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Thermochromic Multicolored Photonic Coatings with Light Polarization- and Structural Color-Dependent Changes

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ABSTRACT: Both temperature-responsive and patterned photonic coatings are receiving a lot of attention from a fundamental and application point of view. However, the fabrication of multicolor patterns, which are also temperature-responsive, remains challenging. The current work presents a thermochromic photonic coating with light polarization-dependent multiple structural colors from a polymer-stabilized cholesteric liquid crystal siloxane oligomer. It is found that the structural color and the temperature response can be manipulated by varying the polymerization temperature, allowing the fabrication of thermochromic multicolor patterns. During heating, each color starts shifting at a different temperature until the sample is entirely blue at a high temperature. Upon cooling, the multicolor pattern returns, showing that the thermochromic response is reversible. On top of that, linearly polarized light dependency is also obtained, showing different reflection colors when varying the incident light polarization angles. A multicolor, thermochromic photonic coating with light polarization- and structural color-dependent features is fabricated for demonstration.

KEYWORDS: cholesteric liquid crystals, thermochromic, linearly polarized light, photonic structures, 3D helical structures, patterning

INTRODUCTION

Photonic structures that present selective reflection colors based on alternating refractive indices inside the material are widely found in nature. Multicolor patterns as well as light polarization-dependent color are present in many species. Dynamic photonic structures are also found, like chameleons adapting their skin color to the environment for camouflage and body temperature control. Inspired by nature, research on artificial photonic structures is dedicated to creating colorful patterns as well as thermochromic and light polarization-dependent colors. Such materials are interesting for applications ranging from temperature control in buildings to security features. However, a photonic material combining all these features has not been realized yet.

Cholesteric liquid crystals (ChLCs) are a popular photonic material for fabricating temperature-responsive devices. The nanostructure of ChLCs is helicoidal, organized by the rotation of the anisotropic liquid crystal moieties giving rise to their structural color. The reflection wavelength ($\lambda$) of the structural color is a function of the rotational pitch length ($p$) of the helical structure. Molecular design of ChLC materials allows for $p$ to be responsive to temperature through various mechanisms, with a corresponding shift of the reflection band. ChLC materials can be obtained by industrially feasible coating processes, such as bar coating and printing. Temperature-responsive ChLC coatings have been fabricated with versatile processabilities, a wide color tuning range, variable working temperatures, controllable response speed, reversibility, and good stability. Patterned coatings with static colors have previously been created by photocross-linking thermochromic oligomeric ChLC precursors to fix the color at different temperatures using photomasks or by painting ChLC polymer particles of different colors, with the latter example even presenting an irreversible color shift upon heating. However, a reversible thermochromic reflection band shift in a multicolor patterned ChLC coating has not been realized so far. The difficulty lies in
the conflicting roles of the cross-linked network: the mobility of the mesogens, which is required for a reversible response, is easily lost by cross-linking the material, but at the same time, the cross-linking is used to fix the helical pitches into different colors. Although the printing of encapsulated ChLC droplets containing various ingredients might be viable, it would be more convenient if the thermochromic response can be manipulated within a single formulation. So far, the scientific challenge to fabricate thermochromic coatings with multicolor patterns remains. Linearly polarized light (LPL)-dependent structural colors in ChLCs by deforming the circular helices into elliptical shapes have been reported using achiral oligosiloxane LCs and chiral network monomers (>15 wt %), which feature reflection color dependency of LPL with patternability. However, this system is also not thermochromic.

In this work, we show a thermochromic patterned coating consisting of ChLC siloxane oligomers (ChSiloxane12, Scheme 1a) stabilized by a diacylate LC network. The ChLC siloxane possesses a thermochromic response originating from a smectic A-to-cholesteric (Sm−Ch) pretransitional effect. Structural color patterns can be made via polymerization at different temperatures using photomasks, and a three-color patterned coating was fabricated for demonstration. Remarkably, when heated, each color has its own temperature shift onset, and the initially multicolored coating becomes a single color at higher temperatures. In addition, linearly polarized light dependency can be obtained, and these stratified thermochromic coatings show changing colors when altering the polarization angle of incident LPL. We created a stratified two-color patterned coating for demonstration of its potential toward aesthetics and security applications.

**EXPERIMENTAL SECTION**

**Materials.** The network monomer was purchased from Merck. The initiator 1 (Irgacure 184) was purchased from Ciba Specialty Chemicals, Inc. The dichroic photoinitiator (1-({4′,heptyl-[1,1′:4′,1″-terphenyl]-4-yl})-2-methyl-2-morpholinopropan-1-one) was obtained from Merck. The surfactant (2-(N-ethylperfluoroctanesulfonamide) ethyl methacrylate) was purchased from Acros. The polyimide Optmer AL 10S1 or alternatively AL 1254 was purchased from JSR Micro.

**Synthesis.** The cholesteric ChSiloxane12 was lab synthesized following literature methods and materials. The feed ratio of P1/P2 precursors in the reaction was adjusted to be 88/12 in mol. Yield: 3,219 g, 69.2%.

**Preparation of the Coatings.** In a glass vial were weighted ChSiloxane12 and the network monomers. The surfactant and the initiator were then added into the mixture via diluted tolune solution in volumetric flasks. More tolune was added to dissolve all the components homogeneously. The total solute comprised about 45 wt % of the mixture.

**Preparation of ChLC Mixtures.** A glass vial was weighted ChSiloxane12 and the network monomers. The composition for nonstratified coatings was as follows: ChSiloxane12/network monomer/surfactant/initiator 1 = 94.6/4/1/1 in weight ratio. The composition for stratified coatings was as follows: ChSiloxane12/network monomer/surfactant/initiator 1 = 94/4/1/1 in weight ratio.

**Preparation of Rubbed Polyimide-Coated Glass Substrates.** The glass substrates were of 9 × 9 cm² size, and the procedure of applying a polyimide layer followed the exact same method as reported in the previous study.

**Preparation of the Coatings.** An RK PrintCoat Instruments K control coater was used to prepare all coatings. The example of nonstratified, nonpatterned coatings is given below: On a 9 × 9 cm² size glass substrate with rubbed polyimide was loaded 60 µL of the mixture, and it was placed at 100 °C for 40 min to evaporate the solvent. Afterward, the substrate was transported to the preheated coater, at 52 °C on the substrate surface. The coating was applied using a 60 µm gap (4-sided applicator, 1107/80/1, Sheen), which was pushed forward over the mixture automatically by the coater. The speed of the applicator movement was about 0.5 cm/s in the direction antiparallel to the rubbing direction of the polyimide layer. The coating was then cooled down to the desired temperature (recorded at the substrate surface) and was UV-cured using an EXFO Omnicure S2000 mercury lamp at an intensity of 4.5 mW/cm² (unpolarized) for 1 min in a nitrogen environment followed by postcuring for 4 min. The coating was cooled to room temperature. The entire piece of coating was cut into several pieces for different tests using a glass
For the stratification process, a high-contrast linear polarizer (PUV260C35S from LOT-QuantumDesign GmbH) was placed between the coating and the UV light source during the curing. The polarization direction was perpendicular to the coating direction. The intensity of polarized UV light that reached the coating surface was ~1.5 mW/cm².

For the patterned coatings, a photomask was placed above the coating during each temperature stage of UV curing (Scheme 1a). The photomasks were made from commercial polyimide tape (Kapton) stuck to glass plates and were laser-cut into different patterns. To avoid direct contact with the coating surface, the photomask was attached to the glass substrate using double-sided tape, whose thickness is larger but very close to the coating thickness.

The thickness of all obtained coatings was 22−27 μm as determined by interferometry.

Characterizations. Proton nuclear magnetic resonance (1H NMR) spectra were recorded on a 400 MHz Bruker Avance III HD spectrometer. Gel permeation chromatography (GPC) was performed on a Shimadzu LC-2030C.3D instrument equipped with a PDA-254 nm detector, using tetrahydrofuran (THF) as the eluent and monodisperse polystyrene calibration standards. Differential scanning calorimetry (DSC) curves were measured with a DSC Q2000, TA Instruments. Thicknesses of the coatings were examined using a Forgale Zoomsurf 3D interferometer.

Transmission spectra of unpolarized light were measured using a PerkinElmer LAMBDA 750 UV/vis/NIR spectrophotometer with a 150 mm integrating sphere detector. Temperature control of the samples was realized via a Linkam THMS600 hot stage with a customized aperture diameter of 6 mm. Heating and cooling of the hot stage were programmed using a Linkam T96 controller. For linearly polarized light transmission spectra and the polar plots, the spectrometer was equipped with a linear polarizer.

Transmission spectra of circular polarized light were measured on a Shimadzu UV-3102 PC equipped with a linear polarizer in combination with a quarter-wave plate.

Temperature calibrations for transmission spectra and the photographs followed the literature methods. Due to the differences in calibration methods and due to changes in the environmental temperature, a 1−3 °C difference in temperature designation among each test was expected.

RESULTS AND DISCUSSION

Thermochromic and Multicolor Pattern Coatings. Mixtures containing an oligosiloxane ChLC and a diacrylate LC monomer were prepared for the fabrication of thermochromic, multicolor patterned coatings (Scheme 1a). The siloxane random co-oligomer of mesogenic (P1) and chiral dopants (P2) was synthesized according to a literature method. The molar ratio of P1/P2 is 88/12, which is fine-tuned to make sure that the thermochromic response covers the full range of the visible part of the electromagnetic spectrum, where the shortest wavelength limit is decided by the content of P2. The average backbone length is found to be approximately 6 (Figure S1), with a polydispersity of 1.21 (Figure S2). The oligomer undergoes a ChLC−isotropic transition (Tc) at 65 °C, an Sm−Ch transition (Tc(Sm−Ch)) around 40 °C, and a glass transition (Tg) around ~2 °C (Figure S3). When brought into a planar alignment cell, strong thermochromic behavior can be observed within the cholesteric temperature range until becoming scattering near the end of the DSC transition peak, showing that the Sm−Ch transition takes place (Figure S3). We chose the optimized network ratio of 4 wt %, referring to the amount of diacrylate LC in the mixture, to prepare thermochromic photonic coatings. Furthermore, 1 wt % methacrylate surfactant was added to the mixture to assist planar Ch alignment of the coating at the coating−air interface, as well as 0.4 wt % photoinitiator 1 to initiate network formation. Upon barrier-coating the mixture at 52 °C on a glass substrate with a rubbed polyimide layer, a planarly aligned cholesteric coating was obtained. The Sm−Ch thermochromic response of the oligomer remains, showing blue (470 nm), green (520 nm), orange (585 nm), and red (630 nm) reflection at 52, 47, 42, and 40 °C, respectively (Figure 1a). Via subsequent polymerization of the diacrylate LC monomer under UV light irradiation at these temperatures, polymer-stabilized ChLC coatings are formed. We initially prepared individual single-color coatings cured at 52, 47, 42, and 40 °C, in order to investigate the thermochromic response of the different structural colors.

At room temperature (RT), the polymer-stabilized ChLC coating appears with blue, green, orange, and red color, respectively (Figure 1b). The slight scattering at room temperature can be explained by the formation of twisted smectic A domains, where the smectic order presents helical twists on the larger scale. A slightly longer reflection wavelength than in the uncured state is observed in the spectra due to the broader shape of the reflection bands (Figure 1a and Figure S4). This might be due to a certain degree of phase separation between the siloxane oligomer and the network. DSC curves of the cured coatings all reveal an isotropic transition peak around 63 °C and Sm−Ch transitions around 39 °C, regardless of the curing temperature (Figure S5).

Heating and cooling cycles were performed on these coatings to examine their thermochromic responses. Cycles between 20 (Sm) and 61 °C (isotropic) demonstrate a fully reversible thermochromic shift of these coatings, with each structural color showing its own unique thermochromic responsiveness (Figure 1 and Figure S6). For convenience,
we first discuss the cooling cycles. When cooling from 61 °C (isotropic) to 55 °C (cholesteric), a reflection band appears centered at 402−440 nm (blue) for all these samples. Therefore, the planar Ch alignment is not lost due to the memory of the liquid crystal network. When the samples are further cooled, the reflection color redshifts, following the same response as the uncured mixture until reaching the corresponding curing temperatures. Below that temperature, the shift versus temperature slows down and stalls at some point, except for the red sample, which starts to slow down at 42 °C and only stalls below 35 °C (Figure 1a). Below 35 °C, upon entering the Sm phase, no further response of the band center is observed, but an increase in scattering takes place, thus sealing the plateau of the color shift at blue, green, orange, and red. It seems that the presence of the polymer network inhibits the pretransitional effect, which is present in the uncured mixture at lower temperatures. Interestingly, the RT-red sample that was cured at 40 °C has the largest color change (∼210 nm) changing from red to blue, while for the RT-blue sample cured at 52 °C, the range is the smallest (∼90 nm, light blue to deep blue). During heating, the reverse thermochromic response is observed, reaching a blue reflection at 54 °C and showing disappearance of the color when heated further. However, the major band shift starts later than the curing temperature and has a steeper trend near 54 °C (Figure S7).

Thermochromic color patterns are created via polymerization at different temperatures using photomasks (Scheme 1b). The photomasks were made from polyimide tape (Kapton), which has excellent absorption over the full UV range, via laser-cutting into different patterns. The RT-blue patterns were fabricated at 52 °C using a mask containing “square”, “5”, and “fire (in Chinese)” shapes, while the RT-green patterns were created at 47 °C using a mask having “circle”, “triangle”, and “E” shapes. Finally, the RT-red background was polymerized at 40 °C to receive a three-color pattern at room temperature (Figure 2). Upon increasing the temperature, the RT-red background changes color around 40 °C, while the RT-green and the RT-blue patterns retain their colors until the steep blueshift above 52 °C. At 55 °C, just below the isotropic transition temperature, the background and the patterns appear in a blue color of different shades. Upon reaching the isotropic temperature at 61 °C, the patterns turn mostly colorless with a slight purple reflection, which is assumed to originate from the polymer network mesogens.
under unpolarized light and linearly polarized light at
without postshot adjustments. A polarizer was mounted on top of the coating when taking polarized images.

The colors reappear with cooling from the isotropic, and the reversed thermochromic response of “square-5-fire”, “circle-triangle-E”, and the background is observed, until they settle in blue, green, and red at 52, 47, and 35 °C, respectively. The thermochromic response in each part of the patterned coating’s different areas is identical to the corresponding nonpatterned coatings.

Thermochromic Patterns with Light Polarization-Dependent Color. We introduced LPL dependency features to our photonic coating by stratification using dichroic initiator 2 following our earlier reported method. The dichroic initiator aligns with the liquid crystal molecules and preferentially absorbs light with a polarization direction parallel to its molecular orientation (Scheme 2). As a result, when irradiated by linearly polarized UV, polymerization is initiated in the regions with the same molecular alignment director as the light polarization, which is every half a pitch length. The diacrylate network monomers are polymerized in these initiated regions, causing a diffusion of monomers from the noninitiated regions. The stratification process distorts the helical structures by local variation of the concentration of chiral molecules and forms alternating layers of network-rich and network-poor moieties with different refractive indices along the thickness.

The coating was polymerized with linearly polarized UV light with the polarization direction perpendicular to the coating direction, which from this point on is defined as 0/180° (Figure 3a). When stratified at 42 °C, a red reflecting coating is obtained at room temperature. The unpolarized transmission spectrum of the coating demonstrates an asymmetric reflection band centered at 600 nm with a minor notch at 650 nm (Figure 3b). Compared to the symmetric reflection band of the nonstratified sample cured at the same temperature, this asymmetric reflection band is broader and inclined to higher wavelengths (Figure S9), and therefore, its color at RT appears to be red (Figure 3d). Linearly polarized light transmission of 600 nm at varied polarization angles shows a “peanut-shaped” polar plot (Figure 3b,c, red curve).

Transmission at −45° is the highest (60%), while at +45° transmission is the lowest (17%). The brightness contrast is clearly seen in photographic images under a linear polarizer, at the directions of −45 and +45° (Figure 3d). The transmission data also indicates a reflection of over 50% at +45° polarization, which exceeds the usual reflection limit of a single cholesteric layer. This is attributed to the distortion of the helices via stratification adding additional photonic effects. As the network-rich region is presumably achiral, there is an anisotropy of the refractive index between the x and y axis (Scheme 2). Such anisotropy could function as a retardation layer, which would transform LPL into circular polarized light (CPL). This would cause a decrease or increase of the transmission in the reflection band, depending on the handedness of transformed light. In our case, the transformed light could be ellipsoidal such that the reflection did not reach the 0/100% extremes. Apart from the brightness, the reflection color also slightly changes with the polarization angle due to the changed ratios of reflection band intensity between the two edges of the reflection band (Figure 3b). The RT-red coating retains high reflection of left-handed CPL, while also giving weak reflection of right-handed CPL (Figure S10). The reflected colors under right- and left-handed CPL are different as well.

RT-green and RT-blue samples were successfully prepared as well, via stratification at 47 and 53 °C (Figure 3d), where the uncured mixture appears in green and blue reflection, respectively. The reflection centers at RT in the unpolarized
Figure 4. Images of the stratified patterned coating, showing (a) red and green patterns at room temperature. The red patterns, which were stratified, are hidden into the unstratified background of the same color under unpolarized light and are revealed under a linear polarizer. Both the stratified red and green patterns show variation in optics with rotating the polarization angle, between the darkest reflection angle of −45° and the brightest reflection angle of +45°. (b) At an elevated temperature (46 °C), the angular dependency of the patterns is diminished. The hidden pattern is therefore not clearly revealed with a polarizer. (c) Thermochromic response of the coating at various temperatures during heating and cooling between 20 and 61 °C, under unpolarized light.

spectra are at 525 and 498 nm, respectively. However, the LPL angle dependencies of these samples are less prominent compared to the red sample (Figure 3c and Figure S11). The lowest and highest transmissions of the green coating at its reflection center are 40 and 57%, respectively, whereas for the blue coating, they are only 46 and 50%, respectively. The LPL angular contrast of the RT-green sample is minor, and that of the RT-blue sample is hardly distinguishable by the naked eyes (Figure 3d). The angles showing the darkest and brightest reflection are still −45° and +45°, respectively. We propose that proper stratification is more difficult to achieve for shorter pitches due to a shorter diffusion distance. As the distance between two initiated regions becomes shorter, less monomers are present within half a pitch, and thus, the gradient of network concentration is less between network-rich and network-poor regions, resulting in a reduced LPL dependency.

The stratified coatings present a similar type of thermochromic response compared to the unstratified coatings when viewed with unpolarized light. Major reflection band shifts start above the curing temperatures, and they are fully reversible upon cooling from isotropic (Figure S12). We investigated the LPL dependency at elevated temperatures as well. It is found that the LPL dependency of the RT-red and RT-green samples diminishes at temperatures higher than 35 °C but fully recovers when cooled to room temperature (Figure S13). The temperature at which LPL dependency disappears is around the Sm–Ch transition of the coating. Therefore, it can be inferred that the LPL dependency is highly related to the smectic transition, in which the formed smectic clusters tend to align along the anisotropic direction of the network, extending the deformed areas in the helices.

A patterned coating with stratified structures was fabricated using the combination of a linear UV light polarizer and photomasks. The patterns of “circle”, “triangle”, and “E” were exposed at 47 °C (green color) with LP UV light; then, the patterns of “square”, “S”, and “fire” were exposed at 42 °C (red color) with LP UV light, and finally, the background area was cured at 42 °C with unpolarized UV (Figure 4a). As the “square-S-fire” patterns were cured at the same temperature as the background, these patterns have almost the same color as the background and are therefore hidden with unpolarized light. The hidden red patterns can be clearly revealed under a linear polarizer due to the contrast in color and brightness against the unstratified background. Both the RT-red and RT-green patterns show various shades of color when rotating the LPL direction between −45° and +45°, with good resolution. At elevated temperatures where thermochromic shifts take place, the LPL dependency of both patterns is largely weakened, and the “square-S-fire patterns” are not clearly revealed by LPL anymore (Figure 4b and Figure S14).

The coating shows diverse thermochromic response for the different colored patterns, in the same way compared to the unstratified coatings (Figure 4c). Throughout the temperature cycle, the RT-red patterns remained almost invisible under unpolarized light, proving the highly similar thermochromic response between stratified and unstratified parts. In particular, at 54 °C during cooling, all three parts show a uniform shade of the blue color, and therefore, all patterns are hidden. This is different from the unstratified patterns (Figure 2). We attribute this phenomenon to the broader reflection bands after stratification, whose reflection colors are more impure making it difficult to distinguish among different shades. At the isotropic temperature of 61 °C, the remaining blue color seems to be brighter than the unstratified patterns, which might be due to slight scattering in the sample. The stability of thermooptical performance is proven by repeatable results over three temperature cycles. As the hidden patterns could only be clearly revealed under a linear polarizer at room temperature, an interesting scenario of interactive art and information encryption by this patterned sample is presented.

CONCLUSIONS

We have developed thermochromic photonic coating devices that combine the features of patternability and LPL-dependent structural color. By performing polymerizations at different temperatures (40–52 °C), various reflection colors from red to blue can be obtained. A three-color patterned coating could be prepared, and the initially multicolored coating becomes blue
at higher temperatures, with each color shifting in sequence instead of all colors shifting simultaneously. Linear polarization-dependent color can be achieved, which shows color and brightness contrast of the reflection at room temperature when viewed with LPL between the polarization angles of $-45^\circ$ and $+45^\circ$. The LPL dependency mostly disappears at elevated temperatures, but this is fully reversible. We demonstrate an encrypted coating with some stratified patterns hidden into the unstratified background when viewed with unpolarized light at any temperature. The patterns are only revealed when viewed with LPL at room temperature. These multifunctional coatings would be of interest for products in aesthetics or security purposes. Protective topcoats could feasibly be applied to this ChLC coating to enhance the mechanical strength of the devices.\textsuperscript{38,61,62}

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsapm.1c01434.

Oligomer characterizations, detailed spectral measurements, and photos of the fabricated coatings (PDF)

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Author Contributions

W.Z. performed the investigation, methodology, visualization, and writing of the original draft; A.P.H.J.S. performed conceptualization, supervision, and review and editing of the manuscript; A.J.J.K. performed conceptualization and review and editing of the manuscript; G.Z. performed funding acquisition and project administration; L.T.d.H. performed conceptualization, supervision, and review and editing of the manuscript.

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Notes

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