



Self-shading by optical or thermal control of transmittance with liquid crystals doped with push-pull azobenzene

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ABSTRACT

Self-switching smart windows, which can be switched between the transparent and opaque states by ambient conditions, have applications in buildings, automobiles, and switchable sunglasses. In particular, technologies for transmittance control without light scattering are highly desirable owing to their ability to control the throughput of sunlight and solar heat through a window without blocking the view. In this study, a self-shading window based on liquid crystals doped with push-pull azobenzene with a haze-free opaque state for transmittance control is demonstrated. For the realization of the haze-free opaque state (or a low-transmittance state), the isotropic phase was used, which can be induced either thermally or optically.

1. Introduction

Technologies for transmittance control with switchable windows have been widely studied for architectural glazing owing to their ability to control the throughput of sunlight and solar heat into a building [1–3]. The switchable windows can provide a comfortable environment for occupants while saving energy for heating, cooling, and artificial lighting [4–9]. In particular, transmittance control without light scattering is high in demand for various applications in buildings, automobiles, and switchable sunglasses. These windows allow people to see through them, which implies that the view is not blocked even when it is darkened.

Switchable windows with electrochromic materials [10,11], liquid crystals (LCs) [12–14], and suspended particles [15,16] can be switched between states electrically. On the other hand, photochromic [17], thermochromic [18,19], and phase change materials [20–22] can be switched between states non-electrically. Among these glazing technologies, windows based on photochromic and thermochromic materials have been widely used as smart windows in architectural glazing and switchable sunglasses because they can be switched automatically by the ambient conditions without any externally applied power or signal [17–19]. However, those chromic glasses have a critical issue in terms of the slow response time, which can vary from several minutes to hours, depending on the panel size. Recently, we developed sunlight-switchable windows using liquid crystals (LCs) doped with push-pull azobenzene materials [23–25]. However, although the transmittance of these devices can be controlled automatically by the ambient conditions within 30 s, they cannot provide a haze-free opaque state because they

scatter the incident light in the translucent and opaque states, which blocks the view.

In this study, a self-shading window with a haze-free opaque state for transmittance control without light scattering is demonstrated. It is based on LCs doped with push-pull azobenzene, which can speed up the thermal relaxation. For the realization of the haze-free opaque state, the isotropic phase was used, which can be induced either thermally or optically. This window can be used for energy saving; it can save the energy used for heating and lighting in the high-transmittance nematic (N) phase and for cooling in the low-transmittance isotropic (I) phase.

2. Cell fabrication

In our self-shading approach for a light shutter, control of the transmittance is achieved by adding push-pull azobenzene HABA (2-(4-hydroxyphenylazo)benzoic acid) to the LC mixture. This azobenzene, which contains two benzene rings with a donor-acceptor group, undergoes a transition from *trans*- to *cis*-azobenzene when triggered by ultraviolet (UV) light; an opposite transition occurs in a dark environment [23–28]. Once the LCs, doped with azobenzene, are irradiated by UV light and the *trans*–*cis* photo-isomerization occurs, the bent shape of the *cis*-azobenzene introduces a molecular disorder in the LC mixture.

A nematic LC, 4'-Pentyl-4-biphenylcarbonitrile (5CB), was mixed with push-pull azobenzene HABA (Fig. 1). A surface-active agent, hexadecyltrimethylammonium bromide (HTAB), was used for the homeotropic boundary condition. In order to achieve absorption of the incident light, the LC mixture was doped with a black dichroic dye mixture S-428, which comprises red azo, yellow azo, and blue

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Nomenclature

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| α_{\parallel} | absorption coefficient parallel to the molecular axis [μm^{-1}] |
| α_{\perp} | absorption coefficient perpendicular to the molecular axis [μm^{-1}] |
| c | dye concentration [wt%] |
| d | cell gap [μm] |
| T_0 | transmittance constant includes effects of the losses [dimensionless] |
| T_h | transmittance of the LC cell in the homeotropic state [dimensionless] |
| T_i | transmittance of the LC cell in the isotropic phase [dimensionless] |

anthraquinone dyes. Measured absorption coefficients α_{\parallel} and α_{\perp} were $1.431 \mu\text{m}^{-1}$ and $0.195 \mu\text{m}^{-1}$ at 550 nm, respectively. All chemicals were purchased from commercial suppliers and used without a further purification. The materials were mixed in a glass vial by stirring continuously for 24 h at room temperature. The mixture was filled to a 20- μm -thick cell through capillary action at room temperature.

Although the planar state of a dye-doped chiral-nematic LC cell has a low transmittance, a chiral-nematic LC cell suffers from several disadvantages including a complex drive scheme, high operating voltage, and slow response time [13]. The I-phase of the dye-doped LC cell can be a better candidate to achieve the haze-free low-transmittance state, compared with the planar state. Although the transmittance of the I-phase is higher than that of the planar state, it is significantly lower than those of other states, such as the homogeneous and twist states [13,29,30].

The transmittances of the LC cells in the homeotropic state and I-phase were calculated as a function of the cell gap, and compared with those measured by using an unpolarized white light source (Xenon lamp light source, Hamamatsu). The transmittances of the LC cells in the homeotropic state and I-phase can be expressed as:

$$T_h = T_0 \exp(-\alpha_{\perp} cd) \quad (1)$$

$$T_i = T_0 \exp\left[-\frac{(\alpha_{\parallel} + 2\alpha_{\perp})}{3} cd\right] \quad (2)$$

respectively [31,32]. The concentration of dye was assumed to be 1.5 wt% in our numerical calculation. The difference in the transmittance between the two states was the highest when the cell gap was $d = 20 \mu\text{m}$, as shown in Fig. 2. Therefore, the cell gap was set to 20 μm .

3. Results and discussion

Our approach to a self-shading light shutter is shown in Fig. 3. The

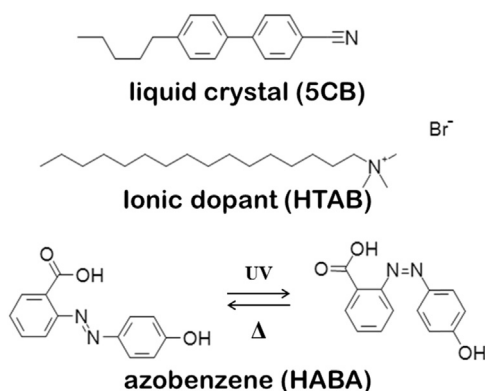


Fig. 1. Chemical structures of the used materials.

phase of the LCs in the fabricated cell can be changed from the N-phase to the I-phase by heating, and vice versa by cooling. Moreover, using UV irradiation, the phase of the LCs can be changed from the N- to the I-phase by *trans-cis* photo-isomerization of the azobenzene molecules. Once the UV irradiation is removed, the LC rapidly relaxes from the I-phase to the N-phase through *cis-trans* back-isomerization of the azobenzene molecules, as the LC mixture contains push-pull azobenzene which exhibits a fast thermal relaxation. In the initial N-phase, the LC and dye molecules are aligned perpendicular to the substrate. This phase is used as a high-transmittance state as the light absorption is minimized. In both thermally- and optically-induced I-phases, the LC and dye molecules are randomly oriented, and both phases can be used as low-transmittance states.

First, the thermal switching of the self-shading light shutter was confirmed. Fig. 4(a) shows the temperature dependence of the transmittance of the fabricated LC cell measured by an unpolarized white light source (Xenon lamp light source, Hamamatsu). The order parameter of the LCs was ignored in Eqs. (1) and (2); however, a change of the order parameter can affect the transmittance of the homeotropic state. With the increase of the temperature, the order parameter decreases, which can decrease the transmittance of the fabricated LC cell. The transmittance abruptly decreased at $\sim 35^\circ\text{C}$, which is close to the N-I phase transition temperature (switching point between the high- and low-transmittance states). The measured transmission spectra of the fabricated LC cell measured using a spectrometer (MCPD 3000, Photal), in the range of 400–700 nm, are shown in Fig. 4(b). A decrease of the transmittance for shorter wavelengths was observed in all states, owing to the absorption properties of push-pull azobenzene HABA. Photographs of the fabricated self-shading light shutter placed on a printed sheet of paper, which are dependent on the temperature, are shown in Fig. 4(c). For the N-phase at 25°C and 30°C , characters behind the panel can be clearly identified, as the fabricated LC cell is in a high-transmittance state. In the I-phase, the fabricated LC cell can provide a low-transmittance state as it absorbs the arbitrarily polarized light without light scattering.

Next, the optical switching of the self-shading light shutter was confirmed. The fabricated LC cell was exposed to unpolarized UV light (365 nm) using a mercury arc lamp (Osram HBO 103 W/2). The N-I phase transition temperature (clearing point) of the LC cell was measured as a function of the HABA concentration, as shown in Fig. 5(a). The measurement of the clearing point was carried out using a Mettler FP82 hot-stage and a Mettler FP90 controller. Before the UV irradiation, the clearing point does not change with the increase of the HABA concentration. Once the mixture is irradiated by UV light and the *trans-cis* photo-isomerization occurs, the bent shape of the *cis*-azobenzene introduces a molecular disorder in the mixture. Therefore, the clearing point is lowered. When exposed to UV light, the clearing point of the mixture (HABA 2 wt%) changes from 35°C to 27°C . It should be noted that the switching temperature of the light shutter depends on the LC material.

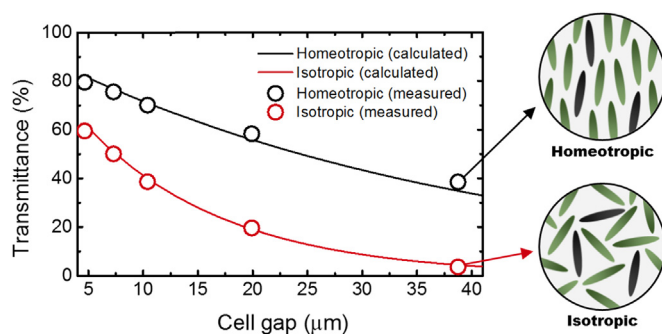


Fig. 2. Transmittances of the LC cells in the homeotropic state and I-phase as a function of the cell gap. The dye concentration was 1.5 wt%.

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