Liquid Crystalline Cholesteric Reflective Layers for Colored Silicon-Based Solar Cells

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Abstract: The performance of a prototype opaque-type colored silicon-based solar cell integrated with liquid crystalline cholesteric layers is investigated. These devices were developed using only organic components and wet processes, without complicated vacuum processes. The evaluated performances of the prototype solar cells were inferior to those of the other types of previously reported colored solar cells because of the inherent limitations of the cholesteric layers, such as the limited reflectance (~50%), narrow color gamut, and viewing angle-dependent color changes. We propose effective strategies for improving the performance of colored solar cell modules integrated with cholesteric layers.

Keywords: liquid crystals; cholesteric; photovoltaic; solar cells

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Photovoltaic solar cells (SCs) are environment-friendly energy generation devices that have attracted global attention as a solution to environmental issues arising from the use of fossil fuels [1]. Recently, the concept of building-integrated photovoltaics (BIPVs) [2] has been proposed to realize ultimate energy harvesting, thus achieving net-zero energy consumption [3–5]. For BIPV applications, SCs should cover all installable areas on building exteriors, such as windows, rooftops, and facades [6]. Because the colors of building exteriors strongly affect the esthetic and artistic impressions of cities, the color of the SCs is important for BIPV applications [4].

Colored SCs can be categorized into two types: Semitransparent and opaque. In general, semitransparent-colored SCs can be realized by tuning the absorbance of the photoactive layer of the SCs. Thus, SCs such as organic PVs and perovskites can readily tune the absorbance of the photoactive layer. These types of SCs have focused applications such as in energy-harvesting windows and glass roofs [7–10]. Compared to the conventional uncolored black SCs, these semitransparent ones inevitably exhibit a significant power conversion efficiency (PCE) loss owing to the absorbance tuning of their photoactive layers. However, for applications on rooftops and building walls, mainly silicon-based and high-efficiency opaque-type SCs are expected to be used [11].

Opaque SCs with a colorful appearance can be realized by integrating external colored or nanostructured layers with standard SCs [4,12,13]. For example, colored opaque SCs were demonstrated by attaching plasmonic color filters, consisting of an array of metallic subwavelength nanowires, on a transparent substrate of opaque SCs [13]. The reflective colors were modulated by the thickness or width of the metal gratings. Similarly, colored SCs attached to one-dimensional (1D) multi-nanolayered oxide reflective filters of opaque SCs have been reported [4]. These studies motivated us to explore colored opaque SCs with a color filter composed of liquid crystalline materials. In this study, we fabricated a prototype version of colored silicon-based SCs by integrating a reflective color layer using liquid crystalline cholesteric (Ch) phases and performed feasibility tests on these devices. These colored SCs were developed using only organic components and wet processes,

and no complicated vacuum process was employed contrary to the previously reported 1D photonic crystal layer fabrication method. The liquid crystalline Ch phases display a periodic structure originating from self-assembled helical structures and, consequently, can be considered as 1D photonic crystals [14,15]. Such Ch phases selectively reflect circularly polarized light (CPL) with the same handedness as that shown by helical structures. Therefore, within the reflective bandwidth, the Ch phase with right (left)-handed helicoidal structures transmits the left (right)-handed CPL and reflects the right (left)-handed CPL, as illustrated in Figure 1. The selectively reflected light within the reflective bandwidth was observed as colors [16]. Owing to these phenomena, the liquid crystal Ch phases can be used as reflective color filters.

Figure 1. Illustration of the reflective and transmissive optical characteristics of Ch phases.

First, three colored Ch mixtures were fabricated by doping a chiral dopant (Iso-(6OBA)₂ [17], synthesized in our laboratory) in a calamitic nematic liquid crystal (NLC) mixture (HTW109100-100, HCCH). The host NLC showed the following phase sequence during the heating process: Crystal (−40 °C) nematic (112°) isotropic liquid. Red (R), green (G), and blue (B) colored Ch mixtures were obtained by blending the NLC mixture with 4.9, 5.5, and 6.4 wt% chiral dopants, respectively. All the prepared Ch mixtures exhibited a Ch phase below 105 °C. The Ch mixtures were injected into sandwiched cells (cell gaps: 10 μm) comprising two glass substrates. To realize planar alignment in the Ch phases, the glass substrates were subjected to surface rubbing processes. Figure 2a shows the typical polarized optical microscopy images of the R, G, and B Ch layers, captured in the reflective mode. As shown in Figure 2a, the “oily streak” defects indicate planar textures in the R, G, and B Ch layers. Figure 2b shows the typical reflectance curves for the G Ch layers, obtained at different temperatures. The reflectance curves for the R, G, and B Ch layers exhibited constant profiles within the evaluated temperature range (25–65 °C), as shown in Figure 2b. These phenomena were produced by the host NLC mixture that exhibited a wide temperature range of the nematic phase and the constant birefringence within the nematic phase. The reflectance spectra exhibited a full width at half maximum (FWHM) of 75–90 nm, and the peak wavelengths of the R, G, and B Ch layers were 650, 540, and 465 nm, respectively. All the R, G, and B Ch layers showed ~50% reflectance at their respective peak wavelengths. Figure 2c shows the color coordinates of the reflectance spectra of the fabricated R, G, and B Ch layers. The color gamut of the fabricated R, G, and B Ch layers was narrow because of the limited reflectance (~50%) and wide FWHM.

Figure 2. (a) Typical polarized optical microscopy images of the red (R), green (G), and blue (B) Ch layers. The images were captured in the reflective mode. (b) Typical reflectance curves for the G Ch layers, obtained at different temperatures. (c) Color coordinates of the reflectance spectra of the fabricated R, G, and B Ch layers. Here, the reflectance spectra of the fabricated R, G, and B Ch layers were obtained by illuminating the layers with normally incident light.

Figure 3 shows the gradual changes in the reflective spectra of the R, G, and B Ch layers with an increasing viewing angle. In general, 1D phonic crystals change their reflective color when the viewing angle is changed. Therefore, all the spectra were blue-shifted at higher viewing angles, resulting in the realization of iridescence. However, the reflective color change at different viewing angles is not always desirable from an esthetic viewpoint when Ch reflective filters are used in BIPVs. Thus, for utilizing Ch layers as reflective filters, addressing the associated drawbacks such as the limited reflectance, narrow color gamut, and viewing angle dependence of the reflective colors is critical.

Figure 3. Reflectance spectra of the red (R), green (G), and blue (B) Ch layers with an increasing viewing angle.

Subsequently, we integrated the film with a commercially available silicon-based SC module (itemSchool) to investigate the applicability of the fabricated R, G, and B Ch

layers in colored SC modules. To evaluate the device performance, we prepared four modules: SC-Ref and three colored SCs (SC-R, SC-G, and SC-B). Schematic illustrations of the four modules are presented in Figure 4a. SC-Ref, encapsulated on a printed circuit board together with a glass cover substrate (refractive index, $n = 1.55$), was the reference module without the fabricated Ch layers. SC-R, SC-G, and SC-B were the silicon-based SC modules with R, G, and B Ch layers, respectively, integrated onto their front surface using a refractive index-matching fluid ($n = 1.56$). The device performance was evaluated against that of SC-Ref using a solar simulator under AM 1.5 G conditions (100 mWm^{-2}) with a scan rate of 50 mV s^{-1} and scan step of 10 mV at room temperature and relative humidity of 35%. The typical current–voltage (J–V) curves are shown in Figure 4b, along with the transmittance curves of the fabricated R, G, and B Ch layers in the inset. The open-circuit voltage (V_{oc}), short-circuit current density (J_{sc}), fill factor (FF), and PCE of the four SC modules are summarized in Table 1.

Figure 4. (a) Schematic illustrations of the four modules investigated in this study. (b) Typical current–voltage (J–V) curves of the four modules. Transmittance curves of the fabricated R, G, and B Ch layers are depicted in the inset. (c) EQE spectra of the fabricated R, G, and B Ch layers. Photos of the SC-R, SC-G, and SC-B modules are presented in the inset. The EQE spectra were evaluated using the QE measurement system (Newport), which records J_{sc} by applying a monochromatic light on the samples under ambient conditions.

Table 1. Performances of the four modules investigated in this study.

	V_{OC} [V]	J_{SC} [mA/cm ²]	FF [%]	PCE [%]	Relative PCE [%]	PCE Loss [%]
SC-Ref	2.25 ± 0.05	4.65 ± 0.02	70 ± 1	7.43 ± 0.22	100	-
SC-R	2.25 ± 0.03	3.33 ± 0.03	71 ± 1	5.38 ± 0.14	72	28
SC-G	2.27 ± 0.02	3.45 ± 0.02	71 ± 1	5.60 ± 0.12	75	25
SC-B	2.22 ± 0.02	3.57 ± 0.02	71 ± 1	5.68 ± 0.08	77	23

Compared to the SC-Ref, the colored SC modules (SC-R, SC-G, and SC-B), with the fabricated R, G, and B Ch layers attached onto their front surfaces, exhibited large PCE losses of 28%, 25%, and 23%, respectively. The external quantum efficiency (EQE) spectra presented in Figure 4c indicate that these large PCE losses originate from parasitic losses such as the wide reflectance bandwidth of the Ch layers and strong absorption of the Ch layers below 350 nm. Thus, the PCE losses can be significantly reduced by simultaneously reducing the parasitic losses. First, a smaller birefringence of the host NLC is required to reduce the bandwidth. The bandwidth ($\Delta\lambda$) of the selective reflection is denoted as [18,19]

$$\Delta\lambda = \Delta n P \quad (1)$$

where $n = (n_o + n_e)/2$ is the average of the ordinary (n_o) and extraordinary (n_e) refractive indices of the uniaxial host NLC mixture, $\Delta n = n_e - n_o$ is the birefringence, and P is the helical pitch. If P is fixed, then the bandwidth can be tuned to a certain extent by controlling the birefringence of the host NLC mixture. Second, the absorption band of the host NLC mixture should be eliminated to reduce the strong absorption below 350 nm (inset of Figure 4b). That is, the π -conjugation of the constituent molecules in NLC should be shorter. When the Ch layer is sandwiched between two glasses owing to the liquidity of the host NLC mixture, which has a low molecular weight, unexpected reflection at the additional glass interfaces affects the device performance. In addition, the form factors (e.g., shape, size, and other hardware designs) are considerably limited. To address these problems, the Ch layers should be fabricated as thin films without liquidity. Therefore, it is favorable that the host NLC possesses a photoreactive polymerizable moiety that aided in the fabrication of a thin Ch film. Based on our preliminary approaches for optimizing the Ch layers intended for colored SC applications, flexible Ch film integrated-colored SCs with significantly improved PCEs have been developed, and the details will be reported in a separate article.

In summary, a prototype version of colored SCs integrated with liquid crystalline Ch layers was fabricated, and the device performance was evaluated. Liquid crystalline Ch layers can selectively reflect light within the reflectance bandwidth owing to spontaneously organized helical structures. This feature renders the Ch layers as promising candidates for reflective color filters that are applicable in colored SCs. However, the intrinsic shortcomings of the Ch layer, such as the limited reflectance (~50%), narrow color gamut due to a wide bandwidth, and viewing angle dependence of the reflective colors, are unfavorable from an esthetic viewpoint. Moreover, attaching the fabricated R, G, and B Ch layers onto the front surface of the SC modules causes large PCE losses originating from parasitic losses such as the wide reflectance bandwidth of the Ch layer and strong absorption of the Ch layer below 350 nm. Effective strategies for reducing parasitic losses in the Ch layers are suggested in this paper.

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