Proposal of Novel Temperature-Independent Zero–Zero-Birefringence Polymer for Real-Color Display

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ABSTRACT

In a simple binary copolymerization process, we synthesized temperature-independent zero–zero-birefringence polymer (TIZZBP) films with high heat resistance, sufficient mechanical strength and high transparency. The novel TIZZBP film will be widely used to achieve real-color images not only for vehicle-mounted displays but also flexible displays.

1 INTRODUCTION

Some transparent polymeric materials are used in displays of liquid crystal displays (LCDs) and organic light-emitting displays (OLEDs). Polymeric materials generally have advantage of light weight, flexibility, lower manufacturing cost and easier processing than inorganic glasses. However, polymeric materials exhibit birefringence, which degrades image quality of displays. Therefore, polymeric materials with no birefringence are significantly required to develop real-color displays.

Birefringence is typically classified into two types of orientational birefringence and photoelastic birefringence. Orientational birefringence is caused by orientation of polymer main chain in heat stretching process above glass transition temperature (T_g). On the other hand, photoelastic birefringence is caused by elastic deformation below T_g. Both types of birefringence are compensated by copolymerization of positive birefringent monomers and negative birefringent monomers. We have designed and synthesized zero-zero-birefringence polymer (ZZBP) in which both types of birefringence are compensated to be zero, by using a random copolymerization method[1].

Recently, we demonstrated that orientational birefringence depends on temperature. Temperature dependency of orientational birefringence becomes a problem in vehicle-mounted displays because vehicle-mounted displays are used in the environment in which an ambient temperature changes largely. Therefore, it is required to suppress temperature dependency of orientational birefringence in addition to both types of birefringence. We successfully synthesized the polymers that exhibit no birefringence over a wide temperature range: temperature-independent zero–zero-birefringence polymers (TIZZBPs)[2]. However, these conventional TIZZBPs are not practical because of too complex composition and insufficient mechanical strength.

In this research, we propose a very practical TIZZBP in a simple binary copolymerization system. The novel TIZZBP exhibits high heat resistance, sufficient mechanical strength and high transparency. The TIZZBP film will be invaluable not only for vehicle-mounted displays but also for flexible displays.

2 EXPERIMENT

2.1 Preparation of polymer films

We synthesized a novel TIZZBP in a binary copolymerization system of commodity monomers by bulk polymerization method. In this binary system, its photoelastic birefringence is decreased by specific interactions between M1 unit and M2 unit. This synthesized polymer was dissolved in dichloromethane and precipitated in methanol for purification. We fabricated polymer films by using solvent casting method. To compare with this novel TIZZBP film, two kinds of polymer film, poly (methyl methacrylate) (PMMA) and polycarbonate (PC), were also fabricated by using solvent casting method. PMMA and PC are widely used in optical devices for reasons such as high transparency and excellent mechanical strength.

2.2 Evaluation of birefringence characteristics

Polymer films were uniaxially heat drawn above their T_gs by using a film biaxially stretching machine (IMC-C513, Imoto Machinery Corp., Ltd.). The degree of orientation of main chains was determined from an infrared dichroic ratio measured by using a polarized Fourier-transform infrared spectrometer (7000e FT-IR, Varian Inc.). The orientational birefringence of the uniaxially drawn films was measured by utilizing an optical heterodyne interferometry at a wavelength of 633 nm using a birefringence measurement device (ABR-10A, Uniopt Corp., Ltd.). Photoelastic birefringence of polymer films was measured at various levels of uniaxial tensile stress (about 1% strain) at room temperature by optical heterodyne interferometry at a wavelength of 633 nm using the ABR-10A.

To evaluate the temperature dependency of orientational birefringence, we measured the orientational birefringence of the unixially drawn films by utilizing the ABR-10A when the temperature was changed from 15°C to 70°C at an interval of 5°C.

2.3 Evaluation of heat-resistance, mechanical strength and transparency

Grass transition temperature (T_g) of polymers, which is one of the indexes of heat-resistance, was measured by using a differential scanning calorimeter (DSC-60, Shimadzu Corp.). Tensile strength of polymer films, which is one of the indexes of mechanical strength, was measured by utilizing a universal tensile testing machine (Tensilon RTC-1210A, A&D Co., Ltd.). Total light transmittance and HAZE value of polymer films, which are indexes of transparency, were measured by utilizing a spectral haze meter (SH 700, Nippon Denshoku Corp.).

3 RESULT AND DISCUSSION

3.1 Analysis of birefringence characteristics

We evaluated orientational birefringence (Δn_{or}) and photoelastic birefringence (Δn_{ph}). Figure 1 (a) shows the relationships between Δn_{or} and degree of orientation of a novel TIZZBP, PMMA and PC. Figure 1 (b) shows the relationships between Δn_{ph} and stress of a novel TIZZBP, PMMA and PC. As shown in Figs. 1 (a) and 1 (b), the novel TIZZBP exhibited a very low orientational birefringence and a very low photoelastic birefringence. Intrinsic birefringence (Δn_0) is the indicators of Δn_{or}. The Δn_0 of each copolymer is defined by the equation (1).

$$\Delta n_{or} = \Delta n^0 \cdot f \qquad (1)$$

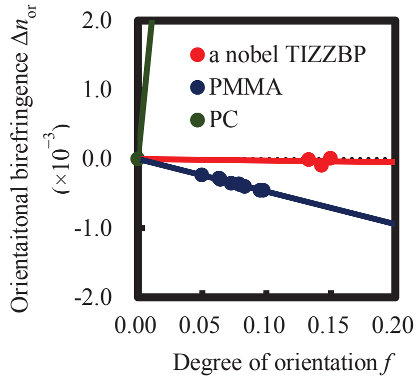
Here, Δn_{or}, Δn^0 and f are orientational birefringence, intrinsic birefringence and degree of orientation, respectively. Photoelastic coefficient (C) is the indicators of Δn_{ph}. The C of each copolymer is defined by the equation (2).

$$\Delta n_{ph} = C \cdot \sigma \qquad (2)$$

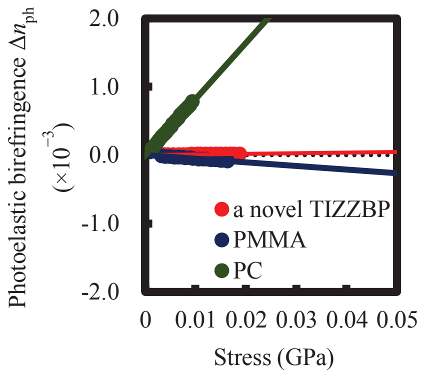
Here, Δn_{ph}, C and σ are photoelastic birefringence, photoelastic coefficient and stress, respectively. We estimated Δn^0 and C of each polymer film from the results of Figs. 1 (a) and 1 (b). The results are shown in Table 1. As shown in Table 1, $|\Delta n^0|$ and $|C|$ of the novel TIZZBP are less than 1×10^{-3} and 1×10^{-12} Pa^{-1}, respectively; these are negligible levels for displays.

Furthermore, we evaluated temperature dependency of Δn_{or}. As shown in Fig. 2, the novel TIZZBP exhibits low temperature dependency of intrinsic birefringence in comparison with PMMA and PC, and its Δn^0 is suppressed to a negligible level over wide temperature range. Temperature coefficient of intrinsic birefringence ($d\Delta n^0/dT$) is the indicator of temperature dependency of Δn_{or} and $d\Delta n^0/dT$ is defined as a change amount of intrinsic birefringent per unit temperature. We estimated $d\Delta n^0/dT$ of each polymer films from the results of Fig. 2. The results are shown in Table 1. A $|d\Delta n^0/dT|$ of the novel TIZZBP is less than 1×10^{-5}°C^{-1}; this is negligible level for displays as with Δn^0 and C.

From the above, the novel TIZZBP exhibits very low birefringence and very small temperature dependency of birefringence. Therefore, the novel TIZZBP can achieve a real-color display over a wide temperature range.



(a) The relationships between orientaitonal birefringence and degree of orientation.



(b) The relationships between photoelastic birefringence and stress.

Fig. 1 Birefringence properties of a novel TIZZBP, PMMA and PC. (a) The relationships between orientaitonal birefringence and degree of orientation. (b) The relationships between photoelastic birefringence and stress.

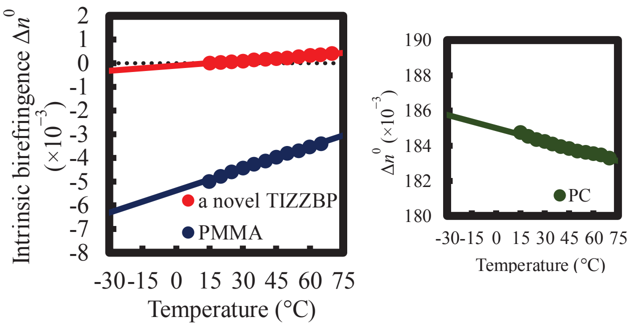


Fig. 2 The relationships between intrinsic birefringence and temperature of a novel TIZZBP, PMMA and PC which degree of orientation f at room temperature are 0.15, 0.07 and 0.14, respectively.

Table. 1 Birefringence characteristics of a novel TIZZBP, PMMA and PC.

Polymer	Δn^0 ($\times 10^{-3}$)	C ($\times 10^{-12}$Pa^{-1})	$\mathrm{d}\Delta n^0/\mathrm{d}T$ ($\times 10^{-5}$°C^{-1})
a novel TIZZBP	−0.23	0.90	0.72
PMMA	−4.88	−5.31	3.11
PC	189.31	85.73	−3.87

3.2 Evaluate of heat-resistance, mechanical strength and transparency

We evaluated glass transition temperature (T_g) of a novel TIZZBP, PMMA and PC. The results are shown in Table 2. The T_g of the novel TIZZBP is about 50 °C higher than PC, which is generally used as a high heat resistant polymer. Based on the result, it is considered that the novel TIZZBP has a very high heat resistance.

Table. 2 The T_gs of a novel TIZZBP, PMMA and PC.

Polymer	T_g (°C)
a novel TIZZBP	194
PMMA	126
PC	147

Tensile strength (TS) of a novel TIZZBP is 32 MPa. It is a little weaker than PMMA and PC (TS (PMMA) = 42 MPa, TS (PC) = 45 MPa). However, we have assumed that the mechanical strength of a novel TIZZBP is enough for optical polymer films of displays.

Total light transmittance and Haze of a novel TIZZBP are 91.5% and 1.52%, respectively. Those of PMMA are 92.3% and 0.86% and, those of PC are 90.7% and 1.02%, respectively. The results show that a novel TIZZBP has high transparency approximately equivalent to PMMA, PC. Figure 3 shows transparency of a polymer film and a polymer bulk of the novel TIZZBP. As shown in Fig. 3, the novel TIZZBP exhibits a very excellent transparency.

This novel TIZZBP exhibits very high heat resistance, sufficient mechanical strength and high transparency. Therefore, this polymer film is applicable to various displays such as vehicle-mounted displays and flexible displays.

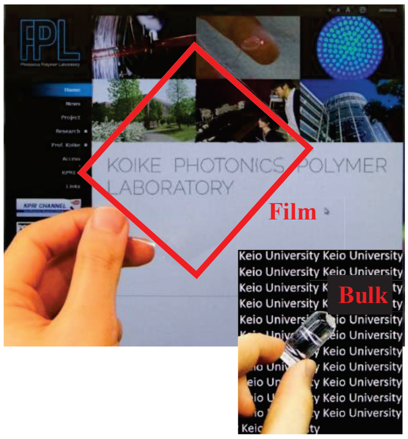


Fig. 3 The image of a polymer film and bulk of a novel TIZZBP.

4 CONCLUSIONS

We proposed the simple binary TIZZBP, which exhibits no birefringence over wide temperature range. Moreover, the novel TIZZBP exhibits high heat resistance, sufficient mechanical strength and high transparency. Therefore, the novel TIZZBP will achieve real-color images not only for vehicle-mounted LCD and OLED but also for flexible LCD and OLED.

REFFERENCES

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