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Wrinkling-to-delamination transition in thin polymer films on compliant substrates

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Abstract

Compressing a thin, stiff film attached to a thick, compliant substrate can lead to a number of different modes of mechanical deformation depending upon the material properties of the system. In this article we explore direct transitions from surface wrinkling to buckle delamination, and provide a theoretical framework for understanding the conditions under which such transitions take place, as well as the resulting dimensions of the wrinkling-induced delamination. A key conclusion of this work is that the width of the delamination blister formed from a wrinkled film is relatively strain-independent, suggesting that delaminations can be used in such systems to measure the adhesion energy at the film-substrate interface. In addition, we demonstrate how the length and width of delaminations can be tailored through straightforward control of the substrate and film properties in the system, illustrating how wrinkling delaminations can be used for both thin film metrology and patterning applications.

1 Introduction

In-plane compression of a thin film attached to a substrate can result in a state of mechanical instability, resulting in the film and/or substrate deforming out of the plane in order to accommodate the strain energy in the system.¹ Two classes of behavior are generally observed: buckling instabilities (wrinkling and delamination) and strain localizations (folding and creasing). Among the buckling instabilities, surface wrinkling occurs when the film remains attached to the substrate and adopts a sinusoidal wrinkling morphology. Film buckling delamination (referred to simply as “delamination” throughout this text) is observed when some portion of the film-substrate interface fails and the film detaches from the substrate, buckling upward. A schematic illustrating both surface wrinkling and delamination is shown in Fig. 1a. Although mechanical buckling instabilities such as wrinkling and delamination have traditionally been considered as “failure modes,” recent

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Conflicts of Interest

There are no conflicts to declare.

work has demonstrated that these instabilities can be harnessed to provide a facile route towards novel surface patterning,² as well as to measure the Young's modulus³ and the adhesion energy of very thin films and 1-D structures such as nanotubes⁴ and nanowires⁵—these measurements would be difficult or impossible using other techniques.

The mathematical expressions that describe wrinkling and delamination are unsurprisingly similar and have led researchers to investigate the evolution and possible co-existence of these two deformation modes.^{6,7} It has been shown that wrinkling is dictated by the thickness of the stiff film and the ratio of the plane strain moduli of the film and substrate (modulus mismatch).^{1,8} Delamination must also factor in the energy of adhesion between the film and substrate in addition to the aforementioned parameters that govern wrinkling.^{9–11} Thus, the transition between these two states can be described in terms of film geometry, modulus mismatch, and adhesion properties.¹² Several studies have demonstrated, for example, that a polystyrene (PS) film mounted on a poly(dimethylsiloxane) (PDMS) substrate will transition from a wrinkled to a delaminated state when strains are high enough to induce delamination at the film-substrate interface.^{13,14}

The purpose of the current paper is to explain the fundamental system parameters that govern the transition from wrinkling to delamination in thin films on compliant substrates. We demonstrate how the presence of wrinkles in a delaminating film can lead to unique scaling relationships that differ from previous treatments of film delamination from both rigid¹⁵ and compliant¹⁰ substrates. We utilize these relationships to obtain a measurement of the adhesion energy of a thin polymer film on a soft elastic foundation, and demonstrate how wrinkling-induced delaminations can be controlled to pattern films. These results are of both theoretical and practical interest, especially given the growing interest in bonding thin films of stiff materials to compliant substrates for applications such as stretchable electronics^{16,17} and organic photovoltaics.¹⁸

2 Discussion

2.1 Wrinkling vs. delamination

For a thin film on a relatively rigid substrate, in-plane compression will result in delamination of the film when the applied strain, ϵ , reaches a critical value:^{13,19–23}

$$\epsilon_{c,d} = \frac{\pi^2}{12} \left(\frac{2h}{L} \right)^2 \quad (1)$$

where h is the film thickness and L is a pre-determined delamination length along which the film is not adhered to the substrate. Yu and Hutchinson¹¹ have shown that the critical strain for delamination of a stiff film from a compliant substrate can be significantly lower than that predicted by eqn (1). Knowing that, Mei et al. presented a “buckling mode selection map” by plotting eqn (1) along with the expression giving the critical strain to the onset of surface wrinkling:^{13,24}

$$\varepsilon_{c,w} = \frac{1}{4} \left(\frac{3E_s}{E_f} \right)^{\frac{2}{3}} \quad (2)$$

where E_s and E_f are the plane-strain Young's moduli of the substrate and film, respectively.[†] Their map predicted whether a given film would wrinkle or delaminate upon compression, but it required one to know the value of L , which for most systems is practically impossible to estimate *a priori*. Indeed, in most systems delamination manifests as a spontaneous event at locations where there is presumably a small region of weak adhesion at the film-substrate interface. For such systems, once a delamination forms its width, L , will continue to grow until reaching an equilibrium value. Bazhenov et al., for example, showed that after formation of a buckle delamination, L will continue to increase as long as the energy penalty associated with debonding the film is outweighed by the energy gain from strain relaxations in the film in its new buckled morphology.¹⁵ Another key conclusion of these authors' work was that delaminations can only form when the debonding energy is less than a certain strain parameter involving the modulus and thickness of the film:^{12,25}

$$G < \varepsilon^2 E_f h \quad (3)$$

where G is known alternatively as the fracture toughness, debonding energy, energy release rate, or adhesion energy. Eqn (2) and (3) can be equated in strain to yield a function that defines a phase map for when wrinkling or delamination would be expected by a given system (Fig. 1b).

Fig. 1b illustrates that high debonding energies, low film moduli, and small film thicknesses all lead to higher values for the dimensionless parameter, $G/(E_f h)$, and favor wrinkling, as do low values of the substrate to film modulus ratio, E_s/E_f . In other words, wrinkling is the preferred buckling instability mode when a film that can be relatively easily bent is well-adhered to a substrate that has a significantly lower modulus. Alternatively, delamination will be preferred when the substrate modulus is closer to the film modulus, or if the work per area required to bend the film ($\sim E_f h$) becomes greater than the energy released during delamination (G). The recent work of Cordill et al.,⁹ Mei et al.,¹³ and Vella et al.¹⁰ provide validation tests for these predictions. Cordill et al. observed buckle delamination for a tungsten film deposited on SiO₂ ($E_f \approx 400$ GPa, $E_s \approx 40$ GPa, $G \approx 2$ J/m²), while Mei et al. (for small strains) observed wrinkling in PS on PDMS ($E_f \approx 4$ GPa, $E_s \approx 2$ MPa, $G \approx 50$ mJ/m²). For each of these systems, $h \approx 100$ nm. Vella et al. utilized a polypropylene (PP) on a polyvinylsiloxane (PVS) ($E_f \approx 2$ GPa, $E_s \approx 1$ MPa, $G \approx 50$ mJ/m²), for which buckle delamination was observed. This latter system has material properties similar to the case of PS on PDMS, but the researchers utilized films with $h \approx 90$ μ m. As can be seen in Fig. 1b,

[†]Unless otherwise specified, E in this paper denotes a plane-strain Young's modulus, which is equal to the Young's modulus divided by the quantity $(1 - \nu^2)$, where ν is the Poisson's ratio and is approximately 0.33 for PS and 0.5 for PDMS.

the locations of the above systems on the instability map are consistent with the observations of either wrinkling or delamination in each case.

While Fig. 1b indicates the energetically preferred instability mode for a given system, it should be noted that a key condition of small compressive strain near the onset of buckling is assumed here. Because delaminations take a finite amount of time to form and propagate, wrinkling may indeed be seen briefly at high strain rates in certain systems that should only display delamination.[‡] Also, Fig. 1b only indicates the mode a system would be expected to enter first—at higher strains, as suggested by the inequality in eqn (3), wrinkled systems may begin to delaminate, as has been observed in certain cases.¹³ Because the wrinkling-to-delamination transition results in significant strain relaxation in both the film and the substrate, the formation of “wrinkling delaminations” cannot be described by the conventional theories that assume delamination from a rigid, flat substrate (Fig. 1a).¹⁵ Even recent treatments of delamination from compliant substrates¹⁰ are incapable of fully describing the wrinkle-to-delamination transition, since it occurs upon a substrate that is sinusoidally deformed and consequently capable of storing elastic deformation energy both within and out of the plane of the film. In the following section, we develop a theory for delamination from a pre-wrinkled film that accounts for these unique system attributes.

2.2 Delamination from a wrinkled film

When a film-substrate system is strained uniaxially (see Fig. 2a), it undergoes wrinkling with a sinusoidal morphology represented by:²⁶

$$y = A_w \sin \left(\frac{2\pi}{\lambda} x \right) \quad (4)$$

where y is oriented normal to the plane of the film, and x is oriented along the direction of strain (perpendicular to the wrinkle crests). The amplitude of the wrinkling instability, A_w , is strain-dependent, increasing in proportion to the film thickness as:^{27–29}

$$A_w = h \sqrt{\frac{\varepsilon - \varepsilon_{c,w}}{\varepsilon_{c,w}}} = h \sqrt{\varepsilon_0} \quad (5)$$

where ε_0 is known as the overstrain.³⁰ The wavelength of wrinkling, λ , is given by:^{26,31,32}

$$\lambda = 2\pi h \left(\frac{E_f}{3E_s} \right)^{\frac{1}{3}} \quad (6)$$

[‡]Wrinkling may also be sustained in any localized areas of the film surface where the adhesion energy is higher, or where buckle delamination events have relaxed the local strain below the nominal value applied to the substrate.

Before delamination occurs, the wrinkled system stores energy both in the bending of the film ($U_{b,w}$) and in the sinusoidal deformation of the substrate above and below its neutral plane (U_s). These energies (per unit width of film, w) are given by:^{15,24,26}

$$U_{b,w} = \frac{E_f I}{2} \int_0^\lambda \left(\frac{\partial^2 y}{\partial x^2} \right)^2 dx = \frac{4\pi^4 E_f I A_w^2}{\lambda^3} = \frac{\pi^4 E_f A^2 h^3}{3\lambda^3} \\ = \frac{\pi E_s h^2 \varepsilon_0}{8} \quad (7)$$

$$U_s = \int_0^\lambda \int_0^{A_w \sin(\frac{2\pi}{\lambda} x)} k y \partial y \partial x = \frac{k A_w^2 \lambda}{4} = \frac{\lambda E_s h^2 \varepsilon_0}{4} \quad (8)$$

where $I = h^3 w/12$ is the second moment of area for the film, $k = \pi E_s w/\lambda$ is the Winkler modulus of the substrate²⁶, and w represents the width of the film in the direction into the page (Fig. 2a). The energy expressions in eqns (7) and (8) have been calculated over one wavelength of film length.

The formation of a buckle delamination of half-height, A_b , and width, L , is associated with its own bending energy ($U_{b,d}$), as well as with the energy required to form new surfaces (U_{surf}) as the film separates from the substrate:

$$U_{b,d} = \frac{\pi^4 E_f h^3 A_b^2}{3L^3} \quad (9)$$

$$U_{surf} = GL \quad (10)$$

As Vella et al. have indicated, delamination from compliant substrates also results in a concentration of strain energy in the substrate under the free surface of the delamination, leading to an additional energy term (U_{sub}):¹⁰

$$U_{sub} \approx \frac{1}{2} E_s \varepsilon_b^2 L^2 \quad (11)$$

where ε_b is the strain accommodated by the delamination.

As can be seen from Fig. 2b, it cannot be assumed that all strain in the substrate is accommodated by the delamination event. Certainly, there is significant relaxation immediately surrounding the delamination, and this behaviour has been observed in 2D finite element models.²⁵ The presence of the seemingly unperturbed wrinkling pattern

beginning at a distance of $\approx R/2$ on either side of the delamination illustrates that the strain outside this region eventually returns to the nominal applied value (ε) of the system (from a value of $\varepsilon = 0$ at the delamination edge), similar to the way in which strain is accommodated around cracks in a film-substrate system held in tension.¹⁸ Thus, the total energy associated with the delamination process ($\bar{U}$) can be written as the sum of the energies required for creation of the buckle *minus* the energies for relaxation of wrinkles within the region of R :

$$\bar{U} = \bar{U}_{b,d} + \bar{U}_{surf} + \bar{U}_{sub} - \frac{R}{\lambda} \cdot (\bar{U}_{b,w} + \bar{U}_s) \quad (12)$$

where R/λ is the number of wrinkles that relax upon the delamination event and is a characteristic ratio of two lateral length scales. Because λ can be directly measured through experiments, it is a convenient descriptive length scale.

To simplify these equations further, expressions are needed for A_b and ε_b since these quantities are not measured directly. If we assume that the contour length of the film does not change during the transition from a wrinkled to a delaminated state (as illustrated in Fig. 2a), one can derive the following relationship for the contour length of the film within the region R :

$$R \left(1 + \frac{\pi^2 A_w^2}{\lambda^2} \right) = (R - L) + L \left(1 + \frac{\pi^2 A_b^2}{L^2} \right) \quad (13)$$

where the left side of the equation describes the contour length of a wrinkled film and the right side of the equation describes the contour length of a delaminated film. Rearranging eqn (13) and inserting A_w from eqn (5) yields:

$$A_b^2 = \frac{LR A_w^2}{\lambda^2} = \frac{LR h^2}{\lambda^2} \varepsilon_0 \quad (14)$$

Moreover, we can approximate the strain accommodated by the buckle within the delaminated region as:¹⁰

$$\varepsilon_b = \frac{\Delta d}{L} \quad (15)$$

where Δd is the end-to-end displacement of the film within the delaminated region. One can estimate Δd by calculating the arc length of a sinusoid, which in the limit of small A_b is:

$$\Delta d = \frac{\pi^2 A_b^2}{L} \quad (16)$$

Combining eqns (14), (15), and (16) yields an expression for the strain accommodated by a buckle:

$$\varepsilon_b = \frac{\Delta d}{L} = \frac{\pi^2 A_b^2}{L^2} = \left(\frac{R}{L}\right) \frac{\pi^2 h^2}{\lambda^2} \varepsilon_0 \quad (17)$$

Substituting eqns (14) and (17) into the energy expressions and minimizing eqn (12) with respect to R :

$$\frac{\partial \bar{U}}{\partial R} = \frac{\pi E_s h^2 \lambda \varepsilon_0}{8 L^2} + E_s \varepsilon_0^2 \varepsilon_{c,w}^2 R - \frac{3 \pi E_s h^2 \varepsilon_0}{8 \lambda} = 0 \quad (18)$$

$$R = \frac{3 \pi h^2}{8 \lambda \varepsilon_{c,w}^2 \varepsilon_0} \left(1 - \frac{1}{3} \left(\frac{\lambda}{L}\right)^2\right) \cong \frac{3 \pi h^2}{8 \lambda \varepsilon_{c,w}^2 \varepsilon_0} = \frac{h}{\varepsilon_0} \left(\frac{E_f}{E_s}\right) \quad (19)$$

and minimizing eqn (12) with respect to L yields:

$$\frac{\partial \bar{U}}{\partial L} = G - \frac{2 \pi E_s h^2 \lambda R \varepsilon_0}{8 L^3} = 0 \quad (20)$$

$$L^3 = \frac{\pi E_s h^2 \lambda \varepsilon_0}{4 G} R = \frac{\pi^2 E_f h^4}{2 G} \left(\frac{E_f}{3 E_s}\right)^{\frac{1}{3}} \quad (21)$$

3 Experimental results[§]

3.1 Strain dependence of L and R

We first tested the derived strain dependence of L and R in eqns (21) and (19) using polystyrene (PS) films on PDMS, prepared using a previously described procedure.³ A PS film with thickness $h = 98 \text{ nm} \pm 5 \text{ nm}$ was transferred to a semi-infinite slab of PDMS ($E_s \approx 2 \text{ MPa}$, $h_s \approx 1.1 \text{ mm}$) pre-strained from 50 mm to 61.0 mm in length. Next, the strain was relaxed in 0.5 mm steps, corresponding to strain increments of 0.82 %, to induce wrinkling and eventually delamination. Optical microscopy images were taken at each step in strain, and average values of L and R were obtained from the images. The values of L were measured here by taking the optically-determined width of the buckled region, as shown in Fig 2b. We note that while it is difficult to determine the exact point of the buckled film detachment from the substrate (crack tip) with this method, the fairly uniform contrast/

[§]Equipment and instruments or materials are identified in this work in order to adequately specify the experimental details. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials are necessarily the best available for the purpose.

intensity across the entire width of the buckle delamination suggests that the film is not in contact with the substrate between the dark borders of the buckle delamination. This suggests that our optical measurement of L is an accurate measure of the delamination width. The results are shown in Fig. 3 and confirm the strain-dependence predictions for L and R given by eqns (21) and (19). While L remains essentially constant, R decreases in inverse proportion to the applied strain. It should be noted that as compressive strains beyond the critical delamination strain were applied, L decreased slightly as can be seen in Fig. 3. However, this effect was quite small relative to the observed change in R .

Because L is essentially strain-independent for small deformations, the system must instead accommodate strain by reducing the delamination relaxation length, R . The result that L is strain-independent for wrinkling delamination is somewhat non-intuitive, since previous results examining buckle delamination from rigid¹¹ and compliant⁸ substrates have predicted that $L \sim \epsilon^{1/2}$ and $L \sim \epsilon^{1/3}$, respectively. However, in the system described here, wrinkling and buckle delamination co-exist and interact (i.e. the compressive strain need not be relaxed by growing the width of the delaminated region (L), but rather by reducing the relaxation region (R) along with an increase in the amplitude of the wrinkles). It is also interesting to note here that L and λ are not directly proportional but their ratio is weakly dependent on h , G , and E_f as shown in Table 1. As we will discuss shortly, the strain independence of L makes wrinkle delamination a particularly attractive way of measuring the adhesion energy (G) for certain polymeric systems.

3.2 Scaling relations for L and R

We next tested the derived scaling relationships for L and R in eqns (21) and (19) using PS films on PDMS. A series of five PS films ranging in thickness from ≈ 100 nm to ≈ 500 nm was prepared, and each film was diced into two pieces. One film of each thickness was then transferred to both a high ($E_s \approx 2$ MPa, $h_s \approx 1.1$ mm) and a low ($E_s \approx 0.5$ MPa, $h_s \approx 1.5$ mm) modulus PDMS substrate (i.e. a single PDMS substrate had five PS films of different thickness on it). Prior to film transfer, the PDMS substrates were pre-strained from 72 mm to 77 mm in length and held fixed during the film transfer. The strain in the PDMS was then released in 0.2 mm steps, corresponding to strain increments of 0.26 %, and the PS films were imaged using an optical microscope after each step in strain. Measurements of λ , L , and R were obtained via direct measurement in an image analysis program for each film at the lowest possible strain, and the film modulus (E_f) calculated according to eqn (6).³ The data are presented in Table 1. The applied strain values were subsequently adjusted according to linear composite plate theory in order to determine the actual strain experienced for each film based on the film-to-substrate ratios of thickness and Young's modulus.

The data in Table 1 can be used to test the predictions for L and R that were developed above. Graphing the right-hand side of eqn (21) versus L yields the plot shown in Fig. 4a, which illustrates that the data are well-described by a power law relationship with an exponent of $1/3$, consistent with the L^3 scaling predicted by eqn (21). On the other hand, a plot of the right-hand side of eqn (19) versus R should yield a slope of 1. The data from Fig 3 (R vs. ϵ) are included in Fig 4b to extend the range of data included in the fit. A cursory assessment of Fig 4b would indicate a slope of less than unity, if fitting all data sets together.

Upon closer examination, however, one can discern that the open circle data appears to have a systematic error in one of the parameters resulting in an offset that lowers the slope when all data are fitted together. If we fit the two data sets represented by closed symbols, we obtain a slope of 0.91, while the slope of the data represented by open symbols fitted alone exhibits a slope of 0.95. One possible source of error for the data represented by the open symbols may be in the measurement of the substrate modulus (E_s) for that data set. This would also result in a higher deduced film modulus (E_f) via eq 6, and a cursory examination of E_f for samples 6–10 in Table 1 indeed suggest this may be the case.

The scaling relationships for wrinkle delamination given in eqn (19) and (21), and illustrated in Fig. 4, differ from those given for conventional unwrinkled systems. For example, for a given film-substrate system (both E_s and E_f constant), $L \sim h^{4/3}$ for wrinkling delamination, whereas for buckle delamination from flat foundations $L \sim h^{3/2}$ when the substrate is rigid¹¹ and $L \sim h^{6/5}$ when it is compliant.⁸ We note that the film thickness scaling exponent of 4/3 is intermediate to the scaling exponents for rigid and compliant substrate buckle delamination. In some sense, this is due to the fact that wrinkling delamination encompasses elements of both theories. Similar to the theory for compliant substrates, wrinkling delamination incorporates the local deformation of the substrate beneath each buckle into the energy calculations. These substrate deformation events are isolated, however, for reasonable strains due in part to energy accommodation via the relaxation of the wrinkled film and substrate in the vicinity of the buckle. This energetic isolation is reminiscent of buckle delamination from relatively rigid substrates where the lack of substrate deformation leads to mechanical isolation of the individual buckle events.

3.3 Determination of G

The ability of buckle delaminations in wrinkled systems to accommodate additional strain through shrinking of the relaxation region surrounding each buckle [eqn (19)] enables the system to establish buckles with widths (L) that are essentially strain-independent [eqn (21) and Fig 3]. Because the exact strain on a system can be challenging to measure directly, wrinkle delamination can function as a relatively straightforward method for determining the interfacial toughness (G) of thin polymer films on compliant substrates. One approach would be to rearrange eqn (21) in terms of G :

$$G = \frac{\pi^2 E_f h^4}{2L^3} \left(\frac{E_f}{3E_s} \right)^{\frac{1}{3}} \quad (22)$$

A more statistically robust approach is to take advantage of the large data set in Fig 4a, where the slope of the power law fit to the data is:

$$\text{slope} = \left(\frac{\pi^2}{\sqrt[3]{3} \cdot 2 \cdot G} \right)^{\frac{1}{3}} \quad (23)$$

Using eqn (23) and the slope in Fig 4a, we estimate G between PS and PDMS to be 52.2 mJ/m². This value for G is in excellent agreement with previous measurements of the work of adhesion between PS and PDMS ($G \approx 49$ mJ/m² to 55 mJ/m²).^{34–36} The agreement between our measurement of G and literature values demonstrates that wrinkle delamination adds distinctive measurement capability to the surface wrinkling platform.

3.4 Controlling wrinkle delamination via control of G

In addition to providing a method for measuring the adhesion energy in thin films systems, the scaling relationships for L and R provide insight into how the film and surface properties can be judiciously tailored to yield patterned delaminations that could be useful in a number of scientific contexts, such as the fabrication of microfluidic channels.²¹ For example, by masking a portion of the PDMS substrate and exposing it to a UV-ozone (UVO) treatment, we selectively increase the surface energy and hence the adhesion energy (G) upon film transfer on the exposed portions of the substrate. Because larger G values require higher strains for transition from wrinkling to delamination, we can control the spatial placement and length of wrinkle delaminations by arresting the delamination process at the boundary between regions of lower and higher adhesion, as illustrated in Fig. 5.

Conclusions

We have developed a theory for treating wrinkle delamination of thin, stiff films on compliant substrates under compressive strains. Such systems display surface wrinkling, buckle delamination, or wrinkling followed by delamination at higher strains (wrinkle delamination). We provide a theoretical framework for understanding transitions from wrinkling to delamination, and we derive predictive equations for the size of wrinkle delamination blisters (L) and the length scale of strain relaxation (R) accommodated by each delamination event. Our predictions for whether a system will initially display surface wrinkling or buckle delamination agree well with the reported results of other researchers, and we have found excellent agreement between our theoretical predictions and experiments using PS films on PDMS substrates.

A key conclusion of our work is that L is relatively strain-invariant at small strains in wrinkle delamination systems. This result, which is due to the system's ability to accommodate strain via the relaxation of neighbouring wrinkles, provides an extremely straightforward approach for measuring the adhesion energy (G) associated with debonding thin polymer films of known thickness from compliant substrates. One need only make direct optical microscope observations of L and λ —more difficult measurements of the delamination amplitude and system strain are unnecessary.

This work forms a bridge between the numerous previous studies treating buckle delamination from both rigid and compliant substrates, and those which have investigated wrinkling instabilities in which the film remained bonded to the compliant substrate. We have demonstrated that wrinkle delamination exhibits a number of unique physical characteristics that allow it to function as a fast and simple metrology tool. In addition, fairly straightforward steps to control the film and surface properties can yield a high degree of patterning control over the resulting delaminations. Such techniques to measure and pattern

coatings will become increasingly important as stretchable electronics and other such stiff-compliant composites see growing use in advanced applications.

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Notes and references

1. Bowden N, Brittain S, Evans AG, Hutchinson JW, Whitesides GM. *Nature*. 1998; 393:146–149.
2. Chung JY, Nolte AJ, Stafford CM. *Adv Mater*. 2009; 21:1358–1362.
3. Stafford CM, Harrison C, Beers KL, Karim A, Amis EJ, VanLandingham MR, Kim HCC, Volksen W, Miller RD, Simonyi EE. *Nat Mater*. 2004; 3:545–550. [PubMed: 15247909]
4. Xiao J, Jiang H, Khang DY, Wu J, Huang Y, Rogers JA. *J Appl Phys*. 2008; 104:33543–7.
5. Gunawidjaja R, Ko H, Jiang C, Tsukruk VV. *Chem Mater*. 2007; 19:2007–2015.
6. Wang Q, Zhao X. *MRS Bull*. 2016; 41:115–122.
7. Wang Q, Zhao X. *J Appl Mech*. 2014; 81:0510041–5100410. [PubMed: 25278631]
8. Chung JY, Nolte AJ, Stafford CM. *Adv Mater*. 2011; 23:349–368. [PubMed: 20814918]
9. Cordill MJ, Bahr DF, Moody NR, Gerberich WW. *Mater Sci Eng A*. 2007; 443:150–155.
10. Vella D, Bico J, Boudaoud A, Roman B, Reis PM. *Proc Natl Acad Sci U S A*. 2009; 106:10901–6. [PubMed: 19556551]
11. Yu HH, Hutchinson JW. *Int J Fract*. 2002; 113:39–55.
12. Tarasovs S, Andersons J. *Microelectron Reliab*. 2012; 52:296–299.
13. Mei H, Huang R, Chung JY, Stafford CM, Yu HH. *Appl Phys Lett*. 2007; 90:151902–4.
14. Ebata Y, Croll AB, Crosby AJ. *Soft Matter*. 2012; 8:9086–9091.
15. Bazhenov SL, Myagkov VG, Zhigalov VS, Volynskii aL. *Dokl Phys*. 2002; 47:128–131.
16. Kim DH, Ahn JH, Choi WM, Kim HS, Kim TH, Song J, Huang YY, Liu Z, Lu C, Rogers JA. *Science* (80-). 2008; 320:507–511.
17. Ahn BY, Duoss EB, Motala MJ, Guo X, Park SI, Xiong Y, Yoon J, Nuzzo RG, Rogers JA, Lewis JA. *Science* (80-). 2009; 323:1590–1593.
18. Lewis J. *Mater Today*. 2006; 9:38–45.
19. Hutchinson JW, Suo Z. *Advances in Applied Mechanics*. 1991; 29:63–191.
20. Chai H, Babcock CD, Knauss WG. *Int J Solids Struct*. 1981; 17:1069–1083.
21. Chen S, Gao H. *Int J Mater Res*. 2006; 97:584–593.
22. Chen S, Gao H. *Proc R Soc A Math Phys Eng Sci*. 2006; 462:211–228.
23. Waters JF, Kalow J, Gao H, Guduru PR. *J Adhes*. 2012; 88:134–144.
24. Genzer J, Groenewold J. *Soft Matter*. 2006; 2:310–323.
25. Mei H, Landis CM, Huang R. *Mech Mater*. 2011; 43:627–642.
26. Volynskii AL, Bazhenov SL, Lebedeva OV, Bakeev NF. *J Mater Sci*. 2000; 35:547–554.
27. Huang R, Stafford CM, Vogt BD. *J Aerosp Eng*. 2007; 20:38–44.
28. Chung JY, Youngblood JP, Stafford CM. *Soft Matter*. 2007; 3:1163–1169.
29. Chen D, Yoon J, Chandra D, Crosby AJ, Hayward RC. *J Polym Sci Part B Polym Phys*. 2014; 52:1441–1461.
30. Cai S, Breid D, Crosby AJ, Suo Z, Hutchinson JW. *J Mech Phys Solids*. 2011; 59:1094–1114.
31. Allen, HG. *Analysis and Design of Structural Sandwich Panels*. Pergamon press; Oxford: 1969. p. 156–189.
32. Stafford CM, Harrison C, Beers KL, Karim A, Amis EJ, VanLandingham MR, Kim HC, Volksen W, Miller RD, Simonyi EE. *Nat Mater*. 2004; 3:545–550. [PubMed: 15247909]

33. Chung JY, Chastek TQ, Fasolka MJ, Ro HW, Stafford CM. ACS Nano. 2009; 3:844–852. [PubMed: 19298053]
34. Crosby AJ, Karim A, Amis EJ. Polymer (Guildf). 2001; 42:645–646.
35. Mangipudi VS, Huang E, Tirrell M, Pocius AV. Macromol Symp. 1996; 102:131–143.
36. Yurdumakan B, Nanjundiah K, Dhinojwala A. J Phys Chem C. 2007; 111:960–965.
37. Rogers JA, Huang Y. Proc Natl Acad Sci. 2009; 106:10875–10876. [PubMed: 19567838]

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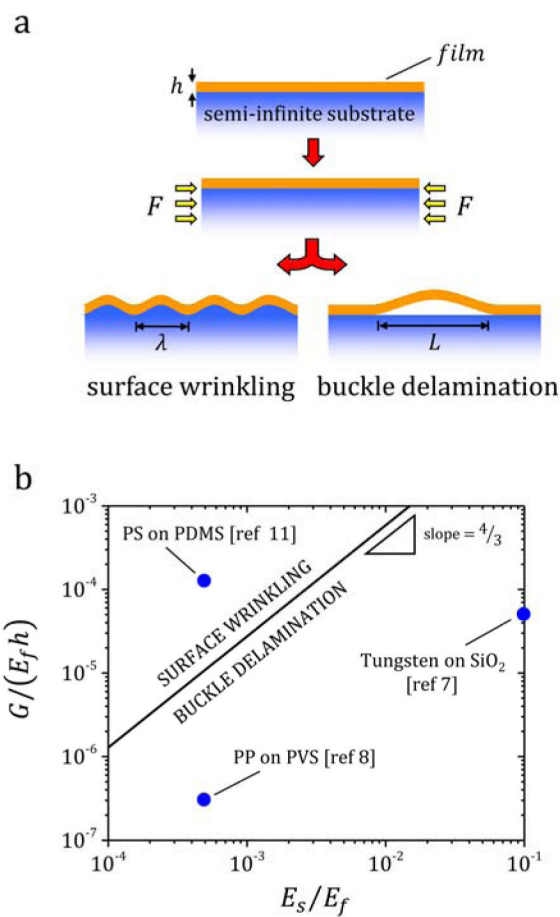


Fig. 1.

(a) A thin film on a compliant substrate undergoes either surface wrinkling or buckle delamination in response to in-plane compression. (b) An instability “map” that predicts the preferred mode for a given system. Upon in-plane compression, the film will prefer to either wrinkle or delaminate from the substrate per the system parameters. The circles indicate the approximate locations for three specific systems obtained from the references as indicated.

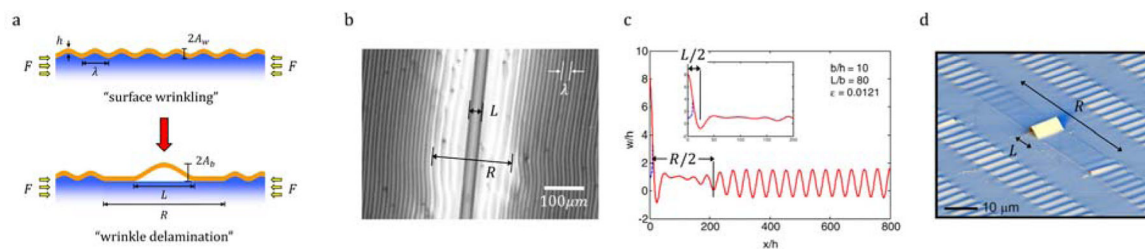


Fig. 2.

(a) Delamination in a wrinkled film-substrate system. The formation of a delaminated region of width, L , results in the relaxation of compressive strain in the film over a region with characteristic width, R , which in turn causes a significant decrease in wrinkle amplitude within the relaxation region. (b) An optical micrograph showing a wrinkled PS film ($E_f \approx 4$ GPa, $h \approx 300$ nm) on PDMS ($E_s \approx 0.5$ MPa) following delamination. The strain in the film at delamination was $\approx 2\%$. (c) Modeled relationship demonstrating the profiles of the delamination, relaxation zone, and wrinkled regions.²⁵ (d) Scanning electron micrograph showing oblique angle of delamination and relaxation zone.³⁷ (c–d) Used with permission from Mechanics of Materials and Proceedings of the National Academy of Sciences.

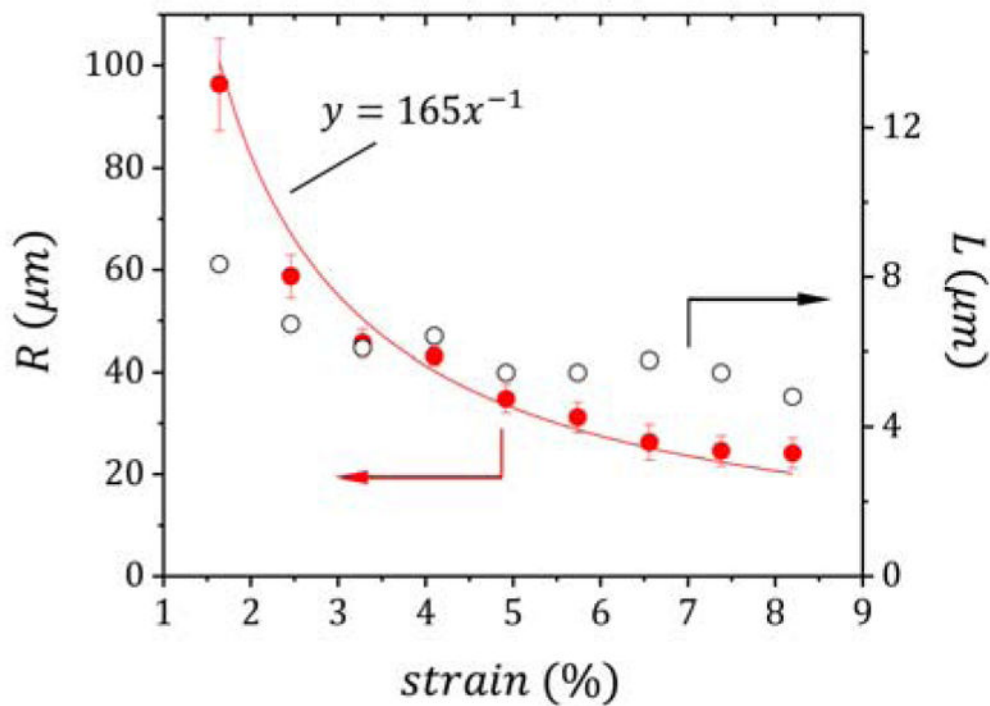
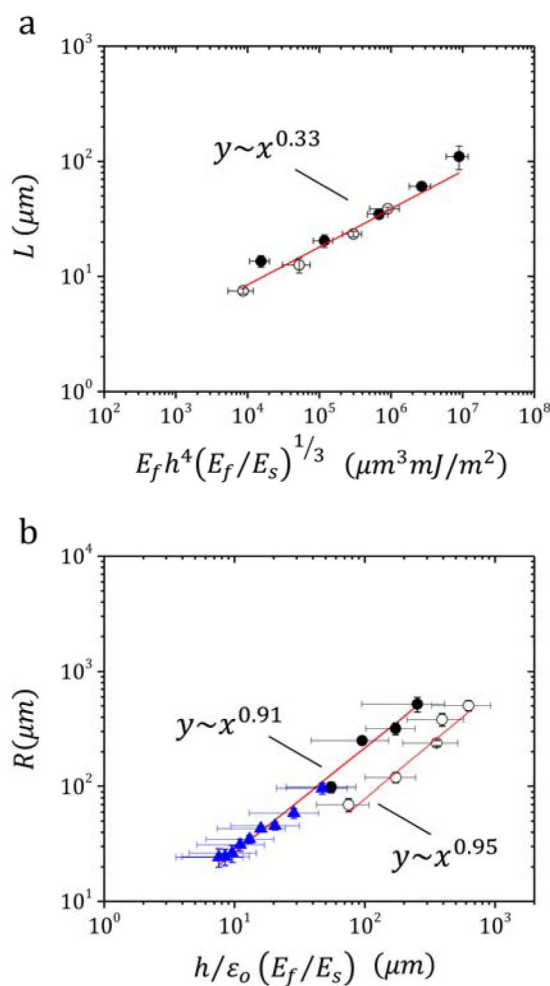


Fig. 3. Relaxation length (R) (right y-axis, red solid circles) and buckle delamination length (L) (left y-axis, black open circles) as a function of strain for a PS film ($h \approx 100$ nm) on PDMS. R is inversely proportional to strain, while L is essentially constant. The line is a power law fit to the R data with the exponent constrained to a value of -1 . The error bars represent one standard deviation of the data, which is taken as the experimental uncertainty of the measurement.

**Fig. 4.**

Scaling relationships for L and R . Samples 1–4 (high E_s) are plotted with solid circles, and samples 6–10 (low E_s) with open circles. In (b), data from Fig. 3 are plotted as solid triangles. The error bars represent one standard deviation of the data, which is taken as the experimental uncertainty of the measurement.

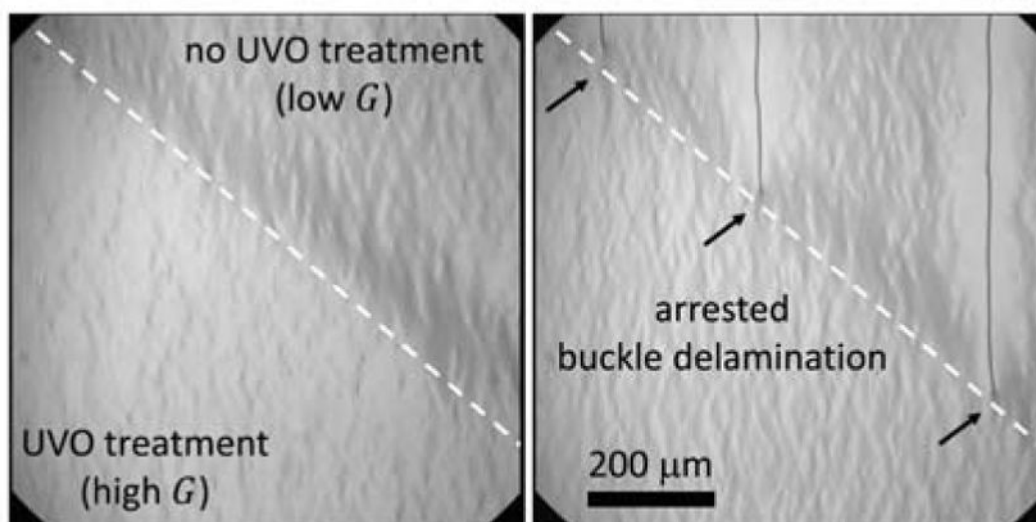


Fig. 5.

A PS film bonded to a selectively UVO-treated PDMS, showing the initiation of buckle delaminations on the untreated region (low G) and their arrest at the boundary between the treated (high G) and untreated (low G) regions.

Table 1

Measured values of h , λ , L , R , ϵ , and E_s for PS films on PDMS, and calculated values of E_f and L/λ .

Sample ^c	h (nm) ^a	λ (μm) ^b	L (μm) ^b	L/λ ^b	R (μm) ^b	ϵ (%) ^d	E_s (MPa)	E_f (GPa)
1	119 ± 6	6.0 ± 0.4	7.5 ± 0.4	1.3 ± 0.1	99 ± 10	1.69	2.35 ± 0.12	3.72 ± 0.91
2	177 ± 9	9.5 ± 0.7	12.6 ± 1.9	1.3 ± 0.2	250 ± 17	1.53	2.35 ± 0.12	4.36 ± 1.20
3	274 ± 14	14.6 ± 0.2	23.5 ± 1.2	1.6 ± 0.1	319 ± 38	1.36	2.35 ± 0.12	4.34 ± 0.72
4	398 ± 20	19.3 ± 1.6	38.6 ± 1.3	2.0 ± 0.2	518 ± 75	1.33	2.35 ± 0.12	3.23 ± 0.96
5	536 ± 27	--	--	--	--	--	--	--
6	119 ± 6	9.8 ± 0.3	13.6 ± 1.6	1.4 ± 0.2	70 ± 9	1.70	0.61 ± 0.03	4.09 ± 0.73
7	177 ± 9	16.2 ± 0.3	20.5 ± 2.6	1.3 ± 0.2	119 ± 12	1.24	0.61 ± 0.03	5.71 ± 0.96
8	274 ± 14	25.2 ± 0.8	34.8 ± 1.8	1.4 ± 0.1	238 ± 13	0.96	0.61 ± 0.03	5.75 ± 1.06
9	398 ± 20	35.4 ± 1.4	60.6 ± 4.9	1.7 ± 0.2	503 ± 51	0.80	0.61 ± 0.03	5.23 ± 1.03
10	536 ± 27	47.9 ± 2.1	110.5 ± 25.9	2.3 ± 0.6	382 ± 47	1.58	0.61 ± 0.03	5.28 ± 1.09

^aUncertainty taken as 5 % of the measurement value

^bUncertainty taken as the standard deviation of at least 4 measurements

^cFor Sample 5, λ could not be accurately measured due to buckle delamination.

^dThese *film* strain values have been adjusted from the controlled strain values applied to the *film-substrate* systems according to linear composite plate theory. The values given are the strains at which the measurements of L and R were made on a given sample. This was done since multiple PS films were tested on a single PDMS substrate simultaneously.