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Effects of cyclic deformation on conductive characteristics of indium tin oxide thin film on polyethylene terephthalate substrate

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ABSTRACT

Effects of cyclic deformation and annealing treatment on conductive durability of indium tin oxide (ITO) thin film deposited on polyethylene terephthalate (PET) substrate are investigated. In-situ electrical and mechanical tests of ITO/PET sheet under various combinations of cyclic and static loadings are conducted at room temperature. Experimental results show that the number of cycles to failure is significantly decreased with an increase in displacement amplitude, given a specific extent of electrical resistance change of ITO/PET sheet. A static holding period of 1000 s in various loading modes plays a role in influencing the failure of ITO/PET sheet. Cyclic bending combined with a static tensile holding generally generates more damages and a smaller number of cycles to failure than does that combined with a compressive holding, neutral holding, or no holding. Under a small fatigue loading, the conductive durability of ITO/PET sheet is increased with an increase in annealing temperature. However, there is little effect of annealing temperature on ITO/PET fatigue life under a larger displacement amplitude of fatigue loading. Using a surface-based cohesive modeling technique, a simplified three-dimensional finite element analysis micromodel subjected to tensile and compressive loadings is numerically analyzed to clarify the failure mechanism of interfacial and buckling-like delamination which governs the change in electrical conductivity of ITO/PET sheet. Modeling results indicate that buckle height of the ITO/PET micromodel subjected to tensile loading is significantly greater than that of compressive loading, providing more evidence of the aforementioned effect of loading mode.

1. Introduction

In recent years, there has been increasingly considerable interest in flexible optoelectronic devices which have significant advantages, such as flexibility, light weight, low cost, large screen, and mass roll-to-roll manufacturing capability [1–5]. In flexible optoelectronic technology, indium tin oxide (ITO) on polyethylene terephthalate (PET) thin film is being commonly used as a transparent electrode in organic light emitting diode (OLED), organic photovoltaic (OPV), organic field effect transistor (OFET), organic photo detector (OPD), flexible liquid crystal display (LCD), and touch panel due to its low electrical resistivity, extreme transparency, and high flexibility of substrate [1–11]. However, as a natural brittle material, durability of ITO thin film is the main challenge in the flexible optoelectronic technology development, which is in need of consideration.

In general, there are no testing standards for mechanically durable behavior of conductive thin films. The most commonly-used testing methods of mechanical durability are uniaxial stretching test and compressive test [12–17]. In addition, various mechanical testing methods such as scratching test, twisting test, and impact test are also designed and performed to investigate the failure mechanism of flexible thin films [18–21]. It is found that electrical resistance of ITO thin film changes dramatically at a high strain due to an increasing number of cracks on coating surface or interfacial and buckling delamination of bonded layers [22]. Cyclic bending tests are widely introduced to address the damage accumulation of brittle thin film on polymeric substrate [23–34]. Some cyclic bending tests are also incorporated with an in-situ electrical resistance measurement technique during deformation of samples [24–29,32]. Under applied loading, PET substrate undergoes distortion which leads to the deformation of ITO thin layer. It is found that the number of bending cycles has a dominant influence on the failure of ITO thin film [23]. Controlling environmental condition is also crucial on quality of product during roll-to-roll process due to the influence of temperature and humidity on electrical failure of flexible thin films [23]. It is noted that ITO thin films also significantly degrade when subjected to a combined effect of corrosion and fatigue by immersing them in various concentrations of acid solution, for example, acrylic acid environment [20,35–38]. For ITO thin film under static loading at different bending curvatures, it is reported that conductivity of ITO on plastic substrate is prone to decreasing even with no change in surface profile [39]. The situation becomes even worse for a critical bending cylinder with a diameter of 15 mm [39]. During the roll-to-roll manufacturing process and/or routine use of flexible devices,

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these thin films are repeatedly subjected to cyclic bending and static loading. Such loadings could produce and accelerate cracking leading to a significant electrical resistance change of ITO thin film. For these conditions, the effect of combined cyclic and static loading on electrical conductivity may enhance degradation of ITO thin film performance and should be considered. Although there area variety of mechanical testing methods in characterizing failure mechanism of ITO thin film on flexible substrate under various types of loading [11–41], however, studies on the effects of combined cyclic–static loading and annealing treatment on the conductive durability of ITO thin film are limited.

The coupling of interfacial and buckling delamination is a typical failure mechanism of ITO on polymeric substrate [22,40,42]. Aiming to investigate the buckling mechanism of these thin films under large deformation, some studies have been conducted to characterize the effect of interfacial and buckling delamination on cracking behavior of brittle film on polymeric substrate due to mismatch of stiffness or modulus between adjacent materials [19,22,40,42,43]. It is shown that inserting a specific conductive interlayer such as Ag and durable carbon nano-tube (CNT) or a buffer layer of silicon dioxide (SiO_2) may enhance resistance for cracking and delamination [24–26,34]. So far, a number of analytical studies have been conducted to predict interfacial and buckling failure mechanism of thin film structures under mechanical loading [42–46]. Incorporating with experimental work, the failure mechanism of thin film under compressive and bending deformation has been explored by numerical modeling [16,42–47]. In reality, ITO thin film on PET substrate often experiences an external and/or residual tensile stress during the roll-to-roll manufacturing process and/or routine services. Under tensile and/or compressive loading, cracks may initiate at and propagate from the initial defects on ITO thin film leading to an electrical resistance change of ITO thin film. It is attributed to the coupling of interfacial and buckling-like delamination. Most of the previous studies were mainly focused on the failure mechanism of ITO thin film under compressive and bending deformation [16,42–47]. However, there is still lack of study on the interfacial and buckling-like delamination of ITO thin film under tensile loading, in particular, when subjected to combined cyclic–static loading.

The purpose of this study is to investigate the long-term durability of ITO thin film on PET substrate under various combinations of cyclic–static bending. In particular, the change of electrical resistance is characterized so as to clarify the effects of deformation on electrically conductive properties of the ITO film under applied cyclic loadings. In addition, the effects of annealing temperature on the deformation-induced change of electrical conductivity are also characterized for a certain type of cyclic loading. A finite element analysis (FEA) technique is developed to simulate the mechanical behavior and offer more understanding of the failure mechanism that governs the change in electrical conductivity of ITO thin film using a surface-based cohesive model. Fractography and microstructural analyses are conducted with scanning electron microscopy (SEM) to find the failure mechanism after mechanical tests. It is hoped that this study may provide helpful information for users to prevent the failure of ITO thin film on polymeric substrate and to reduce the operational cost of flexible optoelectronic devices as well as the cost in roll-to-roll manufacturing process.

2. Experimental procedures

2.1. Material and sample preparation

The as-received ITO thin film on PET substrate (ITO/PET) sheets in the present study was commercially purchased from Win-Optical Technology Co., Ltd. (Tao-Yuan, Taiwan). These ITO/PET sheets consist of a 100-nm thick ITO layer deposited on a 125-μm thick PET substrate. According to the manufacturer's datasheet, no heat treatment has been performed on the as-received sheets. In order to determine the effect of annealing temperature on the mechanical durability of ITO thin film under mechanically cyclic loading, the as-received ITO/PET sheets were separated into several batches and annealed in air at selected temperatures of 95 °C, 125 °C, and 150 °C for 30 min. In this study, the ITO/PET sheets annealed at 125 °C were selected to characterize the effects of bending displacement amplitude and loading mode. From these annealed sheets, rectangular samples in physical dimensions of 150 mm (length) × 12 mm (width) were cut out to conduct mechanical tests. The thermal annealing treatments were all carried out in a temperature-controlled furnace. X-ray diffraction (XRD) analyses of the as-received and annealed ITO/PET films were then performed using an XRD instrument (D2 PHASER, Bruker AXS Corporation, Karlsruhe, Germany). The diffraction pattern was obtained at room temperature with a wavelength $\lambda = 0.15406$ nm over scanning angle (2θ) from 10° to 80°.

2.2. Mechanical testing

In the current study, a bending fixture is designed and made to apply cyclic and static loading on the ITO/PET samples. Each sample was firstly clamped to the fixture at both ends by two plates, as shown in Fig. 1. In order to avoid initial damages of clamping, electrical resistance of the investigated sample was monitored to ensure no artificial effect while setting up the experiment. In dynamic bending tests, the investigated specimens were pushed up and down for various numbers of cycles at a frequency of 1 Hz by two small pins (diameter of 6 mm). This would produce periodic compressive-tensile stress on the ITO layer, as shown in Fig. 1. In addition to pure cyclic bending (Mode I) test, three other combined cyclic–static bending tests were conducted to characterize the interaction effects of cyclic and static loading, as shown in Fig. 2. When the sample is bent in a concave downward position (Fig. 2c), ITO thin layer is under compression, which is defined as static

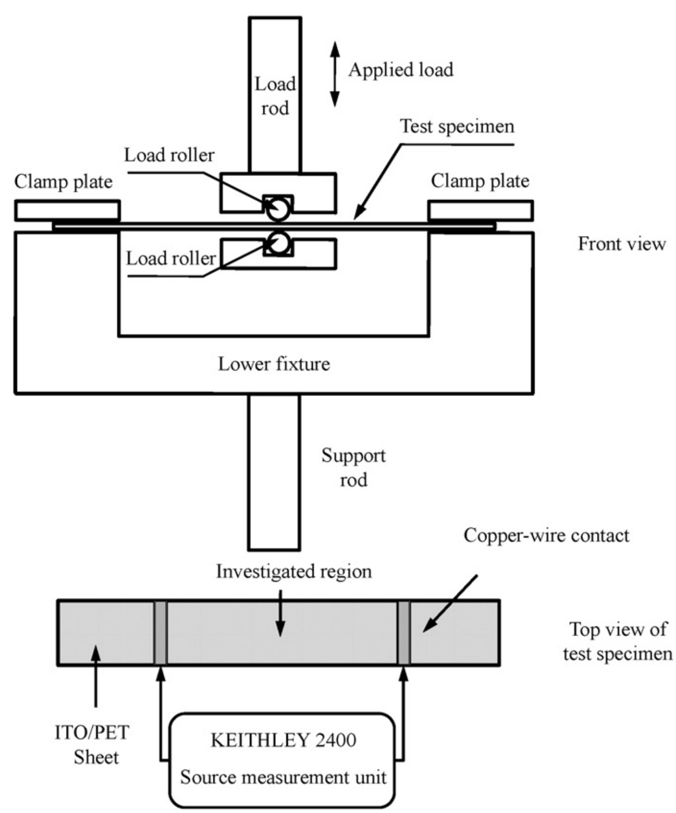

Fig. 1. Schematic of cyclic bending test and in-situ measurement of electrical resistance.