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Buckling and cracking of thin films on compliant substrates under compression

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Abstract. It is shown that unless the substrate is at least as stiff as the film, the energy stored in the substrate contributes significantly to the energy release rate of film delamination under compression either with or without cracking. For very compliant substrates, such as polyethylene terephthalate (PET) with a indium tin oxide (ITO) film, the energy release rate allowing for the deformation of the substrate can be more than an order of magnitude greater than the value obtained neglecting the substrate's deformation. The argument that buckling delaminations tunnel at the tip rather than spread sideways because of increase in mode-mixity may need modification; it is still true for stiff substrates, but for compliant substrates the average energy release rate decreases with delamination width and the limitation in buckled width may be due to this stability as much as the increase in mode-mixity.

Key words: Thin film, delamination, buckling, cracking, compliant substrate.

1. Introduction

The study of delamination under compressive strain was originally motivated by the buckling of sandwich panels in aircraft structures (Gough et al., 1940) and later by the delamination of laminated composites (Chai et al., 1981). More recently the interest has been in the delamination and buckling of thin films deposited on substrates for a wide variety of reasons such as wear resistant coatings to metal-cutting tools, hard transparent coatings on optical polymers, and ceramic thermal barrier coatings (Giola and Ortiz, 1997). Compressive residual stresses are induced in many of these systems either because the deposition takes place at high temperature and there is a mis-match in the thermal expansion coefficients, or intrinsic stresses, induced in sputter-deposited thin films due to lattice mis-match. High compressive residual stresses can cause a blistering of the film in a variety of patterns. Some of the morphologies, such as the 'telephone cord' have been observed in many different film/substrate systems. An exhaustive review of such buckled delaminations has been given by Giola and Ortiz (1997). Hutchinson and Suo (1991) have given a convincing explanation of the development of the 'telephone cord' from an initially near axisymmetric blister based on the assumption of a stiff substrate.

The motivation for the present paper is the behaviour of indium-tin oxide (ITO) coatings which have application in optoelectronic and display devices. ITO is used in organic light emitting displays (OLED) as a transparent anode. There is much current interest in flexible OLED. ITO is a brittle ceramic which can crack under tensile strain and lose its functionality. Thus delamination and cracking of ITO under tension or compression is one limit to the flexibility of such devices. Polyethylene terephthalate (PET) 0.2 mm thick films with a coating of ITO approximately 100 nm thick are commercially available and the present study arose from a need to determine the conditions under which the ITO would delaminate or crack. The

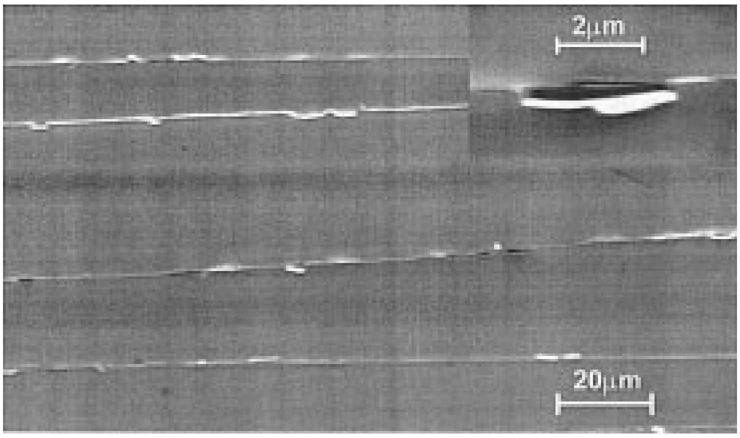

Figure 1. SEM pictures of compression cracks in ITO film on PET substrate.

ITO on these films is deposited at near room temperature, consequentially the residual stresses are low and there is no spontaneous blistering. When the ITO coating is on the tension side of the PET film, cracks channel through the coating as described by Hutchinson and Suo (1991) and Beuth (1992) and their analyses provide a good estimate of the fracture toughness of the ITO. If the PET film is bent cylindrically so that the ITO coating is in compression, cracks appear in the ITO coating, aligned in the direction of the cylindrical axis, that are superficially similar to the tension cracks (see Fig. 1). However, closer examination shows that the cracks grow behind delaminated buckles that propagate parallel to the cylindrical axis of bending (see Fig. 2). Because the stress is uniaxial, there is no telephone cord-like meandering that is observed in delaminations under biaxial residual stresses. Thouless (1993) has calculated the energy release rate for the combined buckling and cracking of a film assuming that the substrate is stiff and the buckled deformation is large compared with the thickness of the film so that the position of the line of force at the cracked section is unimportant. We initially used the analysis of Thouless (1993) to provide an estimate of the delamination toughness of the ITO coating, but the value obtained (5 J/m^2) was clearly too small. The analyses of Thouless (1993), and Hutchinson and Suo (1991), for the propagation of a delaminated buckle without cracking, are accurate if the substrate is stiffer than the film, but are inaccurate if the substrate is very much more compliant than the film. In the case of an ITO coating on PET, the substrate is about 60 times more compliant than the ITO coating. We present the analysis for steady-state delamination and buckling with and without cracking where the energy stored in the substrate is considered.

2. Buckling of films on compliant substrates without cracking

Under applied or residual compressive strain, a delaminated buckle can propagate from an edge (Thouless et al., 1992, 1994) or central flaw. Hutchinson and Suo (1991) have shown that for a stiff substrate, the average energy release rate for a straight-sided buckle increases with the width of buckled delamination. The sideways propagation is limited because the mode-II component of the energy release rate increases with the width of the buckle and becomes pure mode II when the applied strain to the critical strain ratio (ϵ/ϵ_c) is large. Under pure mode II loading compressive contact occurs between the delaminated film and the substrate causing

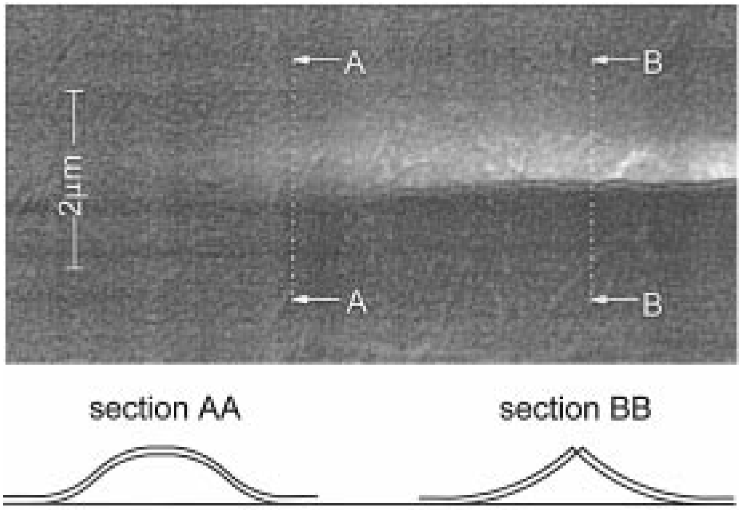

Figure 2. SEM picture showing tunnelling delamination-buckle-crack.

frictional effects to occur (Thouless et al., 1992; Stringfellow and Freund, 1993) which are not considered in this paper. Under uniaxial strain a delaminated buckle will propagate normal to the strain with stable straight sides (Jensen, 1993).

The buckled delamination is analysed as a plate under plane strain small deformation. The analysis follows that of Hutchinson and Suo (1991), but deformation in the substrate is taken into account. The substrate is assumed to be very thick compared with the film. As the delaminated film buckles, the normal axial compressive force at the edge of the delamination relaxes from the applied value (N) to the critical buckling value (N_c). If the substrate is not stiff compared with the film, the relaxation in the force in the film, under fixed strain, causes an axial displacement, u_0, at the edge of the delaminated film. At the same time a bending moment, M_0, arises to hinder the buckling and causes a rotation, ϕ, at the edge of the delaminated film. In addition the change in the axial force causes a rotation and the end moment causes an axial deformation (see Fig. 3). The compliances of the delaminated and buckled film are defined by

$$\begin{aligned} u_0 &= \frac{(N - N_c)}{\bar{E}} A_{11} + \frac{M_0}{\bar{E}h} A_{12}, \\ \phi &= \frac{(5n - N_c)}{\bar{E}h} A_{21} + \frac{M_0}{\bar{E}h^2} A_{22}, \end{aligned} \tag{1}$$

where $\bar{E}$ is the plane strain elastic modulus of the film and h is the film thickness. If the half width, b, of the delamination is large compared with the film thickness, h, then the compliance is virtually independent of b/h because the relaxation in the film stress only causes deformation in the region of the delaminated tip. The finite element method has been used to calculate the compliance assuming b/h is very large (see Appendix 1). The compliance depends upon the two Dundurs (1969) parameters, α, β, defined by

$$\begin{aligned} \alpha &= \frac{(\bar{E} - (\bar{E}_s)}{\bar{E} + \bar{E}_s}, \\ \beta &= \frac{1}{2} \frac{\mu(1 - 2v_s) - \mu_s(1 - 2v)}{\mu(1 - v_s) + \mu_s(1 - v)}, \end{aligned} \tag{2}$$

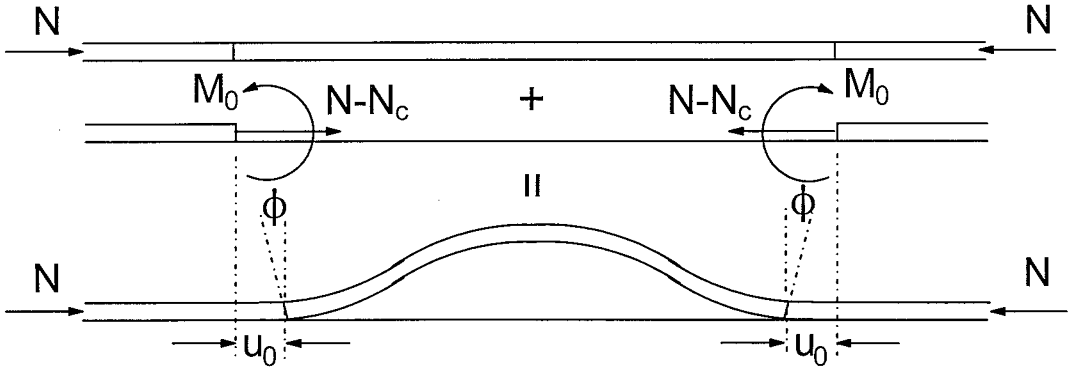

Figure 3. Schematic illustration of superposition to obtain the deflection at the tip of the delamination.

where μ and v are the shear modulus and Poisson's ratio respectively; constants without a subscript are for the film and those with a subscript s are for the substrate. The first Dundurs parameter, α, is the most important. Though Appendix 1 gives the compliances for $\beta = 0$ and $\beta = \alpha/4$ the analyses have only been made for $\beta = 0$ since the energy release rate, G, is little affected by β (Suo and Hutchinson, 1989). In this case the stress intensity factors at the tip of the delamination are real and are given, in terms of the bending moment M_0 and change in axial force $N - N_c$, by Suo and Hutchinson (1990) for an infinitely thick substrate, as

$$\begin{aligned} K_I &= \sqrt{\frac{h}{2}}\left[2\sqrt{3}\frac{M_0}{h^2}\cos\omega - \frac{(N-N_c)}{h}\sin\omega\right], \\ K_{II} &= -\sqrt{\frac{h}{2}}\left[2\sqrt{3}\frac{M_0}{h^2}\sin\omega + \frac{(N-N_c)}{h}\cos\omega\right], \end{aligned} \tag{3}$$

where ω is a function of α and β.

The buckled deflection, w, of the delamination can be written in terms of the end bending moment, M_0, as

$$\frac{w}{h} = \frac{12M_0}{\bar{E}\lambda^2}\left(\frac{b}{h}\right)\left[1 - \frac{\cos\lambda(1-x/b)}{\cos\lambda}\right], \tag{4}$$

where $\lambda = \sqrt{12\epsilon_b}b/h$ and ϵ_b is the strain in the buckled film. The bending moment can be found by equating the rotation at $x = 0$ obtained from Equation (4) to the rotation obtained from Equation (1). The eigen value of λ (for $\lambda \leq \pi$) is found by equating the change in length of the buckled delamination, due to shortening because of compressive and lateral deformation, with that obtained from Equation (1) with the constraint that $\phi \geq 0$. The compliance A_{11} has the most effect on the buckling. In the limit for a rigid substrate, and for large values of b/h for other substrates, $\lambda = \pi$. The Hutchinson and Suo (1991) result is the asymptotic value as the ratio, b/h, tends to infinity for all substrates. However, only for very weakly bonded films will the film delaminate to large widths and in practice the Hutchinson and Suo (1991) result is only accurate for substrates that are at least as stiff as the film.

For steady-state tunnelling delamination and buckling, the energy released is the difference between the energy stored well ahead of the buckled delamination and that well behind it where the width of the delamination is constant. In this steady-state calculation, which is the same as that used by Hutchinson and Suo (1991), it is not possible to describe the growth of the delamination and buckling at the tip of the tunnelling delamination. Unless the effect of mode-mixity on the delamination energy is known, the average delamination energy is a more important measure of toughness than the local value. However, it is true that the sideways spread of a buckled delamination may be restricted because the of the increase in local toughness due to the increasing local mode II component. The local energy release rate can be obtained either from the stress intensity factors given in Equation (3) or by differentiating the average energy release rate. For a rigid substrate the energy release rates are dependent only on the ratio, ϵ/ϵ_c. Thus it is convenient to use this ratio as the dependent variable. In all our figures we non-dimensionalise the applied strain by the critical strain for buckling on a rigid substrate. Thus ϵ_c is given by

$$\epsilon_c = \frac{\pi^2}{12}\left(\frac{h}{b}\right)^2. \tag{5}$$

In fact we think that it is more convenient to use $(\epsilon/\epsilon_c)^{1/2}$ rather than the ratio itself, because if the square root is used, the parameter is proportional to width of the delamination for a given film thickness and strain. Following Hutchinson and Suo (1991) we non-dimensionalise the energy release rates by, $G_{0,}$ the energy release rate if all the strain energy stored in the film ahead of the buckle is the energy released by the buckle, where

$$G_0 = \frac{\bar{E}\epsilon^2 h}{2}. \tag{6}$$

The energy release rates both average and local are shown in Fig. 4 for a range of substrate stiffnesses ($\alpha = -1$ to $\alpha = 0.99$) and applied strains ($\epsilon = 0.01\%$ to $\epsilon = 1\%$). Fig. 4d, for a rigid substrate ($\alpha = -1$), contains the same results as previously given by Hutchinson and Suo (1991). The energy release rate is always greater than that for a rigid substrate and for a very compliant substrate (for ITO on PET $\alpha = 0.97$) the energy release rate can be more than an order of magnitude larger. In addition the average, as well as the local, energy release rate shows a maximum value for large values of α. Even if the delamination energy does not depend upon mode-mixity, the delamination is unlikely to be much wider than the width determined from the value of $(\epsilon/\epsilon_c)^{1/2}$ at the maximum in the average energy release rate[1].

There are a number of limitations to the results presented in Fig. 4. The calculation of the compliances in Appendix 1 assumes that b/h is large. An estimate of the limitation in the smallest valid b/h has been obtained in Appendix 1 by calculating the position where the axial strain in the substrate decays to 5% of its maximum value. An upper limit to b/h occurs when there is compressive contact between the delaminated film and the substrate when friction shields the delaminated tip which has been discussed by Thouless et al. (1992) and Stringfellow and Freund (1993). This compressive contact can arise from two sources: the mode I component of the stress intensity factor becomes negative, or the eigen value for λ is greater than π. For delaminated buckling without cracking, the first of these always occurs at a smaller delamination width than the second and is thus the limitation on the accuracy of the

[1]It cannot be said that the delamination width *cannot* be wider than the value that gives the maximum energy release rate since the maximum width of the delamination and buckle is determined at the tip of the propagating delamination where the steady-state solution is not valid. Obviously the delamination cannot spread sideways as it becomes left behind the tip of the delamination when the width of the delamination is on the stable portion of the steady-state energy release rate curve.

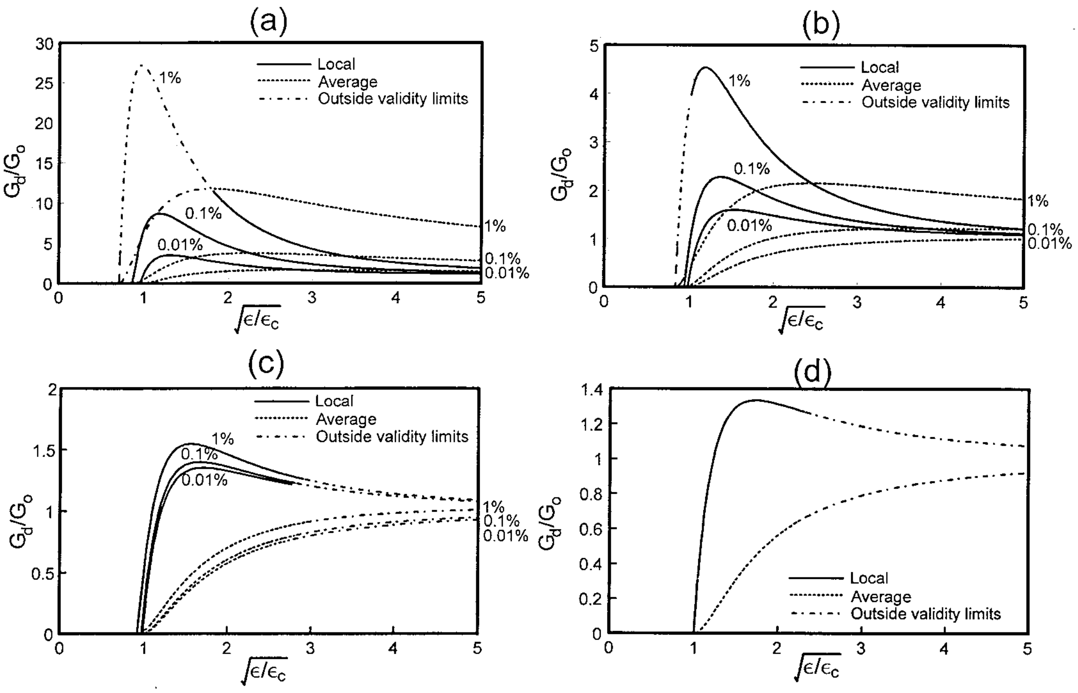

Figure 4. Average and local delamination energy release rate for delamination without cracking as a function of $(\epsilon/\epsilon_c)^2 = (b/h).(12\epsilon)^2/\pi^{1/2}$ for a range of indicated applied strains. (a) $\alpha = 0.99$, $\beta = 0$; (b) $\alpha = 0.9$, $\beta = 0$; (c) $\alpha = 0$, $\beta = 0$; (d) $\alpha = -1$, $\beta = 0$ (for all applied strains).

estimates for the delamination toughness. In our analysis the eigen value for λ is constrained to be less than or equal to π, but no account is taken of shielding of the delaminated tip by friction. The minimum and maximum limits to valid delamination toughnesses are indicated in Fig. 4.

The mode-mixity angle defined by

$$\psi = \tan^{-1}(K_{II}/K_I) \tag{7}$$

has been calculated from Equations (3) and is shown in Fig. 5. For compliant substrates the mode-mixity angle only increases relatively slowly with $(\epsilon/\epsilon_c)^{1/2}$, which is proportional to b/h, as compared with stiff substrates where the fracture mode rapidly tends to mode II. On the other hand the average energy release rate does show a maximum for compliant substrates. Hence it is suggested that the sideways spreading of the buckled delamination for compliant substrates is as much limited by the decrease in energy release rate with delamination width as it is by the increase in mode-mixity. However, if the substrate is as stiff as the film, there is no maximum in the average energy release rate and the Hutchinson and Suo (1991) explanation that it is the increase in the mode II component with increase in the delamination width that stabilises the width, is the most satisfactory.

3. Buckling of films on compliant substrates with cracking

The analysis of delamination-buckling-cracking follows closely on the work of Thouless (1993). However, since we are interested in comparatively small b/h ratios, we assumed that

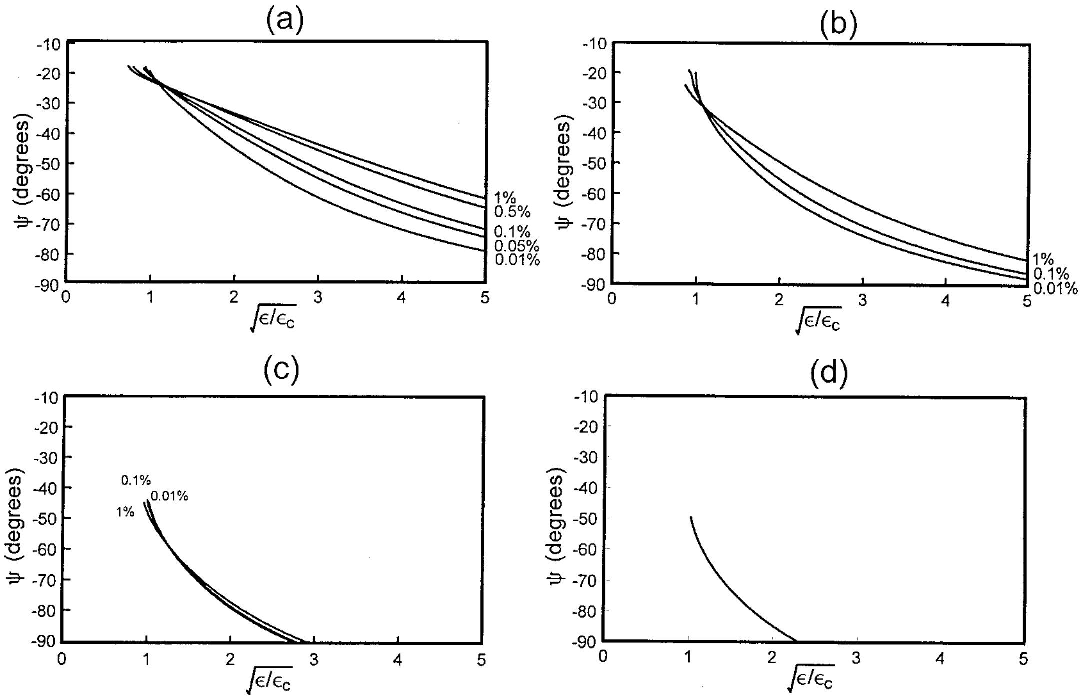

Figure 5. Mode-mixity for delamination without cracking as a function of $(\epsilon/\epsilon_c)^2 = (b/h).(12\epsilon)^2/\pi^{1/2}$ for a range of indicated applied strains. (a) $\alpha = 0.99$, $\beta = 0$; (b) $\alpha = 0.9$, $\beta = 0$; (c) $\alpha = 0$, $\beta = 0$; (d) $\alpha = -1$, $\beta = 0$ (for all applied strains).

when cracking is complete the buckling load acts through the bottom edge of the completed crack unlike Thouless (1993) who assumes the buckling force acts through the neutral axis. Naturally the two solutions are asymptotically the same for infinite delamination widths. Also, as described in Section 2, we take into account the compliance of the substrates. Under these assumptions the buckled form of the cracked delamination can be written as

$$\frac{w}{h} = \frac{\phi}{\lambda}\left(\frac{b}{h}\right)\sin\lambda x/b + \left(\frac{\delta}{h} - \frac{1}{2}\right)(1 - \cos\lambda x/b), \tag{8}$$

where δ is the deflection of the buckle at the cracked centre. The values of the edge rotation, ϕ, the deflection, δ, and the eigen value, λ, can be found from Equations (1) and from considering the change in length of the buckled delamination, due to shortening because of compression and lateral deformation. The edge rotation, ϕ, is positive for $(\epsilon/\epsilon_c)^{1/2} \leq 5$. The average energy release rate, G_t, is a combination of the energy release rates for delamination, G_d, and cracking, G_c, and is given by

$$G_t = G_d + G_c\left(\frac{h}{2b}\right). \tag{9}$$

The local energy release rate is not considered. Further delamination is possible, but unlikely, after the film cracks. The total energy release rate, G_t, non-dimensionalised by G_0 is shown in Fig. 6 for a range of substrates. The same limits are indicated as for Section 2. The averaged energy release rate shows a much more pronounced maximum for substrates that are more compliant than the film for the cracked buckled delamination as compared with the uncracked.

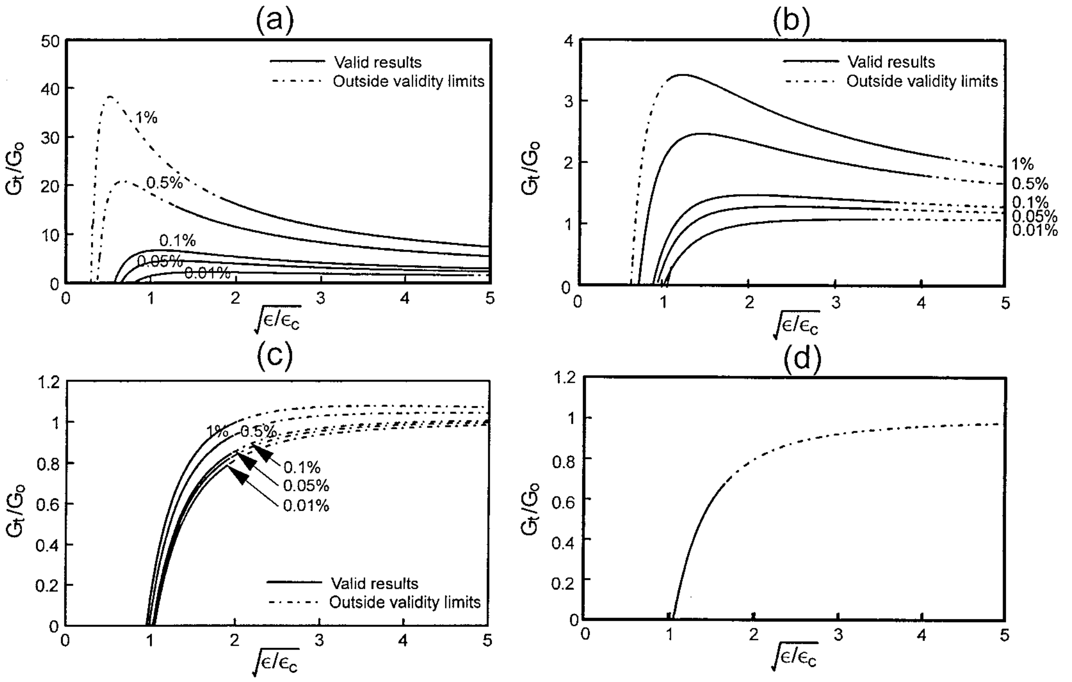

Figure 6. Average delamination energy release rate for delamination with cracking as a function of $(\epsilon/\epsilon_c)^2 = (b/h).(12\epsilon)^2/\pi^{1/2}$ for a range of indicated applied strains. (a) $\alpha = 0.99$, $\beta = 0$; (b) $\alpha = 0.9$, $\beta = 0$; (c) $\alpha = 0$, $\beta = 0$; (d) $\alpha = -1$, $\beta = 0$ (for all applied strains).

If the fracture toughness of the film is known from independent tests, then the interfacial delamination toughness can be found. Naturally the energy released by a buckled and cracked film is always greater than a film that delaminates and buckles without cracking for the same buckled width. Whether the film cracks is determined largely by the maximum tensile strain induced in the buckled film. That is it is an initiation criterion that determines whether the film cracks under compressive loading. A robust criterion for the delamination of films under compression, in the spirit advocated by Hutchinson (1996) , would be to assume all the energy goes into delamination even when the film cracks. Such an approach is not necessarily that conservative because the contribution to the total energy release rate in Equation (9) from the cracking of the film is unlikely to be large.

The mode-mixity has also been calculated and is shown in Fig. 7. The same general trend noted for the uncracked case is observed but pure mode II being established at smaller values of $(\epsilon/\epsilon_c)^{1/2}$. Because the average energy release rate shows a much more pronounced maximum and the mode tends to pure mode II at smaller values of $(\epsilon/\epsilon_c)^{1/2}$, the width of a buckled and cracked delamination is more restricted than a delamination that buckles without cracking.

4. Conclusions

When thin films delaminate and buckle from compliant substrates under compressive strain, with or without cracking, the energy released from the substrate can be very much more than the energy stored in the film itself. Thus using the above analysis, the delamination energy for

Table 1. Compliances and accuracy assessment

α	$\beta = 0$					$\beta = \alpha/4$				
	A_{11}	A_{12}	A_{21}	A_{22}	x/h	A_{11}	A_{12}	A_{21}	A_{22}	x/h
0.99	298.7	2.56	2.58	45.3	16.5	299.4	5.04	5.06	44.3	12.7
0.95	78.6	1.95	1.99	25.7	11.0	79.0	2.96	3.02	25.0	8.5
0.90	44.6	1.83	1.90	19.9	9.2	44.9	2.53	2.60	19.4	7.6
0.80	24.8	1.76	1.84	15.3	8.1	25.0	2.20	2.29	14.9	6.8
0.50	10.5	1.68	1.77	10.7	7.1	10.6	1.85	1.95	10.5	6.1
0.00	4.71	1.64	1.74	8.09	6.0	4.71	1.64	1.74	8.09	6.0
−0.50	2.48	1.62	1.73	6.87	4.7	2.40	1.54	1.64	6.92	5.9
−0.90	1.47	1.62	1.73	6.23	4.2	1.33	1.49	1.33	6.29	5.2

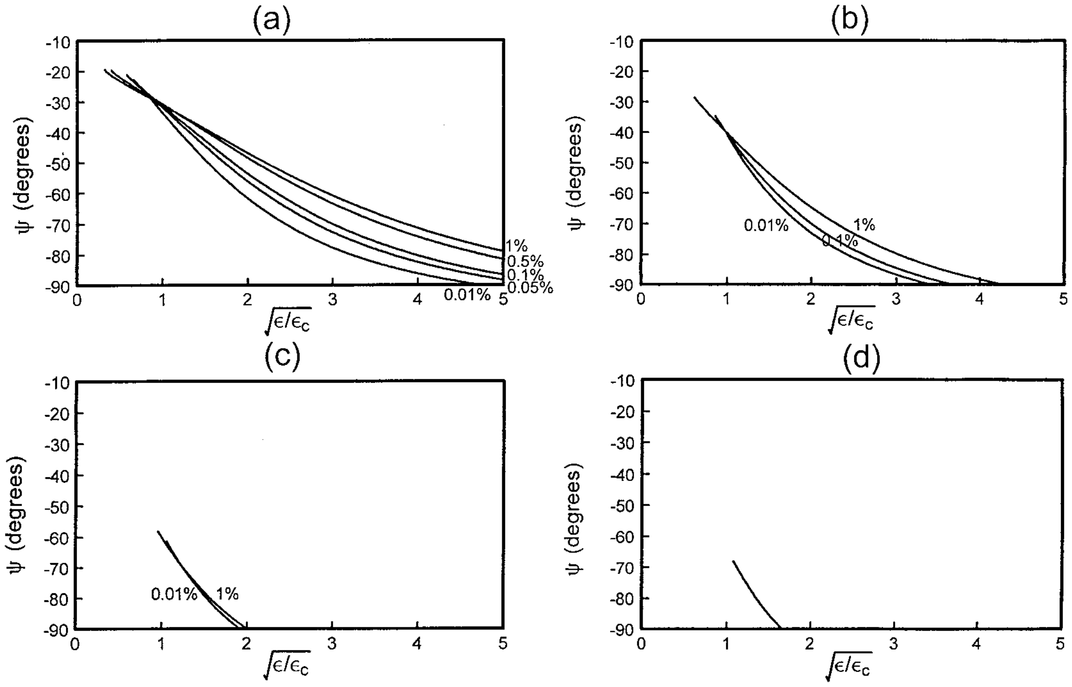

Figure 7. Mode-mixity for delamination with cracking as a function of $(\epsilon/\epsilon_c)^2 = (b/h).(12\epsilon)^2/\pi^{1/2}$ for a range of indicated applied strains. (a) $\alpha = 0.99$, $\beta = 0$; (b) $\alpha = 0.9$, $\beta = 0$; (c) $\alpha = 0$, $\beta = 0$; (d) $\alpha = -1$, $\beta = 0$ (for all applied strains).

ITO coatings on PET substrates ($\alpha = 0.97$) was estimated to be 32–36 J/m^2 as compared with 5 J/m^2 using the solution of Thouless (1993) for a rigid substrate.

The explanation that delaminating and buckling films tunnel without increasing the width of the delamination due to increased mode-mixity is probably correct for films on stiff substrates. However, the change in mode-mixity with increase in width of the delamination is much less pronounced for films on compliant substrates. On compliant substrates, the average energy release rate does exhibit a maximum with increase in width and hence in this case the width of the delamination is also limited because of the decrease in energy release rate.

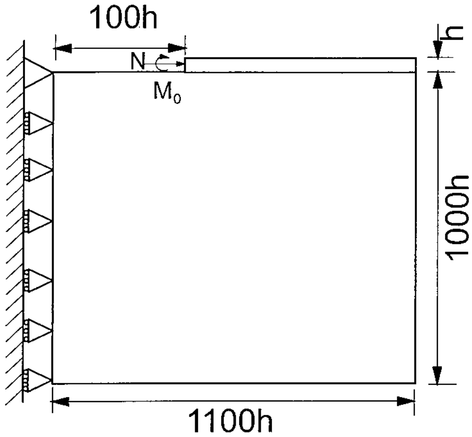

Figure 1a. The dimensions and loading of the model used to obtain the compliance.

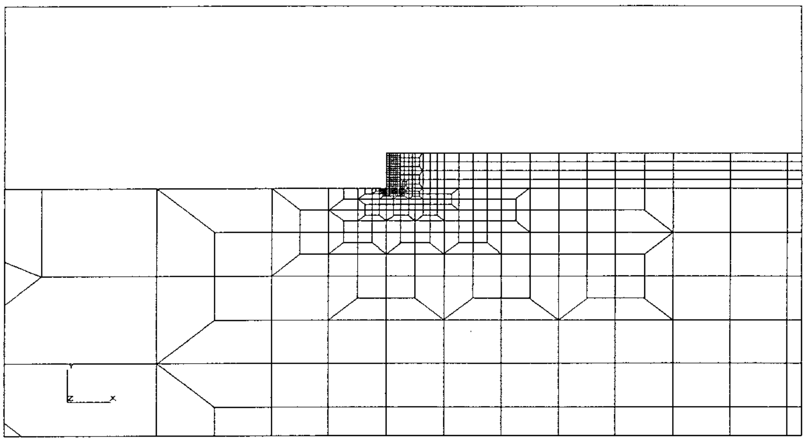

Figure 2a. The finite element mesh used in the compliance calculations.

If the film cracks as well as buckles, the maximum in the energy release rate shows a much more pronounced maximum and the mode-mixity reaches pure mode II at smaller values of b/h. Hence it is suggested that buckled and cracked films will only be observed for at moderately small values of the ratio b/h. In the experiments with ITO coatings on PET films, the value of b/h was 10–12.

Appendix 1

The compliances [A] given by Equation (1) were calculated using the finite element code ABAQUS. The problem analysed is shown in Fig. 1A. It was necessary to analyse such a thick substrate to avoid a contribution to the compliance by the deformation of the substrate as a whole especially in bending for the very compliant substrates. Fig. 2A is part of the FEM model. In all there are 1757 first-order plane strain elements in the model. There are 16 elements across the thickness of the film at the point where the loads are applied.

The axial force, N, is uniformly distributed along the edge nodes except for the two surface nodes which carry half the force of the other nodes. The moment, M, is applied by simulating a linearly distributed stress about the centre of the film. Within each element the nodal forces are determined by matching the moment and force with that due to true linear stress variation. The axial displacement, u_0, and the rotation, ϕ, are given by

$$u_0 = \frac{1}{h}\int u \, \mathrm{d}y, \qquad \phi = \frac{12}{h^3}\int uy \, \mathrm{d}y. \tag{1A}$$

The FEM results have been integrated numerically to obtain u_0 and ϕ.

Thus the compliances, as defined by Equation (1) have been calculated and are given in Table 1 as a function of the two Dundurs parameters α and β. By the reciprocal theorem $A_{12} = A_{21}$. The small discrepancy is the result of mainly of the numerical integration to obtain u_0 and ϕ. Since we assume that b/h is large we have also found the value of x/h where the axial strain is 5% of its maximum value.

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