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The Fracture of Brittle Thin Films on Compliant Substrates in Flexible Displays

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Abstract

One mechanical issue in flexible Organic Light Emitting Displays (OLED) is the fracture of extremely thin brittle conducting transparent oxide films deposited on thin flexible substrates. Understanding the behaviour of these films under flexed condition is essential for designer of flexible OLED. Controlled buckling experiments on the film and substrate have been designed to study the fracture of the films under both tension and compression. Fracture of the film is superficially similar in both tension and compression. However, under tension a channelling crack is formed, while under compression, the film delaminates, buckles and cracks in a tunnelling motion. The fracture toughness of the film and the delamination toughness have been estimated from these experiments. Design to maximise the flexibility of the device is discussed.

Key Words: Thin Film, Fracture toughness, Delamination, Indium Tin Oxide, Flexible displays

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1. Introduction

There are many thin, layered devices in electronics, such as liquid crystal displays, organic solar cells and organic light emitting displays (OLED). Often advantage is gained by making them flexible. The OLED has a promising future in panel display industry not only because it can be made flexible, but also because it possesses other unique properties, such as lightweight, wide display angle, and high resolution [1-3]. One of the obvious ways to make flexible displays is to use polymer as the carrying substrate. In OLED, the anode materials are usually transparent conducting oxides (TCO) such as Indium-Tin Oxide (ITO). These materials can be either crystalline or glassy, depending on the deposition temperature, and are brittle by nature, which limits the flexibility of the devices. Depending on the design and materials selection, there may be other brittle components in the device but the use of TCO is inevitable. Our priority is to study the failure strain of this type of material as deposited on the substrate. The motivation for this work comes from the need to maximise the flexibility of such a device in which thin brittle layers are deposited on polymeric substrate.

In order to design flexible devices, it is necessary to know the flexing limit of the brittle component, the TCO films. Depending on the relative position of the film to the substrate, there are two possibilities of film failure: the film may fracture when subjected to tension and delaminate (with or without fracturing) when subjected to compression. The materials properties controlling the failures are the fracture toughness of the TCO films and the interfacial toughness between the TCO and its substrate, respectively. The fracture toughness of thin film depends upon the deposition process and cannot be obtained from bulk properties. Adhesion between the film and its substrate is another important issue and is often cannot be easily measured. In this paper it is shown how both the fracture toughness and the interfacial toughness of an ITO film can be measured using a novel-testing scheme.

2. Experimental Details

Controlled buckling is the most appropriate means of straining the thin ITO coated polymer substrate because it is able to attain large curvatures (see Figure 1). The samples can be simply supported or, to achieve even larger strains, clamped. The most suitable way of clamping can be selected according to the flexibility of the specimen. In our particular case, we found clamped test is more appropriate.

Commercially available ITO-on-PET polymer substrates have been tested. A typical ITO coated polymer sheet has a total thickness of about 0.18 ~ 0.20 mm, while the ITO's thickness is between 80 to 140 nm. The plane strain moduli of the ITO film and the polymer substrate materials are very disparate (approximately 250 and 4 GPa respectively). The modulus of ITO has been obtained the nano-indentation method, and the modulus of the PET from published values.

The tests were carried out in an Instron machine, where both the load and the displacement were recorded. Since the film is conducting and the substrate is non-conducting, cracking of the ITO film can be determined from a loss in electrical conductivity. Thus an ohmmeter was used to monitor the resistance in the film along the loading direction during both the tension and compression tests. As will be shown later, a sudden loss of conductivity in the resistance - strain curve indicates that film has cracked.

The testing scheme in Figure 1 can be analysed as a plane strain beam loaded along its axis. Large deformation buckling theory of beams gives [4]:

$$\lambda = 2\left[1-\frac{E(k)}{K(k)}\right]; \quad \frac{l}{R} = 4K(k)k \tag{1}$$

where $K(k)$ and $E(k)$ are complete elliptic integrals of the first and second, $k=\sin(\theta/2)$, L the original length of the beam, R the radius of curvature, and $\lambda=e/L$ the contraction ratio. For the two schemes in Figure 1, $l=L$ for simple support and $l=L/2$ for built-in ends. Therefore from

measuring the geometry, the radius of the bending can be calculated by measuring the shortening of the beam, *e*. Since the ITO film is so thin compared with the substrate (see Table 1 below), the neutral axis of the composite is very near the centre of composite. Thus the strain in the ITO is given by

$$\varepsilon = \frac{\left(h_s + h_f\right)}{2R} \tag{2}$$

where h_s and h_f are the thickness of the substrate and film respectively. Samples were tested with the ITO film placed on both tension side and compression side. Resistance - strain curves were recorded. Scanning electron microscope (SEM) and atomic force microscope (AFM) observations were made on the films after testing.

Three types of commercially available ITO/polymer samples were tested. The specifications are listed in Table 1. Sample 1 was tested with the film in both the tension and compression. It was found that the test results were very stable, i.e., the measured critical strains for all the specimens from the same sample were very close. Therefore three specimens from each sample were enough and we took the average value as the critical strain for the following calculation.

3. Results and Analysis

Table 1 shows the critical strains of the three different types of ITO/polymer samples. Figure 2a shows one typical test record from sample S1 of the resistance (normalised by the resistance reading in the relaxed state) versus the strain on the ITO film when the ITO is in tension. Figure 2b shows a record from sample S1 for the ITO in compression. In both plots a sudden increase of the resistance can be clearly identified indicating that the film has been broken at that particular applied strain. The value at the point just before the sudden rise in resistance is taken as the critical strain. The critical strain under compressive loading for sample S1 was larger than its value under tension (1.7% and 1.1% in Table 1). Thus for this

particular combination of film and substrate, tension is the more critical. However there may be occasions that compression failure occurs at the lower strain than tension, which we will discuss later. The thickness of the film has a large influence on the critical strains as will be shown below in the analysis.

The SEM image shown in Figure 3a exhibits a typical fracture pattern obtained from the ITO film surface under tension. This kind of channelling crack has been studied before by Beuth [5], Hutchinson [6], and Hutchinson and Suo [7]. For film channelling cracks, steady-state propagation establishes quickly after the crack advances for a few film thicknesses. It has been shown that for an existing partially-through defect in a stiff film on a compliant substrate it is energetically favourable for the crack to run through the whole thickness once the initiation starts [5]. How the crack is initiated is unimportant since, following Hutchinson [6], we propose a conservative fail-safe design against propagation rather than attempt to determine initiation, which would depend upon very variable defects. The essence of this design approach is to prevent the steady-state crack propagation rather than initiation. This approach initially proposed by Hutchinson [6] has been widely accepted.

It was observed that once a channelling crack was formed, it and many others propagated almost simultaneously for large distances across the specimen to form multiple parallel cracks. These cracks do not interfere with each other until the spacing becomes very close (8 times the film thickness for the case studied by Thouless [8]). The steady state energy release rate, G_c is given as [5,6]

$$G_c = \frac{1}{2}\overline{E}_f \varepsilon^2 \pi h_f \, g(\alpha,\beta) \tag{3}$$

The factor $g(\alpha,\beta)$ is a function of the Dundurs' parameter, α and β, which for plane strain condition are given by

$$\alpha = \frac{\overline{E}_f - \overline{E}_s}{\overline{E}_f + \overline{E}_s}; \qquad \beta = \frac{\overline{E}_f \left(\frac{1-2\nu_s}{1-\nu_s} \right) - \overline{E}_s \left(\frac{1-2\nu_f}{1-\nu_f} \right)}{2\left(\overline{E}_f + \overline{E}_s \right)} \tag{4}$$

Of these two parameters α is much more important. The steady state fracture toughness, G_c^{cri}, is obtained by equation (3) when the applied strain, ε, reach its critical value. The value of the g factor can be computed by finite element method [5]. For our ITO-polymer combination, α=0.968 and β=0.158. The values of G_c^{cri} calculated from the average fracture strains in tension range from 50 to 60 J/m^2, see Table 1. This fracture toughness can be used as a conservative, defect-tolerant design parameter for the deposited ITO film. Note from equation (3) that if the fracture toughness is unaffected by the deposited thickness, the critical strain is inversely proportional to the square root of the film thickness.

Compressive cracking of the ITO films produces a similar parallel pattern to the tension cracking under lower magnification (cf. Figures 3a & 3b). However when we employed a high-resolution scanning electron microscope (field emission SEM in this case) and an atomic force microscope (see Figures 4a & 4b) to observe the details of the fracturing, it was revealed that the failure mechanism is buckling delamination closely followed by film cracking. As in tensile cracking, we concentrate on the steady-state delamination and cracking. In such a case, the total energy release rate, G_t, is given by

$$G_t = G_d + \left(\frac{h_f}{2b} \right) G_c \tag{5}$$

where G_d and G_c are for the energies released by delamination and film cracking respectively and 2b is the delamination width.

Hutchinson and Suo [7] have calculated the energy released during delamination and buckling of a thin film under compression. Thouless [9] extended this analysis to include cracking of the film. However, both of these analyses tacitly assume that the substrate is stiff.

Provided the substrate is as stiff as the film, these analyses are accurate, but if the substrate is more compliant than the film they are inaccurate. For example a preliminary estimate of the delamination toughness of ITO on a PET substrate of S1 sample using these analyses gave a value of 4.9 J/m^2 which is clearly too low. Realising that a compliant substrate releases significant energy when a film delaminates and buckles, we performed an analysis [10] that included the deformation of the substrate.

Based on the new analysis [10], the delamination toughness is estimated to be about 35 J/m^2. Fortunately the energy release rate is not very sensitive to the width of the delamination which has to be estimated from Figures 4. In this case the delamination toughness would be very much underestimated if the substrate effect were ignored. The new analysis also shows that if the substrate is compliant, the energy release rate reaches a maximum as the delamination spreads, in addition to an increase in mode II component of the delamination. Thus the stabilisation of the delamination, for compliant substrates, may be as much due to a maximum being reached in the energy release rate as to the increase in delamination toughness due to the mode II component. This finding is interesting but will not be discussed in further details in this paper. Interested readers please see reference [10].

4. Discussion

We have demonstrated that a controlled buckling test is suitable for testing thin films on substrate, especially when the film's electrical resistance can easily monitor the cracking. When the film is not conducting, other parameters such as the acoustic emission signal may be monitored. In theory there will be a curvature change as well as a load drop after the film cracking. However the change in bent angle is proportional to $\left(\overline{E}_f / \overline{E}_s\right)\left(h_f / h_s\right)^2$. From this expression it is clear that the thickness ratio (h_f/h_s) dominates the effect even when the film's Young's modulus is much higher than the substrate. For our testing samples, the film is much

thinner than the substrate (~ 1:1800), the load drop or displacement shift was unnoticeable even when there is multiple cracking. We have also attempted to use the sheet resistance approach on non-conducting film by coating an ultra-thin conducting layer.

It has been shown that a brittle stiff film on a soft compliant substrate cracks under both tension and compression. Under tension the failure mechanism is by channelled cracking of the film while in compression it is tunnelled buckling delamination and cracking. The fracture toughness and the delamination toughness derived from the tests provide vital information for the device designers. In our particular case, the critical strain under tension is less than the one under compression. Clearly the ratio of the film cracking toughness to the one of delamination decides which mode is more critical[2]. When the adhesion toughness is small, compressive failure may occur at a lower strain than under tension. Knowing the critical energy release rate for cracking, G_c^{cri}, and for delamination, G_d^{cri}, a simple comparison will tell which mode will fail first. The criterion is that when the ratio of delamination toughness to the cracking toughness is larger than the one of their energy release rates, i.e. when $G_d^{cri}/G_c^{cri} > G_d/G_c$, tension is more critical for the same amount of strain.

From design point of view, it is always possible to maximise the flexibility by placing the most critical component near the neutral axis of the lamination in order to minimise the strain in the critical film. When there are only one layer of film and one layer of substrate, the film is at the edge of the composite. Placing a "capping" layer on top of the film will push the film toward the neutral axis of the new composite where the strain is a minimum. To place the neutral axis at the centre of the film, the choice of the capping layer should follow the rule of

[2] Notice in equation (5), when the ratio of film thickness to the delamination width ($h_f/2b$) is small, the contribution from the film cracking, G_c, to the total energy release rate, G_t, is insignificant. This is indeed true in our case where the delamination width is about 2 μm and the film thickness is only 0.1 μm.
The required delamination plus cracking energy is not much more than delamination alone.

$\overline{E}_c/\overline{E}_s = (h_s/h_c)^2$, in which the subscripts *c* stands for the cap layer and *s* for the substrate. Based on this principle, minimisation of the risk of failure for any number of layers is always possible. Another advantage of sandwiching the brittle film, compared with the film-on-side design, is that the cracking energy release rate will be lower when the strain in film is the same, provided that the adhesion between layers is perfect. Therefore the allowed bending will be higher.

Reducing the film or device thickness will increase the allowable strain in the film, and thus higher bending strain of the device. When the thickness is reduced to half, the allowed bending strain will be 40% higher, and so does the bending curvature. Therefore within performance requirements thin film is preferred. The modulus ratio $\overline{E}_f/\overline{E}_s$ plays its role through the *g*-factor in Equation 3. When the two moduli are closer to each other (smaller α), the value of *g* is smaller [6] and so is the energy release rate. The *g*-factor rises sharply for $\alpha>0.8$. For ITO/polymer combination, there is little that can be done to reduce the film modulus so the only way may be selecting a polymer or polymeric composite of higher modulus.

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Sample	Test Mode	Film thickness (nm)	Substrate thickness (μm)	Critical strain (%)	Toughness[a] (J/m^2)
S1	Film in tension	108	181	1.1	62
S1	Film in compression	108	181	1.7	35
S2	Film in tension	139	187	0.9	53
S3	Film in tension	109	181	1.0	52

[a]The value is the film cracking toughness when in tension and the delamination toughness when in compression.

Table 1

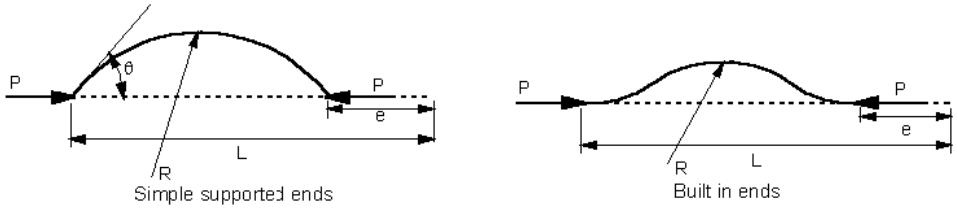
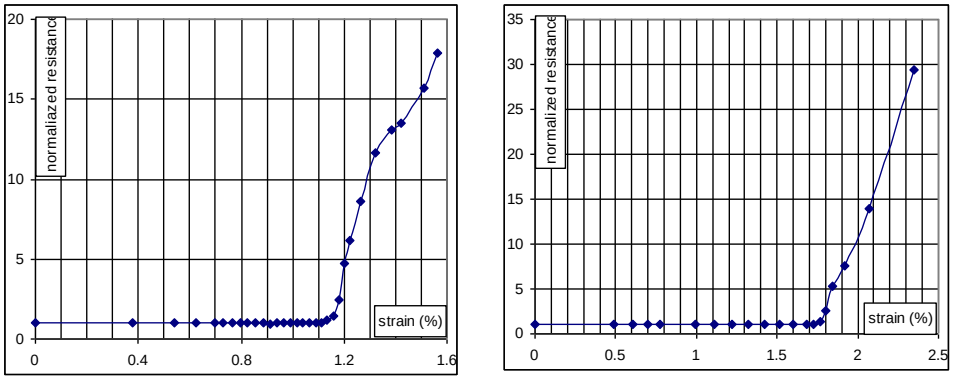


Figure 1



(2a) (2b)

Figure 2

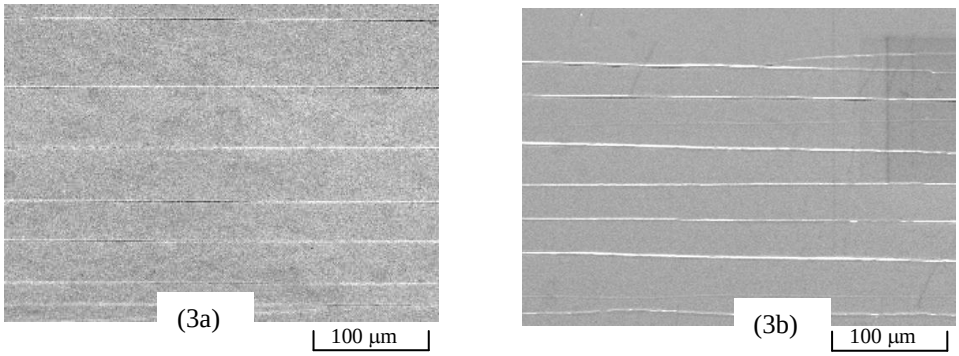
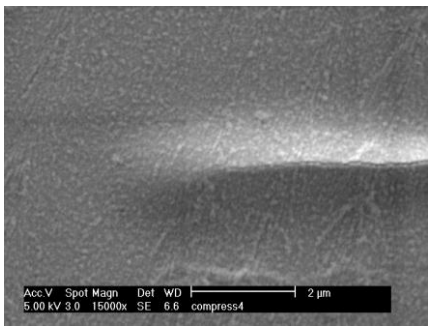
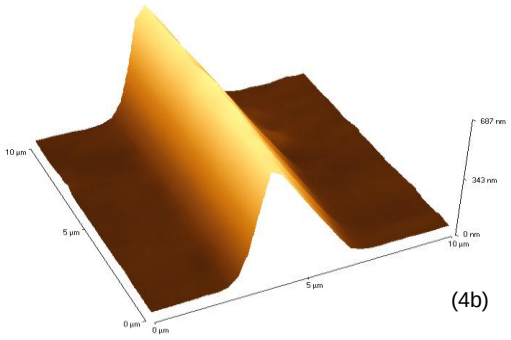


Figure 3



(4a)



(4b)

Figure 4