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Mechanical properties of amorphous indium–gallium–zinc oxide thin films on compliant substrates for flexible optoelectronic devices

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ABSTRACT

Amorphous indium–gallium–zinc-oxide (a-IGZO) thin films were deposited using RF magnetron sputtering on polyethylene naphthalate (PEN) and polyethylene terephthalate (PET) flexible substrates and their mechanical flexibility investigated using uniaxial tensile and buckling tests coupled with *in situ* optical microscopy. The uniaxial fragmentation test demonstrated that the crack onset strain of the IGZO/PEN was ~2.9%, which is slightly higher than that of IGZO/PET. Also, uniaxial tensile crack density analysis suggests that the saturated crack spacing of the film is strongly dependent on the mechanical properties of the underlying polymer substrate. Buckling test results suggest that the crack onset strain (equal to ~1.2%, of the IGZO/polymer samples flexed in compression to ~5.7 mm concave radius of curvature) is higher than that of the samples flexed with the film being in tension (convex bending) regardless whether the substrate is PEN or PET. The saturated crack density of a-IGZO film under the compression buckling mode is smaller than that of the film under the tensile buckling mode. This could be attributed to the fact that the tensile stress encouraged this crack formation originating from surface defects in the coating. It could also be due to the buckling delamination of the thin coating from the substrate at a lower strain than that at which a crack initiates during flexing in compression. These results provide useful information on the mechanical reliability of a-IGZO films for the development of flexible electronics.

1. Introduction

Electronic devices fabricated on flexible plastic substrates are expected to find a variety of new applications due to their attractive advantages, such as being mechanically robust, light weight and potentially having simple roll-to-roll-based fabrication and mass production [1,2]. Transparent oxide semiconductors, such as zinc oxide (ZnO), gallium-doped zinc oxide (GZO), indium tin oxide (ITO), zinc tin oxide (ZTO), and indium gallium zinc oxide (IGZO) have attracted many researchers with their large potential in flexible optoelectronic applications, such as transparent electrodes in solar cells, rollable displays, and channel layers in transparent thin film transistors TTFTs [1,3], because of the capability of large-area, uniform deposition at low temperatures [4]. In particular, IGZO has drawn considerable attention as an extremely promising alternative to hydrogenated amorphous silicon (a-Si:H) for thin-film transistors (TFTs) due to its high electron mobility, processing compatibility with plastic substrates, good-uniformity and high transparency in the visible wavelength region (400–700 nm) [4,5].

Polyethylene terephthalate (PET) and polyethylene naphthalate (PEN) have been widely used as the base substrates in the fabrication of flexible optoelectronic devices. This is due to their satisfactory optical transmittance, mechanical flexibility, light weight, transparency, low cost and ability to be manufactured through roll-to-roll processing. However, due to the low melting point of the polymers, transparent oxides must be fabricated on polymer substrates at a lower deposition temperature. The RF magnetron sputtering technique is one of the most commonly used methods to deposit a-IGZO thin films on flexible substrates at room temperature [6]. Many research groups have reported using radio frequency (r.f.) sputtering to fabricate a-IGZO layers for use in thin film transistors (TFTs) on flexible substrates [6].

However, transparent conducting oxide films are brittle by nature, and susceptible to cracking and/or buckling delamination under externally applied mechanical deformation, which significantly limits the flexibility of the devices [7]. Consequently, failure behaviour of the films under various loading modes such as stretching, bending, or twisting becomes a critical issue during both manufacturing processes and in service conditions [8]. Hence, this gives rise to the motivation for predicting the onset of failure such as critical strain and critical radius of curvature to provide this information to optoelectronic device designers.

Uniaxial tensile and buckling tests, coupled with *in situ* optical microscopy are commonly used to determine the failure strains of thin coatings adhering to a compliant substrate, where cracks first initiate, as well as any subsequent buckling and delamination that may occur

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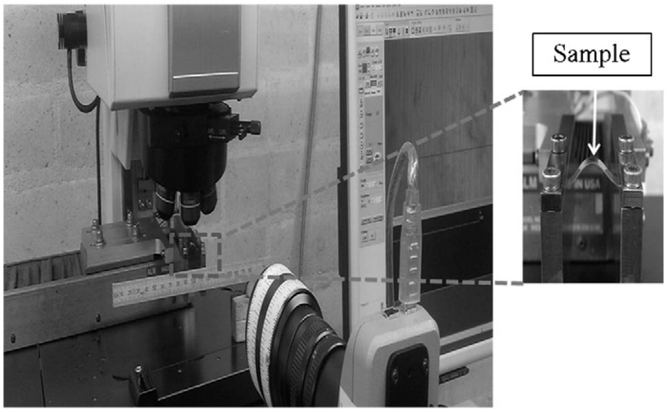

Fig. 1. The bending test set-up used in this study.

[9]. For example, the mechanical behaviour of ZnO coated polymer substrates was investigated under compression tests by Sierros et al. and the critical onset strain for cracking was found to be approximately 2% [10]. Chen et al. [11] reported buckling experiments carried out on ITO, reporting that the crack onset strain (COS) of the film under tension at 1.1% was less than its value under compression. In addition, Ni et al. [12] investigated the fracture properties of AZO-coated on PET substrates under simple-support bending conditions. It was reported that the coating damage, under tensile strain, is caused by the creation of channel cracks, while under compression the film specimen may first delaminate from the polymer substrate and then buckle before the initiation of a crack. They explained that in tension, on the outer surface, the stored elastic energy went solely into crack formation; however in compression the elastic energy was distributed between crack formation and delamination, so the crack density is usually lower for bend-testing on the compression side.

However, relatively little research to date has been reported about the mechanical behaviour of a-IGZO coated on polymers. Cherenack et al. [2] investigated the performance of amorphous thin film transistors (a-IGZO) under mechanical bending. It was pointed out that the films can be flexed down to 10 mm radius of bending curvature and, still, can remain functional. Also Munzenrieder [13] investigated the behaviour of a-IGZO TFTs on flexible substrates under tensile and compressive stress conditions. It was found that the mechanical stress has considerable impact on the TFT mobility and threshold voltage. Gleskova et al. [14], demonstrated that the amorphous silicon TFTs fabricated on polyimide foil can be strained more in compression than in tension. No mechanical failure was noted in compression for strains smaller than 2%, while in tension, mechanical failure was found at a strain of 0.5%. They also pointed out that the failure mode was the formation of periodic cracks perpendicular to the straining direction. Such cracks interrupt the current path if the source-drain current path and the strain direction are parallel.

In this work, we report on the mechanical flexibility of a-IGZO thin film grown on PET and PEN substrates *via* RF magnetron sputtering at room temperature for use in flexible optoelectronics. We utilize two different polyesters in an effort to elucidate the effect(s) of the underlying substrate on the resulting mechanical performance of the sputtered films since the applied stresses are transferred from the substrate to the coating through the interface. We examined the mechanical integrity of a-IGZO by using uniaxial tensile fragmentation and buckling tests coupled with *in situ* optical microscopy to further understand the failure mechanism under different mechanical deformation modes. Although the performance of the coated substrates will be modified once they are in a complete device, with potentially several other layers above or below, an understanding of the mechanical properties of the IGZO/polymer layers remains of fundamental importance.

2. Experimental procedure

The polymer substrates used were two semi-crystalline polyesters, 0.125 mm thick polyethylene terephthalate (PET Melinex ST 504) and 0.125 mm thick polyethylene naphthalate (PEN Teonex Q65FA). Samples of both were supplied in the form of A4 sheet (DuPont Teijin Films, UK). Thermal and mechanical properties of both substrates were measured by using differential scanning calorimetry and uniaxial tension (Instron 4410) respectively. a-IGZO film of thickness ~50 nm was deposited on to polymer substrates using RF magnetron sputtering from a In_2O_3:Ga_2O_3:ZnO (1:1:1) target (99.99% purity), the samples having a dog-bone shape (50 mm length, with 18 mm gauge length and 4 mm gauge width). The substrates were cut from sheets using a Moore Hydraulic Press. Prior to introduction inside the sputtering chamber, the polymer substrates were ultrasonically cleaned in acetone, ethanol, and then in deionized water for 5 min each. Deposition was performed (without heating the substrate) in an argon atmosphere and without an oxygen feed. A 4-in. diameter ceramic target, 20 cm from the substrate, was used under a base pressure of 5.1×10^{-6} Pa; constant RF power of 55 W; deposition pressure of 0.5 Pa; power density of 0.7 W/cm^2; Ar flow rate of 50 sccm (sccm denotes standard cubic centimetre per minute at STP conditions) and deposition rate of ~3.3 nm/min. In order to remove contaminant on the surface of the target, the a-IGZO target was presputtered for 5 min before the deposition of the film. The optical transmittance of the films was measured in the visible range from 400 to 800 nm using a Jenway 6310 spectrophotometer. Moreover, X-ray diffraction was used to examine the structural properties of the a-IGZO films. The IGZO deposition conditions were previously optimized to produce thin film transistors with a high mobility, low threshold voltage and large switching ratio. Examples of TFT characteristics have been reported previously in [15].

The mechanical flexibility of the a-IGZO films deposited on the polymer substrate was evaluated by uniaxial tensile and buckling tests. The uniaxial fragmentation test was performed using a Miniature Material testing machine. The test was coupled with *in situ* optical microscopy; images were taken every 3 s during the test in order to monitor the critical onset strain and development of the cracking of thin film as the applied tensile strain increases. Equipment originally designed to determine the critical failure strain of optical fibres was slightly modified to test the a-IGZO/polymer samples. Fig. 1 shows the experimental set-up used in this case. The sample is clamped between the two parallel plates, where one plate is movable, while the other plate is fixed. The distance between the two parallel plates was measured using a side-view digital imaging system and image analysis software (Image J). Crack initiation and propagation were carefully monitored by using confocal microscopy. *In situ* optical observation was used as a simple and effective way to determine the crack-initiation strain of thin film, particularly as the film is non-electrically conductive [16]. The values of the resulting strain from buckling were calculated using the following equation [17].

$$\in = \frac{h_s}{R} \tag{1}$$

where *R* is the radius of curvature and h_s is substrate thickness.

Scanning electron microscopy (SEM) was performed to investigate the microstructure of the IGZO coatings and to characterize the cracking morphology of the a-IGZO films after testing. (The specimens were

Table 1
Thermal and mechanical properties of PET and PEN.

Property	PET Melinex ST 505	PEN Teonex Q65 FA
Glass transition temperature, Tg (°C)	78	119
Melting point, TM (°C)	257	263
Young's modulus, E (Gpa)	3	3.5