Interface and surface roughness of polymer-metal laminates
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Chapter 3

Cohesive zone modeling of laser induced delamination

3.1 Introduction
A careful characterization of the adhesion of polymer coatings to metal substrates is important to evaluate the performance of coatings in practice. Among the various available techniques, the so-called blister test has become well established [1-5]. The method is based on the introduction of a pressure between the coating and the substrate. Usually a hole is made in the substrate and the system is loaded and finally an increasing pressure will produce failure of the coating. A drawback of this approach is the difficulty of creating a hole without damaging the coating or the interface.

An alternative is the Laser Induced Delamination (LID) technique in which blisters are formed with the help of a pulsed infrared (IR) laser. The technique was proposed by Meth et al. in [6], where the adhesion properties of sandwich-like films were measured, and which was developed further by Fedorov & De Hosson in [7] for coatings of polymers on metals. An advantage of this technique is that no special preparation of the samples is required. By increasing the power density of subsequent laser pulses, an increasing pressure of gas (resulting from the evaporation of polymer near the interface) inside subsequent blisters can be attained that is sufficient for delamination [7].

Analysis of the experiments is usually performed by using a simple elastic model [7]. This limits the applicability of the method to situations where several of the assumptions are questionable, i.e. concerning plasticity, complex geometries or fracture at the interface depending sensitively on the mode mixity at the locus of delamination (i.e. the crack tip). A finite element model including a simple description of the interaction across the interface (e.g. in terms of a force-separation law) extends the applicability range of the method to such a situation. Another new interesting aspect is the possibility to incorporate plasticity in the description of the blister.

In addition, the method of analysis presented in [7] is only useful to extract a value of the work of adhesion $G$ and does not address other parameters describing the interaction at the interface such as the maximum interface stress or the range of the interaction potential. The maximum interface stresses may have a large impact on the failure mechanism [8,9]. In soft materials crack blunting could appear [8]. Crack blunting can be included in the theoretical model using the concept of cohesive zones and the theory shows remarkable agreement with experiments. Moreover, cohesive zones are able to describe all kinds of failure [8,9].
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In this chapter, the finite element model described in chapter 2 is used in conjunction with the elastic model to analyze the experimental data obtained with the LID. The objective is to investigate whether the method leads to identical results in situations where the elastic model should hold. This touches upon the applicability of cohesive zone modeling. Another objective is to investigate the effects of plasticity.

The outline of this chapter is as follows. First, a summary will be given of the experimental techniques. Second, a brief overview of the analysis of the experimental results is presented in terms of linear elasticity. It is shown that the method of Fedorov and De Hosson [7] may be further simplified. Third, a finite element model (FEM) has been used to describe the blister geometry. This model will be compared with the linear elastic analytical model to investigate the limits of the validity of the linear elastic analytical model. Fourth, the FEM will be used to mimic the behavior of PET and show the impact of plasticity. Finally to show the accuracy of both models in reality, both models are employed to evaluate several measurements of a PET layer of 40 µm on top of chromium coated carbon steel (ECCS steel).

3.2 Laser induced delamination

The method of LID is illustrated schematically in Fig. 3.1. A sample with an IR transparent polymer coating on a steel substrate is subjected to an IR laser pulse. The interface is heated above 1400 K [7] and a thin layer of coating is evaporated. The evaporated coating material expands and develops a blister.

In the approach detailed in [7], part of the IR laser beam is masked. The unexposed edges will delaminate if the pressure is large enough. By increasing the laser power, the delaminated area increases (shot 1 and 2). At a certain laser power the masked strip is delaminated completely and the two blisters merge to one (shot 3). As will be explained in the thin plate theory (see section below), the blister height $H$ and blister width $b$ are sufficient to determine the work of adhesion $G$ (see also chapter 6), the overpressure $p'$ and the number of moles of gas $n$ inside the blister.

An Expla Nd:YAG IR laser from Continuum was used to produce the IR pulses. The maximum pulse energy is 800 mJ in one pulse of 5 ns. The wavelength of the laser is 1.064 µm. The original beam width is 8 mm, which is expanded three times. The energy density is therefore 1.8 kJ/m². After the expansion a high power attenuator from Del Mar Ventures was used to vary the intensity of the beam. The attenuator consists of a UV grade fused silica wheel with diffraction gratings. Finally, a lens was used to focus the beam onto the sample. The sample can be moved along a track parallel to the beam, resulting in different positions with respect to the focal point. The energy density on the sample can be varied by both the attenuator and the position of the sample with respect
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to the focus point. In order to prevent that the blister cap is blown away by the impact of the shockwave, a glass plate is mounted on top of the sample. After a laser shot, the profiles were measured with a Mahr profilometer. These profiles yield the width \( b \) and the height \( H \) of the blister. After integrating the profile, the cross-sectional area is found.

It is worthwhile to discuss a few aspects of the LID method in more detail since they touch upon the reliability of the analysis.

First, it is assumed that only elastic deformations are introduced in the coating process. As long as the time scales involved in the polymer are much larger than the laser pulse, this is a valid assumption. The IR laser used in this investigation has a pulse length of 5 ns and therefore the deformation can be treated as being elastic [7].

Second, the shape of the blister is predicted by the thin plate theory and defined by \( b \) and \( H \). In [7], the shape of the blister could be fitted within the experimental error.

Third, there is an amount of gas \( n_m \) that should obey the ideal gas law \( (p'V = n_mRT) \). \( b \) and \( H \) are measured at room temperature and indirectly the pressure \( p' \) and volume \( V \) are deduced. These parameters are sufficient to calculate \( n_m \). Agreement between the measured \( n_m \) and the computed \( n_m \) implies that the elastic thin plate description of the blister is correct and furthermore it indicates that plastic deformation does not occur. In [7], 20 to 30 blisters were taken in a vacuum system connected to an absolute pressure meter. The blisters were cut one by one with a razorblade and at the same time the pressure was measured. These pressures, together with the measured volume, were used to compute \( n_m \). The measurements showed a good correlation with the calculated values of \( n_m \).

Fig. 3.1. Schematic presentation of the LID technique. A series of subsequent laser shots are performed through the mask shown at the top-left. The pulse intensity increases with each shot. Shots 1 and 2 correspond to pulse intensities below the intensity needed to delaminate the masked region. Shot 3 corresponds to the intensity at which the masked area just delaminates and both blisters merge together [7].
These arguments show that the theory in [7] describes correctly the geometry within a linear elastic model. So far, this elastic model is used to create an energy balance to find the work of adhesion $G$. The validation of this procedure and the corresponding limits will be presented in this chapter.

### 3.3 Thin plate model

In the past several studies were dedicated to the description of the blister [7,10,11] with different formulas used to calculate the work of adhesion $G$. In this section, a concise derivation of $G$ will be given, leading to a simplified form of $G$.

An infinitely long cylindrical blister aligned with the $x$ axis can be described as a 2D bending plate (Fig. 3.2). The analysis in [7] assumes that the Kirchhoff assumptions [12] can be applied to this problem:

a. Initially the material is a flat, elastic, homogeneous, isotropic plate.

b. The normal deflection (i.e. along $z$, see Fig. 3.2) of the neutral plane $w(y)$ is small compared to the thickness $t$ of the plate. The slope $\frac{\partial w(y)}{\partial y}$ is therefore negligible.

c. The initial thickness of the plate is preserved.

d. Since the displacements of the plate are small, it is assumed that the neutral $w(y)$ axis remains unstrained (thin plate theory) after bending.

We start from equilibrium and apply the Kirchhoff assumptions shown in Fig. 3.2. After assuming that only one force component $F_z$ is applied, the equilibrium conditions for the two-dimensional system become:

$$\frac{\partial \sigma_y}{\partial y} + \frac{\partial \sigma_z}{\partial z} = 0$$

$$\frac{\partial \sigma_z}{\partial z} + \frac{\partial \sigma_y}{\partial y} + F_z = 0,$$

where $\sigma_i$ are the stress components in the different directions (see Fig. 3.2).

![Fig. 3.2. Schematic representation of cylindrical blister showing relevant parameters (see text).](image)
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After application of assumption b, the only nonzero principle strain term is \( \varepsilon_z \).

\[
\varepsilon_z = \frac{\partial w}{\partial z}, \tag{3.2}
\]

where \( w \) describes the deflections along \( z \). After integration of these equations and rewriting, the deflection \( w \) of a thin plate under normal uniform pressure \( p' \) is given by:

\[
d^4w \quad \frac{F_z}{bD} \quad \frac{p'}{D}, \tag{3.3}
\]

where \( D = E t \left(12(1-\nu^2)\right)^{-1} \) is the flexural rigidity, \( E \) is the elastic modulus, \( \nu \) is the Poisson’s ratio and \( t \) is the film thickness. The blisters are overpressurized and the excess pressure with respect to the atmospheric pressure \( p_{\text{atm}} \) is denoted by \( p' \). Then the absolute pressure inside the blister is \( p = p' + p_{\text{atm}} \).

The boundary conditions are as follows:

\[
w = 0, \quad \frac{dw}{dy} = 0, \quad \text{at} \quad y = -\frac{b}{2} \quad \text{and} \quad y = \frac{b}{2}, \tag{3.4}
\]

where \( b \) is the dimension of the blister along the \( y \) axis. The solution for this boundary value problem is

\[
w(y) = \frac{p' b^4}{24D} \left[ \left( \frac{y}{b} \right)^2 - \frac{1}{4} \right]^2 \tag{3.5}
\]

and the corresponding blister shapes are shown in Fig. 3.3. The following parameters are used in the figure: \( t = 30 \mu m, \ \nu = 0.4, \ b = 1 \text{ mm} \) and \( E = 2 \text{ GPa} \) (also used in the following figures). The maximum height \( H \) is reached at the centre of the blister (i.e. \( y = 0 \)):

\[
H = w(0) = \frac{p' b^4}{24D} \frac{1}{16}. \tag{3.6}
\]

The pressure inside the blister can be calculated from \( b \) and \( H \):

\[
p = p_{\text{atm}} + \frac{24D \cdot 16H}{b^4}. \tag{3.7}
\]

The blister shape can be also expressed in terms of \( b \) and \( H \):

\[
w(y) = 16H \left[ \left( \frac{y}{b} \right)^2 - \frac{1}{4} \right]^2. \tag{3.8}
\]
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Fig. 3.3. Analytical blister shapes for increasing pressure $p' = (\cdot) 0.2, (\ldots) 0.4, (\cdots) 0.6$ and $(\cdot) 0.8$ bar. The following parameters are used: $t = 30 \, \mu\text{m}$, $\nu = 0.4$, $b = 1 \, \text{mm}$ and $E = 2 \, \text{GPa}$.

The non-zero stresses obtained from linear elasticity are obtained by integrating the equations of equilibrium:

$$
\sigma_x = -\frac{Ez}{1-v^2} \frac{p'b^2}{6D} \left( \frac{y}{b} \right)^2 \left( \frac{1}{4} \right),
$$

$$
\sigma_{yz} = \frac{E}{2(1-v^2)} \left( z^2 - \frac{t^2}{4} \right) \frac{p'y}{D},
$$

$$
\sigma_z = -\frac{E}{2(1-v^2)} \left( \frac{z^3}{3} - \frac{t^3}{4} - \frac{t}{12} \right) \frac{p'y}{D} - p' \approx 0,
$$

Fig. 3.4. Maximum shear stresses $\sigma_{yz}$ as a function of the blister height for $b = \Box 0.9$, $\ast 1.0$ and $\circ 1.1$ mm. The following parameters are used: $t = 30 \, \mu\text{m}$, $\nu = 0.4$, $b = 1 \, \text{mm}$ and $E = 2 \, \text{GPa}$.

Fig. 3.5. Maximum tensile stresses $\sigma_z$ in the coating as a function of the blister height for $b = \Box 0.9$, $\ast 1.0$ and $\circ 1.1$ mm. The following parameters are used: $t = 30 \, \mu\text{m}$, $\nu = 0.4$, $b = 1 \, \text{mm}$ and $E = 2 \, \text{GPa}$.
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In the interior of the blister the pressure acts as the only force along the $z$ direction. This value is also found as $\sigma_y|_{z=-t/2} = -p^*$. Inside the coating shear stresses operate (Fig. 3.4). The component $\sigma_y$ provides the largest contribution to the strain energy of the film (Fig. 3.5). Due to bending of plate, at the boundary ($y = \pm b/2$ and $z = -t/2$), tensile stresses appear and compressive stresses occur at the center of the bottom of the membrane ($y = 0$ and $z = -t/2$). At the top ($z = t/2$) of the blister, the $\sigma_y$ stress has a negative value, which is opposite to the stresses at the bottom ($z = -t/2$). According to the thin plate theory, the principle stresses at the neutral axis are equal to zero. The maximum stress in the film is reached for the $\sigma_y$ component at the clamped boundaries ($y = -b/2$ and $y = b/2$) at the interface with the substrate ($z = -t/2$):

$$\sigma_{y\text{max}} = \frac{Et}{(1-v^2)24D} p^* b^2.$$

The separate contributions to the elastic energy $U_{el}$ stored in the blister can be calculated as follows using plane strain conditions:

$$U_y = \iint_A \frac{(1-\nu^2)\sigma_y^2}{2E} = \frac{1}{2} \left( p^* \right)^2 \frac{b^5}{30 \cdot 24D}$$

and similarly

$$U_z = 0 \text{ and}$$

$$U_{yz} = 0.$$

Then the total elastic energy $U_{el}$ becomes:

$$U_{el} = U_y + U_z + U_{yz} = \frac{1}{2} \left( p^* \right)^2 \frac{b^5}{30 \cdot 24D}$$

The volume per unit length for the blister has the following expression:

$$V = \frac{p^* b^5}{30 \cdot 24D}.$$

As a consequence of the evaporation of part of the coating the pressure increases, which might lead to partial delamination of the coating. The work done by the gas at constant $T$ is given by the change in the Helmholtz free energy $dF = -pdV$. Due to the delamination the pressure will drop. The condition for delamination is as follows:

$$G_{db} = p_{in} dV - (dU_{el} + dF),$$

where $db$ is the delaminated area.
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During delamination, the blister width changes from $b_0$ to $b$. $n_w$ is assumed to be constant ($pV = const$). Therefore, $p'$ can be written as a function of the blister width:

$$p'(b) = -\frac{p_{am}}{2} + \sqrt{\left(\frac{p_{am}}{2}\right)^2 + \left(p_{am} + p'_0\right) p'_0 \left(\frac{b_0}{b}\right)^3}, \quad (3.19)$$

where $p'_0$ is the initial overpressure in the blister. The change of the strain energy in Eq. (3.13) can be written as a function of the change in blister size:

$$dU_{el} = \frac{\partial U_{el}}{\partial b} db + \frac{\partial U_{el}}{\partial p'} dp'. \quad (3.20)$$

Because the blister increases in size, the first term at the right hand side is positive. The second term will be negative because of the pressure drop:

$$\frac{dp'}{db} = \frac{5 p'(p_{am} + p')}{b(p_{am} + 2p')}. \quad (3.21)$$

For reasons of simplicity the partial derivative is expressed in terms of the original function $p'$ given in Eq. (3.19).

Using Eq.(3.16), (3.19) and (3.21), Eq. (3.20) can be written as follows:

$$\frac{dU_{el}}{db} = -\frac{b^4 (p')^3 p_{am}}{12 \cdot 24 D (2 p' + p_{am})}. \quad (3.22)$$

The second term necessary to compute the work of adhesion $G$ is

$$\frac{dV}{db} = \frac{\partial V}{\partial b} + \frac{\partial V}{\partial p'} dp'. \quad (3.23)$$

Performing the partial derivatives one finds:

$$\frac{dV}{db} = -\frac{b^4 (p')^3}{6 \cdot 24 D (p_{am} + 2 p')} \quad (3.24)$$

The next step is to rewrite Eq. (3.18) such that the calculated derivatives can be implemented:

$$G = -p_{am} \frac{dV}{db} - \left(\frac{dU_{el}}{db} - (p' + p_{am}) \frac{dV}{db}\right) = p' \frac{dV}{db} - \frac{dU_{el}}{db} \quad (3.25)$$

Combining this equation with Eq. (3.22) and (3.24) yields the following relation for $G$:

$$G = \frac{(p')^2 b^4}{12 \cdot 24 D}. \quad (3.26)$$
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Since the only terms measured by the stylus profiler are \( H \) and \( b \) it is convenient to use Eq. (3.7) leading to:

\[
G = \frac{512D}{b^2} H^2. \tag{3.27}
\]

The next section is focused on the FEM and we will adopt Eq. (3.27). It is for that reason that a couple of aspects should be clarified. First, the Kirchhoff assumptions are valid for blisters with height \( H \leq 0.3 \cdot l \) [7]. Higher blisters have a strained neutral axis. Second, \( G \) determines the shape of the blister and therefore implicitly the tensile stresses in the coating \( \sigma_y \) (Fig. 3.6). Third, a larger blister implies a smaller pressure (Fig. 3.7). Fig. 3.7 shows an asymptotic like relationship (Eq. (3.26)). For constant pressure, initial fracture results in fracture of the whole interface. From pressure point of view, this makes the fracture process unstable. Fourth, the amount of gas molecules \( n_m \) inside the blister at \( p^* = p \) is a linear function of \( G \) and \( b \):

\[
n_m = \frac{p^* V}{R \cdot T} = \frac{12G \cdot b}{30R \cdot T}. \tag{3.28}
\]

Since the laser spot size \( b \) and the amount of moles \( n_m \) show a linear relationship, one could not expect that changing the laser spot size leads to delamination. To start delamination, it is necessary to increase the power density to create a situation of delamination.

Fig. 3.6. Relation between the work of adhesion and the tensile stress in the coating at the moment of delamination. The calculations are carried out for three different blister width: \( b = \square 0.8, \star 1.0 \) and \( \circ 1.2 \) mm.

Fig. 3.7. Relationship between the pressure and the width of the blister for different \( G \): (\( \cdot \)) 1, (\( \ldots \)) 5, (\( \sim \)) 10 and (\( \sim \)) 30 J/m\(^2\).
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3.4 Finite element model versus analytical model

First we will study the validity of the finite element model, showing that it approaches the analytical solutions. Second, we use the finite element model to study the behavior of blisters beyond the range of applicability of the analytical model.

3.4.1 Boundary conditions

Fig. 3.8 schematically shows the boundary conditions. The laminate consists of a substrate and a coating. The substrate mimics steel and is defined as a purely elastic material with an elastic modulus $E = 200 \text{ GPa}$ and Poisson’s ratio $\nu = 0.3$. The coating will have various properties e.g. purely elastic or elastic-plastic. The attachment of the coating to the substrate is described by a stress-separation law or cohesive zone. The cohesive zone has been explained in chapter 2 and is defined by the interface energy $G_{c2}$, the coupling between shear (Mode II) and normal opening (Mode I) and the characteristic interaction distance $\Delta_0$.

Fig. 3.8 also shows the boundary conditions. On the left and right, the displacements are suppressed in the horizontal direction and are free in the vertical directions. The bottom of the finite element model is restricted in both horizontal and vertical direction. The pressure is implemented by applying a force perpendicular to the coating surface at each node (in Fig. 3.8 the crossing of two lines) of the inner blister surface. The magnitude of the force is computed from the surface area and the applied pressure. Therefore, the simulation is pressure controlled.

The blister becomes unstable when fracture occurs (see Fig. 3.7) and the pressure is kept constant. Because our interest is focused on the fracture process, this instability should be avoided. In the first stage of the simulation, the coating behaves elastic and the pressures are not high enough to initiate fracture. In this stage, the blister is pressure controlled and the pressure is increased with fixed steps $dp'$. After a certain number of steps, the initiation of fracture, the next stage of the simulation, starts. Stage two can be characterized as mole controlled. The pressure does therefore not increase linearly and the instability is avoided. At each step $n$, the ideal gas law is assumed to be obeyed:

$$p_n' V_n = c_n$$

where $c_n$ is $n_nRT$. The pressure of the step $n + 1$ is calculated as follows:

$$dp_{n+1}' = \frac{dc_{n+1}}{V_n} - \frac{c_n dV_{n+1}}{V_n^2}.$$  \hspace{1cm} (3.30)

The differences are calculated as $dx_n = x_n - x_{n-1}$, where $x$ is $c$, $p'$ or $V$. The volume change $dV_{n+1}$ can be approximated (assuming constant $p'$) by:

\begin{align*}
\end{align*}
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\[ dV'_{n+1} = dV_n \frac{dc_{n+1}}{dc_n}. \]  

(3.31)

Combining Eqs. (3.29)-(3.31) the new pressure becomes:

\[ p'_{n+1} = p'_n + dp'_{n+1} = p'_n + \frac{dc_{n+1}}{V_n} \left( 1 - \frac{c_n}{V_n} \frac{dV_n}{dc_n} \right) \]  

(3.32)

The new pressure is now expressed in known parameters and the applied molar change \( dc_{n+1} \).

In general, the first pressure step \( dp'_1 \) dictates the simulation process. This has the disadvantage that after fracture, several cohesive zone elements are able to fracture in one step. This implies that the cohesive zone elements are not able to follow the defined stress-displacement curve. The integral of the stress-displacement path is the dissipated energy, which should be equal to \( G_{CZ} \). Therefore, when many cohesive zone elements fracture in one step, the results will not be reliable.

To avoid this problem, \( dc_{n+1} \) is divided by 10 when an element reaches an normal opening \( \Delta_n > \Delta_n \). On the other hand, if the fracture stops, \( dc_{n+1} \) is increased slowly.

Fig. 3.8. Schematic representation of the boundary conditions of a finite element calculation of a blister. The gray area is enlarged schematically on the right.

3.4.2 The interface

As discussed in chapter 2, the cohesive zone represents the interface. Since the concept of the cohesive zone is used here for the first time, a few examples will be given to clarify it. Fig. 3.9 shows a selection of different interaction lengths \( \Delta_0 \) with the same work of adhesion of the cohesive zone \( G_{CZ} = 2.5 \text{ J/m}^2 \). A short \( \Delta_0 = 50 \text{ nm} \) and strong interaction represents a brittle failure and a long \( \Delta_0 = 500 \text{ nm} \) and weak interaction describes ductile fracture. The interaction length is a crucial parameter, which should be chosen with special care. Another interesting aspect is the mix of modes. In Fig. 3.9,
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Fig. 3.9. Normal (left) and tangential (right) traction components calculated for the cohesive zones with various characteristic normal traction separation length $\Delta_0$. Tangential interaction length is fixed at $\sqrt{2}\Delta_0$.

both interactions are shown separately. In reality interaction between both modes occurs. To illustrate this, Fig. 3.10 shows the displacements at the crack-tip of a blister. As the blister is not symmetrical at the crack-tip, a mode mixity is expected (Fig. 3.11). For increasing pressures and displacements, the stresses increase until the maximum appears and the next nodes take over the load. When the stresses return to $(0,0)$ the full amount of energy $G_{c2}$ is dissipated and the cohesive zone is fractured. After fracture, further separation will not lead to stresses.

Fig. 3.10. Evolution of the normal and tangential displacement of the nodes located at the blister boundary during delamination. The blister pressure is the running parameter with direction as indicated. The legends of the nodes are given schematically in the insert. The used interaction length is $\delta_n = 200$ nm (see Fig. 3.9).

Fig. 3.11. Evolution of the normal and tangential stressed calculated at the nodes located at the blister boundary during delamination (see Fig. 3.10). The blister pressure is the running parameter with direction as indicated. The maximum normal and tangential traction components at pure loading mode are shown with the broken line.
3.5 Comparison of analytical and finite element model

3.5.1 Stresses

For the sake of comparison first the situation of an infinitely strong cohesive zone \((G_c \rightarrow \infty, \Delta_n \rightarrow 0\) and \(\sigma(\Delta_g) \rightarrow \infty)\) is chosen and the same elastic coating \((E = 2\ \text{GPa}, \ \nu = 0.33\ \text{and} \ t = 40\ \text{µm})\). In this case, both models should behave similarly. Nevertheless, the boundary conditions of the models are different: clamped (thin plate) or not clamped (FEM), and therefore comparison of both models is not possible at the crack-tip. Nevertheless, each graph contains the stresses according to both the FEM and the analytical thin plate model (see Eq. (3.9)-(3.11)).

The largest stress in the system is \(\sigma_y\) (Fig. 3.12). Even close to the boundaries, the elastic model is able to predict the stresses very well. In practice, this stress is only indirectly responsible for delamination. The easiest way for delamination is a mode I fracture \((\sigma_y)\). The less preferred mode II \((\sigma_{yz})\) due to the asymmetry at the crack-tip is also important. In the analytical model, both stresses are zero at the crack-tip. Still, comparison of the shear stresses shows that the models yield similar results (Fig. 3.13). An interesting aspect is the existence of stresses outside the boundary conditions of the analytical model.

Fig. 3.12. Comparison of the tensile stresses \(\sigma_y\) between (left) analytical model and (right) finite element model.

Fig. 3.13. Comparison of the shear stresses \(\sigma_{yz}\) between (left) analytical model and (right) finite element model.
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3.5.2 Shape

In the analysis of the experiments it is convenient to fit the blister shape with the equation \( w(y) \) (Eq. (3.8)) to determine \( b \) and \( H \). It is found that blister shapes resulting from the finite element model can be fitted very well with Eq. (3.8). Nevertheless, a couple of aspects are noteworthy.

Fig. 3.14 shows \( b/b_0 \) as a function of the pressure, where \( b_0 \) is the initial applied width. Due to the geometry of the blister and the measurement location at the free polymer surface, \( b/b_0 \) is slightly larger than 1. With increasing blister height (and pressure) this error increases somewhat. Since \( G \propto b^{-2} \), this results in an underestimation of \( G \) at least 12% for a 1 mm blister. Fig. 3.14 indicates that in general the width of the blister is overestimated by roughly the thickness \( t \) of the coating. Because the shape of the blister is measured at a distance \( t \) from the inner side of the blister, it is reasonable to express the overestimation in terms of the thickness.

Fig. 3.15 displays the height \( H \) as a function of the pressure \( p' \). At first sight, the analytical model overestimates \( H \). In the range of validity (indicated in the graph) of the analytical model (\( H \leq 0.3t \)) both models yield the same results. For smaller blisters, the heights will be smaller and therefore the comparison between FEM and the analytical model will be focused on blisters with an initial width \( b_0 = 0.5 \) mm.

Fig. 3.16 shows two different interpretations of a single a typical FEM result. The smooth curve is calculated from a fit of the surface according to Eq. (3.8). The other curve is taken from the cohesive zone. To determine which part of the coating was still attached, the following fracture criterion was used: \( \Delta_s > 2\Delta_h \). The smooth curve gives an overestimation of the width (see also Fig. 3.14). The other curve shows two stages.
the first stage, the pressure increases inside the blister while the width is constant. Beyond a certain pressure, fracture starts and the width of the blister increases (indicated by the open circle).

Fig. 3.16 gives a description of the behavior of a blister. Using Eq. (3.27), each width-height pair can be converted to a work of adhesion $G$ (Fig. 3.17), which is only valid when fracture takes place. The difference in analysis of the results leads to different values of $G$. The fitting technique leads to an underestimation of $G$ and the cohesive-zone values result in an overestimation of $G$. Independent of the technique, a constant $G$ is not found.

This suggests that the measured curves for $b$ should be corrected for the thickness of the layer to give $b' = b - t$. The calculations of $b'$ are shown here to give a much better estimate of $G$.

![Fig. 3.16. Two different types of analysis of the numerical results of $H(b)$ with $\delta_n = 200$ nm and $G = 2.5$ J/m$^2$. (-) extracted from the cohesive zone and (--) using a fit of the surface.](image)

![Fig. 3.17. The work of adhesion $G$ according to Eq. (3.27) for the curves shown in Fig. 3.16 and (--) for the corrected width $b'$. For the determination of $G$, the kink marked by the open circle.](image)

### 3.6 $G_{CZ}$ versus $G$

In this section the analytical model will be used to “measure” $G$ for blisters simulated with the finite element model. The aim is to compare this “measured” value of $G$ with the value of $G_{CZ}$.

Several parameters determine the value of $G$: the interaction length of the cohesive zone, the geometry of the blister and $G_{CZ}$ of the cohesive zone. Combining all different sources, both the validity window and the quality of the analytical model can be investigated.

The following values will initially be used: $h_0 = 1$ mm, $\delta_n = 200$ nm, $G_{CZ} = 2.5$ J/m$^2$ and $t = 40$ $\mu$m. Each parameter except $t$ will be changed. In Fig. 3.16
and Fig. 3.17, the results of various “measuring” techniques are shown. In this section, the cohesive zone technique is used (fracture criterion $\Delta_n > 2\Delta_0$), because this technique is able to give a unique value of $G$. In both figures, a kink in the curve reveals the initiation of fracture. This condition is used to define the initiation of delamination. In the following figures, an envelope function is added to express the applied $G_{cz}$. In the ideal situation, the measurements (black squares) should be on the envelope.

The first analysis concerns the initial width of the blister. Fig. 3.18 shows a decreasing $G$ as a function of the width of the blister. The thin plate theory should show the best agreement for $H < 0.3t$, which implicitly leads to better agreement for small blisters. However, the smallest blister does not show the best agreement. This is the result of the applied fracture criterion, which implies a certain crack opening. For a small blister, the width might be influenced, resulting in a larger $G$. On the other hand, the error in the estimate of $G$ is less than 20%.

Fig. 3.18. The “measured” $G$ for blister sizes ($b_0$). The dashed line indicates the $G$ of the cohesive zone.

Fig. 3.19 shows the influence of the cohesive zone definition. The following aspects can be inferred from the figure. For large $\Delta_0$, $G$ increases as a function of $\Delta_0$. The smallest $\Delta_0$ value is not obeying the linear relationship. Both responses can be explained by the stresses at the crack-tip, which have the following relationship for constant $G$:

$$\sigma(\Delta_0) \propto \Delta_0^{-1}$$  \hspace{1cm} (3.33)

For $\lim \Delta_0 \to 0$, infinite stresses appear and adhesive failure is impossible. For $\lim \Delta_0 \to \infty$, the fracture criterion ($\Delta_0 > 2\Delta_0$) is never met. Both limits result in an infinite value of $G$. Between both extremes, a minimum measured $G$ would be reasonable. Fig. 3.19 supports this argument with its minimum at $\Delta_0 = 100$ nm. The maximum error in the measured $G$ is less than 10%.
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The last and most important aspect is the sensitivity of $G$ to variations in $G_{CZ}$. Fig. 3.20 shows a remarkable sensitivity of $G$ for small values of $G_{CZ}$. With our analysis of the experimental results we are capable to estimate $G_{CZ}$ within an error of 20%.

In summary, within the used window and combining all errors, the analytical model is able to quantify $G$ within an error of 20% – 30%.

Fig. 3.19. The “measured” $G$ as a function of the interaction length $\delta_*$ of the cohesive zone (see e.g. Fig. 3.9). The dashed line represents applied $G_{CZ}$.

Fig. 3.20. The “measured” $G$ as a function of the applied $G_{CZ}$.

3.6.1 Simulated plastic deformation

In the previous section, the simulations are elastic and have a maximum opening stress below the yield stress of the coating $\sigma(\Delta_0) < \sigma_{yeld}$. In many applications, this assumption could be violated. This section will show what happens when $\sigma(\Delta_0) > \sigma_{yeld}$.

The simulations will be performed on a PET coated steel with a $\sigma_y = 51$ MPa at a strain $\varepsilon = 6\%$. A complete stress-strain curve is shown in Fig. 2.18. For this specific case, the cohesive zone is defined by: $G_{CZ} = 15, 20, 25 \ and \ 30 \ J/m^2$, $\sigma(\Delta_0)/\max(\sigma(\Delta_0)) = 0.75$, $\sigma(\Delta_0) = 30, 40, 50, 60, 70, 80 \ and \ 90 \ MPa$. The coating thickness is chosen at $t = 30 \ \mu m$ and the initial width $b_0 = 1 \ mm$.

This specific case can be solved only with the finite element model. First, the coating has a smaller elastic modulus (823 MPa instead of 2 GPa). Second, the coating is thinner (30 $\mu m$ instead of 40 $\mu m$). Third, the coating adheres better to the substrate. Fourth, plasticity appears in the coating. All arguments lead to a higher blister for the same pressure and therefore they are in conflict with the thin plate criteria. The following graphs will support this.

Fig. 3.21 shows the height of the blister as a function of $G_{CZ}$ for different maximum normal opening stresses $\sigma(\Delta_0)$. All curves increase as a function of $G_{CZ}$. Each height and width combination can be converted to a $G$ value according to Eq. (3.27) (Fig.
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3.22). The agreement for small $G$ in the previous section (Fig. 3.20) is not found in the simulations presented here. There are several reasons for this behavior, which results $H \gg 0.3r$. For larger deflections, the neutral axis will be strained and eq. (3.27) will give an underestimate.

Fig. 3.21. $H$ as a function of $G_{cz}$ for different values of $\sigma(\Delta_h)$ from bottom to top: (▼) 30, 40, (▲) 50, 60, (●) 70, 80 and (■) 90 MPa.

Fig. 3.22. $G$ as a function of $G_{cz}$ for different values of $\sigma(\Delta_h)$ from bottom to top: (▼) 30, 40, (▲) 50, 60, (●) 70, 80 and (■) 90 MPa.

Fig. 3.23 and Fig. 3.24 give another representation of the data of Fig. 3.21 and Fig. 3.22. These figures are added to show the influence of the maximum opening stress $\sigma(\Delta_h)$ at the crack-tip. The height of the blister increases with increasing $\sigma(\Delta_h)$. The stresses in the coating are related with the height of the blister. Therefore, if the blister is high enough, it will delaminate. Fig. 3.23 supports this argument and Fig. 3.24 is a logical outcome. One important aspect should be stressed. According to Fig. 3.21 and Fig. 3.23, it is not possible to define for a certain width-height combination a unique combination of $G$ and $\sigma(\Delta_h)$. For each single $H$, different $G_{cz}$ values are possible.

Fig. 3.23. $H$ as a function of $\sigma(\Delta_h)$ for different values of $G_{cz}$: (○) 15, (●) 20, (□) 25 and (■) 30 J/m$^2$.

Fig. 3.24. $G$ as a function of values of $\sigma(\Delta_h)$ for different $G_{cz}$: (○) 15, (●) 20, (□) 25 and (■) 30 J/m$^2$. 
Fig. 3.25 shows the von-Mises stress at the crack-tip of a highly pressurized blister. The coating starts to neck at the crack-tip. This preferred position is due to the shape local bending. Therefore, this weak spot will fracture cohesively for well adhering specimen.

Fig. 3.25. The Von-Mises stress at the crack-tip of the highest blister of Fig. 3.21
\( G = 30 \, \text{J/m}^2 \) and \( \sigma(\delta_c) = 90 \, \text{MPa} \).

3.7 Experiment, analytical solution and FEM

Here we will make a comparison between the experimental results and numerical analysis [11]. Fig. 3.26a presents the contour plot of the work of adhesion calculated with Eq. (3.27) using a coating thickness of \( t = 40 \, \mu\text{m} \). As the intensity of the laser pulse increases the blister dimensions, blister height \( H \) and width \( b \), should take the following trend. At low laser intensities, when no delamination occurs, the width of the blister is constant and equal to the image of the opening in the mask projected on a sample (see Fig. 3.1). The height of the blister increases gradually with the intensity of the laser beam. As the laser intensity increases the pressure inside the blisters increases and eventually the condition for delamination described by Eq. (3.18) is satisfied. Both the blister width and the height increase and follow one of the contour lines given by Eq. (3.27). Since the work of adhesion is a characteristic property of the interface, all blisters, independent on their dimensions will follow one of the contour lines during delamination. This behavior is observed in Fig. 3.26a, where different series are indicated by different symbols. In order to cover a wide range of the blister dimensions, a series of measurements has been carried out with different sizes of the mask. Most of the data points covering a wide range of blister widths (from 700 to 1400 µm) are found between the contour lines corresponding to 2 and 3 J/m². It can be also observed that as the blister width increases the data points tend to show a lower work of adhesion.
Fig. 3.26. a) Different sets of measurements (symbols) presented in the blister dimensions height $H$ and width $b$. The figure shows also contour lines, which are defined by Eq. (3.27). The broken line is taken from (b). b) Several finite element simulations with the same conditions as the experiment with $G = 3 \text{ J/m}^2$, similar to Fig. 3.17.

The shape of a number of blisters with various initial dimensions has been simulated with the FEM code. The blister pressure is increased gradually until delamination takes place. The corresponding blister dimensions are given in Fig. 3.26b, where the contour lines calculated with Eq. (3.27) are also shown. To compare the simulations with the experimental results the work of adhesion of the cohesive zone is taken at $G = 3 \text{ J/m}^2$. The broken line indicates the critical dimensions of the blisters when delamination is initiated. It is quite interesting that the FEM and the thin plate model show fair agreement with each other.

From a comparison between Fig. 3.26a and b it could also be observed that the results obtained with FEM calculations are more in line with the experimental results than the thin plate model. This can be seen in Fig. 3.26a by comparing the experiments of $b = 1300 \mu \text{m}$ with those of $b = 800 \mu \text{m}$. Taking into account that only during fracture the contour lines have physical meaning, the case of $b = 800 \mu \text{m}$ shows a significantly larger $G$. Comparing these results with the broken line, much better agreement has been found. The reason is that the concepts of the thin plate model used in the elastic description are valid under the condition that $H < 0.3t$ [12], where $t$ is the plate thickness. In case of the 40 $\mu \text{m}$ thick polymer coating used in this work the blister height is limited to $H = 12$ $\mu \text{m}$, which is less than the experimentally observed blisters. Therefore the FEM results show much better agreement with experiments for the highest blisters.
3.8 Conclusion

A Laser Induced Delamination method for the study of adhesion of polymer coatings on metal substrates is analyzed. This technique is suitable for measuring the adhesion of laminated materials without elaborate pre-treatment.

Linear elastic thin plate theory has proven to be sufficient for the determination of the work of adhesion $G$. For coatings with $G \leq 5 \text{ J/m}^2$ and $b = 1 \text{ mm}$, good agreement between elastic thin plate theory and the FEM is found. The approximate error of $G$ is about $20\%–30\%$.

The limitations of the elastic model show up when the blister height is larger than the values in the range $H < 0.3t$, i.e. the thin plate condition. When the coating adheres well to the substrate, it is shown that the stresses in the coating go beyond the yield stress of the coating. This situation results in plastic deformation of the coating prior to delamination. The plastic deformation is concentrated near the crack-tip.

Well adhering coatings cannot be characterized with the thin plate theory. For $G_{cd} \geq 15 \text{ J/m}^2$ and $b = 1 \text{ mm}$, a large discrepancy occurred between the elastic model and the FEM. Furthermore, it was not possible to find a unique combination of $G$ and high maximum adhesion stresses for each height-width combination.

3.9 References

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