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Polymer Distribution Control of Polymer-Dispersed Liquid Crystals by Uni-Directionally Diffused UV Irradiation Process

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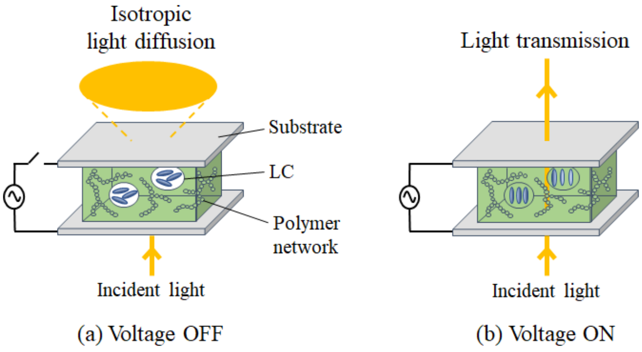
SUMMARY Recently, a control technique of light distribution pattern has become important to improve the functionality and the light utilization efficiency of electronic displays, illumination devices and so on. As a light control technique, polymer-dispersed liquid crystals (PDLCs) have been commonly used so far. However, a precise control of the light diffusion distribution of conventional PDLC has been difficult due to the random polymer network structure, which results in the low light utilization efficiency. On the other hand, reverse-mode PDLCs with homogeneously aligned molecules can anisotropically diffuse light. The reverse-mode PDLC, however, has polarization dependency in the haze value due to homogeneously aligned molecules, which also results in the low light utilization efficiency. Therefore, it is necessary to establish the optimization method of light diffusion distribution without the molecules alignment treatment, and we have proposed a novel PDLC with structure-controlled polymer network which was fabricated by the irradiation with uni-directionally diffused UV light. In this paper, we investigated the effect of the process temperature during UV irradiation on the internal structure and light diffusion distribution of the proposed PDLC. As a result, in case that the mixture during UV irradiation was in isotropic phase, we clarified that the structure-controlled PDLCs with alternating striped LCs/polymer pattern could be obtained because the mixture was sufficiently irradiated with uni-directionally diffused UV light. For the high haze, this structure-controlled PDLC should be fabricated as low temperature as possible with maintaining the mixture in isotropic phase so that the mixture was not a nano-scaled molecular mixing state. Also, this PDLC had no polarization dependency in the haze value and could electrically switch the light distribution pattern between anisotropic light diffusion and light transmission. From the above results, we concluded that the proposed PDLC could precisely control the light diffusion distribution, and realize the high light utilization efficiency.

key words: *light control, polymer-dispersed liquid crystal, polymer network structure, UV curing*

1. Introduction

Recently, a control technique of light distribution pattern has become important to improve the functionality and the light utilization efficiency of electronic displays, illumination devices and so on. In the case of the electronic displays, a wide-distributed light pattern is necessary for the viewing by a plurality of people, whereas a narrow-distributed light pattern is necessary for personal use. The suitable light control for various situations not only enables the protection of user's privacy by the control of viewing angle range but also improves the light utilization efficiency.

As a control technique of light distribution pattern, polymer-dispersed liquid crystals (PDLCs) have been commonly used so far [1]–[4]. This PDLC diffuses the incident light isotropically due to the mismatch of refractive indices between liquid crystal (LC) droplets and polymer network (Fig. 1 (a)). In contrast, it becomes transparent state when an electric field is applied to the PDLC because the ordinary refractive index of the aligned LC molecules along the electric field corresponds with that of the polymer network (Fig. 1 (b)). Based on this principle, PDLCs can electrically switch the light distribution pattern between isotropic light diffusion under voltage off-state and light transmission under voltage on-state. However, a precise control of light diffusion distribution has been difficult due to the random polymer network structure in conventional PDLC [5]–[7], which results in the low light utilization efficiency because it diffuses light to the unnecessary areas. Therefore, the optimization of light diffusion distribution of PDLCs is one of the important challenges to overcome this problem and improve the light utilization efficiency.



(a) Voltage OFF (b) Voltage ON

Fig. 1 Schematic illustrations for operation of conventional PDLC in (a) voltage off-state and (b) voltage on-state.

The reverse-mode PDLCs have been reported so far as a method for the control of light diffusion distribution [8], [9]. The reverse-mode PDLCs are composed of homogeneously aligned reactive mesogens (RMs) and LC molecules. When the electric field is applied to the reverse-mode PDLC, the mismatch of extraordinary refractive indices between RM and LC molecule occurred because the LC molecules are aligned along the electric field. Due to this mismatch of extraordinary refractive indices, the reverse-mode PDLC can anisotropically diffuse light toward the direction perpendicular to optical axes of RM and LC molecules. However, ordinary light straightly transmits since the ordinary refractive index of LC molecule corre-

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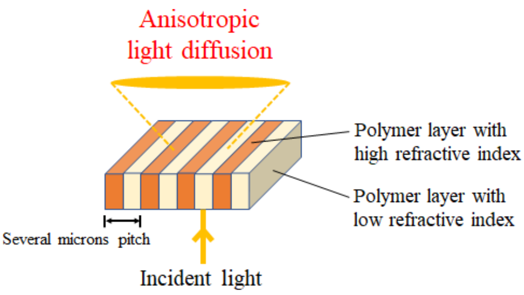


Fig. 2 Schematic illustration of light-diffusing film with alternating polymer layers structure.

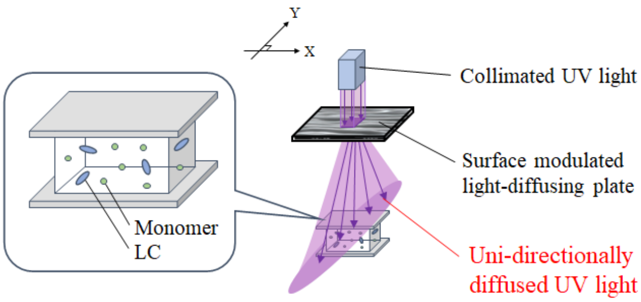
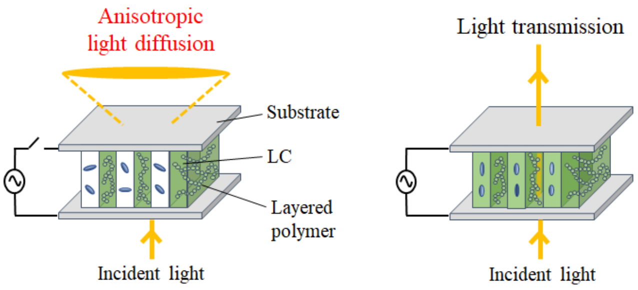


Fig. 3 Schematic illustration of the fabrication method of proposed PDLC.



(a) Voltage OFF

(b) Voltage ON

Fig. 4 Schematic illustration for the operation of proposed PDLC in (a) voltage off-state and (b) voltage on-state.

sponds with that of RM, which results in the low light utilization efficiency.

Therefore, it is necessary to establish the optimization method of light diffusion distribution without the molecules alignment treatment; however, such method has not been established yet. Hence, we have proposed a novel optimization method of the light diffusion distribution of PDLCs by the distribution control of polymer network structure.

2. Distribution Control of Polymer Network Structure

We have previously reported light-diffusing films composed of alternating polymer layers with different refractive indices at a pitch of several microns (Fig. 2) [10]–[12]. This film is fabricated by the polymerization-induced phase separation (PIPS) method and has an optical property of anisotropic light diffusion [13]–[16] due to the diffraction phenomenon in microstructure having the anisotropic distribution of refractive indices [17]. In the PIPS method, a mixture of two kinds of monomers with high and low refractive indices is irradiated with the uni-directionally diffused ultraviolet (UV) light. When the mixture is irradiated with the UV light, the monomer with higher reactivity aggregates at the upper side of the mixture and photopolymerization of the monomer with higher reactivity is promoted under the aggregates. Consequently, the alternating polymer layers structure is formed by separating from the other monomer with lower reactivity [18], [19].

On the basis of this control technique of polymer structure distribution, we have proposed a structure-controlled PDLC with anisotropic distribution of the polymer network structure which was fabricated by the irradiation with uni-directionally diffused UV light (Fig. 3). Due to this anisotropic internal structure, we considered that the proposed PDLC can electrically switch the light distribution pattern between the anisotropic light diffusion under voltage off-state and the light transmission under voltage on-state. (Fig. 4). In general, it has been reported that the phase separation of polymer network and LCs in conventional PDLCs is considerably affected by the temperature of mixture during UV irradiation process [20]–[22]. Therefore, in this paper, we investigated the effect of the process temperature during uni-directionally diffused UV irradiation on the internal structure and light diffusion distribution of the proposed PDLC.

3. Experimental

The fabrication procedure of the proposed PDLC is as follows. We prepared a mixture composed of nematic LCs with a positive dielectric anisotropy (E-7 supplied from Merck Co., Ltd., $n_o = 1.522$, $n_e = 1.746$, $\Delta\varepsilon = 13.8$, $T_{NI} = 59.8$°C) and UV curable monomers (NOA65 supplied from Norland Products Co., Ltd., $n = 1.524$) at a weight ratio of 3 : 2. Next, the mixture was injected to an empty cell which was composed of two polycarbonate substrates by the capillarity phenomenon. The cell gap was 10 μm. The molecules alignment treatment was not carried out on the polycarbonate substrates. Finally, the mixture cell was irradiated with the uni-directionally diffused UV light (non-polarized) adjusted by combination of a collimated UV light source (JATEC Co., Ltd.) and a surface modulated light-diffusing plate (Luminit Co., Ltd.) whose diffusion angle is 60 along the Y-axis direction and 1° along the X- axis direction (Fig. 3). The center wavelength of UV light was 365 nm, and the temperature of mixture during UV irradiation process was controlled from 30°C to 90°C by a hot plate. The integrated amount of UV light was 1.8 J/cm^2 in order to sufficiently polymerize the monomers in mixture.

The internal structure of PDLCs was observed by the polarizing microscope (BX-50, Olympus Co., Ltd.) under crossed-Nicols.

The light diffusion distribution of PDLC was measured by the angle-luminance analyzer (Conoscope, Autronic-

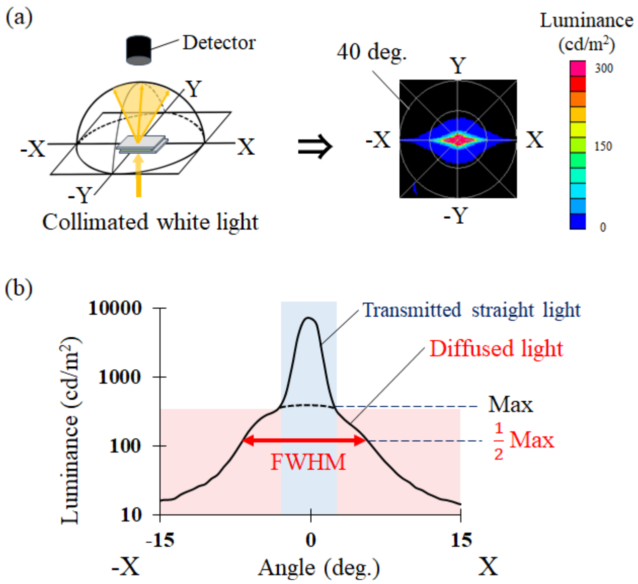


Fig. 5 Evaluation method of (a) light diffusion distribution and (b) FWHM of the diffused light.

MELCHERS GmbH) equipped with a collimated white light source (Fig. 5 (a)). In order to evaluate the anisotropy of light diffusion distribution, we estimated a full width at half maximum (FWHM) from angle-luminance characteristic of the diffused light excluding the transmitted straight light (Fig. 5 (b)), and defined a ratio of FWHM along the X and Y-axis as an ellipticity (Eq. (1)). The light diffusion distribution of PDLC shows isotropic when the ellipticity is equal to 1. If the ellipticity is other value except for 1, the diffused light can be distinguished as anisotropic light diffusion.

$$\text{Ellipticity} = \frac{\text{FWHM along X-axis}}{\text{FWHM along Y-axis}} \tag{1}$$

The amount of the diffused light (haze value) of PDLC was measured by the haze meter (HM-150, Murakami Color Research Laboratory Co., Ltd.). The haze value indicates a ratio of the total light transmittance and the diffused light transmittance and calculated by using following equation (Eq. (2)).

$$\text{Haze (\%)} = \frac{\text{Diffused light transmittance}}{\text{Total light transmittance}} \times 100 \tag{2}$$

4. Results and Discussion

Figure 6 shows the relation between ellipticity of light diffusion distribution and temperature of mixture during UV irradiation process. When the process temperature was lower than 40°C, the light diffusion distribution of PDLCs showed isotropic with the ellipticity of about 1. In contrast, when the process temperature was higher than 42.5°C, the light diffusion distribution of PDLCs showed anisotropic with the ellipticity of more than 1.5. To discuss the relation between light diffusion distribution and internal structure, we observed the PDLC surfaces by using the crossed-Nicols polarizing microscope. The surface images on PDLC are shown in Fig. 7. Here, the presented two kinds of data were examples for isotropic light diffusion at 30°C and anisotropic light diffusion at 60°C. In the case of the process temperature of 30°C, the surface image on PDLC showed the blend structure composed of random polymer network and many LC droplets by polymerization-induced phase separation (Fig. 7 (a)). On the other hand, in the case of the process temperature of 60°C, the surface image on PDLC showed the alternating striped LCs/polymer pattern along the diffusion direction of UV light (Fig. 7 (b)). The alternating dark and bright region of striped pattern under crossed-Nicols polarizer means the presence of many polymers with isotropic refractive index and LCs with birefringence, respectively. Therefore, the formation of layered LCs/polymer structure was indicated by the observation of microscope images.

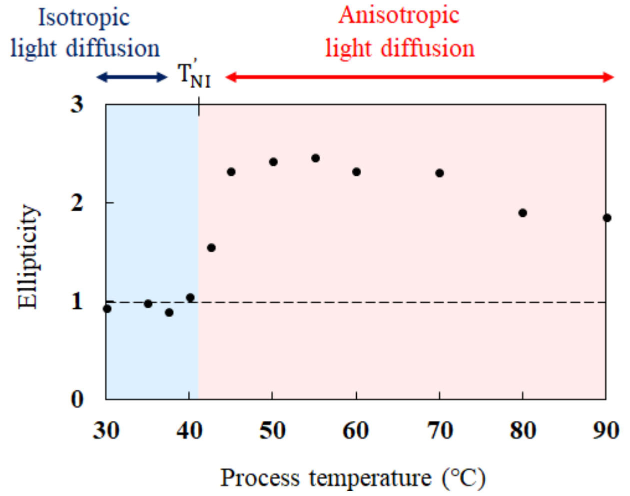
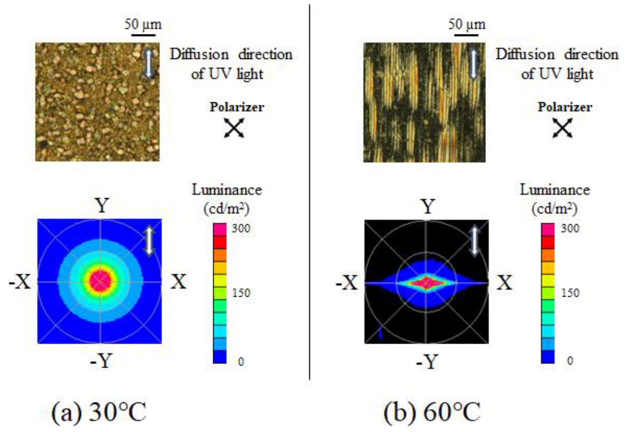


Fig. 6 Process temperature dependency in the ellipticity



(a) 30°C (b) 60°C

Fig. 7 Microscope images of PDLC surface and angle-luminance distributions of the diffused light at process temperature of (a) 30°C and (b) 60°C.

Then, we investigated that the light scattering behavior of mixture during UV irradiation in order to clarify the

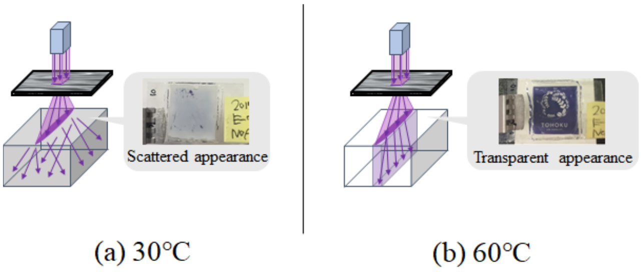


Fig. 8 Appearances of mixture cell during UV irradiation at the process temperature of (a) 30°C and (b) 60°C.

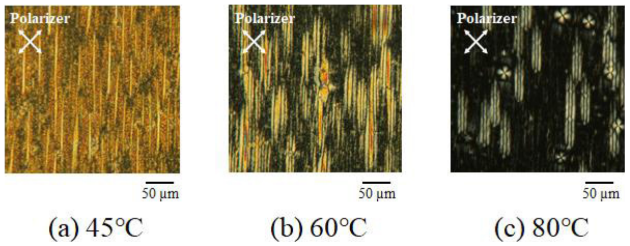


Fig. 10 Microscope images of PDLC surface at process temperature of (a) 45°C, (b) 60°C and (c) 80°C.

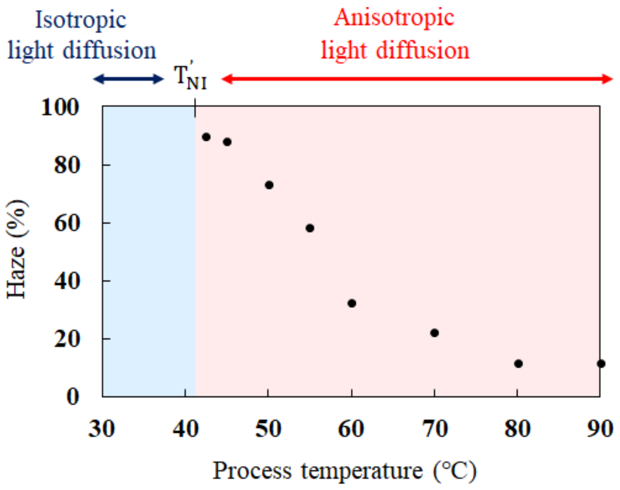


Fig. 9 Process temperature dependency in the haze value.

reason why their structural difference occurred. We placed the mixture cells on a paper with the logo of "*Tohoku University*". When the process temperature was lower than 40°C, the cell was so scattered that the logo could not be seen because the LCs in mixture exhibited nematic phase (Fig. 8 (a)). Therefore, we considered that the structure-controlled PDLC could not be obtained since UV light was scattered and the uni-directionality of UV light was disturbed. In contrast, when the process temperature was higher than 42.5°C, the cell was so transparent that the logo could be seen clearly. This result is attributed to isotropic phase of LC in the mixture (Fig. 8 (b)). Generally, it is reported that the nematic-isotropic phase transition temperature of LCs/polymer (T'_{NI}) is lower than that of pure LCs (T_{NI}) due to the reduction of the order parameter [23], [24]. Hence, we considered that the structure-controlled PDLC could be obtained because the mixture was irradiated with the uni-directionally diffused UV light. From the above results, we clarified that the UV irradiation to the mixture in isotropic phase, here is higher than 42.5°C, enables to control the polymer structure distribution and to optimize the light diffusion distribution.

Figure 9 shows the relation between the haze value of structure-controlled PDLC and the process temperature during UV irradiation. The evaluated haze value of structure-controlled PDLC decreased with increasing the process temperature. To clarify this reason, we observed the PDLC surfaces by using crossed-Nicols polarizing microscope. Three microscopic images at 45°C, 60°C and 80°C are shown in Fig. 10. The microscope image of PDLC at 45°C thoroughly showed the alternating striped LCs/polymer pattern (Fig. 10 (a)). On the other hand, the microscope images of PDLC at higher temperature than 45°C showed the local formation of alternating striped pattern, and the dark region could be quite observed under crossed-Nicols (Figs. 10 (b) and (c)). The dark region on microscope images indicate the transmitted straight light without diffusing, and the wide-distributed dark region resulted in the low haze value.

In general, the compatibility of mixture improves with increasing the ambient temperature. We considered that the polymerization-induced phase separation is hard to occur at high process temperature because LC molecules and monomers become a nano-scaled molecular mixing state by the high compatibility. Consequently, the anisotropic distribution of polymer network structure could not be sufficiently formed, which resulted in the low haze value at high process temperature. From the above results, we clarified that the structure-controlled PDLCs should be fabricated as low temperature as possible with maintaining the mixture in isotropic phase for the high haze value.

Figure 11 shows the polarization dependency in the haze value. The solid line in Fig. 11 indicates the data of structure-controlled PDLC which was fabricated at 45°C. Here, we defined a rotation angle θ as the angle consisted of transmission axis of polarizer and direction along striped pattern of polymer structure. Compared to structure-controlled PDLC, the polarization dependency in the haze value of reverse-mode PDLC (applied voltage is 80 V) is also shown in Fig. 11 as dashed line. In reverse-mode PDLC, we defined the rotation angle θ as the angle consisted of transmission axis of polarizer and alignment direction of molecules. As a result, the structure-controlled PDLC had no polarization dependency in the haze value, whereas the reverse-mode PDLC considerably has polarization dependency in the haze value from 10% to 60%. Therefore, we confirmed that the structure-controlled PDLC enables more efficient light diffusion than the reverse-mode PDLC.

Finally, we demonstrated the electrical switching of light distribution pattern using the proposed PDLC and a monochromatic laser light with a wavelength of 650 nm. Figure 12 shows the photographs of light distribution pat-

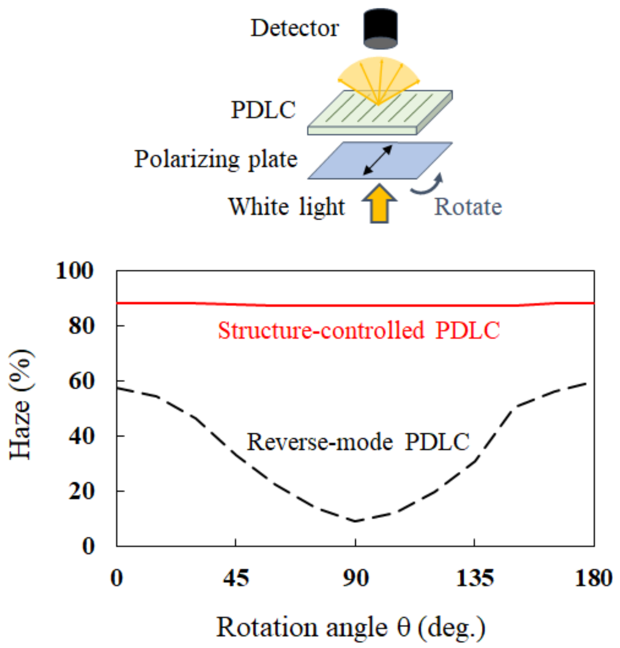


Fig. 11 Polarization dependency in the haze value of structure-controlled PDLC (solid line) and reverse-mode PDLC (dash line).

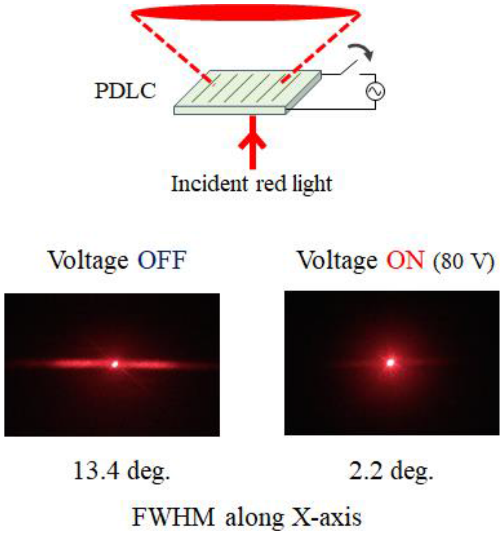


Fig. 12 Electrical switching of light distribution pattern of structure-controlled PDLC

tern of the structure-controlled PDLC under voltage on- and off-states. A red laser light is perpendicularly incident to this PDLC. As a result, the FWHM along X-axis showed 13.4° under voltage off-state and 2.2° under voltage on-state. Therefore, we confirmed that the structure-controlled PDLC can electrically switch the light distribution pattern between the anisotropic light diffusion under voltage off-state and the light transmission under voltage on-state.

5. Conclusions

For a precise control of the light diffusion distribution of PDLC, we have proposed a novel control technique of polymer network structure by the irradiation with uni-directionally diffused UV light, and we investigated the effect of the process temperature during UV irradiation on the internal structure and light diffusion distribution of the proposed PDLC. As a result, we clarified that PDLC with anisotropic distribution of polymer network structure could be obtained by the UV irradiation to mixture in isotropic phase. For the high haze, this structure-controlled PDLC should be fabricated as low temperature as possible with maintaining the mixture in isotropic phase. Also, this PDLC had no polarization dependency in the haze value and could electrically switch the light distribution pattern between anisotropic light diffusion and light transmission. From the above results, we confirmed that the proposed PDLC could precisely control the light diffusion distribution, and realize the high light utilization efficiency.

Acknowledgments

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