Thin Solid Films 518 (2010) 3089–3092

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Plastic substrate with gas barrier layer and transparent conductive oxide thin film for flexible displays

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ARTICLE INFO

Available online 13 October 2009

Keywords:
Plastic substrate
Flexible displays
Transparent conductive oxide
Gas barrier
Sputtering

ABSTRACT

A novel plastic substrate for flexible displays was developed. The substrate consisted of a polycarbonate (PC) base film coated with a gas barrier layer and a transparent conductive thin film. PC with ultra-low intrinsic birefringence and high temperature dimensional stability was developed for the base film. The retardation of the PC base film was less than 1 nm at a wavelength of 550 nm (film thickness, 120 µm). Even at 180 °C, the elastic modulus was 2 GPa, and thermal shrinkage was less than 0.01%. The surface roughness of the PC base film was less than 0.5 nm. A silicon oxide (SiO_x) gas barrier layer was deposited on the PC base film by a roll-to-roll DC magnetron reactive sputtering method. The water vapor transmission rate of the SiO_x film was less than 0.05 g/m^2/day at 40 °C and 100% relative humidity (RH), and the permeation of oxygen was less than 0.5 cc/m^2 day atm at 40 °C and 90% RH. As the transparent conductive thin film, amorphous indium zinc oxide was deposited on the SiO_x by sputtering. The transmittance was 87% and the resistivity was 3.5×10^{-4} ohm cm.

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1. Introduction

There has been growing interest in the use of plastic substrates in the fabrication of future electronic devices such as flexible displays [1–3], photovoltaics, batteries, and sensors. These flexible devices have a high potential to realize thin, lightweight, robust, and bendable electronic products. However, conventional plastic substrates made from commercial polymers are not sufficient to satisfy the demands of display designers; further improvements in birefringence, heat resistance, and gas barrier performance are especially demanded. In the manufacture of liquid crystal displays (LCDs) by using plastic substrates, the retardation value, which is the product of the substrates' birefringence and thickness, should be less than 10 nm to avoid interference colors. In addition, they should be capable of withstanding temperatures of at least 150 °C, and high gas barrier performance against water vapour (≤1 g/m^2/day at 40 °C and 90% RH) and oxygen (≤1 cc/m^2/day at 40 °C and 90% RH) is demanded to ensure endurance and reliability of the final products.

In this study, a high-temperature PC substrate with a gas barrier layer and a transparent conductive oxide thin film was developed to improve the properties that are especially important for flat panel display devices.

A cross-sectional view of the substrate is illustrated in Fig. 1. The substrate was composed of a PC base film (120 µm thick), a silicon oxide gas barrier layer (30 nm thick), a hard coating (2 µm thick), and an indium zinc oxide transparent conductive layer (130 nm thick). The main properties of the substrate are shown in Table 1. Remarkable improvements were achieved in some properties, for instance: the glass transition temperature (T_g) of the base film was 215 °C, the retardation was 1 nm, the water vapor permeation rate was 0.05 g/m^2/day, thermal shrinkage was 0.01% after heating at 180 °C for 2 h, and Young's modulus was 2.8 GPa.

2. Experimental details

The high-temperature PC base film was produced by the solvent casting method from dichloromethane solution. As a gas barrier layer, silicon oxide (SiO_x) was deposited on the base film by DC magnetron reactive sputtering using a silicon target. The sputtering chamber was evacuated to 2×10^{-4} Pa. The substrate temperature was set at 20 °C. Sputtering was carried out under Ar ± O_2 mixed gas. The total gas pressure was 0.1 Pa at a power density of 4 W/cm^2.

A hard coating composed of siloxane-containing material was applied on the SiO_x surface of the substrate, and an acrylic-type hard coating was applied on the surface opposite the SiO_x layer by using a conventional gravure coating method. A transparent conductive film was deposited on the acrylic-type hard coating by DC magnetron sputtering using either In_2O_3–ZnO (Zn: 10 wt.%; IZO) or In_2O_3–SnO_2 (Sn: 5 wt.%; ITO). The sputtering chamber was evacuated to 9×10^{-5} Pa. The substrate temperature was set at 20 °C. Sputtering was carried out under Ar or Ar ± O_2 mixed gas. The total gas pressure was 0.5 Pa at a power density of 4 W/cm^2. The thicknesses of the IZO and ITO films were adjusted to be about 130 nm by controlling the deposition time.

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doi:10.1016/j.tsf.2009.09.166

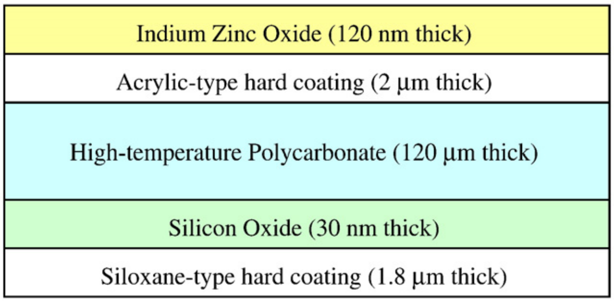

Fig. 1. Cross-sectional view of the plastic substrate developed for flexible displays.

The glass transition temperature T_g and Young's modulus of the PC base film were measured by differential scanning calorimetry (DSC) and dynamic mechanical thermal analysis (DMTA), respectively. The total light transmittance of the substrate was measured by a colorimeter (COH 400, Nippon Denshoku Industries), and the retardation was estimated by spectroscopic ellipsometry. The film thicknesses of the SiO_x and transparent conductive oxides were measured using a surface profiler (Dektak[3], Sloan Tech). To evaluate the gas-barrier properties of the substrate, the water vapor permeability at 40 °C and 100% RH and the oxygen permeability at 40 °C and 90% RH were measured with permeation instruments (OX-TRAN and PERMATRAN, mocon®). The surface roughness of the substrate was analyzed by using tapping mode atomic force microscopy (AFM). The crystallization temperature of the transparent conductive oxides was estimated by DSC measurement of isolated thin films flaked from the substrate.

3. Results and discussion

3.1. Ultra-low birefringence

Large birefringence of the PC base film is undesirable when the plastic substrates are used for LCDs. As the birefringence becomes larger, the display panel tends to have strong interference colors. The birefringence of a polymer film is expressed as $\Delta n_0 = \Delta n_0 f$, where Δn_0 is the intrinsic birefringence of the polymer, and f is the orientation function. According to this equation, Δn_0 and f must be reduced to decrease the birefringence. The base films of conventional substrates are typically made from bis-A type polycarbonate (bis-A PC). This polymer is obtained by reacting 2,2-bis (4-hydroxyphenyl) propane and phosgene in the presence of pyridine. In this polymer, the segment formed from 2,2-bis (4-hydroxyphenyl) propane increases the intrinsic birefringence because the polarizability of the segment parallel to the axis of the polymer chain is larger. In order to reduce the intrinsic birefringence, we designed a new polycarbonate, called "high-temperature PC", for the substrate. A solvent casting process is suitable for reducing the function f. This process yields a highly

Table 1
Properties of the plastic substrate coated with indium zinc oxide (IZO) transparent conductive layer.

Properties	Characteristics (remarks)
Thickness	125 µm
Glass transition temperature	215 °C (basefilm)
Surface resistance	30 ohm/sq.
Light transmittance	87% (91% w/o IZO)
Retardation	1 nm
O_2 permeation rate	0.5 cc/m^2/day at 40 °C/90%RH
H_2O permeation rate	0.05 g/m^2/day at 40 °C/100%RH
Thermal shrinkage	0.01% at 180 °C 2 h
Young's modulus	2.8 GPa
Tensile Elongation	13%

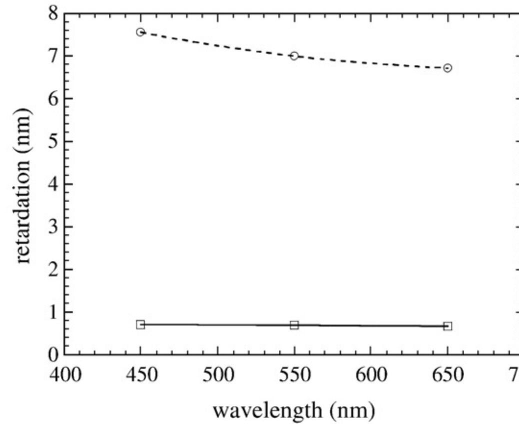

Fig. 2. Retardation of high-temperature PC base film (solid line) and conventional bis-A PC base film (broken line) versus wavelength (thickness 0.12 mm).

amorphous, optically clear, and colorless film. The retardation ($Re = \Delta n \cdot d$, where d is the substrate thickness) of the high-temperature PC base film was less than 1 nm, which is one order of magnitude smaller than that of bis-A PC (Fig. 2). This property is ideal for LCDs. The high-temperature PC base film had not only ultra-low birefringence but also high transmittance of visible light, at more than 90%.

3.2. Improved heat resistance

The most promising way to improve the thermal stability of polymers is to introduce highly stable structural units into the polymer systems, such as aromatic and/or heterocyclic rings. Our new high-temperature PC was designed not only to reduce the birefringence but also to improve thermal stability. Fig. 3 shows the dynamic shear modulus E' and the mechanical damping factor tan δ of the high-temperature PC and conventional bis-A PC versus temperature. The high-temperature PC displayed favorable thermal stability and mechanical properties over a temperature range from 30 to 200 °C. In general, when a plastic substrate is exposed to temperatures close to the T_g of the polymer base film, a relaxation process in the polymer makes the substrate shrink, which can lead to structural distortion, cracks, and film peeling during the process. However, the thermal shrinkage of the high-temperature PC base film was less than 0.01%

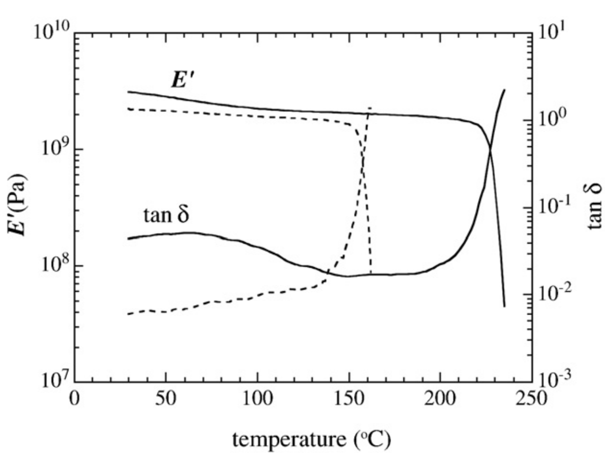

Fig. 3. The shear modulus E' and damping factor tan δ versus temperature of high-temperature PC (solid line) and conventional bis-A PC (broken line). The oscillating frequency in the thermo-mechanical analysis was 1 Hz over the entire temperature range from 30 to 250 °C.

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