

A mechanical assessment of flexible optoelectronic devices

Zhong Chen^{a,*}, Brian Cotterell^b, Wei Wang^b, Ewald Guenther^b, Soo-Jin Chua^b

^a*School of Materials Engineering, Nanyang Technological University, Singapore 639798, Singapore*

^b*Institute of Materials Research and Engineering, 3 Research Link, Singapore 117602, Singapore*

Received 5 November 2000; received in revised form 29 December 2000; accepted 11 April 2001

Abstract

This work has demonstrated novel experimental methods and their relevant analysis to evaluate the fracture properties of thin brittle films on compliant substrates for flexible optoelectronic devices. Based on the understanding of the failure mechanisms, mechanical calculation has been provided to estimate the thin film fracture toughness and film delamination toughness. These values serve as design parameters on the device flexibility and reliability. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Flexible devices; Indium tin oxide; Interfaces; Optoelectronic devices

1. Introduction

In recent years there has been a growing interest in flexible optoelectronic devices such as: liquid crystal displays (LCD); solar cell panels; and organic light emitting displays (OLED) [1–4]. Many advantages are obtained when these devices are made conformable so that they are portable and can be rolled. For example, one possible application is to cover the roof of a building with flexible solar cells, which would be difficult with rigid ones. Because of their low cost, lightweight and mechanical flexibility, polymeric films are often used as the substrate in such devices to carry the active coatings. Some of these coatings, such as indium-tin oxide (ITO) or indium-zinc oxide (IZO) which are used as transparent anodes, are brittle and can withstand only quite limited strains. When a composite film is bent, one surface will be in tension while the other is in compression. Somewhere towards the middle of the film there will be a neutral axis where the strain, ε , is zero. At any other layer in the composite

film a distance y from the neutral axis, the strain is given by

$$\varepsilon = \frac{y}{R} \quad (1)$$

where R is the radius of curvature of the neutral axis (see Fig. 1). Much can be done to maximise the flexibility of the device by designing so that the brittle components are close to the neutral axis [5] where the strain is low. However, if the component cannot be placed near the neutral axis, the flexibility of the device may be limited by its brittleness.

The cracking strain for a brittle material in general depends upon both the crack resistance energy and the flaw size. It is difficult to quantify the largest possible flaw even in monolithic structures and the difficulty becomes almost insurmountable for the very thin coatings used in OLED. Hutchinson [6] has suggested a simple conservative design method that avoids the necessity of specifying the flaw size. In this concept, instead of designing to avoid the initiation of a crack, one designs to avoid propagation. In such a case, the critical flaw size is the coating thickness, h , which

* Corresponding author. Tel.: +65-790-4256.
E-mail address: aszchen@ntu.edu.sg (Z. Chen).

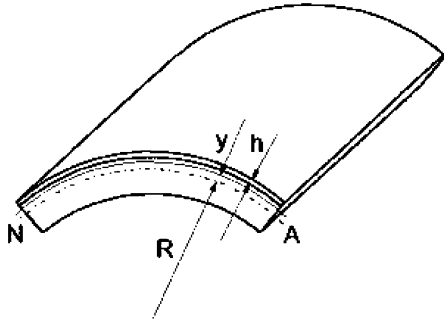


Fig. 1. A composite beam bent to a radius of curvature R .

decides the critical strain, ϵ_c . The critical strain is higher for thinner coatings as shown later.

The very thin brittle coatings used in OLED are very much more robust than the bulk material. In this paper we discuss how to determine the critical strain when the brittle coatings are in tension or compression.

2. Cracking of thin brittle films under tension and compression

When a thin brittle coating on a substrate is uniformly strained, either through residual or external strain, steady-state cracking is possible. That is, the state of strain and stress at the tip of a propagating crack is independent of the crack length and the rate of release of energy is constant. If this release of energy is equal to or greater than the energy required for the crack process, then the coating will fail. The crack process will be different depending upon whether the strain is tensile or compressive, but the principle of the analysis is the same. Once the crack length is more than a few coating thicknesses, propagation reaches a steady-state condition. The energy release rate for tensile film cracking follow the analysis in [6–8] and the one for film on compression will be discussed later. If the coating is isotropic the crack process will take place under the maximum principal strain. It is assumed in the analyses presented here that the substrate is very thick compared with the coating; some results for substrates comparable in thickness with the coating have been given by Hutchinson and Suo [7].

For plane strain elastic interface problems the stress state depends on the two Dundurs parameters α and β given by

$$\alpha = \frac{\bar{E} - \bar{E}_s}{\bar{E} + \bar{E}_s}; \quad \beta = \frac{\bar{E} \left(\frac{1 - 2\nu_s}{1 - \nu_s} \right) - \bar{E}_s \left(\frac{1 - 2\nu}{1 - \nu} \right)}{2(\bar{E} + \bar{E}_s)} \quad (2)$$

where $\bar{E} = E/(1 - \nu^2)$ is the plane strain elastic modulus, ν is the Poisson ratio, and constants without subscripts refer to the coating and with a subscript s refer

to the substrate. The more important of these two parameters is α which is a measure of the mismatch in the modulus between the coating and the substrate. The value of α can vary from -1 for a rigid substrate to 1 for an infinitely compliant substrate. Analyses are simplified if $\beta = 0$ and it has been argued that there is no practical loss in energy if this simplification is used [6,7].

2.1. Steady-state cracking under tensile strain

Under tensile strain a steady-state channelling crack can propagate along the coating. A channelling crack releases strain energy in the coating and substrate adjacent to the crack. The strain energy release rate, G_{ten} , is given by [7,8]

$$G_{ten} = G_0 g(\alpha, \beta) \quad (3)$$

$$G_0 = \frac{1}{2} \bar{E} h \epsilon^2$$

where G_0 is the strain energy per unit width stored in the coating ahead of the crack process, h is the coating thickness and $g(\alpha, \beta)$ is a function of the two Dundurs parameters (see Fig. 2). As the substrate becomes more compliant so the energy release rate increases. In the limit for an infinitely compliant substrate, that is no substrate, steady-state cracking is impossible [6].

A steady-state tension crack process is possible if the crack resistance energy, G_c , required to create a unit fracture area is equal to or less than G_{ten} . By equating G_{ten} in Eq. (3) to G_c , the critical strain, ϵ_c , is shown to be reversely proportional to the square root of coating thickness

$$\epsilon_c = \sqrt{\frac{2G_c}{\bar{E}g(\alpha, \beta)h}} = C \frac{1}{\sqrt{h}} \quad (4)$$

where C is a proportionality constant dependent on all the materials property terms. If the crack resistance,

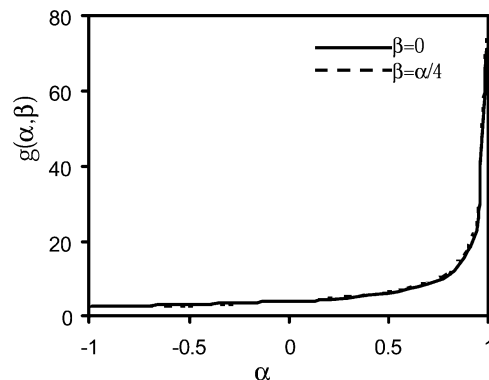


Fig. 2. The function $g(\alpha, \beta)$.

G_c , is known, the proportionality constant in Eq. (4) for the critical strain, ϵ_c , can be determined [7,8].

2.2. Steady-state cracking under compression

Although the brittle coating is in compression, the actual fracture mode for the coating is tensile. A tensile stress is achieved by the coating first delaminating and buckling before cracking as shown in Fig. 3. 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. Following the same line of attack by Thouless [9] and Hutchinson and Suo [7], we had performed a new analysis taking into consideration the substrate deformation [10]. The rate of release in energy by the crack process is given by [10]

$$G_{com} = G_0 f\left(\left[\frac{\epsilon}{\epsilon_b}\right], \epsilon, \alpha, \beta\right) \tag{5}$$

where f is a strong function of ϵ/ϵ_b , the ratio of the strain in the coating to the buckling strain for a coating on a rigid substrate, and α , and a weaker function of ϵ and β . The buckling strain for a coating on a rigid substrate is given by

$$\epsilon_b = \frac{\pi^2}{12} \left(\frac{h}{b}\right)^2 \tag{6}$$

where $2b$ is the width of the delamination. The function f is shown in Fig. 4 for one particular case of $\alpha = 0.9, \beta = 0$. The curves for a range of values of α can be found in [10]. Interested readers please see Cotterell and Chen [10] for more details. In compression the cracking process involves delamination and cracking. The energy release rate, G_{com} , is based on the delamination area and hence the resistance energy for the crack process is $G_d + (h/2b)G_c$ where G_d is the delamination interfacial energy. Hence, if the coating is

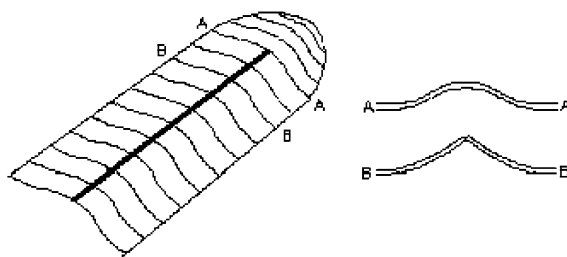


Fig. 3. A tunnelling delamination–buckle crack.

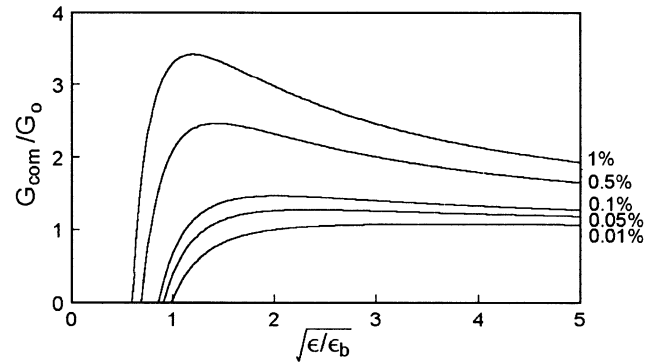


Fig. 4. The function $f([\epsilon/\epsilon_b], \epsilon, \alpha, \beta)$ for $\alpha = 0.9, \beta = 0$.

in compression, the critical strain energy release rate is given by

$$G_{com}^{cri} = G_d + \frac{h}{2b} G_c \tag{7}$$

As in the tensile case the critical strain has the form given by Eqs. (4) and (7) which enables the constant of proportionality to be determined.

3. Experiments to determine the energy resistance for cracking and delamination

The coating of interest was ITO on a polyethylene terephthalate (PET) film. The coating was 108 nm thick and the PET film 181 μm thick. The plane strain elastic moduli of the ITO film and the PET are 250 and 4 GPa, respectively, which gives the Dundurs parameters: $\alpha = 0.97$ and $\beta = 0.16$. The PET film is extremely flexible and the usual three or four-point bend test was inappropriate. The easiest means of straining the PET film is by displacement controlled buckling with either simple supported or clamped ends (see Fig. 5). The

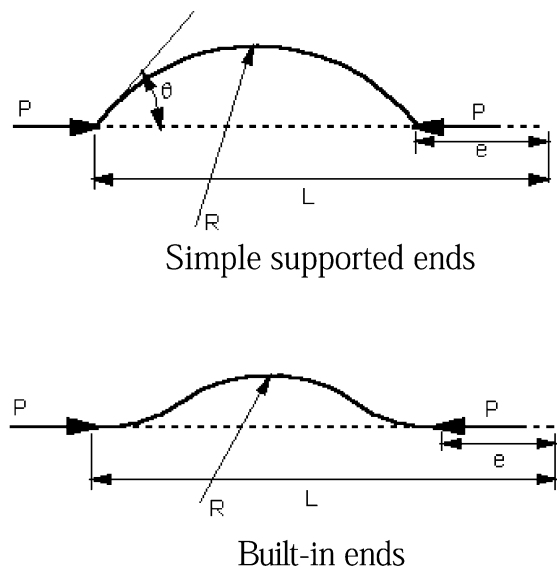


Fig. 5. Buckling test. Top: simple supports; bottom: clamped ends.

deflection of the film at the onset of cracking in the ITO is very large and the usual small deflection theory cannot be used to obtain the radius of curvature of the film. Fortunately there is an analytical large deflection solution for the radius of curvature, which can be expressed in terms of the axial displacement, e , of the ends of the buckled film [11]

$$\frac{L}{R} = NkK(k) \tag{8}$$

$$\frac{e}{L} = 2 \left[1 - \frac{E(k)}{K(k)} \right]$$

where L is the length of the film, $K(k)$ and $E(k)$ are complete elliptic integrals of the first and second kind and $N = 4$ for simple supports and $N = 8$ for clamped ends.

The analysis given was obtained for a coating on a film under uniform strain. However, it can be applied to the present ITO coating on a PET film under bending because the ratio of the thickness of the coating to the thickness of the PET film is very small ($\approx 1:1800$) so that the strain gradient in the coating is very small. Also, since the coating is so thin compared with the substrate, the neutral axis of the composite film is almost exactly at the centre of the PET film so that the strain in the ITO is given by

$$\varepsilon = \frac{h_s}{2R} \tag{9}$$

By buckling the PET film so that the ITO coating is on the convex or concave sides, the coating can be tested in tension or compression. The cracking of the ITO film is particularly easy to monitor because the film is conducting and so loss of conduction marks the onset of cracking. The tests were carried out with in an Instron 5543 testing machine, where both the displacement and resistance of the film were recorded. Two

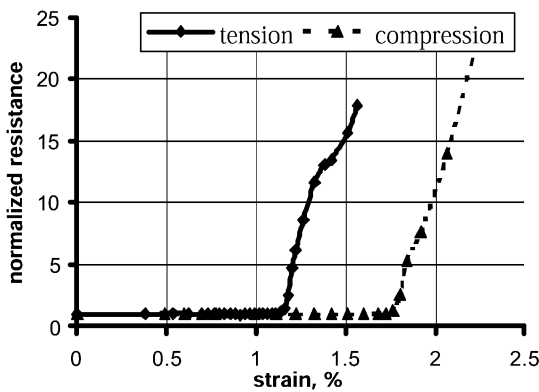


Fig. 6. The resistance as a function of strain for ITO coatings under tension and compression.

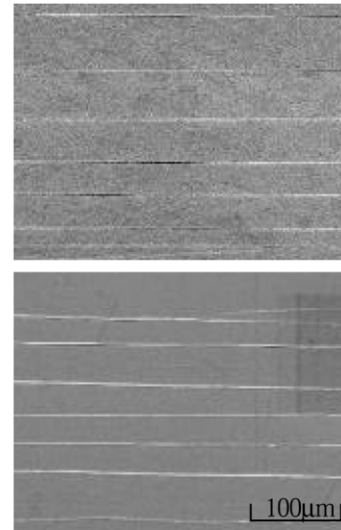


Fig. 7. SEM pictures of cracked ITO coatings Top: ITO in tension; bottom: ITO in compression.

representative normalised resistances against strain curves are shown in Fig. 6 for ITO coatings in tension and compression. The very marked increase in resistance indicates the onset of cracking both in tension and compression. Microscopic observations show that the fracture pattern of ITO under tension is parallel channelling cracks (see Fig. 7). Compressive cracks of the ITO films at low magnification are superficially similar to the tension pattern (see Fig. 7). However, under a high-resolution scanning electron microscope (see Fig. 8) it is revealed that the failure mechanism is by a tunnelling–delamination–buckle–crack as shown schematically in Fig. 3.

Using the critical strain of 1.1% in the tension tests, the crack resistance energy for the ITO coating was found to be 62 J/m² from Eq. (4). The mean critical strain for the compression tests was 1.7%. With this value and the value for the crack resistance of the coating of 62 J/m², and the value of $h/2b$ estimated to be 1/20 from the scanning electron microscope (SEM) studies, Eq. (7) has been used to calculate the delamination interfacial toughness of 35 J/m². Fortu-

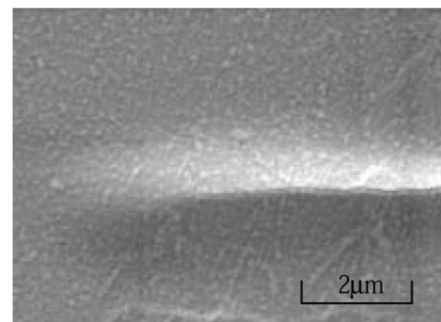


Fig. 8. The front of a channelled delaminating and cracking film under compression.

nately for this particular value of the compressive critical strain, the result is not very sensitive to the value of $h/2b$, in fact if the cracking resistance energy is neglected completely, the delamination interfacial toughness is only overestimated by 3.1 J/m^2 .

4. Discussion and conclusions

We have demonstrated the controlled buckling test and the analysis for the mechanical assessment of flexible optoelectronic devices where the traditional three-point bending or four-point bending test are not suitable. This approach is also suitable for other thin sheet devices, such as credit card, smart card, etc., where flexibility assessment or the reliability of the built-in circuits under repeated flexing is necessary.

Brittle coatings can limit the flexibility of optoelectronic devices. Where possible flexible devices should be designed so that brittle coatings are close to the neutral axis so that the strain is small. A recommended conservative procedure for device safety is to design against steady-state propagation of the coating crack rather than initiation. In which case, the critical strain of brittle coatings under tension or compression is inversely proportional to the square root of the coating thickness. As long as other requirements are met, thinner coatings should be favoured in order to achieve higher flexibility. It has to be pointed out that the proportionality factors in both tensile and compressive energy release rates [Eqs. (4) and (5)] are influenced by the mismatch index, α , especially in the region $\alpha > 0.8$. The energy release rate will decrease when α is closer to zero. However, due to limited choices of materials, it is impractical to try to maximise the flexibility through minimising the mismatch between the coating and the substrate.

The mechanism of ITO coating failure under tension

is by a channelling crack. Under compression, the coating must first delaminate and buckle before it can crack. Whether tensile or compressive strain is the more critical depends upon the ratio of the cracking resistance energy to the delamination toughness, G_c/G_d . The paper shows how the coating crack resistance energy and the delamination toughness can be calculated from tension and compression tests under controlled buckling. Once these values are known the critical cracking strains can be calculated for other coating thicknesses, if the deposition process does not change.

Acknowledgements

The authors would like to express their deep appreciation for the experimental assistance from, and helpful discussion with Dr K.Y. Zeng, Dr J. Li, Dr Y.P. Guo, Mr K.C. Toh and Dr K.T. Tsai.

References

- [1] E. Harlev, T. Gulakhmedova, I. Rubinovich, G. Aizenshtein, *Adv. Mater.* 8 (1996) 994.
- [2] U. Bach, D. Lupo, P. Comte, J.E. Moser, F. Weissörtel, J. Salbeck, H. Spreitzer, M. Grätzel, *Nature* 395 (1998) 583.
- [3] G. Gustafsson, Y. Cao, G.M. Treacy, F. Klavetter, N. Colaneri, A.J. Heeger, *Nature* 357 (1992) 477.
- [4] G. Gu, P.E. Burrows, S. Venkatesh, S.R. Forrest, M.E. Thompson, *Opt. Lett.* 22 (1997) 172.
- [5] Z. Suo, E.Y. Ma, H. Gleskova, S. Wagner, *Appl. Phys. Lett.* 74 (1999) 1177.
- [6] J.W. Hutchinson, *Mechanics of Thin Films and Multilayers*, Technical University of Denmark, Lyngby, Denmark, 1996.
- [7] J.W. Hutchinson, Z. Suo, *Adv. Appl. Mech.* 29 (1992) 63.
- [8] J.L. Beuth, Jr., *Int. J. Solids Struct.* 29 (1992) 1657.
- [9] M.D. Thouless, *J. Am. Ceram. Soc.* 76 (1993) 2936.
- [10] B. Cotterell, Z. Chen, *Int. J. Fract.* 104 (2000) 169.
- [11] S.J. Britvec, *The Stability of Elastic Systems*, Pergamon Press, New-York, NY, 1973.