TIME-DEPENDENT DELAMINATION OF PLASTIC FILM:
OBSERVATION AND MODELING

Haiying Zhang, Zhenwen Zhou and Alexander Chudnovsky
University of Illinois at Chicago, Chicago, IL, USA
Hoang Pham
Avery Dennison Corporation, Mentor, OH 44060, USA

Abstract

The paper is aimed at the observation and modeling of thin plastic film delamination from a substrate and its progression. The authors are addressing the following issues: 1) what physical and geometrical parameters of the film-adhesive-substrate system play the major role in initiation and progression of delamination; 2) what is the root cause of slow delamination growth in time; and 3) formulation of a quantitative model of the process. A brief review of existing approaches, simple experimental setup, observations and the modeling results are reported.

Introduction

The problem of thin film debonding from a substrate in general is quite complex. It is addressed in numerous publications [1-25] that commonly focused on one of the two delamination modes, buckling-delamination and wrinkling. The former considers buckling of the film when it is partly delaminated from the substrate; while the latter assumes no delamination as the substrate deforms coherently with the film. Many theoretical models have been developed to describe the buckling delamination phenomena. These models mainly fall into two categories. The first is based on the Continuum mechanics approach. The fundamental work in that direction was done by Cotterell and Chen [1], Hutchinson et al. [2, 3, 13, 16, 24]. The authors rigorously address the effect of elastic-property contrast between the film and the substrate. They evaluate the elastic energy-release-driving delamination by applying Fracture Mechanics methods. The second category contains a number of so-called “lattice models” representing film and substrate by a lattice of points connected by bonds and springs, such as in Jagla [5] as well as in other works [18-21]. Wrinkling is commonly observed in stiff films deposited on relatively compliant substrates. Several models of wrinkling have been proposed [4, 7, 12, 15, 17, 22, 25]. Some of them employ a complex analytical and semi-analytical technique in dealing with the problem. For instance, Cerda and Mahadevan [4] use a semi-analytical approach to solve the Föppl–von Kármán equations and quantify the wrinkling of a thin elastic sheet under external forces and geometrical constraints.

The main applications of the models mentioned above are for semiconductor devices, magnetic storage media, surface coatings, stretchable electronics, and biomedical systems. The models can successfully deal with the failure of thin films on substrates due to thermal and/or mechanical strain, buckling phenomena of a relatively compliant film on a very rigid substrate or vice versa in which the film-substrate modulus ratio easily surpasses 100 or more, and wrinkling of films on elastomer substrate. In the case of a thin polymeric film bonding to a thicker load-bearing substrate (applications can be found in the pressure sensitive decorations of home care products, food and beverage containers, instruction labels on consumer goods, etc.), the buckling delamination phenomenon is not essentially different, however, a number of issues are not clearly addressed in the literature: 1) what geometrical and material parameters can be directly measured and used to quantify the chances of film delamination in field applications; 2) what is the root causes of time dependency of buckling delamination; 3) what is the role of adhesive properties. The purpose of this paper is to address the issues listed above, especially the time dependency of buckling delamination.

Experimental Observations

There are many variables affecting the buckling delamination process. Among them are film thickness, substrate geometry (surface curvature), visco-elastic properties of film, substrate and adhesive, as well as the adhesive strength. In the case when thickness of the substrate is significantly larger than that of the film, only geometry of the substrate surface (curvature) is important whereas the thickness and mechanical properties are insignificant for the problem at hand. Meanwhile, since for polymers the film-substrate modulus ratio is relatively small (less than 3), the effect of the substrate deformation is negligible as well. A complex substrate surface geometry with non-zero Gaussian curvature make it difficult to quantify the delamination. Therefore, a careful design of experiments is required to simplify the problem and separate the effects of adhesive strength, film rigidity and substrate curvature on the bonding integrity without losing the essential features of film delamination.

A random field of various defects (such as air bubbles, non-wetting spots etc.) is commonly present, when a film is directly applied onto a substrate. Figure 1 illustrates the
delamination progression with time from an edge initiation. The first frame in upper row \((t = 0 \text{ min})\) shows the top view of a thin transparent film applied on a flat horizontal plate (substrate). Then a pure bending was applied to the plate that became a cylinder with generatrix parallel to the \(y\)-axis (vertical axis on Figure 1) and upper surface with the film under compression. Delamination usually starts at the largest defect. In this case, it was air bubble located at the edge directrix (the bubble is pointed by arrow on the second frame of Figure 1 \((t = 1 \text{ min})\). The delamination grew much faster in the generatrix direction (perpendicular to the compression direction). At time \(t = 58 \text{ mins}\), the length of delamination in generatrix direction is four times larger than that in the directrix direction. Such a difference in the delamination growth is determined by the significant difference in the blister curvature in two directions.

![Figure 1. 2-D delamination of a film initiated an air bubble at the edge of the film.](image1)

To reduce the blister growth to a 1-D problem, one can introduce a narrow defect parallel to the generatrix across the entire width of the film. It triggers delamination that grows in directrix, i.e., in compression direction. The setup is shown schematically in Figure 2. The delamination growth is recorded in real time from two angles: 1) top view; 2) side view. Figure 3 shows a sequence of top view (upper sequence) and corresponding side views (lower sequence) frames of the delamination under compression. Some details of experimental setup and observations will be further discussed in the next section.

![Figure 2. Reduction of 2-D problem to 1-D delamination](image2)

The origin of delamination is a natural trend of all elastic system to minimize the potential energy \(\Pi\) that is conventionally defined as the difference between elastic strain energy \(F\) and work \(W\) done by external forces \(\Pi = F - W\). Formation of a blister releases the potential energy of compression that happens rapidly in a purely elastic system. However, a close examination of the delamination profiles at higher magnifications reveals highly stretched and fibrillated viscoelastic adhesive that connects the film with substrate and forms a transition zone between complete delamination and fully attached film domains. Evolution of the blister tip region is shown on Figure 4 (ignore the mirror reflection on the lower portion of the micrographs).

![Figure 3. Delamination evolution in time: upper row shows top views and lower row displays the side views.](image3)

At time indicated as 15 minutes (from the beginning of the test) the adhesive is “pulled” out and forms two cone-shape-like fibers with bright white reflection lines marking the edges on the left side of the micrographs. These two cone-shape fibers continue to grow following the vertical movement of the film that can be seen in the adjacent micrographs taken in 5-minute intervals. The micrograph taken at 30 minutes shows the same fibers completely separated from the substrate and forming two droplets attached to the film (upper left side of the micrograph. Figure 4 shows only a side view of the transition zone. A complex structure of the transition zone is also revealed by the top view shown on Figure 5.

The observations illustrated in Figures 4 and 5 suggest that the viscosity of adhesive may be the main factor controlling the time dependency of delamination and the rate of blister growth. In other words, the energy dissipation that is a part of work \(W_T\) done by the film deflection on a large viscoelastic deformation of adhesive within the transition zone is the source of delay of elastic energy release in buckling delamination.

Mechanical interaction of the film, adhesive layer and substrate is associated with the traction acting between the film and adhesive layer on one side and the adhesive layer and substrate on the other. The traction distribution depends on the shape of the film deflection and the stress-strain relation of pressure sensitive adhesive. The latter is
typically measured in probe-tack test [26-29]. The adhesive is practically incompressible visco-elastic material. Buckling delamination of the film is a manifestation of release of the elastic energy of the compressed film. Deflection of the film applies a tension pull on adhesive leading to a volume increase between the film and substrate, buildup of hydrostatic tension and cavitation within the adhesive layer. With farther increase of film deflection, the free volume within adhesive grows and a “lizard’s feet” pattern of deformation within transition zone is formed (see Figure 5).

![Figure 5](image.jpg)

Figure 5. “Lizard’s feet” pattern of adhesive large deformation within the transition zone between the delamination and fully bonded film domains.

A stress relaxation within the film apparently leads to a reduction of elastic strain energy and therefore to a certain reduction of the delamination driving force. In continuum mechanics modeling the viscoelastic deformation and energy dissipation within adhesive can be presented as the time dependent specific energy required for delamination. A competition between the above two time dependent processes results in a complex time-dependent process of thin film buckling delamination.

**Quantitative Analysis of Buckling Delamination**

Consider a rectangular film sample of length \( L_f \), width \( b \) and thickness \( h \) attached to a flat and relatively thick plate with help of pressure sensitive adhesive. The plate thickness \( H \) is much larger than \( h \). The plate is supported on the two edges labeled as O as shown in Figure 6. A controlled deflection \( \Delta A \) is applied at the two points A with a distance \( L \) apart.

The geometrical notations of this setup are shown in Figure 6. The middle portion of the plate with attached film between the points A-A is under pure bending. The film attached to the compressed side of the plate also experiences compression that induces a tendency for delamination. A delamination across entire width of the film is triggered by initial narrow defect of size \( l \) (see Figure 6) extended over entire film width \( b \).

\[
\varepsilon = \frac{\Delta A h}{a^2 \left( \frac{2}{3} \right)}.
\]  

(1)

A small blister in the vicinity of the narrow defect \( l \) appears upon application of displacements \( \Delta A \). Formation of blister is a classical problem commonly addressed by using von Karman nonlinear plate theory. The solution of the problem gives the following buckling delamination profile approximation \( \nu(x) \):

\[
\nu(x) = \frac{L}{2} \left( 1 + \cos \frac{2\pi x}{L} \right), \quad x \in \left[ -\frac{l}{2}, \frac{l}{2} \right].
\]  

(2)

where \( \nu(x) \left( -\frac{l}{2} \leq x \leq \frac{l}{2}, \; l < L_f \right) \) is the deflection of the debonded film, \( f \) and \( l \) are the height and length of the blister, respectively. The agreement between the model and experiment is illustrated in Figure 7. It shows the blister (white profile) has grown for 15 hours and has reached the length 12.8 mm and height 1.5 mm.

The red solid line is computed using Eq. (2) with measured \( l = 12.8 \text{ mm} \) and \( f = 1.5 \text{ mm} \). Apparently the model curve (red one) and the experimental blister profile (white curve) are practically overlap.

The entire film consists of two parts: attached \((L_f - l)\) and debonded \(l\) one. Accordingly the total strain energy of the film consists of strain energy \( F_c \) of compression (attached) part and bending strain energy \( F_b \) of debonded portion.
The equilibrium of moments at the blister front is schematically shown in Figure 9 (left). From such curves, maximum stresses $\sigma_{\text{max}}$ and maximum strains $\varepsilon_{\text{max}}$ can be extracted. They both highly depend on strain rate: the higher the strain rate, the higher $\sigma_{\text{max}}$ and $\varepsilon_{\text{max}}$. Without a probe-tack test, we assume a linear force-displacement relationship (as shown in Figure 9 right) with unknown peak adhesive force $F^*$ and maximum displacement $\delta^*$ that can be measured from recorded photos.

$$T(x) = F^* - \frac{F^*}{\delta^*} \sigma(x), x \in \left[ \frac{l_{cr}}{2}, \frac{l}{2} \right]$$

Figure 10. Equilibrium of moments at the blister front.

The equilibrium of moments at the blister front is written as:

$$F = \frac{1}{2} E_f \varepsilon^2 hb \left( L_f - l \right) + \frac{\pi^4 E_f b h^3 l^2}{12 I^3}.$$
\[
\int_{t_{cr} / 2}^{l / 2} T(x) \left( \frac{l}{2} - x \right) dx = \frac{p f}{E_f} I' \left( x \right) \bigg|_{x = \frac{l}{2}}
\]

(8)

By substituting \( T(x) \) with Eq. (7), one can solve Eq. (8) for \( F^* \), thus \( T(x) \) can be determined. The result is shown in Figure 11.

![Figure 11. Traction distribution \( T(x) \) over the transition zone.](image)

This traction acting on the adhesive produces work density \( \bar{W}_{TZ} \) on the loading points displacement. Part of this work \( \alpha \bar{W}_{TZ} \) goes into elastic strain energy density of adhesive \( F_{ad} = \alpha F_{TZ} \) and another part \( (1 - \alpha) \bar{W}_{TZ} \) is dissipated in the inelastic processes within the adhesive such as voiding, large viscous deformation and fracture as illustrated in Figure 4.

The work density \( \bar{W}_{TZ} \) of traction \( T \) at a given point \( x \) within transition zone can be computed as follows:

\[
\bar{W}_{TZ} = 2b \int_{t_0}^{t / 2} T(x) \cdot \dot{v}(x) dt
\]

(9)

where \( \dot{v}(x) \) is the velocity of the film vertical displacement at the crack front. Figure 12 shows the velocity of the film vertical displacement at a position \( x \) within the transition zone as a function of time starting with time when the blister tip is at the current crack front position until the time of crack reaching that blister tip position i.e., the transition zone life time.

The total work \( W_{TZ} \) of tractions \( T(x) \) done over transition zone is:

\[
W_{TZ} = \int_{t_{cr} / 2}^{l / 2} \bar{W}_{TZ}(x) dx
\]

(10)

According to Figure 12 there is a short transition zone life at the beginning of blister formation during which the velocity is relatively high; it increases sharply first and then declines. Later in the process transition zone life becomes much longer, the vertical displacement velocity is much smaller than at the beginning, but behaves in the same manner: first increases and then gradually declines. It corresponds to the crack front moving across the position \( x \).

The work density \( \bar{W}_{TZ} \) is computed according to Eq. (9) and the result is shown in Figure 13. Integration of adhesive strain energy density \( F_{ad}(x) \) over the transition zone length gives the adhesive part of strain energy \( F_{ad} \). The sum of strain energies of \( F_f \) and \( F_{ad} \) gives the strain energy of film-adhesive system \( F = F_f + F_{ad} \).

![Figure 12. Velocity of vertical displacement as a function of time within transition zone life time.](image)

![Figure 13. Development of \( W_{TZ} \) and its derivative per width with respect to blister length.](image)

The energy release rate \( G_t \) driving delamination growth is derived as a derivative of the system potential energy \( \Pi \):

\[
G_t = - \frac{\partial \Pi}{\partial l_c t}
\]

Since there is no work of external forces during the blister growth process the potential energy \( \Pi \) coincides with the strain energy of the system. Thus, in this case the energy release rate associated with delamination growth is:

\[
G_t = - \frac{\partial \Pi}{\partial l_c t} = - \frac{\partial F}{\partial l_c t}
\]

(11)

The law of delamination grows can be derived from the second law of thermodynamics similar to that for the crack growth [30]. The entropy production associated with delamination growth is:

\[
T \cdot S_t = \left( G_t - 2\gamma + [1 - \alpha] \frac{\partial W_{TZ}}{\partial l_c t} \right) \cdot l_c \geq 0
\]

(12)
Here a specific energy of delamination, $2\gamma$, is introduced. It is comprised of the work done on voiding and irreversible stretching of adhesive before break, i.e., the energy dissipation within adhesive, and the work $2\gamma_\delta$ required for brittle fracture of stretched adhesive fibers and membranes. The third term in parenthesis $[1 - \alpha]\cdot \frac{\partial W_{zz}}{\partial \epsilon_{tr}} = [1 - \alpha]\cdot \tilde{W}_{zz}$ is an irreversible work density on large inelastic deformation of adhesive at the delamination tip accumulated over the life time of transition zone.

Thus, according to Eq. (12) the thermodynamic force for buckling delamination $X^{(bd)}$ is defined as:

$$X^{(bd)} = G_t - (2\gamma - [1 - \alpha]\cdot \tilde{W}_{zz}) \quad (13)$$

Progression of delamination takes place in a very slow fashion: for example, in the experiment depicted in Figure 3 about 12.5 mm of delamination length is achieved over about 240 minutes, i.e., the average rate of delamination growth is less than a micron per sec in. Thus, it is clearly a quasi-equilibrium process, for which the thermodynamic force of delamination vanishes, i.e., $X^{(bd)} = 0$ at every instance. It corresponds to the necessary condition of blister equilibrium (similar to Griffith condition in Fracture Mechanics).

It shall be noted, the rate of elastic (brittle) fracture advances fast in contrast with observed slow blister growth, since the rate of delamination progression is controlled by the rate of viscous (inelastic) deformation of adhesive. Indeed, considering a large true strain $\varepsilon(x)$ of adhesive one may ignore the elastic part and relate the traction $T(x)$ with an inelastic strain rate $\dot{\varepsilon}^{(in)} = \dot{\nu}/\nu$; $T(x) = \mu \dot{\varepsilon}^{(in)}$, where $\mu$ is viscosity coefficient with dimensions $[\mu] = MPa \cdot sec$. Therefore, $\dot{\nu}/\nu = \mu^{-1} T(x)$ with $T(x)$ determined above.

Conclusions

Observations and a general thermodynamic consideration of buckling delamination of thin film are discussed. The major parameters determining the film-adhesive-substrate system comparability are the film compression rigidity (the product of elastic modulus and thickness), viscoelastic properties of adhesive and the geometry (curvature) of substrate. The mechanical properties of substrate are less important, when the substrate has comparable with film modulus and much larger thickness, i.e., significantly larger rigidity.

The delamination usually starts from a random defect. Therefore, an especially design defect is introduced to control the location and direction of delamination growth. It allows a quantitative characterization of the process.

A thermodynamic force reciprocal to delamination growth is defined as the difference between the elastic energy release rate and the specific delamination energy required to form a unit area of delamination. The accumulation over time work density on voiding and large deformation of adhesive reduce the specific delamination energy at the instance of delamination advance.

A slow delamination growth is in a large extend controlled by viscous deformation of adhesive.

Acknowledgement

The authors wish to acknowledge Avery Dennison as the sponsor for this project. Special thanks go to Andrew Poslinski, Luigi Sarto, Sjaak Elmendorp and Noelle Sieradski.

References