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An electrically switchable visible to infra-red dual frequency cholesteric liquid crystal light shutter

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Herein, an electrically switchable cholesteric liquid crystal (LC) shutter from visible to infrared light was prepared using a dual frequency LC (DFLC). The fabricated device is semitransparent to visible and IR light in the planar state and opaque in the focal conic state. Both visible and IR lights are transmitted in the homeotropic state. Interestingly, switching among these states is possible by just varying the voltage and frequency. In addition, it is possible to switch between the planar (homeotropic) and homeotropic (planar) states without going to the focal conic state. The proposed device can even regulate the ambient temperature by controlling the sunlight without losing transparency. Thus, the device is expected to have potential applications as smart windows of automobiles and buildings to control the interior temperature and save energy.

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1. Introduction

Liquid-crystal light shutters have attracted significant attention because of their potential applications in display devices as well as switchable windows.¹ Technologies for light shutters are based on either light scattering or light absorption. Light shutters that are based on light scattering include polymerdispersed LCs, polymer-networked LCs, and cholesteric LC (ChLC) devices.²⁻⁹ On the other hand, light shutters that are based on light absorption include electro-chromic devices, suspended-particle devices, and dye-doped LC devices.¹⁰⁻¹⁶ Windows serve an important function in homes and commercial buildings. They control not only light but also heat. They control the access of solar energy to regulate indoor temperature, maintain interior illumination level, and allow view of the outdoors. Windows that can reflect (transmit) a large amount of infrared solar energy in summer (winter) appear to be very attractive.¹⁷ These windows should be transparent over the entire visible light region (400-700 nm) such that extra energy is not required to maintain the illuminance level of the room. Infrared light from the sun spans from 700 nm to ~ 1 mm; however, more than 75% of the energy of infrared light lies

between 700 and 1400 nm.¹⁸ Several devices based on liquid crystals have been proposed to control infrared light.^{19–25}

Cholesteric liquid crystals (ChLC) exhibit many remarkable optical properties due to the existence of a macroscopic helicoidal structure.^{26,27} The most striking features are the exceptionally large optical rotatory power and the selective reflections from the planar Grandjean texture. Selective reflection occurs when the incident light wavelength is of the order of the pitch (p) of the helix such that

$\lambda_0 = np\cos\theta$

where λ_0 is the wavelength of selective reflection maximum, p is the pitch of the helix, n is the average refractive index $((n_e + 2n_o)/3)$, and θ is the angle between the propagation direction and the helix axis. The reflection occurs within the bandwidth $\Delta \lambda = \Delta n p \cos \theta$, where $\Delta n = n_e - n_o$, and n_e and n_o are the extraordinary and ordinary refractive indices of the LC medium, respectively. The pitch of a cholesteric liquid crystal can be adjusted by varying the chiral concentration and is given by the formula $p = 1/(\text{HTP} \times c)$, where HTP and c are the helical twisting power and the concentration of the chiral dopant, respectively. HTP is a characteristic parameter of a chiral dopant and depends slightly on the nematic host.

The arrangement of the LC molecules inside the cell determine their collective optical properties. For example, when the molecules are arranged in a planar (P) alignment that is parallel to the substrate, the layer reflects light of specific wavelengths depending on the pitch and is transparent for the rest of the wavelengths. In a focal conic (F) alignment, the helical structure is preserved, but tilted with respect to the substrate; as a result, more scattering of the incident light in the forward direction occurs.

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Paper

In the homeotropic (H) alignment, where molecules align perpendicular to the substrate, the layer is transparent to all the wavelengths of light. Generally, switching from the P state to the F state is driven by an AC signal in square waves. When a high voltage beyond a critical value is applied, the ChLC will go into the H state. Subsequent transition from the H to P state can be achieved if the field is turned off quickly or to the F state if the high voltage is turned off slowly. In this driving scheme, the transition from the F to P state is accomplished through an intermediate state; *i.e.*, the H state, and the transition time is very long, typically a few seconds, that is too slow for many device applications.²⁸ Therefore, a dual frequency cholesteric liquid crystal that can make the transition from H to P faster than the conventional ChLC due to its material property of frequency revertible dielectric anisotropy is needed.

The dual-frequency nematic liquid crystal exhibits dual dielectric anisotropic properties under applied AC electric fields.^{29–31} For low-frequency voltages, the material has a positive dielectric anisotropy, and the molecules tend to align parallel to the applied field. For high-frequency voltages, it has a negative dielectric anisotropy, and the molecules tend to align perpendicular to the applied electric field. The material is switched directly from the planar texture to the focal conic texture by applying a low frequency and low voltage and switched directly back to the planar texture by applying a high frequency and high voltage.

Most of the windows based on liquid crystals reported in the literature are two-step devices, and switching is either in the normal mode or in the reverse mode.³²⁻³⁹ A polymer-stabilized cholesteric texture light shutter (PSCT) with a dual anisotropic liquid crystal has been studied by Ma et al. The device is switched to a transparent state by a low frequency voltage pulse and to an opaque state by a high frequency voltage pulse. The device remains in the transparent/opaque state after the pulse is removed. They successfully demonstrated the bistability of the device with a zero applied field. However, the switching time of the device is highly dependent on the polymerization process and concentration of the polymer.³² Fast switching from the homeotropic texture to the planar texture using a dual frequency cholesteric light shutter has been demonstrated by Xu et al. With the advent of the special waveform drive scheme, they could achieve a turn off time from the homeotropic texture to the planar texture of less than 100 ms, which is much faster than that of the conventional cholesteric light shutter.33 They also demonstrated a bistable cholesteric reflective display using a dual-frequency cholesteric liquid crystal. The device exhibits two stable states at zero field. It can be switched directly from the planar texture to the focal conic texture by applying a low-frequency voltage and back to the planar texture by applying a high-frequency voltage. They have designed a drive scheme for the display that uses simultaneous application of both low and high-frequency voltages and the cumulative effect of the cholesteric liquid crystals.³⁴ A fastswitching bistable optical intensity modulator is demonstrated by Hsiao et al. using a dual-frequency cholesteric liquid crystal. They achieved direct switching from the scattering focal conic state to the transparent long-pitch planar state and vice versa.

They also demonstrated a very short transition time of 10 ms from the focal conic state to the planar state. The device exhibits bistability at zero voltage.³⁵ Kumar *et al.* have also demonstrated direct switching between a planar (P) state and a focal conic (F) state and *vice versa* in a polymer-free bistable cholesteric light shutter. The functioning of the shutter is based on sign inversion of dielectric anisotropy of the dual frequency liquid crystal (DFLC). They also tested the potential applications of the device for energy-efficient display devices and switchable windows.³⁶

In this study, we present a dual frequency cholesteric liquid crystal light shutter that can switch among the various states of the dual frequency cholesteric liquid crystal by just varying the voltage and frequency. Interestingly, the fabricated device switches between the planar (homeotropic) and homeotropic (planar) state without going to the focal conic state. The planar state is semitransparent to both visible and IR light. On the other hand, the focal conic state is opaque to both wavelengths. The homeotropic state is fully transparent to both wavelengths. By switching to various states, the ambient temperature can be regulated in summer as well as in winter without losing transparency. In other words, we demonstrated the transparency and infra-red light regulation together. Most importantly, it does not involve any mechanically movable parts, polymerization process, special drive schemes, and stacking of several left- and right-handed CLCs, provides instant switching, and is haze free. The transition between optical states can be achieved with optimum voltage and response time. However, to maintain a homeotropic state, voltage needs to be applied continuously. They can be used in high-rise buildings, automobiles, and greenhouse windows to control the energy flow.

2. Materials and methods

2.1. Materials

We prepared a dual frequency cholesteric liquid crystal (DFCLC) mixture by mixing a dual frequency nematic liquid crystal (MDA-00-3969, Merck, Germany) and a chiral dopant (S-811, TCI, Japan) in an appropriate concentration. The liquid crystal exhibits dual frequency with crossover frequency at approximately 15 kHz. At a low frequency (≤ 1 kHz), the liquid crystal exhibits a positive dielectric anisotropy ($\varepsilon = +3.2$), and at a high frequency (≥ 50 kHz), it exhibits a negative dielectric anisotropy ($\varepsilon = -2.7$). The liquid crystal aligns either parallel or perpendicular to the applied electric field depending on the anisotropy. The ordinary (n_0) and extraordinary (n_e) refractive indices of the liquid crystal are 1.4978 and 1.7192, respectively, at a wavelength λ = 589.3 nm. The chiral dopant S-811 possesses left-handedness and helical twisting power (HTP) of 11 μ m⁻¹. It was added to the liquid crystal in a way such that it reflect infrared light (1063 nm). We also added 2 wt% of Irgaphor[®] Black X13 DC dye (BASF, Germany) to the DFCLC mixture.

2.2. LC cell fabrication

A liquid crystal cell was prepared using two indium-tin-oxide (ITO)-coated clean glass substrates. The upper surface of the

ITO was coated with a planar alignment layer PI and cured at a temperature of 230 °C for one hour. Subsequently, the substrates were assembled with 10 µm spacers without rubbing them. The thickness of the cell was measured with a UV-visible spectrometer (S-3100, SCINCO Corp.). The thickness of the cell used for the current study is 10 µm. The cell is placed on a temperature controller and filled with the sample in the isotropic state.

2.3. Characterization

The alignment of the liquid crystal was examined using a polarizing optical microscope (POM, Eclipse E600 POL, Nikon). The images of the bulk cells were obtained by a digital camera (NX1000, Samsung). The wavelength-dependent transmittance was measured by a UV-visible spectrometer (S-3100, SCINCO Corp.) from ultraviolet to near infrared region. An arbitrary function generator (33120A, Hewlett Packard) in conjunction with a high voltage amplifier (HSA 4011, NF Corp.) was used to supply the AC electric field. All electro-optical measurements were performed at a low frequency (1 kHz) and a high frequency (50 kHz). In this study, we present the results of DFCLC with a 2 wt% dye.

Results and discussion

The dye-doped dual frequency cholesteric liquid crystal (DFCLC) is either transparent or opaque to visible and infrared light depending on the voltage and the frequency applied to the cell. The various alignment states of the DFCLC with various voltages and frequencies are shown in Fig. 1. In the planar state, the dye molecules align in the direction of LC. In addition, the dye molecules form a periodic structure with the LC and absorb the arbitrarily polarized light because they are twisted around the helical axis. Therefore, the dye molecules in the planar state absorb more visible light. On the other hand, in the planar state, the DFCLC helix is perpendicular to the substrate, as shown in



Fig. 1 Schematic showing the various states of the dual frequency cholesteric liquid crystal under various voltages and frequencies. (a) Planar state (P), (b) focal conic state (F), and (c) homeotropic state (H).

Fig. 1(a). In this state, it reflects infrared light due to Bragg's reflection. Therefore, when no voltage is applied, the planar state is semitransparent to visible light and infra-red light. When the planar state is subjected to a low voltage and low frequency (1 kHz), it changes to the focal conic state. If the applied voltage is not enough to unwind the helix, the LC molecules try to align in the direction of the applied electric field. As a result, the helices tilt away from the substrate without unwinding the helix. Since the helix is randomly oriented, it scatters visible and infrared light. Therefore, the focal conic state is opaque to visible and infrared light, as shown in Fig. 1(b). When the focal conic state is subjected to low voltage and high frequency (50 kHz), it traces back to the planar state. Since at a high frequency, LC exhibits a negative dielectric anisotropy, the LC molecules orient perpendicular to the applied vertical electric field direction. Hence, it is easy to achieve the planar state from the focal conic state by just decreasing the voltage. In a similar fashion, when a high voltage and low frequency is applied to the focal conic state, it changes to the homeotropic state, where the LC and dye molecules orient perpendicular to the substrates, as shown in Fig. 1(c). In this homeotropic state, the cholesteric helix is unwinding, and LC molecules align in the direction of the applied electric field. In addition, in this state, the LC and dye molecules are aligned perpendicularly to the substrate, allowing most of the arbitrarily polarized light to pass through. Thus, the homeotropic state is transparent to both visible and infrared light. When the voltage is suddenly turned off, the homeotropic state directly reverts to the focal conic state.

On the other hand, when a high voltage and high frequency is applied to the homeotropic state, the homeotropic state directly switches to the planar state without going to the focal conic state. At a high frequency, the LC exhibits a negative dielectric anisotropy. As a result, the LC molecules try to orient perpendicular to the applied field. In other words, the LC molecules orient in the plane of the substrate. Therefore, the planar state is possible when a high voltage and high frequency is applied to the homeotropic state. Interestingly, when a high voltage and low frequency is applied to the planar state, it can directly go to the homeotropic state without going to the focal conic state. At a high voltage, the helix can unwind, and at a low frequency, the LC exhibits a positive dielectric anisotropy. Therefore, it is possible to achieve the homeotropic state from the planar state without going to the focal conic state, as shown in Fig. 1.

The cholesteric textures of the DFCLC with a 2 wt% dye were observed using an optical polarizing microscope and are presented in Fig. 2. All the photomicrographs were obtained at room temperature. The planar state appears light bluish in colour, as shown in Fig. 2(a). It transmits some portion of visible light and infrared light. As the voltage increases to 14 V (f = 1 kHz), the DFCLC changes from the planar state to the focal conic state. At a voltage of 30 V (f = 1 kHz), the whole sample exhibits a focal conic state, as shown in Fig. 2(b). It is evident from the photomicrograph that the focal conic state scatters visible and infra-red light due to randomly oriented helixes. Upon further increasing the voltage, the focal conic



Fig. 2 Photomicrographs of the DFCLC with 2 wt% dye (a) planar state at 0 V, (b) focal conic state at 30 V (f = 1 kHz), and (c) homeotropic state at 70 V (f = 1 kHz).

state becomes a homeotropic state, where the unwinding of the helix takes place. As a result, all the molecules align in the direction of the field and perpendicular to the substrate. In this state, the DFCLC transmits more visible and infrared light as compared to the planar state. The homeotropic state of the DFCLC at a voltage of 70 V (f = 1 kHz) is shown in Fig. 2(c). Various state transitions are observed by increasing and decreasing the voltage and frequency.

3.1. Effect of dye concentration on transmittance

We also studied the effect of the concentration of dye on the transmission of light using a UV-vis spectrometer at room temperature. We measured the transmission spectra of three samples: DFLC with 4 wt% dye, DFCLC with 2 wt% dye, and DFCLC with 4 wt% dye. In case of DFLC with 4 wt% of dye, a homogeneous cell was used. Herein, the director of the nematic DFLC is parallel to the glassy substrate near the walls and in the bulk. The transmission spectra of the three samples are plotted as a function of wavelength and are shown in Fig. 3. The cell filled with DFLC with 4 wt% dye shows absorption in the visible region. It does not show any reflections in the IR region. On the other hand, the cell with DFCLC with 2 wt% dye shows almost the same absorption in the visible region as that with the DFLC with 4 wt% dye. In addition, it shows reflection in the IR region at around 1050 nm. This is due to the Bragg's reflection from the chiral pitch. It is evident from the spectrum that the DFCLC with 2 wt% dye shows a higher transmittance around 800 nm as compared to other samples. In addition, the transmittance of



Fig. 3 Wavelength-dependent transmission of light: DFLC with 4 wt% dye (black line), DFCLC with 2 wt% dye (red line), and DFCLC with 4 wt% dye (blue line).

the sample at 400 nm is higher than that of the other samples. The transmittance of light in the infrared region is nearly 40% higher than that in the visible region. The addition of 4 wt% of dye to the DFCLC significantly reduces the transmittance level in the visible region. However, there is no change in IR reflection. The reason for choosing 2 wt% dye is that it can provide a semitransparent state in a planar state, an opaque state in a focal conic state, and a transparent state in a homeotropic state. If 4 wt% dye is used, the transparency in the planar state is poorer than that obtained with the 2 wt% dye, and if 1 wt% dye is used, the focal conic state is more transparent than that obtained when 2 wt% dye is used. Therefore, we chose 2 wt% dye to achieve three states for different seasons to control the visible and infra-red light simultaneously.

3.2. Voltage-dependent transmittance

We also measured the voltage-dependent average transmittance of the DFCLC in the visible region (400-700 nm) and infrared (900-1100 nm) region. Herein, the average transmittance is the overall transmittance of all the wavelengths of visible light ranging from 400 nm to 700 nm and similar is the case for infra-red light ranging from 900 nm to 1100 nm. The voltage-dependent transmittance in the visible region with the increasing/decreasing voltage and frequency is presented in Fig. 4. When the sample is in the planar state, the transmittance of the visible light is around 20% up to a voltage of 12 V, as shown in Fig. 4. After this, the sample changes from the planar state to the focal conic state. As a result, transmission decreases to around 10%. Since the helixes are randomly oriented in the focal conic state, scattering is greater than transmission. The sample remains in this state between 14 and 40 V. After this, it slowly changes to the homeotropic state. At a voltage of 70 V, it is completely in the homeotropic state, and transmission of light is around 50% in the visible region, which is 30% higher than that in the planar state (the transmittance of light in an LC cell without dye is 71% in our measurement system). When this homeotropic state is subjected to a high frequency, it immediately changes to the planar state due to the negative dielectric anisotropy of the LC. The transmittance



Fig. 4 Voltage-dependent average transmittance of DFCLC with 2 wt% dye in the visible region (400-700 nm).

of the planar state remains constant upon decreasing the voltage, as shown in Fig. 4.

On the other hand, in the infrared region, the transmittance in the planar state is around 35% in the voltage range of 0-12 V, as shown in Fig. 5. After 12 V, the planar state changes to the focal conic state. In this state, the sample scatters more light because of the randomly oriented helices. As a result, the transmittance reaches nearly 10%, as shown in Fig. 5. This behaviour is visible until 40 V. Beyond this voltage, the sample exhibits a homeotropic state. In this state, it transmits nearly 60% of infrared light, which is 25% higher than that in the planar state. When the homeotropic state is subjected to a low frequency with the decreasing voltage, it changes to the focal conic state. As a result, the light transmittance gradually decreases to 10%, as shown in Fig. 5. When a high frequency is applied to the homeotropic state with the decreasing voltage, the sample immediately changes to the planar state. It remains in the same state and shows the same transmittance even upon decreasing the voltage up to 0 V, as shown in Fig. 5.

It is clear from our experimental observation that the transition between optical states can be achieved with an optimum voltage and response time. The voltage required to drive the P state to the F state and the F state to the H state is similar to that reported in previous studies on the same DFCLC or different DFCLC.^{35,36} However, the transition times are different. The turn ON time is defined as the time taken to achieve 10-90% transmittance change with the increasing field, and the turn OFF time is defined as the time taken to achieve 90-10% transmittance change with the decreasing field. We measured the switching time at 1 kHz frequency using the LCMS-2000 instrument (Sesim Photonics Technology Inc., Korea). The measured transition time from the P state to F state is 25 ms and it is the same as reported by Hsaio et al.35 The transition time from the F state to P state is 120 ms, and it is guite slow, as expected, as compared to that from the P state to F state. The transition times between the F and H state and the H and F state are 160 ms and 7 ms, respectively. The transition time from H to F is much faster than that from the F to H state. In this case, we achieved a fast transition time as



Fig. 5 Voltage-dependent average transmittance curves of DFCLC with 2 wt% dye in the infra-red region (900–1100 nm).



Fig. 6 Wavelength-dependent transmittance of DFCLC with 2 wt% dye concentration under an applied voltage.

compared to the results reported by Ma *et al.*³² and Hsaio *et al.*³⁵ However, it is quite slow when switching from F to H as compared to the fast switching of long pitch cholesteric liquid crystal device demonstrated by Kim *et al.*⁹ On the other hand, the transition from the P state to H state takes 108 ms, and that from the H state to F state takes only 2 ms, which is quite fast. Therefore, the switching of the device to various states is much faster than that of the the conventional DFCLC and polymer-stabilized DFCLC.

To study the various working modes of the DFCLC, we measured the transmittance behavior of light under various voltages, as shown in Fig. 6. When there is no voltage, the sample exhibits a planar state, and it is semitransparent to visible and IR light. On the other hand, some portion of the IR light is reflected due to Bragg's reflection from the DFCLC. However, this property disappears when the voltage increases to 30 V (f = 1 kHz). At this voltage, the sample exhibits a focal conic state. As a result, it scatters the infrared and visible light. As the voltage increases to 70 V (f = 1 kHz), the sample will be in the homeotropic state. As a result, it transmits more infrared and visible light as compared to the planar state. The transmittance in the IR is slightly higher than that in the planar state, and the transmittance in the visible range is nearly 25% higher than that in the planar state.

The transparency of various states of DFCLC is observed by obtaining an image of a building through the cell using a camera, as presented in Fig. 7. The building's structure is clearly seen in the planar state, as shown in Fig. 7(a). In the focal conic state, several features of the same building are missing. The building structure is not clearly visible in this state, as shown in Fig. 7(b). In the homeotropic state, the structure of the building is clearly visible as compared to that in the planar state. The structure of the building seen through the homeotropic state is presented in Fig. 7(c).

3.3. Measurement of temperature variations

We also measured the temperature variations in the three states of DFCLC. For this experiment, we fabricated a 3 cm \times 3 cm \times 3 cm miniature house. A laser beam with a wavelength



Fig. 7 The transparency of various states of DFCLC with 2 wt% dye: (a) planar state, (b) focal conic state, and (c) homeotropic state.

of 1064 nm was used to expose the cell inside the house. A thermocouple was used to measure the temperature inside the house. The temperature variations observed in three states of DFCLC as a function of time are presented in Fig. 8. When the cell is in the voltage-off state (that is in the planar state), the temperature inside the house is 20 °C up to 4 min. After this, the temperature steeply increases up to 15 min. Finally, the temperature reaches around 40 °C, and it is maintained until 50 min. It means that the maximum temperature increased by the planar state is 40 °C. The focal conic state (20 V, f = 1 kHz) of the DFCLC shows the same behaviour as the planar state except that the increase in temperature is nearly 2 °C high. In the homeotropic state (60 V, f = 1 kHz), the temperature linearly increases up to 75 °C in 23 min. After this, no change in the temperature is found. It remains the same until 50 min. When the device is in a planar state, it maintains an interior temperature of around 40 °C with good transparency. A similar value of temperature has been reported by Choi et al. with a single CLC cell.²² On the other hand, the focal conic state keeps the interior temperature almost the same as that in the planar state with little transparency.

The homeotropic state increases the temperature up to 75 $^{\circ}$ C with excellent transparency. The device with the planar state is useful in the summer as it keeps the temperature low without losing transparency. In addition, it does not require any additional power. On the other hand, the same device can be used in the homeotropic state during winter. It increases the temperature of the interior and increases the transparency. It only needs power to maintain the homeotropic state. There is no need for gas or electricity-operated floor heaters, which consume a lot of time and money for installation and regular maintenance.



Fig. 8 The time-dependent variation of temperature of DFCLC with 2 wt% dye in various states.

With the advantage of the DFCLC, it is easy to switch between planar and homeotropic states and *vice versa* without going to the focal conic state.

4. Conclusions

We have demonstrated a switchable light shutter that can switch between visible to infra-red and infra-red to visible using a DFCLC. We could control the light transmission and transparency by adding a suitable concentration of dye and chiral dopant to a dual frequency liquid crystal. When it is in the planar state, it is semitransparent to visible and infra-red light, and when it is in focal conic state, it significantly reduces the visible and infra-red light. In addition, in the homeotropic state, it transmits visible and infra-red light. By switching to either planar or focal conic state, it can reduce the light and heat during summer. As a result, the surrounding environment will be cool. Further, in winter, it can switch to the homeotropic state, in which light and heat is transmitted. By switching to various states, we can control the light and heat of the living area. Most importantly, it can also significantly reduce the electricity consumed by air conditioners in the summer and by heaters in the winter. It can be used as windows for cars, high-rise buildings, and homes.

Conflicts of interest

There are no conflicts of interest to declare.

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