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Numerical–experimental assessment of roughness-induced metal–polymer interface failure

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ABSTRACT

A numerical–experimental method is presented to study the initiation and growth of interface damage in polymer–steel interfaces subjected to deformation-induced steel surface roughening. The experimentally determined displacement field of an evolving steel surface is applied to a numerical model consisting of a polymer coating and interface layer. The measured displacement field is obtained with a Finite Element based Digital Image Correlation method.

The resulting simulations provide novel insights into the mechanical behaviour of the polymer–steel interface during interface roughening. The appearance of local hills and valleys on the evolving steel surface results in local bands of intensified stress in the polymer layer. These localized deformation bands trigger interface damage, which grows as the surface deformation increases. Polymer ageing initially delays the initiation of interface damage. However, for increased polymer ages the average interface damage increases. Likewise, the critical strain, at which the interface integrity is locally compromised, decreases.

The presented method allows for a detailed study of the interface integrity during deformation-induced steel surface roughening. With properly identified material parameters, it becomes possible to tailor the polymer–steel material properties to minimize interface damage during production and storage of cans or canisters, e.g. for food and beverages.

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1. Introduction

1.1. Metal–polymer laminates

In recent years, metal–polymer laminates of electrolytic chromium coated steel (ECCS) sheets coated with a

Abbreviations: ECCS, electrolytic chromium coated steel; FE-DIC, Finite Element based Digital Image Correlation; RD, rolling direction; TD, transverse direction; DRD, deep-draw, deep-redraw; EGP, Eindhoven Glassy Polymer.

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polymer layer (see Fig. 1(a)) have become increasingly popular for packaging of food and beverages. Producing cans or canisters from these pre-coated steels offers several advantages over the traditional production method. In the latter, a can is made from blank steel sheet after which it is lacquered with a protective coating on the inside and a decorative coating on the outside. Compared to this traditional production process the use of pre-coated packaging steel leads to a reduction of energy consumption and CO₂ emission with one third. Moreover, the process water used and the resulting solid wastes are reduced to practically zero (Van der Aa et al., 2000).

The use of polymer coated steels entails a number of challenges, since the coating is applied before the actual

Fig. 1. (a) Different material layers in a polymer coated ECC steel (after Van den Bosch et al., 2008) and (b) experimentally observed interface damage (after Boelen et al., 2004).

can forming process. This implies that the coating undergoes the same deformation steps as the ECCS substrate, i.e. deep (re-) drawing (DRD) and wall-ironing. Large deformations are induced at high strain rates, pressures and temperatures. During this production process, the coating must fully adhere to the ECCS substrate. Boelen et al., 2004 have shown experimentally that damage is introduced at the interface during production and sterilization, see Fig. 1(b). After product fabrication, damage is often not visible for the human eye, however it becomes apparent during the prolonged shelf-life of the product. Application for food packaging demands a material that does not exhibit any visible or even measurable corrosion on the inside, even after a relatively long shelf-life period (Boelen et al., 2004; Van den Bosch et al., 2008).

Several authors studied the influence of deformation on the adhesion between the polymer coating and the steel substrate. Boelen et al., 2004 and Van den Bosch et al., 2008 performed peel-off experiments and found that the bonding between the polymer and steel is reduced by (pre) deformation of the steel substrate. Laser-induced delamination experiments by Fedorov et al., 2007 showed a similar result. Recent results from Faber et al., 2014 revealed the presence of interface damage in DRD materials by exploring the interface in cross-sections using Focused Ion Beam milling. These papers suggest that the deformation-induced roughening of the interface is the cause of the loss of adhesion and possible delamination.

1.2. Challenges

It is well known that a polycrystalline metal, such as ECCS, roughens at its free surface when deformed, due to

crystallographic differences between grains and other plasticity related phenomena. An example of the deformation-induced surface roughening for an ECC steel in tension is shown in Fig. 2. The initial height profile (Fig. 2(a)) reveals the presence of an initial rolling profile (grooves along the y direction, i.e. in the rolling direction (RD)). Tension was applied along the x direction (transverse direction (TD)). Upon deformation, the initial profile transforms to a new roughness profile during the tensile test (Fig. 2(b)). Clearly, this roughening occurs over a wide range of length scales, ranging from the scale of individual dislocations to the size of multiple grains. The resulting profile depends on the deformation conditions and the detailed material properties (Raabe et al., 2003).

The roughening phenomenon occurs at the polymer-steel interface during can production, which may result in interface damage. To predict the deformation and potential damage of these polymer-coated steels, an appropriate model has to be developed that incorporates the micro-scale phenomena governing the interface roughening.

In previous research, Van Tijum et al. (Van Tijum et al., 2007; Van Tijum et al., 2007) studied the influence of roughening on the interface properties by numerically generating a roughness profile. While these simulations give valuable insight into the influence of the out-of-plane displacements that can be expected during deformation of a polymer-steel interface, they do not include local in-plane deformations. Furthermore, the numerically generated surface profiles only resemble the average values of experimental measurements. Van den Bosch et al., 2008 also studied the effect of a change in interface roughness by including a roughness parameter, based on the Root-Mean-Square (RMS) roughness, in the adopted exponential

Fig. 2. Initial and final surface profile of an ECC steel during a uniaxial tensile test, measured with a confocal optical profiler; X direction is the tensile direction; colors indicate the local surface height in microns.

cohesive zone law. This method reduced the work-of-separation of the cohesive zone elements as a function of the applied pre-deformation, but did not include the actual physical process of roughening. Hence, while these studies provide valuable knowledge on the effect of the change in interface roughness on the interface integrity, they do not incorporate the actual full-field displacement field that accompanies the roughening phenomenon.

In this paper, a numerical–experimental methodology was developed to study the effect of deformation-induced steel surface roughening on the polymer–steel interface. A methodology to experimentally quantify the three-dimensional displacement field of an evolving surface profile was recently developed and discussed in Van Beeck et al., 2014. A Finite Element based Digital Image Correlation (FE-DIC) methodology was developed to extract the full-field displacements of an evolving surface. The FE-DIC method is applied here to obtain the full-field surface displacements of an ECC steel deformed in tension. The displacement field is applied to a two-dimensional plane strain simulation of a polymer-coated steel. As a result, the steel does not have to be modeled explicitly, since the measured experimental displacements are directly applied to the interface. A sketch of the computational model is shown in Fig. 3. The PET coating is modeled using the parameters for a different PET grade to predict realistic PET behaviour. The parameters for the interface are close to the expected real parameters.

The paper is organized as follows. The experimental methodology to extract the full-field surface deformations is explained in Section 2. The constitutive and computational models are discussed in Section 3. The results of the simulations are given in Section 4 and discussed in Section 5.

The following notations are adopted throughout the paper. A second-order tensor is denoted by $\mathbf{A}$. The inner product is defined as $\vec{a} \cdot \vec{b} = a_i b_i$, $i = 1, 2, 3$. The transpose of a tensor is denoted by $\mathbf{A}^T$.

2. Steel surface deformation

Quantitative predictions of the interface integrity during deformation-induced steel surface roughening requires either (a) a steel constitutive model that accurately predicts the roughening process; or (b) the quantified full-field displacement fields that accompany the roughening phenomenon. Since it is very difficult to accurately simulate the roughening process with an advanced constitutive model for the steel, we will here rely on the experimentally

quantified displacement field of the steel surface as it evolves. The method is outlined below.

2.1. Experimental

Point of departure is the newly developed methodology for characterizing an evolving surface profile in previous work (Van Beeck et al., 2014). A Finite Element based Digital Image Correlation (FE-DIC) technique enables extraction of the three-dimensional surface displacements from measured sequential surface height profiles, taken from confocal optical measurements. The methodology is applied here to quantify the surface displacements during roughening of a packaging steel.

The studied polymer-coated steel is TH340, which is a continuously annealed, single reduced, DWI (draw-redraw-wall ironing) quality Aluminum-killed low carbon ferritic, electrolytic chromium coated steel (ECCS) sheet. The steel layer is 210 μm thick and is lacquered with a protective coating on each side. The coating is a mixture of PET and additives to improve the adhesion to the steel (Van den Bosch et al., 2008). The thickness of the coating is 15 μm on one side of the ECCS and 25 μm on the other side.

Tensile samples (20 $\times$ 4 mm), with the transverse direction (TD) aligned with the tensile axis, were produced by simultaneously milling stacks of 5 samples to reduce edge effects. Prior to the tensile test, the polymer coating was removed in a two-step process using N-Methyl-2-Pyrrolidone (NMP). The NMP was first heated to 80 $^{\circ}\text{C}$ to remove the bulk coating. The coating residuals were removed at 130 $^{\circ}\text{C}$. Removing the coating is essential to improve the accuracy of the confocal height measurement. It can be readily assumed that the absence of the thin compliant coating does not effect the steel roughening process.

Tensile testing was performed on a Kammrath & Weiß-micro tensile stage with a 500 N load cell. The strain rate was set to $5 \times 10^{-4} [\text{s}^{-1}]$. *In-situ* measurement of the surface profile was carried out with Sensofar PL μ 2300 confocal optical profiler using a 50x objective. The experimental setup is shown schematically in Fig. 4. The image pixel dimensions were 332 nm $\times$ 332 nm. The height measurement accuracy is approximately 20 nm. Because the FE-DIC calculations require the difference between two height profiles to be small, an incremental method was used. A new height profile was measured after each strain increment ($\Delta\epsilon = 0.25\%$), during which the progressive changes in height were small (Van Beeck et al., 2014).

The mechanical response of TH340 revealed that the material forms a Lüders band, i.e. plasticity initiates locally

Fig. 3. Sketch of the computational framework, u_{ip} is the in-plane displacement field and u_{op} the out-of-plane displacement field.

Fig. 4. Sketch of the experimental test setup.

on one side of the tensile sample, which extends towards the other side, after which the entire sample deforms plastically. The globally applied engineering strain is then $\varepsilon \approx 5\%$. Note that the local yield strain is much smaller ($\varepsilon_l \approx 0.1\%$). Failure of the material occurs at $\varepsilon \approx 21\%$.

The measured surface height profiles at the start and end of the tensile experiment are shown in Fig. 5(left). Three-dimensional profiles are shown in Fig. 2. Due to Lüdering, plasticity initiates within the measured region after a relatively large global (engineering) strain of $\varepsilon \approx 3.5\%$. A new roughness pattern is formed on top of the existing initial roughness (see Figs. 5(a) and (c)) and it grows upon further deformation, see Fig. 5(e).

From the measured surface height profiles, the full-field displacement field was extracted using FE-DIC with a linear triangular discretization. The discretization size was based on the optimum of the verification analysis by Van Breeck et al. (2014), i.e. approximately 450 pixels per triangular element. The images are analyzed in a sequential manner, i.e. the first and second image are first correlated, followed by the second and third image and so on. The FE-DIC displacements at the start and end of the experiment are shown in Fig. 5(right). Comparison of the discretization at the start (Fig. 5(b)) and end (Fig. 5(f)) shows a positive strain in the x (tensile) direction and a negative strain in the y direction. The roughening process is clearly dominated by local deformation mechanisms, as the deformation is inhomogeneous throughout the surface.

2.2. Surface displacements

Upon closer investigation of the measured displacement fields, small fluctuations between consecutive displacement steps can be identified. These fluctuations are likely due to the adopted incremental calculation procedure, measurement noise and small-scale roughening

Fig. 5. (left) Surface height profiles measured at different strains; (right) the corresponding correlated FE-DIC displacements; the black line is a central deforming line initially parallel to x ; all values are in microns, the tensile (x) direction is horizontal and the colors indicate the local surface height; red square and diamond on the black line are used in Fig. 6. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

events that are not captured by the adopted discretization (Van Beeck et al., 2014). The experimentally acquired displacement fields therefore still require filtering, since these fluctuations may cause non physical behaviour if used directly in numerical simulations.

Singular Value Decomposition (SVD) is an effective tool to filter out spurious fluctuations while preserving the essential kinematics (Geers et al., 1996). Here, SVD was applied to the in-plane displacement fields (u_x and u_y) and the out-of-plane displacement field (u_z). The singular values are truncated when the singular values are less than 1% of the largest singular value. The reconstructed displacement fields sufficiently captured the roughening kinematics while eliminating nonphysical fluctuations at the scale of interest. The unfiltered and filtered displacements in two points are shown in Fig. 6. The fluctuations are especially visible in the out-of-plane displacements (Fig. 6(b)) as the amplitude of the out-of-plane displacements is considerably smaller than the in-plane displacements.

In the two-dimensional plane strain computational model, two scalar displacement fields are prescribed to the polymer–steel interface, i.e. a parallel in-plane displacement and an out-of-plane displacement along a line profile on the initial surface, here a line parallel to the tensile direction. While this line is initially aligned with the tensile direction, it may not stay aligned, due to the three-dimensional nature of the roughening process, e.g. see the line shown in Fig. 5(right). Therefore, the scalar in-plane displacement parallel to the line (u_{ip}) is extracted from the u_x and u_y displacement fields by determining the length change of each line segment, i.e.

$$u_{ip,i} = l_i - L_i, \quad i = 1 \dots n, \quad (1)$$

where n is the number of line segments and L_i and l_i are the previous and new segment length, respectively, and can be determined via the line position and the calculated displacement fields. This is schematically shown in Fig. 7.

The out-of-plane displacement field does not require a projection and is equal to u_z . After applying SVD and determining the in-plane displacement field, the evolution of the black line in Fig. 5(right) can be calculated. The evolving surface profile is shown in Fig. 8. The evolution clearly shows that the steel surface is elongated in-plane. Furthermore, since plasticity initiates locally at an increased strain rate due to Lüdering, a jump in the surface height profile is calculated (see Fig. 8). Finally, the initiation and growth of hills and valleys along the line profile is also visible. It is this evolving surface profile that will be prescribed to the two-

dimensional polymer–steel interface model to study the mechanical response of the polymer coating and interface.

3. Constitutive and computational model

In this section, the material models for the polymer and interface are presented, along with their numerical implementation.

3.1. Polymer model

To accurately predict the mechanical response of the PET, the Eindhoven Glassy Polymer (EGP) model is used. Previous research has shown that the EGP model adequately captures the mechanical behaviour of amorphous polymers (Van Breemen et al., 2011; Klompen et al., 2005). The typical mechanical behaviour of amorphous glassy polymers is shown in Fig. 9(a). During a compression test, the polymer first behaves nearly linear elastic, followed by non-linear visco-elastic behaviour. When the yield point is reached, depending on the polymer age, a softening regime sets in. The softening is overtaken by strain hardening at higher strains. The yield point depends on the applied strain rate. Note that PET is not fully amorphous, but rather semi-crystalline. However, the crystallinity of the used PET coating is small, about 8%. The EGP model is a multi-mode, multi-process polymer constitutive model (Van Breemen et al., 2011). In the present work only one mode and one process is considered, giving a single-mode, single-process approximation (SM-SP). The mechanical analogue of the SM-SP EGP model is shown in Fig. 9(b). The essential equations of the SM-SP EGP model are detailed below.

The EGP model splits the total stress in two contributions, i.e. the driving stress σ_s and the hardening stress σ_r (see also Fig. 9(b)),

Fig. 7. Sketch of the projection method; (top) two-dimensional evolution of the line profile; (bottom) evolution of the projected line.

Fig. 6. Displacements of two points (see red square and diamond in Fig. 5); raw FE-DIC displacements are the dashed lines; displacements after SVD are the solid lines; note that the difference between the unfiltered and filtered in-plane displacements in (a) is negligible. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 8. Evolution of the line profile initially located at $y = 100 \mu\text{m}$.

Fig. 9. Typical mechanical behaviour of amorphous polymers and the mechanical analogue of the SM-SP EGP model (after Van Breemen et al., 2011).

$$\sigma = \sigma_s + \sigma_r. \quad (2)$$

The hardening stress contains the rubber elastic contribution of the oriented entangled network and is described by a neo-Hookean relation,

$$\sigma_r = G_r \tilde{\mathbf{B}}^d, \quad (3)$$

where G_r is the strain hardening modulus and $\tilde{\mathbf{B}}^d$ the deviatoric part of the isochoric left Cauchy-Green deformation (Finger) tensor. The driving stress incorporates the intermolecular interactions and is decomposed in a hydrostatic (σ_s^h) and a deviatoric stress (σ_s^d), i.e.

$$\sigma_s = \sigma_s^h + \sigma_s^d = \kappa(J - 1)\mathbf{I} + G\tilde{\mathbf{B}}_e^d, \quad (4)$$

where κ is the bulk modulus, J is the volume change ratio, $\mathbf{I}$ is the unity tensor, G is the shear modulus and $\tilde{\mathbf{B}}_e^d$ the deviatoric part of the elastic isochoric Finger tensor. The evolution of J and $\tilde{\mathbf{B}}_e^d$ is calculated from

$$\dot{J} = J \text{tr}(\mathbf{D}), \quad (5)$$

$$\dot{\tilde{\mathbf{B}}}_e = (\tilde{\mathbf{L}} - \mathbf{D}_p) \cdot \tilde{\mathbf{B}}_e + \tilde{\mathbf{B}}_e \cdot (\tilde{\mathbf{L}}^T - \mathbf{D}_p). \quad (6)$$

Here $\mathbf{L}$ is the velocity gradient tensor and $\mathbf{D}$ the deformation rate tensor. The plastic deformation rate tensor, $\mathbf{D}_p$ is related to σ_s^d via a non-Newtonian flow rule,

$$\mathbf{D}_p = \frac{\sigma_s^d}{2\eta}, \quad (7)$$

with the viscosity

$$\eta = \eta_{0,\text{ref}} \frac{\bar{\tau}/\tau_0}{\sinh(\bar{\tau}/\tau_0)} \exp\left(\frac{\mu p}{\tau_0}\right) \exp(S), \quad (8)$$

where $\eta_{0,\text{ref}}$ is the zero-viscosity defined according to the reference state, τ_0 is the characteristic equivalent stress and μ is a pressure dependency parameter. The total equivalent stress, $\bar{\tau}$ and the pressure, p , are defined as

$$\bar{\tau} = \sqrt{\frac{1}{2} \sigma_s^d : \sigma_s^d}, \quad (9)$$

$$p = -\frac{1}{3} \text{tr}(\sigma). \quad (10)$$

The state parameter S incorporates the thermodynamic history dependency and is related to the equivalent plastic strain, $\bar{\epsilon}_p$, i.e.

$$S(\bar{\epsilon}_p) = S_a \cdot R(\bar{\epsilon}_p). \quad (11)$$

Here S_a captures the initial state of the polymer. R ranges between 0 (rejuvenated) and 1 (fully aged). In the current implementation the time dependence of S_a , i.e.

physical ageing, is not included. Therefore, the yield stress does not increase with time and only the effect of mechanical rejuvenation can be predicted with the current model.

The equivalent plastic strain rate is defined as

$$\dot{\bar{\epsilon}}_p = \frac{\bar{\tau}}{\eta}. \quad (12)$$

The softening function, $R(\bar{\epsilon}_p)$, describes the influence of the strain on the polymer age. Following (Klompén et al., 2005), the softening function is described by a modified Carreau-Yasuda function,

$$R(\bar{\epsilon}_p) = \frac{(1 + (r_0 \cdot \exp(\bar{\epsilon}_p))^{r_1})^{(r_2-1)/r_1}}{(1 + r_0^{r_1})^{(r_2-1)/r_1}}, \quad (13)$$

where r_0 , r_1 , r_2 are fitting parameters.

The material parameters for the PET coating used in this work are listed in Table 1. The parameters were determined by Poluektov et al., 2013 for an amorphous PET grade. These parameters may not be fully exact for polymer-coated steels, e.g. TH340, but they do predict realistic PET behaviour. The corresponding mechanical response during a uniaxial tensile test for an aged ($S_a = 13.3$) and rejuvenated ($S_a = 0$) PET layer is shown in Fig. 10.

3.2. Interface model

The interface between the PET and ECCS is modeled using Cohesive Zone elements. The modified Xu and Needleman exponential traction-separation law developed by Van den Bosch et al., 2006 is used to describe the adhesion between the PET and steel. The law for normal (n) and tangential (t) traction is defined as

$$T_n(\Delta_n, \Delta_t) = \frac{\phi_n \Delta_n}{\delta_n^2} \exp\left(-\frac{\Delta_n}{\delta_n}\right) \exp\left(-\frac{\Delta_t^2}{\delta_t^2}\right), \quad (14)$$

$$T_t(\Delta_n, \Delta_t) = 2 \frac{\phi_t \Delta_t}{\delta_t^2} \left(1 + \frac{\Delta_n}{\delta_n}\right) \exp\left(-\frac{\Delta_n}{\delta_n}\right) \exp\left(-\frac{\Delta_t^2}{\delta_t^2}\right), \quad (15)$$

where Δ is the cohesive zone opening, ϕ is the work of separation and δ is the characteristic opening length. This is a coupled interfacial law, as the traction depends on both opening directions. As shown by Van den Bosch et al., 2006, the coupling ensures a realistic work of separation during multi-directional opening, see also Fig. 11. The maximum reached traction in one direction decreases with prior opening in the other direction.

Because strain reversal may occur during roughening, an unloading behaviour must be included in the cohesive zone model. Van den Bosch et al., 2007 introduced two types of unloading, elasticity-based damage and

Fig. 10. Mechanical response at different strain rates and PET ages (S_a) during a uniaxial tensile test for the EGP parameters of PET shown in Table 1.

elasto-plastic unloading. Here, elastic damage unloading is assumed, see also Fig. 12(c), i.e. during unloading of the cohesive zone (points 2 to 3) the traction decreases linearly back to zero, i.e. with a degraded stiffness.

The interface damage is quantified by an integrity parameter. The total energy ($\Phi = \Phi_t + \Phi_n$) stored in the cohesive zone during opening is calculated stepwise between each deformation increment, i.e.

$$\Phi_n^{i+1} = \Phi_n^i + \delta\Phi_n = \Phi_n^i + \int_{\Delta_n^i}^{\Delta_n^{i+1}} T_n|_{\Delta_t=\Delta_t^{i+1}} d\Delta_n, \quad (16)$$

$$\Phi_t^{i+1} = \Phi_t^i + \delta\Phi_t = \Phi_t^i + \int_{\Delta_t^i}^{\Delta_t^{i+1}} T_t|_{\Delta_n=\Delta_n^{i+1}} d\Delta_t, \quad (17)$$

where i is the increment number and $\Delta_j^{i+1} = \Delta_j^i + \delta\Delta_j$, $j = n, t$. Note that the current opening in the other direction (e.g. Δ_t^{i+1} for Φ_n^{i+1}) is used. This definition requires that the incremental change of the cohesive opening is small. Not all energy is dissipated in the cohesive zone. The stored energy which is returned during unloading is

$$\Psi_i = \frac{1}{2} T_j(\Delta_{j,max}) \Delta_{j,max}, \quad j = n, t. \quad (18)$$

Here, elastic damage unloading is assumed and $\Delta_{j,max}$ is the (absolute) maximum opening reached in the loading history. The total dissipated energy is then

$$\Omega = \sum_{i=n,t} \Phi_i - \Psi_i. \quad (19)$$

The integrity is then defined as the fraction of energy that can still be dissipated, i.e.

$$\xi = 1 - \frac{\Omega}{\phi}, \quad \xi \in [0, 1], \quad (20)$$

where it is assumed that $\phi_n = \phi_t = \phi$. An example of the evolution of the interface integrity during multi-directional opening using the parameters in Table 2 is given in Fig. 12. First, a cohesive zone element is opened and partly closed in normal direction. Hereafter, the element is opened in tangential direction until the traction is almost zero. Fig. 12(d) shows that the integrity never increases and drops to zero when the tangential traction becomes negligible, indicating complete interface failure.

The constants for the interface model have to be determined via dedicated experiments. It is known from literature that different values for the work of separation ϕ may result for different experiments at different scales,

Table 1
Material properties of PET for the EGP model.

G_r [MPa]	κ [MPa]	G [MPa]	$\eta_{0,ref}$ [MPa · s]	τ_0 [MPa]
4.7	1800	812	$3 \cdot 10^8$	1.262
μ	S_a [–]	r_0 [–]	r_1 [–]	r_2 [–]
$4.8 \cdot 10^{-2}$	13.3	0.98	20	–3.5

Fig. 11. The coupled exponential traction-separation law of Van den Bosch et al., 2006, using the parameters of Table 2.

Fig. 12. Evolution of the interface integrity during multi-directional opening; (a) cohesive zone element at the different steps of opening; (b) prescribed cohesive zone opening; (c) resulting normal and tangential tractions, including unloading (2-3); (d) evolution of the interface integrity parameter ξ ; the used cohesive zone parameters are listed in Table 2.

see e.g. Fedorov et al., 2007 versus Van den Bosch et al., 2008. These differences result from the dissipated energy adjacent to the interface (in the process zone), which is typically lumped in ϕ of the cohesive zone. In the present work, small-scale interfacial phenomena are of interest, and values for ϕ have to be characteristic for de-adhesion only. The adhesion energy determined by Fedorov et al., 2007 is used here. The interface parameters are listed in Table 2. Note that here $\phi_n = \phi_t = \phi$ and $\delta_n = \delta_t = \delta$.

3.3. Geometry and boundary conditions

To model the mechanical response of the polymer and polymer–steel interface as a result of deformation-induced roughening of the steel substrate, it is assumed that the steel dictates the bottom interfacial deformation. The steel is therefore not modeled explicitly.

As discussed in the introduction, a two-dimensional plane strain computational model is used, see also Fig. 3. While the full three-dimensional displacement field is available for the numerical simulation, it is computation-

ally expensive to simulate a 3D situation. It is therefore assumed that the displacements perpendicular to the modeled line profile are small, see also Fig. 8.

For the 2D plane strain state considered, the out-of-plane (u_{op}) and in-plane (u_{ip}) displacements are prescribed to the interface elements, see also Fig. 8. The simulated time frame is identical to the experimental one. To simulate more realistic strain rates, the loading time can be reduced. However, the surface roughening mechanisms may also change when the strain rate is increased.

The geometry of the computational model is shown in Fig. 3. In this model, the bottom of the interface has the topology of the initial surface profile obtained from the undeformed height profile. This initial surface profile is complex and therefore a mix of quadrilateral 4-node and triangular 3-node elements is used for the polymer layer. The left and right boundaries are kept straight during the simulation. A mesh convergence study was performed to determine the optimal element size. The cohesive elements have an initial size of 100 nm.

4. Results

4.1. Polymer response

The equivalent Von Mises stress and equivalent plastic strain in the PET coating at a global engineering strain of $\varepsilon = 10\%$ and $\varepsilon = 20.75\%$ are shown in Figs. 13 and 14, respectively. The displacements of the line initially located on $y = 100 \mu\text{m}$ (see also Figs. 5 and 8) are prescribed to the interface elements.

The results reveal the localized stress and strain bands in the PET coating due to the local variations of the steel surface displacements. Interface damage is clearly visible near the regions of high stress/strain in the PET coating, see arrows in Fig. 13. The local nature of the roughening phenomenon results in localization bands with high stresses at the interface. Damage typically initiates at or near these high stress locations. Localization is triggered by the softening branch in the polymer behaviour (see also Fig. 9) (Van Melick et al., 2003). This local necking of the PET coating leads to an accumulation of strains in this region with interface damage as a result. This necked region is initially small, but grows as the deformation is increased due to strain hardening of the PET coating. This

Table 2
Cohesive zone parameters used in the simulations.

ϕ_n [Jm^{-2}]	ϕ_t [Jm^{-2}]	δ_n [μm]	δ_n [μm]
2.5	2.5	0.25	0.25

Fig. 13. Predicted equivalent Von Mises stress and equivalent plastic strain in the polymer coating along the line initially located at $y = 100 \mu\text{m}$ at a globally applied tensile strain of $\epsilon = 10\%$; the failed interface elements are colored black; visible interface damage is indicated by the arrows.

Fig. 14. Predicted equivalent Von Mises stress and equivalent plastic strain in the polymer coating along the line initially located at $y = 100 \mu\text{m}$ at a globally applied tensile strain of $\epsilon = 20.75\%$; the failed interface elements are colored black; visible interface damage is indicated by the arrows.

growth is accompanied by further opening of interface. At $\epsilon = 20.75\%$, the interface is almost fully delaminated, see Fig. 14. The high stresses in the delaminated PET coating near the left boundary are a result of the applied boundary condition.

Next, a rejuvenated PET coating was studied, i.e. $S_a = 0$. By rejuvenating the polymer coating, the softening branch in the mechanical behaviour vanishes (see also Fig. 10). The stress and strain within the rejuvenated polymer at $\epsilon = 20.75\%$ are shown in Fig. 15. Note that the colors represent the same levels of stress and strain as in Figs. 13 and 14. It is clear that the stresses within the PET coating are now smaller and less localized. Only small stress and strain bands appear and no necking is yet seen. Furthermore, no large interface damage is visible. Some minor interface damage can be identified (see arrow in Fig. 15) due to high stresses and strains at the interface as a result of the initial surface profile and the applied displacement field.

Fig. 15. Predicted equivalent Von Mises stress and equivalent plastic strain in the rejuvenated polymer coating along the line initially located at $y = 100 \mu\text{m}$ at a globally applied tensile strain of $\epsilon = 20.75\%$; the failed interface elements are colored black; visible interface damage is indicated by the arrows.

Clearly, the polymer age influences the scale of the predicted damage events due to the softening branch that is formed at increased polymer ages. Furthermore, the formation, stabilization and growth of the localization band results in delamination of the coating. Growth of this band further damages the interface. Thus, the rate at which the interface damage grows depends strongly on the polymer age.

4.2. Interface integrity

Figs. 13–15 allow for a qualitative comparison of damage only since small-scale damage events are not resolved in the discretization used. To clarify the interface damage in these simulations, the interface integrity ξ is studied. The interface integrity at $\epsilon = 10\%$ and $\epsilon = 20.75\%$ is shown in Fig. 16 for both the rejuvenated and aged PET coating. Both profiles show that the interface integrity decreases locally during deformation, but the reduction for aged PET is more severe. More locations along the line profile show a large reduction in interface integrity at a low strain and the integrity becomes zero for most of the interface at large strains. The rejuvenated PET also reveals a local reduction in interface integrity, but the damage is clearly less severe, indicating that damage is delayed due to the absence of strain softening in the PET.

4.3. Parameter sensitivity

The polymer age greatly influences the predicted interface damage. To further study this influence, the polymer age parameter S_a is varied between 0 and 17.5 to study the effect on the predicted interface damage along the line initially located at $y = 100 \mu\text{m}$. The average interface integrity ξ_{av} as a function of the global engineering strain and the polymer age is shown in Fig. 17. The average interface integrity is calculated by averaging the interface integrity of all interface elements at each increment. The difference in interface integrity for low values of S_a is initially small. For $S_a < 5$ the average integrity decreases slightly with S_a . However, for $S_a > 5$, the integrity rapidly

Fig. 16. Interface integrity profile at $\varepsilon = 10\%$ and $\varepsilon = 20.75\%$ for the rejuvenated and aged PET over the line initially located at $y = 100 \mu\text{m}$.

decreases with S_a and the applied engineering strain. The localization band that forms for aged PET drastically increases the size of the delaminated area (see also Fig. 16), thereby lowering the average interface integrity. It can be seen that the simulations were unable to converge in cases where the interface integrity drops rapidly towards zero.

The average interface integrity provides valuable insight on the global influence of polymer ageing. However, this parameter does not indicate when the interface integrity locally drops below a certain value. Descending below a threshold value for the interface integrity is an indication of local interface failure. To study this parameter, 10 different line profiles were simulated and the average global engineering strain was calculated at the moment when the integrity drops below 10%. This critical strain ε_c is an indication for local crack nucleation at the interface. Again, the polymer age parameter is varied to study the effect of ageing.

The result of this analysis is shown in Fig. 18. Note that the average values were obtained using only those line profiles that actually reveal a drop below the critical value of the interface integrity. Two trends can be noticed: (1) the critical strain slightly increases to a maximum around

Fig. 17. Average interface integrity as a function of the globally applied strain and the polymer age for the line initially located at $y = 100 \mu\text{m}$.

$S_a = 2.5$ (this point changes between individual line profiles); (2) beyond this value, the critical strain decreases as S_a increases. The initiation of interface damage is dependent on two phenomena, i.e. (a) the prescribed roughness evolution and (b) localization due to strain softening. For rejuvenated PET, the localization is determined by the initial surface profile and its evolution. For relatively young PET coatings, mild localization occurs and the localization tends to spread more before the critical integrity is reached in the localization band, delaying the initiation of interface damage compared to rejuvenated PET. This explains the upward trend in Fig. 18. However, when the polymer age is increased further the interface at the localization band is already damaged before the band spreads, due to the high stresses within the localized region. Further increase of the polymer age results in a decrease of the critical strain. The explanation of this trend is found in the competition between strain softening and hardening (Van Melick et al., 2003). Strain hardening stabilizes the localization phenomenon, but this effect is decreased since an increased polymer age also entails an increased yield stress. The stabilizing effect of strain hardening is illustrated in Fig. 19. Note that only a single line profile is shown, i.e. the line initially located at $y = 88 \mu\text{m}$ (the line located at $y = 100 \mu\text{m}$ did not show a drop below 10% integrity for low values of S_a). The hardening modulus G_r was increased to study the effect of the strain hardening branch. The optimum value for the polymer age shifts with the hardening modulus. Increasing the hardening modulus increases the optimal S_a value and vice versa, indicating that the presence of strain hardening delays the localization instability to higher values of S_a .

5. Discussion

The simulations predict the initiation and growth of interface damage as a result of the prescribed evolution of the steel surface profile.

The presence of interface damage in these materials was experimentally verified by Faber et al., 2014. They explored cross-sections of the deformed polymer-steel interface in DRD cans. An example of the experimentally identified interface damage is shown in Fig. 20. The experiments revealed that the polymer coating locally delaminates during can production. The damage events are

Fig. 18. Influence of the polymer age parameter on the critical strain ε_c when the integrity locally drops below 10%; bands indicate the standard deviation between the different line profiles passing this threshold.

Fig. 19. (a) Mechanical response of PET for two values of the strain hardening modulus G_r ; (b) corresponding predicted critical strain at which the local integrity first drops below 10% for the line profile initially located at $y = 88 \mu\text{m}$.

Fig. 20. Experimental results of the deformed polymer–steel interface after can production for a pre-coated packaging steel; results obtained via Focused Ion Beam milling (reproduced from Faber et al. (2014); with kind permission from Springer Science + Business Media B.V.).

small and typically occur near highly deformed regions of the steel surface, indicating that deformation-induced steel surface roughening plays an important role in the initiation of interface damage. These experiments verify the presence of damage, however they do not reveal the evolution of the interface roughness or the damage initiation and hence only allow for a qualitative comparison with the simulations. To the best of our knowledge, the *in-situ* observations required for quantitative comparison are not available and challenging to obtain at a micron scale or smaller.

Finally, quantitative prediction of the interface integrity during forming operations requires properly identified parameters for the particular PET grade used. The mechanical response of a thin PET coating with additives to promote adhesion has been studied by Van den Bosch et al., 2008. They performed tensile experiments on a PET coating after the steel substrate was chemically removed. These experiments may be used to identify the model parameters. Furthermore, a truly quantitative study of the interface damage may require the full three-dimensional displacement field, which has to be coupled to a three-dimensional computational model.

6. Conclusion

A numerical–experimental methodology has been presented, using a Global Digital Image Correlation technique combined with a computational model of a polymer

and interface layer. The presented method allows for a detailed study of the mechanical behaviour of a polymer–steel interface during deformation-induced steel surface roughening. The method incorporates the experimentally obtained displacement field into a numerical framework to predict the initiation and growth of interface damage and hence outperforms the use of average surface roughness characteristics.

The numerical predictions show that the change in surface height profile of the steel and the displacements that accompany this process can cause the polymer–steel interface to locally delaminate. Polymer ageing increases the damage and triggers an initial increase, followed by a decrease of the critical strain at which the interface integrity locally drops below a certain value. An optimum for the polymer age is found, where the competition between strain hardening and softening delays interface failure. These results provide valuable insight into the required processing and storage conditions in industrial applications, where these polymer-coated steels are used, e.g. for food and beverage cans. These applications demand that no interface damage is present, even after prolonged shelf-life.

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