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Reliability of thin films: Experimental study on mechanical and thermal behavior of indium tin oxide and poly(3,4-ethylenedioxythiophene)

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A B S T R A C T

In order to improve the performance of various flexible electronic devices, the research on the transparent conductive thin films becomes very intensive in the recent years. In this work, we studied mechanical and thermal behaviors of two types of transparent conductive thin films, Poly(3,4-ethylenedioxythiophene) (PEDOT) as an example on polymer conductive thin film, and indium tin oxide (ITO) as an example on transparent conductive oxide thin film. Both films are deposited on polyethyleneterephthalate (PET) substrate. Two sheet resistances for PEDOT (i.e., 150 Ω/sq and 225 Ω/sq) and one sheet resistance for ITO (i.e., 60 Ω/sq) were involved in the study. PEDOT showed good mechanical properties with a small electrical resistance change and no clear cracks or deformation on the film surface under the condition of cyclic bending. However, the resistance of ITO significantly increased with the cyclic bending and cracks were seen initiated in the center of sample and propagated toward the edges. Further, design of experiment approach was used to study the effect of different cyclic bending parameters, such as bending diameter and frequency. Additionally, damp heat experiment on similar samples was conducted by applying 85 °C temperatures and 85% relative humidity on them for 1000 h. The electrical resistance was dramatically increased for both films. Scanning electronic microscopy (SIM), Energy Dispersive Spectrometry (EDX) and transmission tests were also used to determine the change of films' compositions and transparencies.

1. Introduction

Flexible electronic displays have unique features, such as light weight, low cost, and flexibility. They are used in many applications such as, solar cell, organic light emitting diode (OLED), and electronic sensors [1]. There were many efforts on improving conductive films to increase the solar cell efficiency. Two of the most common conductive transparent thin films are polyethylene dioxythiophene (PEDOT) and tin indium oxide (ITO). They are very common in liquid crystal displays (LCDs), sensors, and emissions devices applications [2,3].

ITO is a brittle transparent conductive oxide, while PEDOT is a ductile polymer. Many researchers paid attention to show the response of the ITO films to mechanical and thermal stresses. For example, Leterrier et al. [4] noticed that process-induced internal stresses were systematically compressive, and the tensile cracks in the ITO layer always start from surface defects. Further, they noticed that transition from stable to unstable crack growth starts at crack length of several hundred times of the coating thickness. Furthermore, they found that polarization of hard coats provides a good quality surface for polymer substrate and enhances cohesive specifications of ITO thin film.

Many researchers studied mechanical properties of different thin film materials that were exposed to cycling bending and thermal stresses [5]. Alzoubi et al. [6] investigated the effect of high and low cycle bending on electrical properties of copper thin film coated flexible substrate. In their study, they observed that the fatigue failure was developed faster at strain of 2.6% or higher. Also, Hamasha et al. [7] carried out an experiment to study the effect of cyclic bending fatigue on the surface integrity of ITO under harsh environmental condition. They conducted, however, the same experiment on similar samples, but without involving any bending to make a comparison and figure out the effect of bending fatigue itself. They found that the combination of high temperature and humidity under bending fatigue condition is critical on the conductivity and the surface integrity of the thin film.

Park et al. [8] conducted a bending test on the single crystalline silicon coated plastic substrate. They observed three types of

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failure modes when crystalline silicon exposed to different strain rates: slipping, cracking, and delamination. Cairns and Crawford [9] studied the effect of cyclic bending and thermal stresses on ITO and PEDOT thin films coated PET. They found that the ITO thin film was susceptible to damage more than the PEDOT thin film under tensile stress. Moreover, it was observed that cracks start to appear in the ITO film at low rate of strain (less than 2%), while PEDOT thin film had great mechanical properties. However, the PEDOT film suffers with a severe degradation under high temperature conditions. The cracks propagation in thin films deposited on flexible substrate causes electrical failure, while the failure in the bulk materials is caused by a single sudden crack [4]. Strain level, number of cycles, material properties and thickness of thin film affect the intensity and size of cracks in the films [10,11]. Stephenson et al. [12] used the photographic technology in manufacturing flexible displays to minimize residual stresses. In their work, two experiments were conducted: cyclic bending and damped heating experiments to investigate behavior ITO and PEDOT coated flexible plastic substrate under mechanical and thermal stresses. Grego et al. [13] used the dry-etch approach to generate cracks at relaxed position. Some research focused on the change in electric resistance of PEDOT and ITO coated flexible substrate under bending fatigue. In this research, reciprocating dies were used to create cyclic bending of the structure of thin film and substrate [9]. Moreover, some studies showed the benefit of PEDOT on organic photovoltaic (OPV) performance. For example, it is used as a buffer layer between ITO layer and photoactive layer (PAL) leading to increase open circuit voltage (VOC) and power conversion efficiency [14–16].

High temperature generates thermal stresses leading to a weakness in the cohesion between thin film and substrate, and it affects the structural and the physical properties of thin film. Hamasha et al. [17] investigated the behavior of ITO coated flexible PET exposed to fluctuating temperature for a certain period of time. In their study, surface and structure of material were inspected by SEM, and they observed clear thermal damages on the surface. In this paper, cycle bending test was conducted to show effect of bending diameter, sheet resistance, frequency and number of cycles on electrical resistance ITO and PEDOT. Further, damp heat experiment was conducted on the same films to investigate the effect of the constant high temperature and humidity on the films. Furthermore, Christou and Paradee investigated the fatigue behavior and effect of crack propagation in lead free solder in microelectronic packaging [18].

2. Experiments

The thin films used in this work were PEDOT and ITO coated PET. Two different tests were conducted, bending fatigue, and damp heating, to investigate electrical and mechanical properties of these films under bending fatigue, and damp heating conditions.

2.1. Bending fatigue test

Two different PEDOT sheet resistances (150 Ω/sq and 225 Ω/sq) were selected for the test, while only one ITO sheet resistance (60 Ω/sq) was selected. All films were deposited on 127 μm thick polyethylene terephthalate (PET). Each sheet was cut into rectangular stripes with 9.5 cm in length and 1.3 cm in width, as shown in Fig. 1. Both ends of the sample was fixed with a paper strips from the substrate side to make a loop of sample and paper strip. The sample ends from the thin film side were connected with the ohmmeter terminals using copper tapes. The loop of the sample and strip was secured in on the cycle bending device as shown in Fig. 2. A weight of 75 g was clipped on the loop from the ground side to ensure continuous connection between the die and the sample during the bending. The cycle bending device is illustrated in Fig. 2. It consists of electrical motor that provides rotation movement, which is converted using a connector into cyclic vertical displacement or, in other words, reciprocating motion of the die. The device designed to hold a single (see Fig. 2a–c) or double dies (see Fig. 2d–f). In the single die setup, the sample must be bent cyclically around the die during the upward stroke and kept in the flat position during the downward stroke. In the double die setup, the sample must be bent cyclically around the lower die during the upward stroke and around the upper die during the downward strokes.

At upward stroke for any setup (Fig. 2b or e), the upper surface of the sample (thin film side) is exposed to tension stress, while the lower surface (substrate side) is exposed to compressed stress. Tension and compression stresses are formed in the vertical cross sectional direction under bending loads. The maximum tension is formed at the upper surface of the sample (thin film side), while the maximum compression stress is developed at the lower surface. Since the relationship between the stress magnitude and its perpendicular position is linear, a plane of no stress (i.e., neutral axis) on the linear path between the two opposing maxima is formed. The tension or compression stress of a certain point is determined by the classical formula $\sigma = My/Ix$, where M is the force moment about the neutral axis, y is the perpendicular distance between the point and the neural axis (y is zero at the neutral axis, positive in the upward direction, and negative in the downward direction) and Ix is the second moment of area. According to the previous formula, the maximum tension is developed at the thin film and the maximum compression is developed at the lower surface of the substrate. However, in the downward stoke in the double die setup, the upper surface of the thin film side is exposed to compression stress, while the substrate side is exposed to tension stress. The same way of analysis is applied in this case. Thus, the positive direction of y becomes downward so the maximum tension is developed on the lower surface of the substrate and the maximum compression is developed on the upper surface where the thin film is located. However, both surfaces will be at neural stress in the single die setup. If the film is brittle (e.g., ITO), the cyclic tensile stress of the thin film leads to fatigue, and then the cracks are initiated in the area of maximum stress (midsection of the thin film). Then, the cracks will propagate at the line of less energy (perpendicularly to the direction of load toward the edges). In ductile films (e.g., PEDOT), the fatigue and cracks are not expected to be noticed. However, other type of failure like delamination, or damage may be seen.

DOE is considered as an essential key for design improvement and development in manufacturing sector [19]. It is mainly used by production quality engineers to determine significant factors that affect products' defects. Also, electronic packaging scientists use this tool extensively in analyzing parameters and factors that affect electronics resistance aiming to build design with high reliability [20]. For example, Dasgupta et al. [21] studied the factors that affect adhesion compound between liquid crystal polymer and the die using DOE analysis. In addition, nanoparticle conducting films were investigated for resistivity to heat transfer through studying the main effect parameters using DOE analysis [22]. The examples on using DOE procedures to investigate the significant impact of certain parameters on a certain response in the field of material science and thin films technology are so many (e.g., [23–30]).

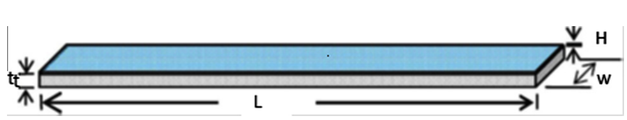

Fig. 1. Sample shape for both PEDOT and ITO coated PET (t: thickness of substrate, H: height of conductive layer, L: length, and w: width).