

Room temperature sputtering deposition of high-haze Ga-doped ZnO transparent conductive thin films on self-textured bio-based poly(ethylene 2, 5-furandicarboxylate) substrates

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ABSTRACT

In this work, bio-based poly(ethylene 2,5-furandicarboxylate) (PEF) films were prepared by drop-casting method and used as substrates for depositing Ga-doped ZnO (GZO) transparent conductive thin films. Results showed that the 300-nm GZO thin films deposited on PEF substrates exhibited haze values above 65% at 550 nm without post-treatment. The high haze value was because of the large surface roughness of PEF films. The total optical transmittance and electrical properties of GZO thin films on PEF were comparable to those of GZO thin films on PET. The present study provides a simple way for the sputtering deposition of high-haze transparent conductive thin films on flexible substrates.

1. Introduction

Presently, the advent of portable and flexible thin film solar cells strongly encourage the use of polymers as lightweight and inexpensive substrates [1–4]. Among polymer substrates, polyethylene terephthalate (PET) is one of the most important thermoplastics used in the photovoltaic field because of its several advantages such as excellent mechanical properties, low cost and high transmittance in a wide spectral range [5–7]. Nevertheless, terephthalate polyesters are immune to microbial attack and are not degradable under normal environmental conditions [8]. Furthermore, their precursors are mostly extracted from fossil fuels [9]. The large-scale use of terephthalate polyesters will increase carbon emissions, thereby accelerating global warming [10]. Thus, there is a growing interest in the preparation of novel polymer substrates because of the need for renewable resources to replace fossil-based materials.

Poly(ethylene 2,5-furandicarboxylate) (PEF) based on furan-2,5-di-carboxylic acid (FDCA) is an important bio-based polymer substrate and is an alternative for PET [11–13]. The glass transition temperature and solvent resistance of PEF are similar to those of PET [14,15]. Moreover, it was reported that PEF exhibits excellent mechanical and barrier properties [16]. Thus, it could be a new type of renewable polymer substrate for a flexible transparent electrode.

Compared with other flexible transparent conductive oxide, zinc oxide (ZnO)-based thin film is a promising alternative to the commercial indium tin oxide film [17,18]. The growth behaviours of ZnO-based thin films on PET substrates were widely studied. For example, Kim et al. [19] systematically studied the effects of O₂ plasma pre-treatment on the properties of Ga-doped ZnO (GZO) films on PET substrate. With appropriate O₂ plasma treatment, the surface energy and adhesion of PET substrate were improved. The optimal resistivity of GZO films on PET substrate was $3.4 \times 10^{-3} \Omega \text{ cm}$. A more recent work by Jia et al. [20] reported that 50-nm ZnO buffer layers restrained the diffusion of moisture and gas from PET substrates to GZO thin films. The resistivity of GZO/ZnO bilayer films decreased considerably more than one order of magnitude than that of GZO film without a ZnO buffer layer. Although some reports have demonstrated the electrical and optical properties of ZnO-based thin films on PET substrates, there have been no reports on the deposition of ZnO-based thin films on PEF substrates.

Therefore, in this study, PEF thin films were synthesized and used as transparent polymer substrates for depositing GZO transparent conductive films (TCFs) and were compared with PET substrates to assess them as alternative materials. Unexpectedly, we found that GZO thin films deposited on PEF substrates exhibited high haze values because of the natural textured surfaces of PEF substrates. Moreover, the

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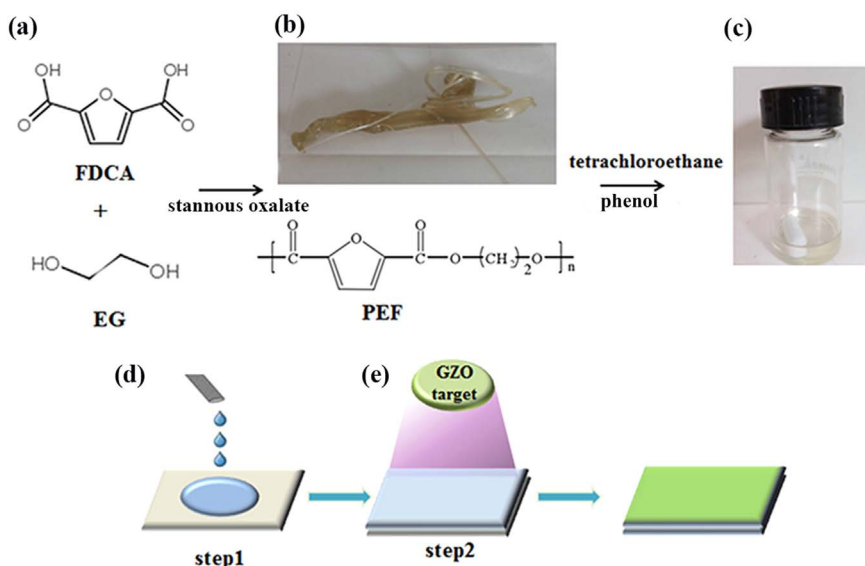


Fig. 1. Fabrication process of GZO/PEF TCFs. (a) Chemical structures of the monomers employed. (b) A photograph and chemical structure of the as-prepared PEF silk product. (c) A photograph of the PEF solution. (d) Drop-casting PEF solution. (e) Sputtering GZO thin film on PEF substrate.

conductivity and transmittance of GZO thin films deposited on PEF substrates were comparable to those of GZO thin films on PET substrates. Therefore, GZO thin films deposited on PEF substrates will have potential applications as front electrodes in flexible thin-film solar cells.

2. Experimental procedures

2.1. Materials

The reagents used in this study were 2,5-furandicarboxylic acid (FDCA), ethylene glycol (EG), stannous oxalate (C_2O_4Sn), phenol (C_6H_5OH) and tetrachloroethane ($C_2H_2Cl_4$). All reagents were of analytical grade and purchased from Sinopharm Chemical Reagent Co., Ltd. All chemicals were used as received without further purification.

2.2. Preparation and fabrication of PEF films

PEF solution was first synthesized using FDCA and EG as monomers. Chemical structures of the monomers are shown in Fig. 1a. Typically, 0.16 mol EG, 0.1 mol FDCA and 0.1 mmol stannous oxalate were added into a 100-ml flask and stirred under nitrogen at 210 °C under atmospheric pressure for no less than 5 h. The solution was then stirred under high vacuum of 70 Pa at 240 °C for no less than 2 h until it turned brown. The precipitate was dried at 60 °C under high vacuum for 48 h and then wire drawn. Photograph and chemical structures of the as-prepared PEF silk product are shown in Fig. 1b. Subsequently, the product was dissolved in a mixture of tetrachloroethane and phenol. The solution was then stirred at room temperature for 5 h. Finally, several drops of PEF solution were drop-coated on five glass substrates ($5 \times 5 \text{ cm}^2$) and held at 100 °C for 3 h in vacuum (100 Pa) to evaporate the solvent. The thicknesses of the PEF films were controlled to be 30, 70, 80, 90 and 110 μm by calculating the volume of solution.

2.3. Fabrication of GZO/PEF TCFs

GZO thin films ($\sim 300 \text{ nm}$) were deposited on five textured PEF films at room temperature by RF magnetron sputtering using a home-made GZO ceramic target containing 95 wt% ZnO and 5 wt% Ga_2O_3 . The RF power was set at 100 W. The deposition chamber was initially evacuated down to $5 \times 10^{-4} \text{ Pa}$. The Ar flow rate was controlled to be 20 sccm. To improve the uniformity of GZO films, the sample holder was rotated at a constant speed of 10 rpm/min. The obtained samples were named as #1, #2, #3, #4 and #5. For comparison, GZO thin films were deposited on low iron glasses and PET substrates under the same sputtering conditions.

2.4. Characterization and measurement

The crystalline phase of GZO/PEF TCFs were analysed by X-ray diffraction (XRD, Bruker, AXS D8 Advance, USA) using a standard θ – 2θ geometry diffractometer with Cu K α radiation ($\lambda = 1.54 \text{ \AA}$). The surface roughnesses of GZO/PEF TCFs were examined using an atomic force microscope (AFM, Veeco Dimension 3100V). The optical properties of GZO/PEF TCFs were measured using an ultraviolet-visible spectrophotometer (Perkin-Elmer, Lambda 950, USA) with an integrating sphere. The sheet resistance of GZO/PEF TCFs was obtained using a four-point probe system (Napson Corp. Cresbox). The surface wettabilities of PET and PEF substrates were measured using a contact angle measurement system (OCA-20, Germany).

3. Results and discussion

3.1. Optical properties

Fig. 2a shows the optical transmittance, reflectance and absorbance of GZO/PEF thin films as functions of PEF thickness. The optical transmittance of the as-deposited GZO/PEF films was critically affected by the thickness of PEF films. As the PEF thickness increased, the transmittance of GZO/PEF films decreased from 83.8% to 70.2% in the range of 400–800 nm. To examine the location of decreasing transmittance, we measured the reflectance and absorbance of GZO/PEF thin films. The results show that the reflectance of these GZO/PEF films in the visible region increased from 14.1% to 19.0% as the PEF thickness increases. The absorbance also increased from 2.1% to 10.8%. Detailed data on this are listed in Table 1. These results show that the decrease in transmittance is a synergistic effect of reflectance and absorbance. However, when the PEF thickness was 30 μm , we were surprised to find that the absorbance of GZO/PEF thin films in the visible region was only 2.1%. This indicates that the absorbance is mainly from GZO thin film, rather than from PEF film [21], which implies that the absorbance of PEF film is negligible when the thickness is less than 30 μm . Fig. 2b shows the transmittance, reflectance and absorbance spectra of PEF thin film (21 μm). The absorbance was 0.43% in the range of 400–800 nm. The inset depicts the cross-section of PEF thin film (21 μm).

As an ideal transparent conductive electrode for thin-film solar cells, the light-scattering capacity of GZO/PEF thin films is a very important factor. Fig. 3 shows the diffuse transmittance and haze values of GZO/PEF thin films. In this figure, the dashed lines indicate the diffuse transmittance of GZO/PEF thin films, and the solid lines indicate the

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