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Scattering birefringence polarizers based on oriented blends of poly(ethylene terephthalate) and core–shell particles

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Here, we present the results on the optical characteristics of polymeric, linear polarizers produced from oriented, two-component, polymer blends. By a proper selection of materials and drawing conditions, the refractive index of the dispersed phase of the blend is matched with the ordinary refractive index of the birefringent continuous phase while, at the same time, a large refractive index mismatch is generated in the perpendicular direction. The films are, therefore, transparent or opaque, dependent on the polarization direction of the incident light and act as linear polarizers with a high contrast ratio (≈ 1000) between the transparent and opaque state. These polarizers are all-polymer-based linear polarizers that operate on the anisotropic scattering of light, instead of anisotropic absorption, which offers some unique features and advantages. © 1998 American Institute of Physics. [S0021-8979(98)02306-8]

I. INTRODUCTION

The insensitivity of the human eye to the polarization direction of incident light^{1,2} is usually circumvented with polarizing filters. The first man-made polarization filters were manufactured from birefringent doubly refracting crystals.^{3–5} A well-known example of this type of polarizing filter is the Nicol prism, which is produced from a birefringent calcite crystal. Unpolarized light entering the crystal experiences two different refractive indices, and consequently, the light is split into two linearly polarized rays with perpendicular polarization directions. One of the light rays, i.e., the so-called ordinary ray, is internally reflected and absorbed. The other (extraordinary) light ray is transmitted and linearly polarized light is obtained.

The first large-area, flexible, thin-film polarizers produced from synthetic polymers were invented by Land in 1928.^{3,6,7} These polarizers are produced from oriented poly (vinylalcohol) sheets, which contain oriented dichroic species such as dichroic dyes. In subsequent studies several other linear sheet polarizers were described, which selectively scatter one polarization direction of light.^{6,8,9} However, especially in the first half of this century, attention was largely focused on dichroic polarizers, which were primarily used in applications such as antiglare sunglasses, photography filters, and optical instruments (microscopes).^{10–12} Nowadays, these linear polarizers are also widely used in flat panel display applications such as twisted nematic (TN) and super twisted nematic (STN) displays,^{13–18} were they serve a dual purpose; a first polarizer is used to generate linearly polarized light. The TN or STN cell contains a low molecular weight, liquid-crystalline material, which modulates the polarization direction of the polarized light dependent on the presence or absence of an electrical field. A second polarizer is used in these electro-optical light shutters to generate visual perception of the modulation of the polarization direction direction of the transmitted light.^{13–18}

A perfect dichroic polarizer with a polarizing efficiency of unity absorbs at least 50% of the incident light, and consequently, the brightness of displays is adversely affected by the polarization filters.^{19,20} This limited light efficiency of dichroic polarizers renewed the interest in nondichroic optical components to produce linearly polarized light.²¹⁻²⁶ In the past decade, several new linear polarizers based on the anisotropic scattering of light were proposed and the operating principle of these polarizers is schematically depicted in Fig. 1. Generally, these polarizers are manufactured from two-phase systems containing at least one birefringent phase. By a proper materials selection, the refractive index of, for instance, the dispersed phase (n_d) is matched with one of the refractive indices of a birefringent continuous phase [the ordinary (n_{ρ}) or extraordinary (n_{ρ}) refractive index] while, at the same time, a large refractive index mismatch is generated in the perpendicular direction (Fig. 1). The materials are, therefore, transparent or opaque dependent on the polarization direction of incident light and act as a linear polarizer. Moreover, devices with a theoretical maximum polarizing efficiency and single piece transmittance of unity were proposed based on these polarizers.^{27,28} These linear polarizers possess some disadvantages such as the use of rather exotic

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FIG. 1. Schematic representation of a linear polarizer based on the anisotropic scattering of light. n_o =ordinary refractive index of the continuous phase; n_e =extraordinary refractive index of the continuous phase; and n_d =refractive index of the dispersed phase.

liquid-crystalline materials and, in most cases, a driving voltage is required to operate these polarizers. Moreover, the production of large-area, flexible films is rather laborious with the proposed materials and experimental procedures.

In this study, it is attempted to produce light-scattering films, which act as a linear polarizer from standard engineering polymers. Conventional processes, such as cast-film extrusion in combination with solid-state drawing, are used to produce these linear polarizers, which enables the production of large-area films in a continuous process.²⁹ Some initial results are presented in this study to illustrate the operating principle of these polarizers.

II. EXPERIMENT

The poly(ethylene terephthalate) grade used in this study (Arnite D02 300, Mw=52 kg/mol) was kindly supplied by DSM (Geleen, the Netherlands). Rubbery core-shell particles (Paraloid EXL3647) with a particle size of 200 nm were purchased from Röhm and Haas (Philadelphia, PA).

Blends of poly(ethylene terephthalate) and core-shell rubbers were prepared on a counter-rotating twin screw extruder at 270 °C. Prior to extrusion, the poly(ethylene terephthalate) was dried for 24 h at 150 °C. Blends were prepared containing 0%, 2%, 4%, 6%, 8%, 10%, 20%, and 30% wt/wt of core-shell particles and after compounding the extrudates were granulated and dried again. Films were prepared from the blends on a conventional cast-film extrusion line at 270 °C. After extrusion, the films were quenched in a water/ ice bath. Solid-state drawing of the films was performed on a tensile tester equipped with a thermostatically controlled oven at 85 °C. The draw ratio of the films was determined by measuring the displacement of ink marks on the films.

The ordinary and extraordinary refractive indices of drawn poly(ethylene terephthalate) films (without core-shell particles) were measured using an Abbe refractometer. The so-called trirefringence technique³⁰ was used to determine the three principle refractive indices of the drawn films.

The structure of the drawn blends was investigated using a scanning electron microscope (SEM). Fracture surfaces of the drawn films were prepared in liquid nitrogen. These frac-



FIG. 2. Schematic representation of the experimental arrangement to measure the light intensity distribution in the forward direction.

ture surfaces were obtained from cross sections of the films both parallel and perpendicular to the drawing direction. The fracture surfaces were etched in an oxygen plasma for 10 min and coated with a gold/palladium layer.

Polarized UV–VIS measurements were performed on a Perkin Elmer Lambda 9 in the wavelength range from 400 to 750 nm. Prior to the measurements, the drawn films were coated with paraffin oil and sandwiched between two glass slides to eliminate the scattering of light at the film surface. The transmittance of the films was measured, respectively, parallel (T_1) and perpendicular (T_2) to the drawing direction. The polarizing efficiency (PE) and single-piece transmittance (T_{sp}) were calculated from these measurements using Eqs. (1) and (2)⁴

$$PE = \frac{(T_2 - T_1)}{(T_2 + T_1)},$$
(1)

$$T_{\rm sp} = \frac{1}{2}(T_1 + T_2). \tag{2}$$

In Fig. 2, a schematic drawing is presented of the experimental setup, which was used to measure the angular intensity distribution of the light passing through the blends in the forward direction. A collimated beam of light (1), originating from a halogen light source, illuminates the sample via an optical fiber. The measurements were performed using a green bandpass filter with a central wavelength of 561 nm and a full width at half maximum of 21 nm. Two linear dichroic polarizers are placed below and above the sample to control the polarization directions of the incident and transmitted light. The light passing through the system is collected by a light collector (4), which is connected to a photomultiplier tube that measures the light intensity in terms of absolute luminescence (Cd/m^2). The microscope can rotate in a vertical plane over an angle (θ) from 0° to 70° (Fig. 2). The sample with the polarizers is mounted on a sample holder, which can rotate over an angle (ϕ) from 0° to 360°. Consequently, the spatial distribution of the light intensity in the forward direction can be evaluated in spherical coordinates (Fig. 3).



FIG. 3. Schematic representation of the conversion of the spherical coordinates θ and ϕ to the Cartesian coordinates x and y.

For a graphical representation of the three-dimensional light intensity distribution, it is essential to convert the spherical coordinates θ and ϕ into Cartesian coordinates x and y with $x = \sin(\theta) * \cos(\phi)$ and $y = \sin(\theta) * \sin(\phi)$. The center of the plot at x=0 and y=0 corresponds to the position of the incident light beam. The drawing direction of the films is always parallel to the x axis and the measured light intensity is plotted on the vertical axis (Fig. 3). The relative light intensity with a sample in the beam at the Cartesian coordinates x and y and the measured light intensity without a sample in the beam at x=0 and y=0.

The ratio of the intensities of light scattered in the forward and backward direction was measured using the experimental arrangement shown in Fig. 4. Light from a laser (Spectra Physics, 2 mW helium neon Class 3 laser, wavelength=540 nm) is polarized using a dichroic polarizer, and subsequently, split into two beams by a beam splitter. One beam is directed to the sample and an integrating sphere and the second reference beam is transmitted directly to a photodiode. The light intensity measured by the photodiode in the integrating sphere is divided by the light intensity of the reference beam measured by the second photodiode to correct for laser intensity fluctuations. The intensity of the



FIG. 4. Schematic representation of the experimental arrangement to measure the light intensity in the forward and backward direction.



FIG. 5. Refractive indices of drawn poly(ethylene terephthalate) films as a function the draw ratio. (\bigcirc) ordinary refractive index (perpendicular to the drawing direction), (\bigcirc) extraordinary refractive index (parallel to the drawing direction), (\bigtriangledown) ordinary refractive index (see Ref. 33), and (\blacktriangledown) extraordinary refractive index (see Ref. 33).

light in the forward direction is measured by placing the sample in front of the integrating sphere [Fig. 4(a)]. Subsequently, the same sample is placed at the rear of the integrating sphere [Fig. 4(b)] and the relative intensity of light scattered in the backward direction is measured. In both the forward and backward directions the measured light intensities are normalized with respect to the light intensity of the incoming beam (i.e., without a sample).

III. RESULTS

In this particular study, poly(ethylene terephthalate) was selected for the continuous phase of the blend because of its excellent solid-state drawing characteristics and its high positive birefringence.^{31,32} The refractive indices of drawn poly(ethylene terephthalate) films are plotted in Fig. 5 as a function of the draw ratio. Literature values for the refractive indices of drawn poly(ethylene terephthalate) films are also included.³³

The extraordinary refractive index of poly(ethylene terephthalate) rapidly increases with the increasing draw ratio and, simultaneously, the ordinary refractive index decreases slightly to a value of approximately 1.53 at a draw ratio of 4–5. So-called core–shell rubbers were selected to match the ordinary refractive index (n_o) of the drawn poly-(ethylene terephthalate) films in the blends. These core–shell rubbers possess a particle size of 200 nm, a refractive index of 1.530, and they consist of a crosslinked styrene–butadiene core in combination with a thin, crosslinked PMMA shell.³⁴

Blends of poly(ethylene terephthalate) and core-shell particles were produced on a conventional cast-film line. Subsequently, these films are drawn uniaxially at temperatures (T_d =85 °C) close to the glass transition temperature of the continuous polyester phase (T_g =70 °C).³² Perpendicular to the drawing direction, the films exhibit a morphology of slightly agglomerated core-shell particles in the poly(ethylene terephthalate) phase [Fig. 6(a)]. In the drawing direction, however, the core-shell particles are aligned and aggregated in pearl chains of arrays of individual particles [Fig. 6(b)].

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(a)



FIG. 7. Photographs of a drawn poly(ethylene terephthalate) blend containing 10% wt/wt core-shell particles. (a) Linearly polarized light perpendicular to the drawing direction, and (b) linearly polarized light parallel to the drawing direction.

FIG. 6. SEM micrographs of drawn blends. (a) cross section perpendicular to the drawing direction, and (b) cross section parallel to the drawing direction.

The separate, spherical core-shell particles are still observed after drawing, which indicates that the particles remain isotropic and nonbirefringent. The dimensions of the strings were measured and it was found that the arrays of particles, typically, possess an aspect ratio of approximately 20.

Photographs of drawn poly(ethylene terephthalate) blends containing 10% w/w core-shell particles are shown in Fig. 7. A conventional dichroic polarizer (analyzer) is used to generate polarized light and to obtain visual perception of the polarizing characteristics of the blends. As expected, the drawn blends are highly transparent for light polarized perpendicular to the drawing direction [Fig. 7(a)] and strongly scatter light in the parallel direction [Fig. 7(b)], which illustrates that a linear polarizer is indeed obtained.

In Fig. 8(a), polarized UV-VIS spectra are shown of drawn poly(ethylene terephthalate) blends. As expected, the transmittance of the blends is low in the scattering state (<5%) and the films are highly transparent (>90%) in the perpendicular direction. The polarizing efficiency (PE) and single-piece transmittance (T_{sp}) of the drawn poly(ethylene terephthalate) blends were calculated from these experimental data and are plotted in Fig. 8(b). Figure 8(b) illustrates that the films combine a high polarizing efficiency (>0.95)with a high single-piece transmittance (>0.35), especially in the region from 450 to 700 nm.

In Fig. 9, the polarizing efficiency of drawn poly(ethylene terephthalate) blends is plotted as a function of the single-piece transmittance (at 600 nm) and a comparison is made with conventional dichroic polarizers.²⁰ The experimental data were obtained from drawn poly(ethylene terephthalate) blends with a varying content of core-shell rubbers (see the experimental section) and it is shown that the performance of the scattering polarizers is virtually identical to conventional dichroic polarizers.²⁰

The optical properties of drawn poly(ethylene terephthalate) blends were further evaluated using polarized lightscattering measurements. The experimental setup that was used is described in the experimental section (Fig. 2). The polarized light intensity distribution in the forward direction of drawn poly(ethylene terephthalate) blends is plotted in Fig. 10 on a logarithmic scale for both the transparent and light-scattering states. In Fig. 10, the ratio of the measured light intensity to the light intensity without a film in the beam (at x=0, y=0) is plotted. A large contrast in light intensity is observed which is, of course, in accordance with previous experimental data. A maximum contrast ratio in light intensity between the transparent and scattering state of approximately 1000 is obtained in direct transmittance at x=0 and y=0. Moreover, it is shown that the light is not randomly distributed in three dimensions, i.e., the light is more intense perpendicular to the drawing direction in both the scattering and in the transparent states.

The above-described experimental data are limited to the intensity distribution of light passing through the polarizer in the forward direction. Potentially, a part of the light is backscattered and, therefore, the ratio of the light intensity in the forward and backward direction was determined (see the experimental section, Fig. 3). In Fig. 11, the relative light intensities in the forward and backward direction are plotted for both the transparent and scattering states as a function of the dispersed particle concentration. As expected, virtually



FIG. 8. (a) UV–VIS spectra of a drawn poly(ethylene terephthalate) blend containing 10% wt/wt core–shell particles (thickness=0.24 mm). (a) Linearly polarized light perpendicular to the drawing direction, and (b) linearly polarized light parallel to the drawing direction. (b) Polarizing efficiency (PE) and single-piece transmittance (T_{sp}) as a function of wavelength.

all the light is transmitted in the forward direction in the transparent state [Fig. 11(a)]. The transmission in the forward direction decreases with an increasing dispersed particle concentration, which indicates that the refractive index match between the dispersed phase and the ordinary refractive index of the continuous phase is not truly obtained. In



FIG. 9. Polarizing efficiency (PE) as a function of the single-piece transmittance (T_{sp}) at 600 nm. (\bullet) scattering polarizers, and (\bigcirc) dichroic polarizers (see Ref. 20).



FIG. 10. Relative light intensity (logarithmic scale) distribution of a drawn poly(ethylene terephthalate) blend containing 10% wt/wt core-shell particles (thickness=0.24 mm). (a) Linearly polarized light perpendicular to the drawing direction, and (b) linearly polarized light parallel to the drawing direction.

the opaque state, a significant portion of the incident light is scattered in the backward direction [Fig. 11(b)]. The back-scattered light intensity increases with increasing concentration of core-shell particles and levels off at a value of approximately 55% of the incident light (at this particular film thickness). The experimental data also suggest that a part of the light is lost, i.e., the sum of the relative light intensities in the forward and backward direction deviates significantly from unity (80%-90%). This originates from light that is not scattered in the forward or backward direction, but propagates within the tape.

IV. DISCUSSION

It was shown that thin-film, linear polarizers based on the anisotropic scattering of light can be produced from conventional polymers provided that special care is taken with respect to materials selection, processing, and solid-state drawing of the films. This particular study was limited to a single-model system based on blends of poly(ethylene terephthalate) and crosslinked core–shell rubbers. Of course, this choice of materials is, to some extent, arbitrary and alternative systems with a similar performance can be easily envisaged if the prime prerequisites with respect to refractive indices, birefringence, solid-state drawing characteristics, and morphology of the materials are met.

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FIG. 11. (a) Relative light intensity in the forward (a) and backward direction (b) in the transparent state as a function of particle concentration (film thickness=0.24 mm). (b) Relative light intensity in the forward (a) and backward direction (b) in the opaque state as a function of particle concentration (film thickness=0.24 mm).

During drawing, the core-shell particles aggregate and align in pearl chains of individual particles. The mechanism for the formation of the pearl chains is somewhat obscure although it seems reasonable to assume that this structure is a consequence of the slightly aggregated structure of coreshell particles in the blends prior to drawing and of the use of crosslinked particles, which resist deformation upon drawing. The light-scattering experiments on drawn blends indicate that the light intensity distribution in the forward direction is anisotropic, i.e., the light scattering is more efficient in the plane perpendicular to the drawing direction in both the transparent and opaque states. This anisotropy also originates from the pearl-chain morphology of the drawn blends. Apparently, the incident light experiences virtual scattering centers with the shape of a cylinder and these aligned cylinders induce excess light scattering perpendicular to their long axis.35-39

It was also shown that the polarizing efficiency and single-piece transmittance of the scattering polarizers are virtually identical to conventional dichroic polarizers. This experimental observation is, however, somewhat misleading because the different materials used and the different operating principle of the scattering polarizers offer some unique features and advantages. For instance, dichroic polarizers





FIG. 12. Schematic representation of a projection system equipped with light-scattering polarizers.

based on drawn poly(vinylalcohol) films exhibit a rather poor temperature and humidity resistance, which is related to the highly hydrophilic character of the host polymer poly(vinylalcohol).⁴⁰ The polymers used for the scattering polarizers are rather hydrophobic and, consequently, an enhanced temperature and humidity resistance is expected. Secondly, dichroic polarizers absorb a part of the incident light and this light is transferred into thermal energy.⁴¹ Consequently, dichroic polarizers exhibit excessive heating in combination with high intensity light sources, which deteriorates their optical performance.⁴¹ The polarizers described in this study scatter the light and excessive heating of the polarizers (and displays) is prevented, which is expected to significantly enhance their lifetime in projection applications such as shown in Fig. 12. In this schematic representation, a projection system is shown consisting of a light source, a first (scattering) polarizer, an electro-optical cell, a second (scattering) polarizer, an aperture stop, projection lenses, and finally, a projection screen. In the transparent state of the display, a highly collimated light beam passes the aperture stop and reaches the projection screen. In the scattering state, light is partly scattered in the forward and partly in the backward direction. The forward scattered light is blocked by the aperture stop and does not reach the projection screen. Thermal energy is now dissipated at the aperture stop and polar degradation is largely prevented.

Finally, a perfect dichroic polarizer (PE=1) absorbs at least 50% of the incident light $(T_{sp}=0.5)$,²⁰ and consequently, the brightness of the transmitted light in displays is reduced. The light-scattering polarizers offer the possibility to recycle the scattered light and to drastically improve the efficiency to produce polarized light.^{27,28} A schematic diagram of a device that recycles scattered light is shown in Fig. 13. Natural light is injected into a waveguide and a reflector is used to transmit the incident light in the direction of a polarizer. In the ideal situation, half of the incident light is



FIG. 13. Schematic representation of an electro-optical light valve in which backscattered light is recycled to enhance brightness and energy efficiency.

transmitted directly as linearly polarized light. The other half is backscattered in the direction of the waveguide and this light is depolarized by a diffusor. Subsequently, the light is redirected to the scattering polarizer and this process is repeated until all the light passes the polarizer. Of course, the theoretical maximum light efficiency of such a device is unity provided that the light, which experiences the opaque state, is completely backscattered in the direction of the waveguide.^{27,28} The polarizers described in this study scatter approximately 50% of the incident light in the backward direction (in the opaque state) and the rest of the light is scattered in the forward direction. This rather low ratio of light scattered in the backward and forward direction limits the efficiency of a device such as shown in Fig. 13. Therefore, it is currently attempted to enhance this ratio and the results of this study will be presented in future reports.

V. CONCLUSIONS

Thin-film linear polarizers were produced from drawn blends of poly(ethylene terephthalate) and core-shell rubbers. The excellent polarizing performance of the blend is attributed to a carefully tuned refractive index match and mismatch between the continuous and dispersed phase in two mutually perpendicular directions. The drawn films exhibit a similar performance as dichroic polarizers with respect to polarizing efficiency and single-piece transmittance. However, the nondichroic character of the polarizers in combination with the selected materials offers some potential advantages with respect to temperature, light, and humidity stability.

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