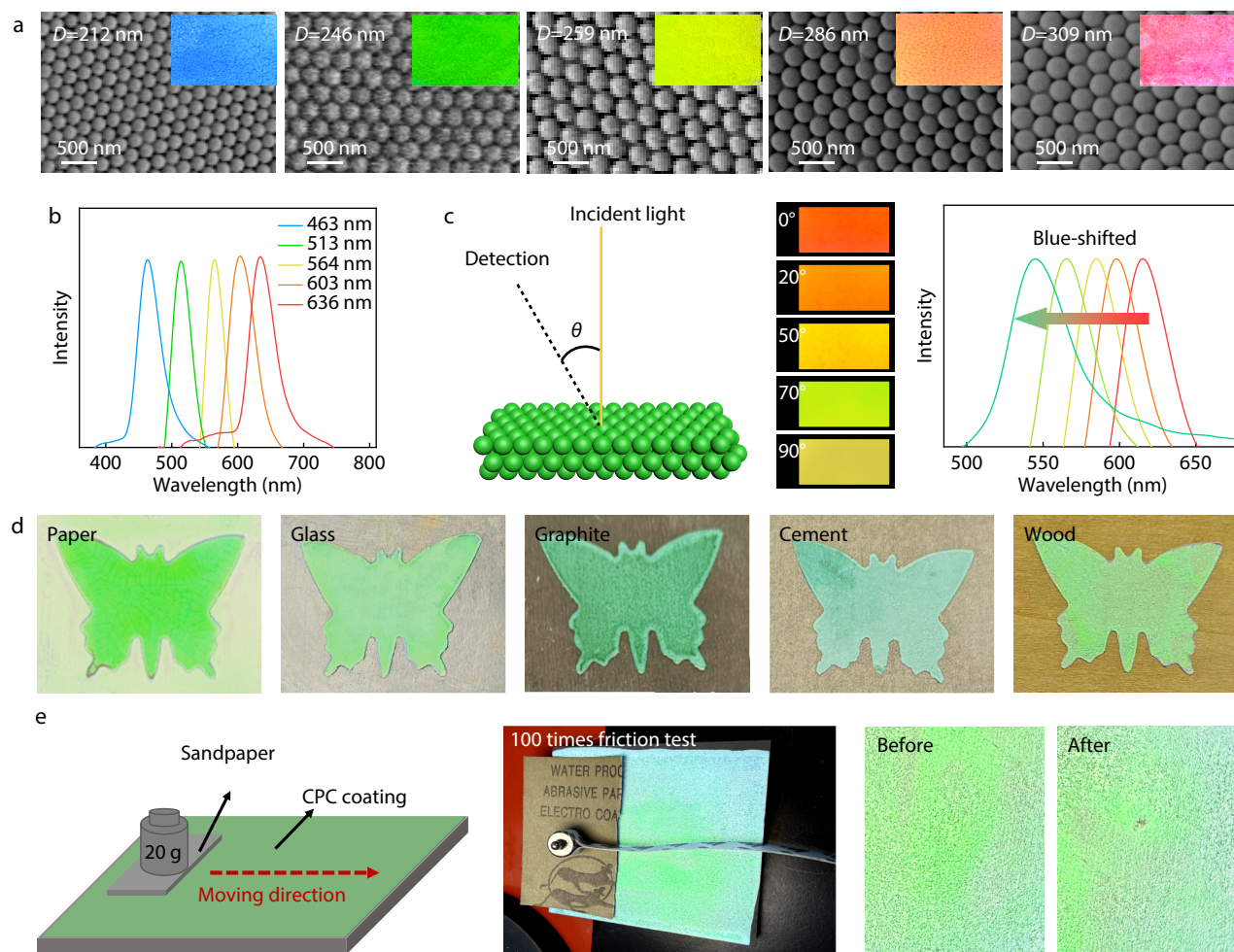


Fig. 3 (a) Optical photographs and corresponding SEM images of CPC/PU films exhibiting blue, green, yellow, orange, and red structural colors by tuning colloidal particles sizes (212–309 nm); (b) Normalized reflection spectra demonstrating bright structural color across the entire visible region; (c) Angle-dependent optical properties of the CPC/PU film and the corresponding reflection spectra; (d) Versatility of the CPC/PU coating applied on diverse substrates: paper, glass, graphite, cement, and wood; (e) Abrasion resistance test of the CPC/PU coating on cement substrate, highlighting its mechanical robustness.

cating large-area, continuous CPC films (Fig. S8 in ESI). Reinforced by the PU network, these films exhibited robust mechanical performance, coupled with brilliant structural coloration. This work provides a viable pathway for the scalable manufacturing of high-performance CPC. The feasibility of fabricating structural color patterns was further explored using a mask-assisted method. As illustrated in Fig. S9 (in ESI), a fine blue dragon pattern and clear green lettering are successfully achieved. Furthermore, the universality of this strategy was demonstrated by extending it to various substrates without any surface pretreatment. As shown in Fig. 3(d), uniform and stable green structural color patterns were successfully constructed on diverse substrates including paper, glass, graphite, cement, and wood. The combination of the mask-assisted technique and PU/CPC system establishes a patterning strategy characterized by operational simplicity, high efficiency, and broad applicability. This approach effectively meets the customized demands for structural color patterns in various application scenarios such as displays, decoration, and anti-counterfeiting.

The mechanical robustness and optical durability of the as-prepared PU/CPC coatings were systematically evaluated through abrasion, water-flushing, and bending tests. As shown in Fig. 3(e), the CPC/PU coating was subjected to 100 reciprocating friction cycles under a 20 g load using #2000 sandpaper. Post-test analysis revealed that the coating largely retained its distinct green structural color. Although slight darkening and minor local scratches were observed, the overall morphology and optical performance remained largely unchanged, indicating that the long-range periodic order of the colloidal lattice is preserved. To further evaluate the mechanical limits of the coating, a more stringent test was performed by increasing the load to 200 g using a #5000 sandpaper (Fig. S10 in ESI). Crucially, no significant delamination or spalling was observed under this elevated stress. These results confirm the excellent abrasion resistance and optical stability of the CPC/PU coatings. Additionally, the adhesion and structural stability under a water-flushing impact were assessed (Fig. S11 in ESI). Samples affixed to the substrate at a 45° angle were washed with a steady water stream for 2 min. During the test, the structural color temporarily shifted from green to yellow-green; however, no delamination or structural damage occurred. Crucially, once the sample was redried, the original green color was fully restored. This reversible color change demonstrates that the microstructure suffered no irreversible damage under the water impact, highlighting the superior waterproofing capability and optical reversibility of the coating. Finally, flexibility and mechanical stability were examined *via* bending tests (Fig. S12 in ESI). The PU/CPC films were subjected to continuous bending at angles ranging from 0° (flat) to 180° (fully folded) for 50 cycles. Throughout the test, the coating remained intact without any signs of cracking, fracture, or delamination. This confirmed that the PU/CPC coating maintained its structural integrity even under severe deformation, exhibiting outstanding flexibility.

To evaluate the long-term stability of the CPC/PU coating, accelerated UV aging tests were conducted in a UV aging chamber (365 nm, 800 W) for up to 6 days (about 144 h) (Fig. S13a in ESI). As illustrated in Fig. S13(b) (in ESI), the experi-

mental setup ensured uniform UV irradiation across the sample surface. The structural and optical stabilities were monitored by tracking the evolution of the reflection peak wavelength and intensity over time (Fig. S13c in ESI). Remarkably, both the peak wavelength (black line) and intensity (red line) remained almost constant throughout the 6-day exposure, indicating that the lattice structure and hydrogen-bonded network remained intact without noticeable photodegradation. Inset of Fig. S13(c) (in ESI) further confirms the visual consistency of the coating before and after aging. Furthermore, the cooling performance of the CPC/PU coating was assessed by monitoring the surface temperature under 1000 W/m² simulated solar irradiation of 1000 W/m² (Fig. S13d in ESI). The temperature profiles of the pristine and aged coatings followed nearly identical trajectories, both stabilizing at approximately 54 °C. This demonstrates that the coating retains its excellent PDC capability and stability even after prolonged UV irradiation. Collectively, these results demonstrate that the PU-enhanced hydrogen-bonding network not only preserves structural color fidelity but also confers exceptional mechanical and environmental robustness, making it a viable candidate for long-term outdoor architectural applications.

PDC Performance Evaluation

As a unique class of photonic metamaterials, CPCs have emerged as promising candidates for PDC by manipulating light-matter interactions at sub-wavelength scales. Ideal PDC coatings must simultaneously reject solar irradiance and enhance thermal emissions.^[40,41] We then characterized the spectral property of the CPC/PU coating in both the solar (0.3–2.5 μm) and atmospheric transparency window (8–13 μm) ranges employing a UV-Vis-NIR and FTIR spectrophotometer, respectively (Fig. 4a). The CPC/PU coating reflected about 63% of the solar radiation based on the narrow-band Bragg reflection of the photonic bandgap. The coating maintained a high emissivity of about 86% in the atmospheric transparency window, which originated from the characteristic IR emission bands. The high reflectivity and IR emissivity of the CPC/PU coating enable it to exhibit exceptional daytime cooling performance. Based on Eqs. (1)–(5), the theoretical cooling power of the CPC/PU coating was calculated to be 28 W/m² under the temperature conditions $T_{\text{atm}}=T_{\text{s}}=298$ K, ignoring thermal convection and conduction ($P_{\text{cond+conv}}=0$). Furthermore, the equilibrium temperature of the CPC/PU coating was obtained by setting $T_{\text{s}}=0$, resulting in 293 K. Consequently, the designed CPC/PU coating could achieve a maximum cooling temperature of 5 °C under idealized, no-convection conditions (Fig. S14 in ESI). In practice, we explored potential cooling applications of the CPC/PU coating. Under simulated solar radiation, we demonstrated the cooling performance of the CPC/PU coating in building cement (Figs. S15 and S16 in ESI).^[42] The surface and chamber temperatures of each sample were monitored using an IR thermal imager during the measurement period. To ensure the reliability of the experimental results, the temperature-tracking data from the IR thermal imager were calibrated against a temperature recorder equipped with a thermocouple. The nearly overlapping temperature profiles from the two independent measurement systems confirmed the high reliability of the experimental results (Fig. S17 in ESI). After exposure to simulated solar irradiation (1000 W/m²) for 80 min, the bare cement substrate reached a

steady-state surface temperature of 73.2 °C. The CPC/PU coating demonstrated 18.9 °C surface cooling for the cement based on its opt-thermal manipulation capability (selective solar reflectance and high infrared emissivity). In contrast, the commercial pigmented coatings brought only 9.5 °C (white) and 6.8 °C (green) cooling temperatures relative to the cement. This result arises from the intrinsic parasitic light absorption and resultant self-heating of conventional pigment-based systems, which compromise their ability to reject solar heat (Fig. 4b and Fig. S18 in ESI). Crucially, this surface cooling could translate into an effective indoor temperature regulation. It was found that the CPC/PU coating effectively cooled the chamber temperatures beneath the cement (Fig. 4c and Fig. S18 in ESI). The chamber temperature of the PU/CPC coating was stabilized at 31.4 °C. This represents a 4.4 °C decrease compared to the bare cement control (35.8 °C) and a 1.3 °C improvement over the commercial

coating (32.7 °C, green). These findings highlight that the integration of CPC/PU coatings onto building envelopes (walls and roofs) can simultaneously satisfy aesthetic color design requirements while substantially mitigating heat transportation, thereby reducing space-cooling energy consumption. Furthermore, we evaluated the actual PDC capability of the CPC/PU coating via outdoor field tests in Nanjing using a self-built setup (Fig. 4d). The CPC/PU coating was placed in an open PS foam box with PE shielding to minimize thermal convection and conduction, and was exposed to direct solar radiation. The ambient temperature was recorded using a thermocouple equipped with shelters to prevent strong convection and solar heating, and the surface temperatures of the CPC/PU coating were monitored using a thermal imager. The apparatus was placed on a platform, 70 cm above the ground.^[43,44] Under an average solar intensity (I_{solar}) of about 538 W/m² in summer, the CPC/PU coat-

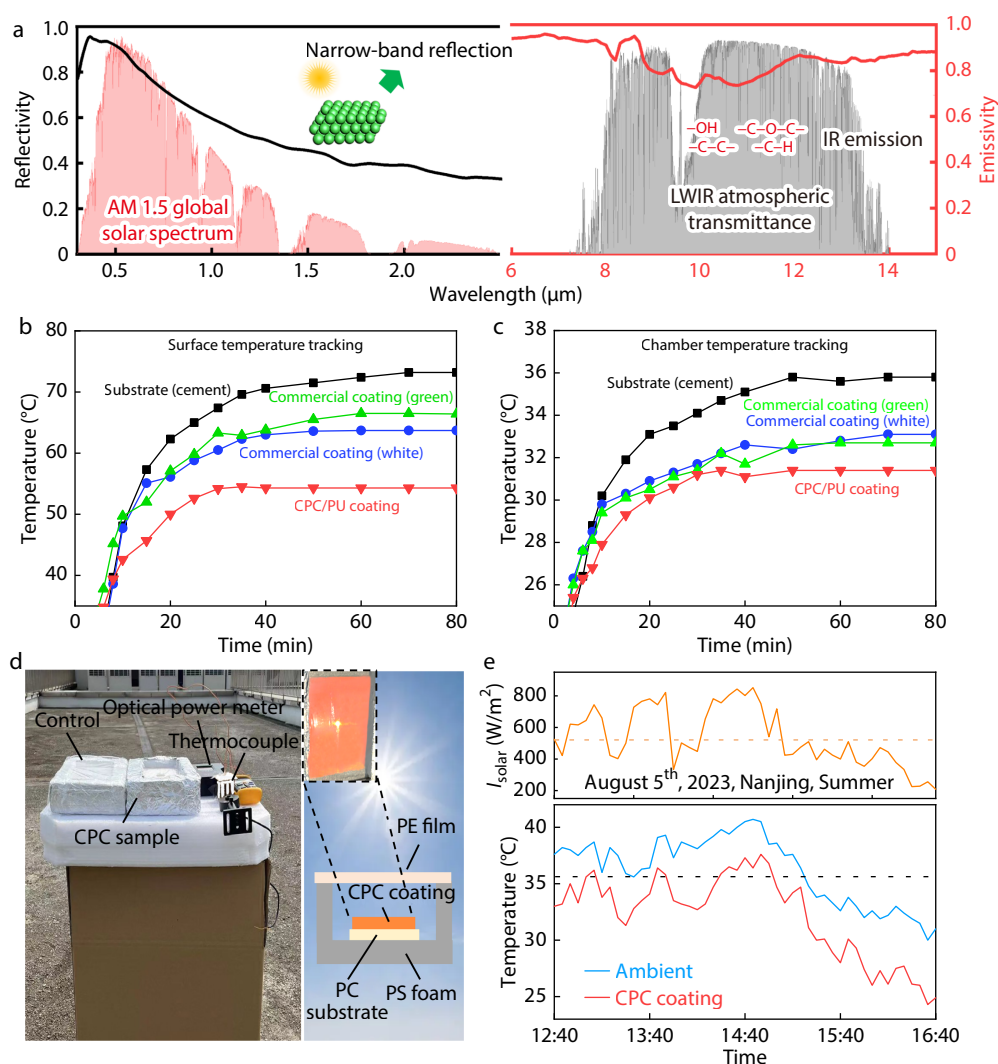


Fig. 4 (a) Solar reflectivity and IR emissivity spectra of the CPC/PU coating across the wavelength range of 0.3–15 μm; Cooling performance evaluation of the CPC/PU coating under 1000 W/m² simulated solar radiation based on (b) surface and (c) chamber temperature tracking; (d) Photograph and schematic of the custom-built setup used for outdoor cooling measurements in Nanjing, China (32°6'N, 118°40'E) of the CPC/PU coating; (e) Real-time temperature profiles of the CPC coating and ambient air during a clear summer day (August 5, 2023). The upper panel shows the I_{solar} with the orange dashed line indicating the average value. The black dashed line represents the ambient temperature recorded by a local meteorological station for reference.

ing achieved a maximum sub-ambient cooling temperature of 6.6 °C (Fig. 4e). This outdoor performance confirms the capacity of the coating for PDC under real-world environmental conditions.

CONCLUSIONS

In this study, we successfully developed a mechanically robust crack-free CPC coating via a hydrogen bond-assisted self-assembly strategy to address the trade-off between optical quality and mechanical durability in large-area CPC. The incorporation of 3 wt% PU establishes a robust hydrogen-bonding network that effectively dissipates capillary stress during the evaporation self-assembly of colloidal particles, yielding centimeter-scale, defect-free films with vivid structural color and retained iridescence. In addition, PU imparts exceptional mechanical stability, enabling the CPC coatings to withstand rigorous abrasion, bending, UV irradiation, and water-flushing impact while preserving the optical and structural properties. Crucially, the CPC/PU coating maintains a high solar reflectance (about 63%) and strong IR emissivity (about 86%), yielding an 18.9 °C surface temperature drop compared to bare cement under 1000 W/m² simulated solar radiation and a maximum sub-ambient cooling of 6.6 °C during outdoor field tests. This study not only provides a facile route to multifunctional photonic materials, but also establishes a viable pathway towards energy-saving building envelopes and sustainable thermal management coatings.

Conflict of Interests

The authors declare no interest conflict.

Electronic Supplementary Information

Electronic supplementary information (ESI) is available free of charge in the online version of this article at <http://doi.org/10.1007/s10118-026-3827-y>.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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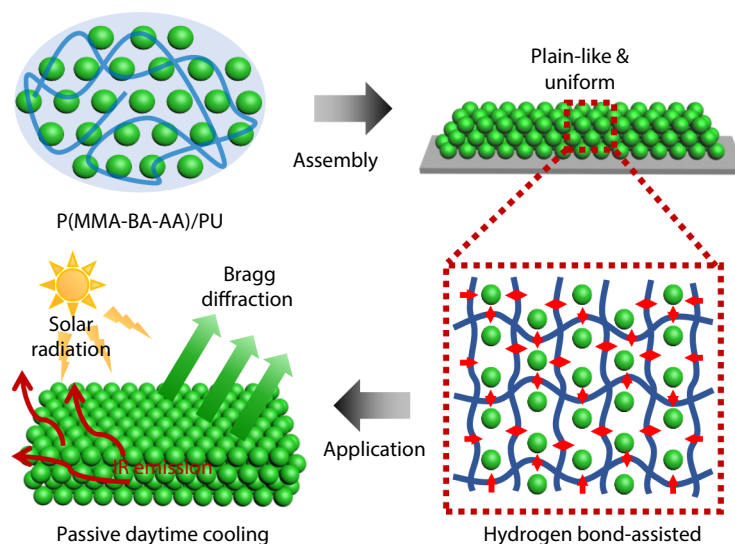
Graphical Abstract

Hydrogen Bond-enabled Crack-free Colloidal Photonic Crystal towards Passive Daytime Cooling

Xiao-Qing Yu, Jia-Xin Hu, Hong-Yuan Zhao, Wei-Lin Wu, Ran Ding, Pei-Li Li, Zi-Rong Li, and Su Chen

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This work reports a facile hydrogen-bond-assisted self-assembly strategy for the fabrication of waterborne polyurethane-enhanced colloidal photonic crystal coatings. This design synergistically delivered crack-free scalability, vivid angle-dependent coloration, robust mechanical stability, and high-performance passive daytime cooling.



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