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Polystyrene Blend Alignment Layers for Pretilt Angle Control of Liquid Crystal

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We prepared blend alignment layer based on polystyrene containing 7-hydroxycoumarin (P7COU) and polystyrene containing 2-naphthoxy (P2NAP) side groups for liquid crystal (LC) alignment. The LC cells made from the rubbed P7COU/P2NAP blended films exhibited a shift of the LC alignment direction by approximately 20° compared with those made from a polystyrene film. Stable and homogeneous planar LC cells having pretilt angles adjustable from 0° to 5° were obtained from these polymer films having molar contents of P7COU and P2NAP in the ranges of 100–80 and 0–20 mole%, respectively. The LC alignment behavior was well correlated with the wettability of the polymer films.

Keywords Alignment; liquid crystal; polystyrene; pretilt angle

Introduction

The pretilt angle control of liquid crystal (LC) has been extensively studied due to the scientific and technical interest in liquid crystal displays (LCDs). In particular, it is desired to improve image quality and electro-optical (E-O) performance. Recently, various techniques have been developed to control the pretilt angle of LCs over a wide range on the alignment layer surfaces, so as to be applied in several LCD modes; rubbing techniques of the polyimide surfaces [1, 2], photoirradiation using the photosensitive alignment layer [3], ion beam techniques of the organic/inorganic alignment layer [4–7], mixing of two kinds of polyimides for planar and vertical LC alignment [8–11], stacking of the alignment layers [12–14], and nano and/or micro-structured patterns using lithographic techniques [15,16]. Recently, Rosenblatt et al. and we reported that pretilt angles on polyimide films can be controlled by changing the baking temperature of the polyimide films from 230 to 290°C, rubbing strength, and composition of the blends [11,17–20].

Recently, we have found that polystyrene derivatives containing 7-hydroxycoumarin (P7COU) exhibits enhanced planar LC alignment on rubbed film [21] and polystyrene derivatives containing 2-naphthoxy side group (P2NAP) show homeotropic LC alignment [22], respectively. In this study, we control the pretilt angle of the LC on these polystyrene

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blend films, P7COU having planar alignment and P2NAP having homeotropic alignment, using the rubbing method. Moreover, changes of the pretilt angles were found to be strongly related to changes of the surface properties. The surface properties of polystyrene films giving different pretilt angles were investigated.

Experimental Part

Materials

Polystyrene derivatives containing 7-hydroxycoumarin (P7COU of $\overline{M}_n = 41\,000$ and $\overline{M}_w/\overline{M}_n = 2.34$, $T_g = 132$) [21] and polystyrene derivatives containing 2-naphthoxy (P2NAP of $\overline{M}_n = 38\,900$ and $\overline{M}_w/\overline{M}_n = 2.21$, $T_g = 82$) side groups were synthesized using same method as described previously [22]. All other reagents and solvents were used as received.

Polymer Blend and Film Preparation

Polymer blends were prepared by precipitating 5 wt% solution containing a certain weight percent of each component (P7COU and P2NAP) into isopropyl alcohol. Polymer blends (P7COU and P2NAP) were prepared from chloroform solutions. Blended polymers were dissolved in CHCl_3 , these solutions were filtered using a PTFE membrane of pore size $0.45\ \mu\text{m}$. Thin films of the polymers were prepared by spin-coating (2,000 rpm, 30 sec) onto $1.5 \times 1.5\ \text{cm}^2$ ITO coated glass substrates.

Alignment Process and LC Cell Assembly

The polymer films were rubbed using a rubbing machine (RMS-50-M, Nam Il Optical Components Corp.). The rubbing density equation is written as $L/l = N[(2\pi rn/60v)-1]$, where L is the total length of the rubbing cloth (mm), l is the contact length of the circumference of the rubbing roller (mm), N is the cumulative number of rubbings, n is the speed (rpm) of the rubbing roller, r is the radius (cm) of rubbing roller, and the v is the velocity (cm/s) of the substrate stage [23]. All of the twisted nematic (TN) and antiparallel LC cells were fabricated using rubbed polymer film onto ITO-coated glass slides. TN and antiparallel LC cell were fabricated by assembling polymer films together orthogonally and antiparallel with respect to the rubbing direction using spacers of thicknesses of 6.5 and 50 μm , respectively. The LC cells were filled with a nematic LC, 4-*n*-pentyl-4'-cyanobiphenyl (5CB, $n_e = 1.736$, $n_o = 1.5442$, and $\Delta\epsilon = 14.5$, where n_e , n_o , and $\Delta\epsilon$ represent extraordinary refractive indexes, ordinary refractive indexes, and dielectric anisotropy, respectively), in an isotropic state in order to avoid creating flow alignment through capillary action, and were then sealed with epoxy.

Instrumentation

^1H NMR measurement was carried out on a JEOL-LA at 300 MHz. Gel permeation chromatography (GPC) was used to measure the molecular weight (\overline{M}_n) and molecular weight distributions ($\overline{M}_w/\overline{M}_n$) of synthesized polymer with respect to polystyrene standards using chloroform (CHCl_3) as an eluent and UV detector at 254 nm. Differential scanning calorimeter (DSC) measurements were carried out on TA instruments 2920 at a heating and

cooling rate of 20°C/min under a nitrogen atmosphere. The E-O property of LC alignment was investigated using optical apparatus equipped with He-Ne laser, polarizer, analyzer, and photodiode detector. The LC alignment direction of the antiparallel LC cells was investigated by measuring the angular dependence of the absorbance of a dichroic dye (disperse blue 1) was dissolved at 1 wt% in 5CB using an optical apparatus equipped with a He-Ne laser, a polarizer, and a photodiode detector as a function of rotation angle of samples [23]. The pretilt angle of antiparallel LC cell was measured by rotating the LC cell along the LC alignment direction (a shift by approximately 20° with respect to the rubbing direction) by the crystal rotation method [24]. The voltage-transmittance (V-T) was measured from the LC cell using the same method as that reported by others [25,26]. The threshold voltage (V_{th}) in the V-T curve are defined as the voltages at which the transmittance was increased to 10% of the initial transmittance value [25,26]. The rising (T_r) and falling (T_f) response times for the white-to-black and black-to-white changes, respectively, are defined as the time to transition from 10% to 90% transmittance and vice versa [25,26]. The total response time (T_t) is determined by the average of T_r and T_f . The cell gap was measured before LC filling using spectrophotometer (Ocean optics Inc., S 2000). Polarized optical microscopy (POM) images of LC cell were taken from an optical microscope (Nikon, ECLIPSE E600 POL) equipped with crossed polarizer and digital camera (Nikon, COOLPIX995).

Results and Discussion

Figure 1 shows the synthetic route of polystyrene derivatives containing coumarin side groups (P7COU) and polystyrene derivatives containing 2-naphthoxy side groups (P2NAP), respectively. Poly(chloromethylstyrene) (PCMS) was obtained by free radical polymerization of 4-chloromethylstyrene using AIBN as an initiator in THF and then P7COU and P2NAP were obtained by the reaction of PCMS with 7-hydroxycoumarin and 2-naphthol in *N,N'*-dimethylacetamide using polymer analogous reactions in previous reports, respectively [21,22]. Molar content of 7-hydroxycoumarin and 2-naphthoxy side group of the polymers were about 82% and 100%, respectively.

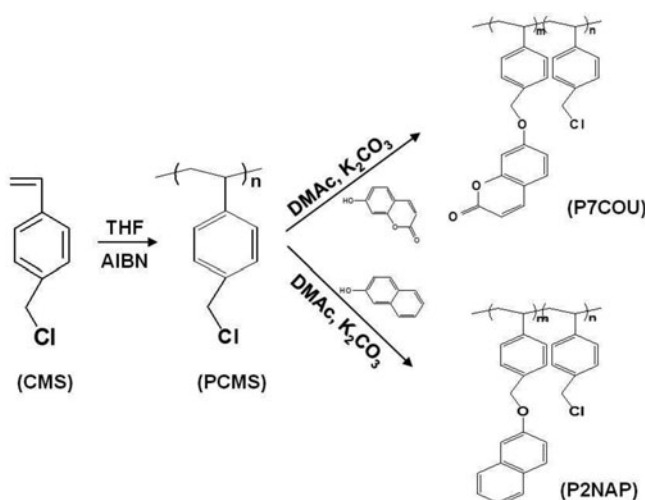


Figure 1. Synthetic route of polystyrene derivatives containing 7-hydroxycoumarin (P7COU) and polystyrene derivatives containing 2-naphthoxy side groups (P2NAP).

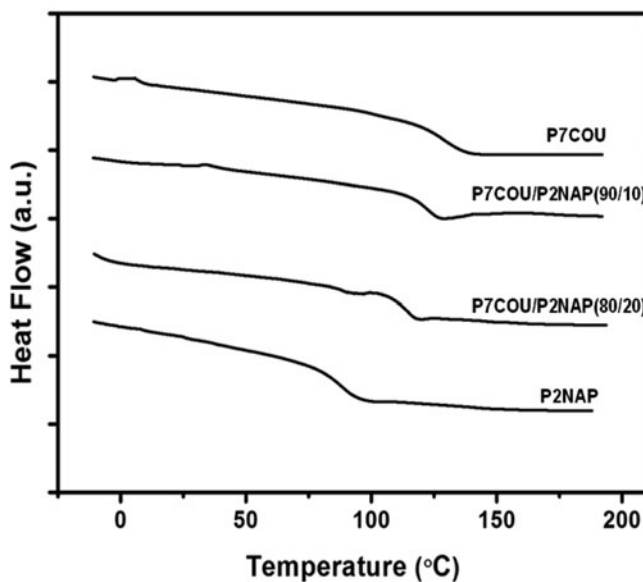


Figure 2. DSC thermogram of the polymers.

As shown in Fig. 2, T_g (132 °C) of P7COU was measured by attaching polar 7-hydroxycoumarin side groups, where the polar coumarin side groups undergo dipole-dipole interactions. T_g (82 °C) of P2NAP was observed by attaching 2-naphthoxy side chains, where the less polar 2-naphthoxy side groups can give a low chance to undergo dipole-dipole interactions, which acts as a plasticizer. The P7COU/P2NAP(90/10) and P7COU/P2NAP(80/20) blend shows one T_g at 113.0 and 122.0 °C, which is situated between the T_g 's of the individual polymers, P7COU and P2NAP, viz. 132 and 82 °C, respectively, indicating that these two polymers are miscible in the amorphous region. When the weight content of P2NAP in the polymers was larger than 20%, the appearance of two distinct T_g 's for the polymer blends exhibit, indicating that these blends are immiscible.

Figure 3 shows POM images of the LC cells made from rubbed polymer blend films, (a) P7COU, (b) P7COU/P2NAP(95/5), (c) P7COU/P2NAP(90/10), (d) P7COU/P2NAP(85/15), and (e) P7COU/P2NAP(80/20) using 5CB, respectively. The planar LC alignment behavior of the P7COU and P7COU/P2NAP blend was observed over the whole area. As the weight content of the P2NAP in the polymer blend was increased, the defect area of the planar LC alignment increased gradually. When the weight content of the P2NAP was larger than 20%, the LC cells fabricated with the polymer blend film showed LC alignment behavior with distinct disclination line. Normally, the homogeneity of a LC cell made from a rubbed polystyrene film is lost after aging for 1 day. These LC cells fabricated from the rubbed polymer blend films have maintained a planar LC alignment for more than 24 months since we first made the LC cell.

The polar diagrams of the absorbance of the dichroic dye (disperse blue 1) in the LC cells fabricated using the rubbed P7COU and P7COU/P2NAP blend films were obtained in order to study the LC alignment direction (Fig. 4). It was found that the maximum absorbances of the LC cells made from the rubbed films of (a) P7COU, (b) P7COU/P2NAP(90/10), and (c) P7COU/P2NAP(80/20) are observed along the 110° \leftrightarrow

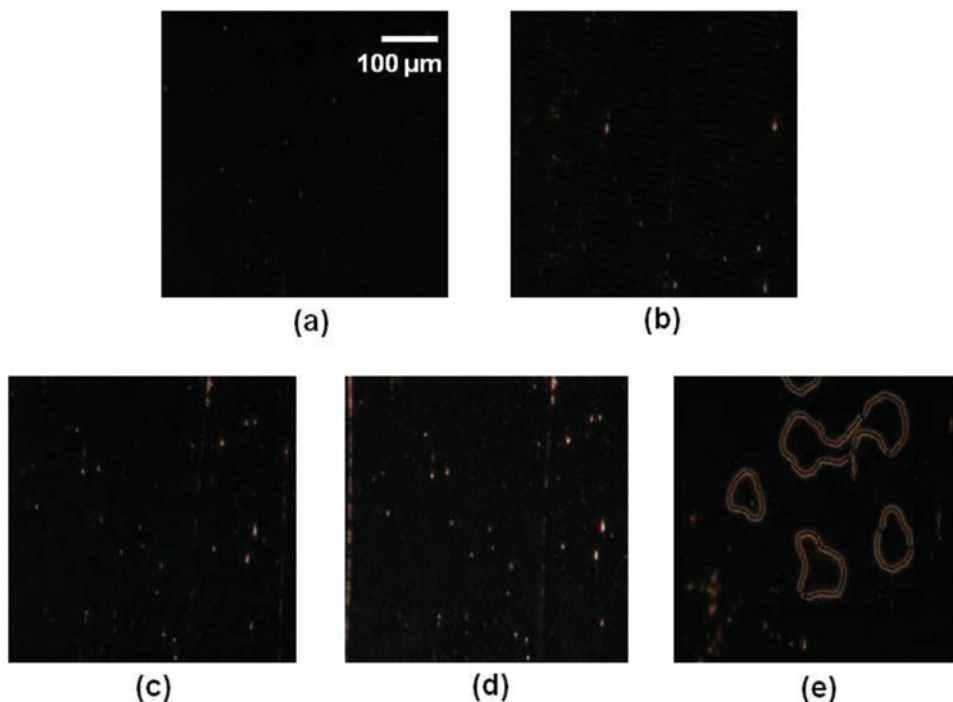


Figure 3. POM images in dark state of the LC cells made from rubbed polymer films ((a) P7COU, (b) P7COU/P2NAP(95/5), (c) P7COU/P2NAP(90/10), (d) P7COU/P2NAP(85/15), and (e) P7COU/P2NAP(80/20)) using 5CB.

290° directions, respectively. The LC cells made from rubbed films of PS showed maximum absorbances along the 90° ↔ 270° direction (data not shown). It has been known that the LC alignment direction depends on the alignment direction of the molecules on the polymer films after the rubbing process [23]. Therefore, the LC alignment (maximum absorbance along the 90° ↔ 270°) perpendicular with respect to the rubbing direction of

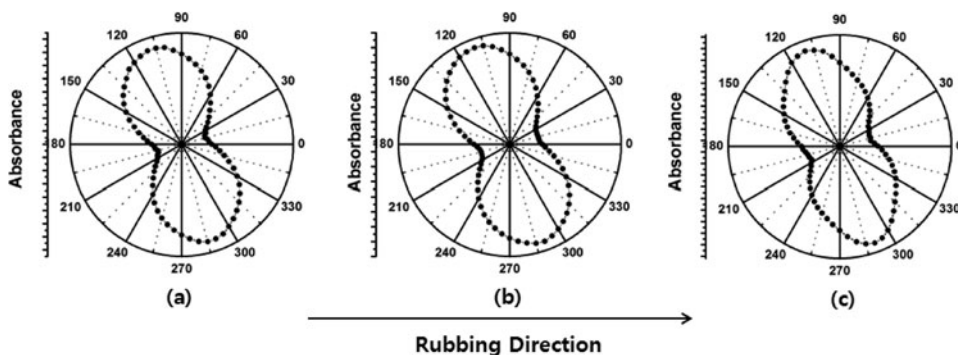


Figure 4. Polar diagram of the absorbance of disperse blue 1 in antiparallel LC cells fabricated with rubbed (a) P7COU, (b) P7COU/P2NAP(90/10), and (c) P7COU/P2NAP(80/20) film as a function of angle of samples.

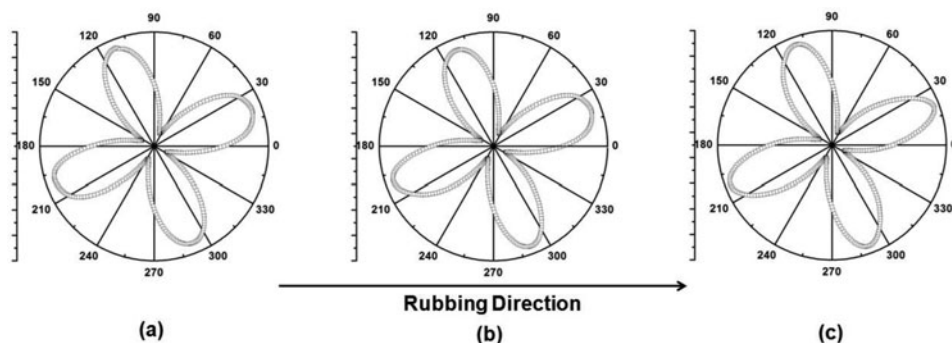


Figure 5. Polar diagram of the light intensity of LC cell made from rubbed (a) P7COU, (b) P7COU/P2NAP(90/10), and (c) P7COU/P2NAP(80/20) film as a function of rotation angle of samples.

the polystyrene film was ascribed to the orientation of the phenyl side groups perpendicular with respect to the rubbing direction reported by others [23]. The absorbance direction of $110^\circ \leftrightarrow 290^\circ$ for P7COU/P2NAP blend, deviated by 20° from that for the polystyrene film, which indicates that the side chain orientation of P7COU/P2NAP blend is not identical to that of polystyrene.

The thermal stability of the LC cells made from the polymer blend films was estimated from the polar diagram for the transmittance of monitoring source through the LC cell after heating the LC cell for 10 min at 200°C (Fig. 5). Figure 5 represents polar diagram for the transmittance of monitoring source through the LC cells made from (a) P7COU, (b) P7COU/P2NAP(90/10), and (c) P7COU/P2NAP(80/20), as a function of rotation angle of samples. As shown in Fig. 5, polar diagram of monitoring source passed through the LC cell made from polymer blend film have maintained at annealing temperature of 200°C above the T_g of polymer blends. Conclusively, LC aligning abilities of annealed LC cells made from polymer blend films have maintained above high temperature, although the molecular motion of main and side chain of polymer blend has been taken place above T_g .

The pretilt angles of the antiparallel LC cells fabricated with the polymer films were measured, in order to determine the effect of the weight content of the P7COU and P2NAP on the LC alignment direction (Fig. 6). We could not measure the pretilt angles of the LC cells made from the polymer blend films having a weight content of P2NAP larger than 20 weight%, due to their poor LC alignment behavior. At first, the pretilt angle of the LC cell from only P7COU was found to be very low about 0° , similar with rubbed polystyrene film [27,28]. When the weight content of P2NAP increases from 0 to 20% for the LC cell made from polymer films, the pretilt angles on the polymer films increase from 0° to 5° .

The contact angle values on polymer blend films according to the weight content of the P7COU and P2NAP were measured to study the effect of the wettability on the pretilt angle of LCs (Table 1). We adopted distilled water contact angles on the blend polymer films in static mode. The contact angle values on polymer blend films increase with the increase of weight content of the P2NAP indicating that the changes of pretilt angles are strongly related with the changes of wettability on polymer films as described by others [29,30]. They are 65° , 66° , 67° , 69° , and 72° at P7COU, P7COU/P2NAP(95/5), P7COU/P2NAP(90/10), P7COU/P2NAP(85/15), and P7COU/P2NAP(80/20), respectively.

The E-O properties of the LC cells made from the polymer alignment films were determined by measuring the voltage-transmittance (V-T) and response time value (Table 1).

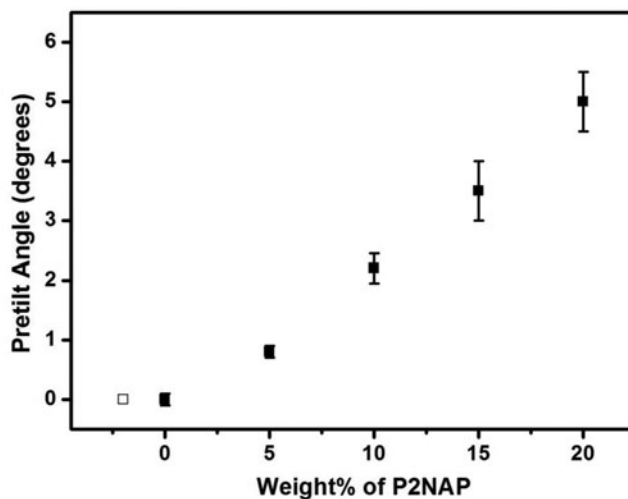


Figure 6. Observation of the pretilt angles of the LC cells fabricated with rubbed blend films based on P7COU and P2NAP using 5CB as a function of blending ratio.

The V_{th} values of this LC cell made from P7COU/P2NAP(80/20) film were found to be 0.92 V, respectively. For comparison, the V_{th} values of the LC cells fabricated using the P7COU films only were found to be 1.02 V, respectively. We also found that the response time (about 26 ms) of the LC cell made from the rubbed P7COU/P2NAP(80/20) is faster than that (about 36 ms) of the LC cell made from the rubbed P7COU film. It has been reported that LC alignment films having larger pretilt angles have faster response times in various LCD modes, such as the TN (twisted-nematic), OCB (optically compensated bend), and NBB (no-bias-bend) modes [31–33]. Therefore, the response time of the LC cell made from the P7COU/P2NAP(80/20) film having a pretilt angle of approximately 5° is faster than that of the LC cell fabricated with the P7COU film having a pretilt angle of about 0° .

Table 1. Surface property and LC alignment property of the polymer blend films

Polymer designation	Water contact angle (o) ^a	Pretilt angle (o) ^b	V_{th} (V) ^c	T_t (ms) ^d
P7COU	65	0	1.02	36
P7COU/P2NAP(95/5)	66	0.8	1.00	33
P7COU/P2NAP(90/10)	67	2.2	0.97	30
P7COU/P2NAP(85/15)	69	3.5	0.95	28
P7COU/P2NAP(80/20)	72	5.0	0.92	26

^aMeasured from static contact angles.

^bMeasured from the antiparallel LC cells made from rubbed polymer films.

^cThe threshold voltage (V_{th}) in the V-T curve are defined as the voltages at which the transmittance was increased to 10% of the initial transmittance value.

^dThe total response time (T_t) is determined by the average of rising and falling response time.

Recently, considerable effort has been made to develop plastic substrates for flexible LC displays [34]. All of the LC cells made from the polymer blend films on PET (polyethylene terephthalate) substrates exhibit similar LC alignment behavior compared with the LC cells made from same alignment films on ITO substrates as previously shown in Fig. 3. We found that the LC cells fabricated using the polymer blend films on plastic **PET** substrates show pretilt angle behaviors. Furthermore, pretilt angle values for this LC cell showed were maintained after bending it 100 times (data not shown). Therefore, these films can be considered as candidate LC alignment layers for flexible LC displays.

Conclusions

We prepared a series of blend films composed of polystyrene derivatives containing 7-hydroxycoumarin and polystyrene derivatives containing 2-naphthoxy side groups. Stable and homogeneous planar LC cells could be prepared from these polymer films through the rubbing process. The pretilt angle, V_{th} , and response time of the LC cell made from the polymer films containing only coumarin groups was about 0° , 1.02 V, and 36 ms, respectively, and those of the LC cell made from the blend alignment layers having weight contents of coumarin and naphthalene groups of 80 and 20 weight% was approximately about 5° , 0.92 V, and 26 ms, respectively. Therefore, the incorporation of P2NAP into the blend polymers increases the pretilt angle of the LC and improves the E-O performance of the LC cell. These polymers could be a good candidate as an alignment layer for flexible LC display applications such as TN, IPS, OCB, and NBB mode because they have low processing temperatures.

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