Modeling the effect of rate and geometry on peeling and tack of pressure-sensitive adhesives

I.K. Mohammed, M.N. Charalambides*, A.J. Kinloch

Department of Mechanical Engineering, Imperial College London, South Kensington Campus, London SW7 2AZ, UK

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ABSTRACT

A model is developed for predicting separation along interfaces of pressure sensitive adhesives. Many authors have used the cohesive zone approach to solve such problems but the parameter calibration of such models remains uncertain. This study reports a novel method for determining such parameters. In addition, it provides crucial evidence for the suitability of the cohesive zone model approach in modelling interface fractures.

Peel tests were performed at various rates using specimens which consisted of a polyester backing membrane supporting an acrylic pressure-sensitive adhesive (PSA) adhered to a polyethylene substrate. Interfacial separation of the PSA from the polyethylene substrate was observed. Finite element (FE) peeling simulations were conducted which modeled the backing-membrane as an elasto-plastic power-law material, the adhesive as a viscoelastic material and the interfacial properties with a cohesive zone model (CZM). The material properties of the backing membrane and the pressure-sensitive adhesive were measured from tensile and stress relaxation experiments. The rate-dependent CZM parameters were measured directly from probe-chip probe-tack tests which were performed at pull-off speeds which corresponded to the rates employed for the peel tests. The effect of the PSA thickness and test rate on both tack and peel was investigated experimentally, as well as modeled numerically. Good agreement was found between the experimentally measured and numerically predicted peel forces for different peel angles, speeds and PSA thicknesses. In addition, it was proven that the rate dependence observed in the peel and probe-tack data was dominated by the rate dependence of the interface properties, i.e. the time dependence of the two CZM parameters of maximum stress and fracture energy, rather than the time-dependent bulk viscoelasticity of the PSA peel arm.

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1. Introduction

Pressure-sensitive adhesives (PSA) are used in a wide variety of applications such as adhesive tapes [1], product labels, postage stamps [2,3], paper note pads, clothing [4] and transdermal patches [5–7]. The transdermal patches consist of the adhesive and the drug sandwiched between an impermeable backing membrane and a release liner, however there are five main designs which deliver drugs in different ways. These are monolithic drug-in-adhesive, multi-laminate drug-in-adhesive, liquid reservoir, polymer matrix and vapor [6,8]. The research conducted by the authors is aimed to develop single-layer drug-in-adhesive patches specifically for the human nail with fungal infections. Previously published work involved characterizing the PSA, backing membrane, PSA-substrate interface and performing peel tests with patches at multiple peel angles, which relates to the force required for removal [9]. The work presented in this paper focuses on testing and modeling the peel test at different speeds and with patches of increasing PSA thicknesses, which directly relate to pain upon removal and the drug-loading capacity, respectively. Although the work had an ultimate aim to develop a pharmaceutical patch for infected nails, the current paper focuses on adhesives with no drug applied to a high-density polyethylene (PE) substrate. Note that PE was selected as the substrate since it possessed a surface energy similar to that reported for the human fingernail plate [10]. The effect of drug-loading and changing the interface to human nails on the peel force will be reported in a future publication [11].

Typical PSA products consist of the PSA material sandwiched between a flexible backing membrane, such as a plastic film, a paper film or a metal foil, and a low surface energy release-liner which in the case of rolls of tapes is laminated to the top surface of the backing material.

* Corresponding author. Tel.: +44 20 7594 7246.
E-mail address: m.charalambides@imperial.ac.uk (M.N. Charalambides).

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Acrylic-based PSAs are bio-compatible with skin [6,12] and unlike other classes of PSAs, such as rubber and silicone, do not require the addition of tackifiers to form a good bond to a substrate surface [1]. Tack is defined as the ability of a PSA to form an instant bond when it is brought into contact with a surface. The quality of the bond is influenced by numerous factors including the surface energies of the adhesive and substrate, dwell time, contact pressure, mechanical properties of the adhesive, temperature and humidity [13]. While tack is necessary to create the bond, it is equally as important when a ‘clean’ separation of the surfaces is desirable such as in the case of drug-loaded patches [12]. The most commonly used testing methods for quantifying tack include the loop-tack [14,15], rolling-ball and probe-tack tests [16]. In the probe-tack test, a flat or spherical probe contacts the PSA surface, compressing it until a specific load is reached (the dwell force) and then the probe is held at that position for a period of time (the dwell time) [17–20]. The probe is then displaced usually at a constant speed in the direction opposite to loading until failure within the PSA or at the interfaces occurs, and the force-displacement history is recorded.

The peel test is a simple experiment in which the force required to separate two surfaces is measured and then used to calculate the energy dissipation [21–24]. The magnitude of the resulting peel force depends on variables such as the peel speed, peel angle, peel arm thickness and adhesive thickness. Modeling of the peeling process accurately is a challenge, requiring the material properties of the entire peel arm and a damage criterion to represent the mode of fracture which can be either failure of the adhesive or debonding at the interface. Numerous authors have modeled the peel test, using various failure criteria such as the cohesive zone model (CZM) [24–28], virtual crack closure [29], xfm [30] or a critical stress at a distance [31,32], but the majority of these studies were conducted at a single peeling speed and with relatively thick metallic peel arms bonded using high-modulus structural adhesives.

There is some literature on investigations involving peeling at different rates. Zhou et al. [33] proposed an extended theoretical peel zone model to determine how the peel velocity impacted the peel strength, shape of the peel zone and angle at the peel point. They tested three commercial tapes at various angles by applying a fixed load to the peel arm to achieve a constant velocity. The results showed that both the peel strength and peel angle increased linearly with peel velocity and the authors were able to validate their theoretical model with the experimental data. Rahulkumara et al. [34] developed a computational model for predicting fracture in viscoelastic materials when peeling at different velocities. A rate-independent cohesive zone model was used to simulate fracture and a dimensional analysis revealed that the thickness, bulk properties of the polymers as well as the cohesive zone parameters influenced the macroscopic fracture energy. However, there was a quantitative discrepancy between the experimental and theoretical peel data which was the result of the macroscopic fracture energy being dependent on the peeling rate, and thus it was concluded that a rate-dependent cohesive law was needed. Du et al. [35] performed peel tests using rubber-based PSAs, which they then modeled using the finite element (FE) software, Abaqus [36], with an elastic energy-density failure criterion to describe the interfacial debonding. Although both the numerical and experimental results gave the same shape for the force-speed master curve, the predicted peel forces were lower than the measured values as the peeling rate increased. The discrepancy was attributed to the fact that the PSA was characterized at very small strains using rheological data. Marzi et al. [37] implemented a rate-dependent bilinear cohesive zone model into the finite element code LS-DYNA to simulate the fracture of tapered double-cantilever beam (TDCB) specimens over six orders of magnitude of test velocity. The model was validated with experimental results and the rate-dependent CZM was then successfully applied to the T-peel test.

In the present work, a cohesive zone approach will be used to model the interface separation and the aim is to prove that the traction-separation law can be calibrated through appropriate, independent tests. Such evidence is lacking in the current literature and therefore will be of significant importance to the research community. Peel tests were performed at different rates using specimens which consist of a polyester backing-membrane supporting an acrylic pressure-sensitive adhesive adhered to a polyethylene substrate. The effect of the geometry of the adhesive layer was investigated experimentally and numerically. Note that the thickness of the adhesive layer in this study is comparable to the thickness of the peel arm, unlike the case of structural adhesive bonds where the adhesive layer is often very thin compared to the peel arm. The peeling model used a cohesive zone failure criterion and has the ability to calculate the peel force at different peeling rates.

The outline of the paper is as follows: firstly, the mechanical characterization techniques, material models descriptions and material constants for the polyester backing membrane and the PSA are given. The experimental poke-chip probe-tack test and peel test methods are then described followed by a description of the cohesive zone model and the finite element models of both tests. Next, the experimental and numerical results of both the probe-tack and peeling tests are presented as well as a discussion of the effect of all the variables investigated such as PSA thickness, velocity and peel angle [9]. Finally the FE peel model is used to perform parametric analyses which investigate the influence of the CZM parameters and the viscoelasticity of the PSA on the peel force. The numerical predictions are compared to the experimental data wherever possible for validation purposes.

2. Experimental

2.1. Materials

In this work, Scotchpak 9757 backing membrane and DuroTak 2852 PSA were used to make peeling samples. The backing membrane was a 20 μm thick polyester film purchased from 3 M while a self-curing acrylic PSA was supplied by Henkel in an organic solvent solution. Before peeling tests were conducted, the backing and PSA were tested individually and characterized with an elastic-plastic and visco-hyperelastic material analytical models respectively [9]. A brief description of the both material models is given below.

The polyester backing membrane was modeled using a simple elasto-plastic power-law, as stated in Eq. (1). This model allows both the initial linear-elastic region and the plastic work-hardenning region of the stress–strain curve to be expressed analytically through:

$$\sigma = \begin{cases} \frac{E\varepsilon}{\varepsilon_y} & (\varepsilon \leq \varepsilon_y) \\ \frac{E\varepsilon}{\varepsilon_y}^n & (\varepsilon > \varepsilon_y) \end{cases}$$

(1)

where is the $\varepsilon_y$ yield strain and $n$ is the power-law constant.

The PSA is a non-linear viscoelastic material, hence its constitutive response under step strain relaxation is both strain- and time-
dependent. The stress, for any strain history, can be evaluated from the Leaderman form of the convolution integral as shown below [38]:

\[
\sigma(t) = g_\infty \sigma_0(t) + \sum_{i=1}^{N} \int_{0}^{t} g_i(t-s) \frac{d\sigma_0(s)}{ds} \, ds
\]

(2)

where \( g(t) \) is the time-dependent function, is represented by the Prony series:

\[
g(t) = g_\infty + \sum_{i=1}^{N} g_i \exp(-t/\tau_i)
\]

(3)

where \( g_\infty \) and \( g_i \) are dimensionless constants, \( \tau_i \) are the relaxation times and \( g_\infty + \sum g_i = 1 \).

The function \( \sigma_0(t) \) is calculated through the Van der Waals hyperelastic potential with the following material parameters: an instantaneous shear modulus, \( \psi \), locking stretch constant, \( \lambda_m \), and the global interaction parameter, \( \alpha \) [39,40]:

\[
\sigma_0 = \frac{dW}{d\lambda} = \psi \left( \lambda - \frac{1}{\lambda} \right) \frac{\sqrt{\lambda^2 - 3} - \frac{\sqrt{\lambda^2 - 3}}{\sqrt{\lambda^2 + 2\lambda^{-1} - 3}}}{\sqrt{\lambda^2 + 2\lambda^{-1} - 3}} - \alpha \frac{\sqrt{\lambda^2 - 3}}{2}
\]

(4)

Therefore, \( \sigma(g(t)) \) represents the instantaneous stress–strain relationship, corresponding to \( t = 0 \), while \( g_\infty \sigma(g(t)) \) is the long-term stress–strain relationship corresponding to \( t = \infty \). A more detailed description can be found in the work published by Goh et al. [41]. It should be noted that Eq. (2) assumes that the strain and time effects on the stress response are separable.

Tensile tests were performed on the backing membrane, while both tensile and relaxation tests were performed on the PSA. The backing membrane was found to have an elastic modulus, \( E \), and yield stress, \( \sigma_y \), of 4.44 GPa and 70 MPa respectively, while the power-law constant, \( n \), was calculated to be 0.287. The Van der Waals hyperelastic material constants were determined as: \( \psi = 0.411 \text{ MPa}, \lambda_m = 8.56 \) and \( \alpha = 0.361 \). The Prony series parameters are summarized in Table 1 where the subscripts represent the relaxation times; these were set to range from 0.1 to 1000 s and spaced such that they were an order of magnitude apart. All material parameters for the backing membrane and the PSA were obtained from previous experiments conducted by the authors [9].

2.2. Poker-chip probe-tack tests

Poker-chip probe-tack tests were performed to determine the interface properties between the PSA and substrate, which in this case was high-density PE. The PSA was dissolved in a dichloromethane solvent to reduce the viscosity and then cast on a fluoropolymer-coated release-liner purchased from 3 M. After the solvent evaporated fully, a soft-solid acrylic PSA sheet was formed and then another release-liner was placed on top of the PSA to prevent contamination of the PSA surface. It should be noted that although this sample preparation method is relatively simple, it is difficult to control the final PSA film thickness, resulting in a range of thickness throughout the solid acrylic sheet. Circular samples of 13 mm diameter were cut from the release-liner/PSA sandwich and subsequently applied to the PE substrate. A 15.6 mm diameter steel probe, attached to a Zwick Roell Z1.0 testing machine, was brought into contact with the PSA surface as seen in Fig. 1(a). The PSA was then compressed, Fig. 1(b), to a set dwell force and held, Fig. 1(c), for a fixed dwell time before being pulled off, Fig. 1(d), at a constant crosshead speed. Upon pulling off the steel probe and the attached PSA, there was no observable fibrillation of the PSA and failure occurred at the PSA-PE interface, Fig. 1(e). The area under the resulting load-displacement curve, between stages 1(d) and 1(e), was calculated and is referred to as the tack energy, or the tack work of adhesion, \( W_a \), while the peak load was converted into a tack strength, or maximum stress, \( \sigma_{\text{max}} \), by dividing the peak load by the cross-sectional area of the PSA film. In the initial set of tests [9], the dwell force and dwell time were both varied between 1–20 N and 10–300 s respectively, in order to determine the minimum threshold value of each variable. The threshold dwell force and dwell time, defined as the values beyond which there was no increase in either the tack strength or tack energy, were found to be 10 N and 60 s respectively. Probe-tack tests were performed at pull-off speeds of 1, 10 and 100 mm/min. For each parameter which was varied, between five to seven replicate tests were performed and all experiments were performed under environmental conditions of 21 °C and 50% humidity.

2.3. Peel tests

Peel test specimens were prepared by casting the acrylic PSA onto the polyester backing membrane and allowing the dissolved solvent to evaporate. The release-liner was placed on the PSA to protect its bonding surface and the assembled tape was cut into 15 mm wide and 80 mm long specimens for the peel tests. The release-liner was subsequently removed from the surface of the PSA, the latter having an average thickness of 250 μm. Approximately 40 mm of the tape length was then applied to the PE substrate using a roller, of mass 2.5 kg and diameter 60 mm, which ensured that a high enough dwell force was achieved to produce complete bonding. The free-end of the peel arm was fixed to a tensile grip on a Zwick Roell Z1.0 mechanical testing machine. The PE substrate was attached to a 80 mm × 40 mm IKO precision linear slide with a stroke length of 47 mm, which ensured that a constant peel angle was maintained during the test. Peel tests were performed at a constant peel angle of 90° and constant speeds of 1, 10 and 100 mm/min. A schematic of the peel test attached to the roller and the testing machine is shown in Fig. 2. At each speed, the peel test was repeated four times and all experiments were performed under environmental conditions of 21 °C and 50% humidity. The resulting peeling force history at each constant speed and angle was recorded.

3. Numerical

3.1. Cohesive zone model

In this section both the probe-tack and peel tests were modeled using the finite element (FE) software, Abaqus [36]. The backing membrane and the PSA were described by elastic-plastic and visco-hyperelastic material models respectively, and the material parameters are given in Section 2.1. A rigid body was used to represent the PE substrate and a failure criterion was implemented.
at the PSA-PE interface as was observed experimentally. In this work a cohesive zone model (CZM) was used with a bi-linear traction-separation law as shown in Fig. 3. The penalty stiffness value, $k$, used was $5 \times 10^{11}$ Pa/m, which was sufficiently high to ensure that the compliance at the interface was negligible. The other two parameters, namely the fracture energy, $G_0$, and maximum stress, $\sigma_{\text{max}}$, were assumed to be equivalent to the experimentally measured tack work of adhesion, $W_\text{tack}$, and tack strength, respectively (see Section 2.2). This traction-separation curve was used for both normal and shear failure modes. Note that previous simulations showed that the Mode II effect in peeling was minimal [9].

### 3.2. Poker-chip probe-tack model

The probe-tack test was simulated with a 2D axisymmetric finite element model. The probe and substrate were modeled as analytical rigid bodies while the PSA was modeled with the visco-hyperelastic material model presented in Section 2.1. A tie constraint was applied between the probe and the PSA while cohesive contact was implemented at the substrate-PSA interface. The substrate was fixed while the probe was given a displacement boundary condition to match the experimental pull-off speeds. As already mentioned, the CZM parameters used were taken from the experimental probe-tack tests (see Section 3.1). A mesh convergence study was performed, from which it was determined that the minimum element size needed was 50 $\mu$m.

### 3.3. Peeling model

A 2D simulation of the peel test with linear, non-reduced integration, plane strain elements was performed. The entire assembly consisted of two parts: an analytical rigid-body representing the PE substrate and a 2D deformable body for the peel arm, which was then partitioned into the polyester backing membrane and the PSA adhesive components. The backing membrane and the PSA were modeled using the elastic–plastic and visco-hyperelastic material models detailed in Section 2.1. A CZM was implemented at the interface between the PSA and the PE substrate, to simulate interfacial failure, as was indeed observed experimentally. The free end of the peel arm was displaced in the required loading direction, in this case 90° to the horizontal, while the rigid PE substrate was restrained both horizontally and vertically.

In the present work, cohesive contact was used instead of cohesive elements. Previous peeling simulations, not presented here, showed that for a given traction-separation law, both methods gave identical results. However, cohesive contact was an easier and a more logical approach to implement because failure via peel crack growth was observed to occur at the interface between the PSA and the PE substrate.

A mesh-sensitivity analysis, using linear elements, was performed in order to determine the optimal level of mesh refinement which was computationally efficient without compromising the accuracy of the results. This was found to be two and three elements thick for the backing membrane and the PSA, respectively, with an element length of 50 $\mu$m. This mesh density was used in all models and is shown in Fig. 4.
4. Results and discussion

4.1. Poker-chip probe-tack

Before any probe-tack experiments were conducted at variable pull-off speeds, a simulation was performed using the data from a previous experimental study [9]. In that work, the tack tests were carried out at a constant pull-off rate of 100 mm/min and using CZM parameters of $G_0 = 70$ J/m$^2$, $\sigma_{\text{max}} = 0.3$ MPa. A parametric study was performed to investigate the effect of increasing the PSA thickness, $h_a$, on the probe-tack test output. The thicknesses simulated varied between 50 and 1500 $\mu$m, and the output reaction force history was used to calculate the global stress. Fig. 5 shows the global stress–displacement from the FE model along with the input traction-separation law.

It can be seen that the stress–displacement curve closely agrees with the input traction-separation law as the PSA thickness decreases. For the thicker PSAs the maximum stress diverges from the input values of $G_0$ and $\sigma_{\text{max}}$. An indication of the additional influence of the deformation of the PSA on the global response. For the simulations with $h_a$ less than 300 $\mu$m, the stress–displacement responses were very similar to each other whereas as $h_a$ increased, such as at 1500 $\mu$m, the adhesive deformed without debonding at the CZM interface.

4.1.1. Experimental probe-tack results

The effect of PSA thickness, $h_a$, on both the tack work of adhesion, $W_a$, and tack strength, $\sigma_{\text{max}}$, was considered for the data in which probe-tack tests were performed with a dwell force of 10 N, pull-off speed of 100 mm/min and dwell times above the threshold value of 60 s. The thickness of each PSA sample was measured with a digital micrometer before each test. The thickness of each sample, after being applied to the PE substrate, was also measured using the Zwick Roell Z1.0 testing machine by recording the position of the probe at the point of initial contact. There was good agreement between the values measured using the two independent methods. For these samples, the thickness varied between 80 and 200 $\mu$m and no correlation was found between the thickness and either $W_a$ or $\sigma_{\text{max}}$ as can be seen in Fig. 6. It was therefore concluded that both parameters were independent of PSA thickness within the range tested which was in agreement with the results of the numerical study outlined in Section 4.1.

The pull-off speeds were then varied at 1, 10 and 100 mm/min while the initial compression speed remained constant at 0.1 mm/min (stage 1a – 1b) in Fig. 1). By performing these tests at different rates, it would be possible to obtain CZM parameters to be used in peeling simulations performed at these corresponding speeds. Fig. 7 shows that both the $\sigma_{\text{max}}$ and $W_a$ increase with pull-off speed. The average thickness of the samples tested at 1 and 10 mm/min was 250 $\mu$m but the values ranged between 230 and 260 $\mu$m. The numerical study in Section 4.1 indicated that this thickness value fell within the range in which there was no significant effect. These samples were also tested at the threshold dwell force and dwell time of 10 N and 60 s respectively. The samples tested at 100 mm/min had an average thickness of approximately 150 $\mu$m and the values of $W_a$ and $\sigma_{\text{max}}$ were calculated using a higher number of datasets (see Fig. 6) for several dwell times above the threshold value.

4.1.2. Probe-tack test contact area

For the probe-tack test, it was assumed that the probe made perfect contact with the PSA. To test this assumption, a special pressure sensitive paper film from Sensor Products Inc. was placed between the probe and PSA which was then compressed to 100 N. An example image of the film after compression is shown in Fig. 8.

The more intense red regions indicated where the applied pressure was greater and from these tests there appeared to be an uneven stress distribution on the PSA, possibly due to probe misalignment. Image analysis was performed on three different samples to measure this area. It was found to be approximately 80% of the full area of the 13 mm diameter circle. Therefore, the original
tack values shown in Fig. 7 were recalculated using this actual area and are given in the last two columns of Table 3.

4.1.3. Numerical probe-tack results

In this section, the FE model discussed in Section 4.1 is also used. The PSA thickness was kept constant and selected to be 150 μm since this value was the measured thickness of the PSA in the tack experiments conducted with a pull-off speed of 100 mm/min [9]. Fig. 9 shows the output stress–displacements obtained from the FE models at the three pull-off speeds, along with each of the corresponding input traction-separation laws. Table 2 gives the $G_a$ and $\sigma_{\max}$ values used at each speed in the CZM which result in the numerical stress–displacement responses shown in Fig. 9. From Table 2 it may be seen that the values of $G_a$ and $\sigma_{\max}$ associated with the CZM for the interface increase with increasing rate, and this arises from the viscoelastic nature of the PSA in the deformation zone at the peel front.

| Pull-off speed, $\dot{\alpha}$ [mm/min] | Input maximum stress, $\sigma_{\max}$ [MPa] | Input fracture energy, $G_a$ [J/m²] |
|----------------------------------------|------------------------------------------|----------------------------------|
| 1                                      | 0.1                                      | 20                               |
| 10                                     | 0.2                                      | 40                               |
| 100                                    | 0.3                                      | 70                               |

As can be seen, the global stress–displacement profile was not in perfect agreement with the input traction separation law with the maximum stress being somewhat less than the input $\sigma_{\max}$ for all the tack simulations. This was attributed to the deformation of the visco-hyperelastic elements which added an extra compliance to the model. These results thus implied that the tack strength and tack energy measured from the probe-tack experiments were not the true interface properties, since the PSA accounted for some of the overall mechanical response. The CZM parameters at the interface could thus be ‘corrected’ (by increasing both $G_a$ and $\sigma_{\max}$) in order to more closely reproduce the stress–displacement response obtained experimentally. These corrected pairs of values could be obtained with a numerical inverse analysis, but this correction procedure was not pursued further as the error was less than 8%. This was small compared to the 20% error in the tack parameters due to the non-uniform stress distribution, as evidenced by Fig. 8.

As a numerical case study, two pairs of CZM parameters ($G_a = 70$ J/m², $\sigma_{\max} = 0.3$ MPa and $G_a = 40$ J/m², $\sigma_{\max} = 0.2$ MPa) were implemented into the probe-tack model for each of the three pull-off speeds. The output stress–displacement graphs for a constant speed were almost identical to each other (results not shown), with less than 1% difference in the tack strength and tack energy. These results indicated that for a relatively thin PSA film, the rate-dependency observed in the probe-tack experiments was due to the rate-dependency of the CZM properties rather than the effect of strain-rate on the deformation of the bulk PSA. This effect will be investigated further with peeling simulations in Section 4.2.2.4.

The experimental probe-tack data shows a large scatter possibly due to imperfect contact but the method does show great potential if a more precisely designed test rig is used.

4.2. Peeling

4.2.1. Experimental peeling results

The steady-state forces from the peel tests were recorded in Table 3 where it can be seen that the peel force increased with crosshead speed. In earlier work, an analytical peeling model developed by Kinloch et al. [42,43] was used to calculate the fracture energy. The peel forces from the experiments performed at different speeds were used in the analytical peeling model to determine the corresponding fracture energies. As expected, the analytically calculated fracture energy, $G_a$, also increased with speed, which is in agreement with the findings of increasing $W_a$ with rate, as obtained from the probe-tack tests. However, the $G_a$ values were much higher, by 50% at the fastest speed and by 120% at the slowest speed, than the corrected probe-tack work of adhesion, $W_a$, shown in Table 3. This trend was also observed previously when peel tests were conducted at various peel angles and a fixed crack speed [9]. The analytical model was therefore not pursued further.

In Table 3, two more pairs of $W_a$ and $\sigma_{\max}$ are shown corresponding to 5 and 50 mm/min. These values were calculated through interpolation of the experimental data points at 1, 10 and 100 mm/min. The interpolations were performed by generating best fit curves which were found to be a power law and a logarithmic function for the $W_a$ and $\sigma_{max}$ respectively (fits not shown).
The interpolated data will be used to enable peel simulations at the additional speeds of 5 and 50 mm/min in Section 4.2.2.

4.2.2. Numerical peeling results

4.2.2.1. Effect of test rate. The measured tack CZM parameters, which have been corrected where appropriate as described above, are shown in Table 3, and were used as the input parameters into the peeling models for the corresponding peeling speeds. For each peeling speed, three different pairs of CZM parameters were used based on the minimum, maximum and average tack values and, hence error bars could be included for the numerical results. The peeling angle was 90° for each simulation. As shown in Fig. 10, there was good agreement between the experimentally measured and numerically predicted peel forces. At the slowest speed of 1 mm/min, the upper bound of the numerical peel force tended to more closely match the measured peel force. At 10 and 100 mm/min, the average predicted forces fell within the experimental range although there was a larger variation in the numerically-predicted force at 100 mm/min due to the bigger spread in the tack CZM parameters. These results are encouraging in showing that peeling could be modeled at different speeds using material parameters which could be measured independently for the backing, bulk adhesive as well as the PSA-substrate interface.

In Fig. 10 a power law curve is shown which was fitted to the three ‘Numerical’ points at speeds of 1, 10 and 100 mm/min. The numerical forces from the peel simulations at 5 and 50 mm/min, using the corresponding interpolated CZM parameters in Table 3, are shown in Fig. 10 for comparison. As can be seen, these two points closely follow the trend predicted by the power law, giving extra credibility in both the experimental data and modeling methods.

4.2.2.2. Effect of peel angle. Peel tests were previously performed and modeled at three peel angles with a crack speed of 100 mm/min [9]. There was good agreement between the numerical and experimental peel forces. In light of the contact area issue in the tack tests, the corrected CZM parameters of 0.35 MPa and 85 J/m² were implemented into the model. The new predicted peel forces are shown in Fig. 11. These are seen to be somewhat higher than previously reported [9] but are still in very good agreement with the experimentally measured peel forces.

4.2.2.3. Effect of thickness. A numerical investigation was undertaken to examine the effect of the geometry on the peel force. Multiple simulations were run with PSA thicknesses between 10 and 500 μm while the peel speed, angle and CZM parameters were kept constant. These values were 100 mm/min, 90°, 0.35 MPa and 85 J/m² respectively. The predicted peel forces are compared to the experimental data in Fig. 12.

Table 3

| Peel speed, \( \dot{\gamma} \) [mm/min] | Experimental peel force, \( F \) [N] | Corrected maximum stress, \( \sigma_{\text{max}} \) [MPa] | Corrected tack energy, \( W_{\text{t}} \) [J/m²] |
|----------------------------------------|-----------------------------------|------------------------------------------|----------------------------------------|
| 1                                      | 0.96 ± 0.11                       | 0.14 ± 0.04                              | 24.8 ± 7.3                             |
| 10                                     | 1.37 ± 0.22                       | 0.25 ± 0.05                              | 46.9 ± 9.0                             |
| 100                                    | 2.18 ± 0.11                       | 0.35 ± 0.10                              | 85.2 ± 34.7                            |
| 5+                                     | N/A                               | 0.22                                     | 38.4                                   |
| 50+                                    | N/A                               | 0.32                                     | 71.2                                   |

Fig. 10. The numerical and experimental steady state peel forces for different peel speeds at a peeling angle of 90°.

Fig. 11. The numerical and experimental peel forces for different peel angles at a crack speed of 100 mm/min. The \( \sigma_{\text{max}} \) and \( G_\text{c} \) values were obtained from the current probe-tack test and were 0.35 MPa and 85 J/m², respectively.

Fig. 12. The numerical and experimental peel forces for various PSA thicknesses at a peel speed and angle of 100 mm/min and 90° respectively.
The results showed that the peel force increased with PSA thickness and also confirmed that the CZM parameters were dependent only on the PSA-substrate interface, hence they are true interface material properties and independent of the geometrical configuration of the PSA over the range studied in the present work. Although the tack tests indicated that the PSA thickness had a minimal effect on the CZM parameters, there was a noticeable effect of peel force with increasing PSA thickness. This can be attributed due to the additional deformation mechanism associated with the bending of the PSA peel arm, see Fig. 2.

In order to investigate this, additional FE simulations were performed where a linear elastic model ($E = 4.44$ GPa) was used to represent both the backing membrane and the PSA. The results are shown in Fig. 13, where it can be seen that the thickness of the PSA did not have an effect on the peel force. Further by dividing the output peel force of 1.4 N by the width of the peel arm (20 mm), a value of fracture energy equal to $70$ J/m$^2$ is obtained which agrees with the input value of $G_c$ in the model and also with analytical predictions for cases where no plastic or viscous dissipations take place [19,42]. Next, the backing was set as a linear elastic material ($E = 4.44$ GPa) and the PSA was viscoelastic with the parameters given in Section 2.1. There was still found to be an increase in peel force with thickness, thus highlighting the importance of bulk viscoelastic dissipation in the PSA peel arm and its effect on the peel force. Additionally, it can be seen that when an elastic and an elastic–plastic backing membrane are assumed (with a viscoelastic PSA in both cases), the predicted peel forces were similar, thus indicating that the plastic energy dissipation in the polyester backing membrane was minimal at a $90^\circ$ peel angle. Note that this was not the case for a $135^\circ$ peel angle [9].

### 4.2.2.4. Parametric study of the CZM parameters

A further parametric study was performed by varying the CZM parameters and the peeling speed. The pairs of $G_c$ and $\sigma_{\text{max}}$ used for these simulations are given in Table 4 and the predicted peel forces are shown in Fig. 14.

As expected, for any given speed, the peel force increased when either of the CZM parameters was increased. When the pair of CZM values was kept constant and the speeds are varied, there was a minimal increase in peel force with peeling speed. This small change was due to the viscoelasticity in the PSA. The study shows that the observed rate-dependency in the peel test was dominated more by the rate-dependence of the CZM properties rather than by the bulk viscoelasticity of the PSA peel arm. This agrees with the reported results from the probe-tack simulations in Section 4.1.3. It should also be noted that as the $\sigma_{\text{max}}$ and $G_c$ values increased, the simulations at $1$ mm/min aborted since the PSA stretched without reaching the stress or energy required for crack propagation to occur at the peel front.

### 4.2.2.5. Parametric study of the PSA modulus

In order to study whether a simpler model than the visco-hyperelastic model could have been used for the PSA, an inverse analysis was undertaken. A simple linear-elastic material model, with modulus values over five orders of magnitude, was implemented into the PSA section. The resulting peel forces are shown in Fig. 15.

The peel force decreased as the material becomes stiffer, however the peel force which was closest to the experimental value corresponded to around $1$ MPa, which is the order of magnitude typically associated with PSAs [44,45] and close to three times the instantaneous shear modulus ($\psi = 0.411$ MPa) calculated in Section 2.1. Although these simulation results show that it was possible to accurately model the peeling using a simple linear-elastic material model for the PSA, it is not straightforward to directly determine what value this should be for any given rate. This

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**Table 4**  
The CZM parameters used for peeling simulations at three speeds.

| $G_c$ [J/m$^2$] | $\sigma_{\text{max}}$ [MPa] |
|-----------------|--------------------------|
| 20              | 0.1                      |
| 50              | 0.1                      |
| 50              | 0.15                     |
| 80              | 0.15                     |
| 20              | 0.2                      |
| 50              | 0.2                      |

**Fig. 13.** The numerical peel forces for various PSA thicknesses and material models at a peel speed and angle of 100 mm/min and 90° respectively, using CZM parameters of 0.3 MPa and 70 J/m$^2$.

**Fig. 14.** The numerical peel forces predicted for various combinations of CZM parameters at different speeds for a fixed PSA thicknesses and peel angle of 90°.

**Fig. 15.** The numerical peel forces predicted for different PSA linear elastic moduli for a fixed PSA thicknesses of 150 &mu;m, peel angle of 90°, peel speed of 100 mm/min.

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is because experimental data showed that, even at small strains, the stress–strain response was rate-dependent.

The peeling model was effective and from the results obtained a great deal of information was gained about the peeling process. The numerically-predicted peel forces from the simulations were in good agreement with the experimentally measured values at different peel angles, peel speeds and PSA thicknesses. Parametric studies showed that rate-dependency in the peel test was dominated by the values of $C_T$ and $\sigma_{\text{max}}$ associated with the CZM for the interface increasing with increasing rate and this arose from the viscoelastic nature of the PSA in the deformation zone at the peel front. However, the bulk viscoelastic energy dissipation in the PSA was still a contributing factor on the peel force as the PSA thickness increased and this was attributed to the additional deformation mechanism associated with the bending of the PSA peel arm. Although the probe-tack test gave reasonably accurate results, the scatter was rather high. Improvements to the test should be sought through better probe-PSA surface alignment.

5. Conclusions

Fixed arm peel tests were performed using specimens in which the peel arm consisted of a polyester backing membrane supporting an acrylic pressure-sensitive adhesive (PSA) adhered to a PE substrate. The tests were undertaken at various peel speeds, at a fixed peel angle and for a specific peel arm geometry. Additionally the thickness of the PSA layer was varied and tests were conducted (at multiple peel angles) with a constant rate of crack growth. In each case, the peel forces needed to cause crack growth along the PSA-PE substrate interface were measured.

Tack tests with a flat steel probe were performed on PSA films adhered to a PE substrate to investigate the interface properties. A previously determined threshold dwell force and dwell time were used while the pull-off speed was varied and then the force strength and work of adhesion was measured. These parameters were applied directly into the traction-separation law of the cohesive zone model used in the numerical modeling of peeling and probe-tack.

Both the peel and tack tests were simulated with the finite element method by implementing a visco-hyperelastic material model and a cohesive zone model to represent the PSA and the PSA-substrate interface respectively. The CZM parameters obtained from the tack experiments at each pull-off speed were used in the peel and tack simulations executed at the corresponding rate. The CZM can be used to model peeling, however the values are rate-dependent. The two required parameters, $\sigma_{\text{max}}$ and $C_T$, can be measured directly from probe-tack tests at pull-off speeds which match the peeling rate. The peeling model was also used for geometrical considerations and predicted the peel force for different PSA thicknesses. The predicted peel forces from the FE simulations were in good agreement with experimentally measured values at different peel angles, peel speeds and PSA thicknesses. The validation of the model against such a wide range of experimental data makes this a unique study and offers significant evidence for the suitability of the cohesive zone approach in modelling fracture in these materials. In the future, a single rate-dependent cohesive zone model should be developed which automatically changes the CZM parameters for any given peeling speed.

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References

[1] C. Creton, Pressure-sensitive adhesives: an introductory course, MRS Bull 28 (2003) 434–439.
[2] S. Zhao, R. Pelton, Peel adhesion to paper—interpreting peel curves, J. Adhes. Sci. Technol. 17 (2003) 815–830.
[3] B. Zhao, L. Anderson, A. Banks, R. Pelton, Paper properties affecting pressure-sensitive tape adhesion, J. Adhes. Sci. Technol. 18 (2004) 1625–1641.
[4] J. Zhou, W. Yu, S.P. Pfeifer, Interfaces of breast motion in sports bras: a review, Textile Res. J. 81 (2011) 1234–1248.
[5] S. Venkataram, R. Gale, Skin adhesives and skin adhesion: 1. Transdermal drug delivery systems, Biomaterials 19 (1998) 1119–1136.
[6] H.S. Tan, W.R. Pister, Pressure-sensitive adhesives for transdermal drug delivery systems, Pharm. Sci. Technol. Today 2 (1999) 60–69.
[7] R.H. Plauta, Two-dimensional analysis of peel adhesive tape human skin, J. Adhes 86 (2010) 1086–1110.
[8] L.K. Lende, N.D. Grampurohit, D.D. Gaikwad, M.V. Gadhave, S.L. JadHAV, Transdermal patches : a review, Int. J. Pharm. Res. Dev. 4 (2012) 96–103.
[9] I.K. Mohammed, M.N. Charalambides, A.J. Kinloch, Modelling the interfacial peeling of pressure sensitive adhesives, J. Non-Newton. Fluid Mech 222 (2015) 141–150.
[10] S. Murdan, C. Poojary, D.R. Patel, J. Fernandes, A. Haman, P.S. Saundh, Z. Sheikh, In vivo measurement of the surface energy of human fingernail plates, Int. J. Cosmet. Sci. 34 (3) (2012) 257–262.
[11] In vivo adhesion of novel anti-fungal ungual patches and correlations with peel tests. K. Rizi, I.K. Mohammed, K. Xu, A.J. Kinloch, M.N. Charalambides, S. Murdan
[12] F. Ciliberto, C.G.M. Gennari, P. Minghetti, Adhesive properties: a critical issue in transdermal patch development, Expert Opin. Drug Deliv. 9 (2012) 33–45.
[13] W.K. Chiang, E. Ghassemies, R. Lewis, J. Rowson, C. Thompson, Comparison of tack of pressure-sensitive adhesives (PSAs) at different temperatures, J. Adhes. Sci. Technol. 24 (10) (2010) 1949–1971.
[14] A. Kowalski, Z. Czech, L. Ryczek, How does the surface free energy influence the tack of acrylic pressure-sensitive adhesives (PSAs)? J. Coat. Technol. Res. 10 (6) (2013) 879–885.
[15] A. Kowalski, Z. Czech, The effects of substrate surface properties on tack performance of acrylic pressure-sensitive adhesives (PSAs), Int. J. Adhes. Adhes. 60 (2015) 9–15.
[16] B. Duncan, S. Abbott, R. Roberts, Measurement Good Practice Guide No. 26: Adhesive Tack, National Physical Laboratory, Teddington, United Kingdom, 1999.
[17] Y. Nakamura, K. Inamura, K. Yamamura, S. Fujii, Y. Urahama, Influence of crosslinking and peeling rate on tack properties of polyacrylic pressure-sensitive adhesives, J. Adhes. Sci. Technol. 27 (2013) 1951–1965.
[18] Y. Nakamura, K. Inamura, K. Ito, S. Nakano, A. Surooka, S. Fujii, M. Sasaki, Y. Urahama, Contact time and temperature dependencies of tack in polyacrylic block copolymer pressure-sensitive adhesives measured by the probe-tack test, J. Adhes. Sci. Technol. 26 (2012) 231–249.
[19] C. Creton, L. Leibler, How does tack depend on time of contact and contact pressure? J. Polym. Sci. Part B: Polym. Phys. 34 (1996) 545–554.
[20] P. Tordjeman, E. Papon, J.-J. Villenave, Tack properties of pressure-sensitive adhesives, Polym. Sci. Part B: Polym. Phys. 38 (1999) 1201–1208.
[21] K. Kendall, Thin-film peeling-the elastic term, J. Phys. D: Appl. Phys 8 (1975) 1449.
[22] D.R. Moore, An introduction to the special issue on peel testing, Int. J. Adhes. Adhes. 28 (2008) 153–157.
[23] D.R. Moore, J.G. Williams, A protocol for determination of the adhesive fracture toughness of flexible laminates by peel testing: fixed arm and t-peel methods. An ESIS Protocol (2010)
[24] B.R.K. Blackman, H. Hadavinia, A.J. Kinloch, J.G. Williams, The use of a cohesive zone model to study the fracture of fibre composites and adhesively-bonded joints, Int. J. Fract 119 (2003) 25–46.
[25] J.G. Williams, H. Hadavinia, Analytical solutions for cohesive zone models, J. Mech. Phys. Solids 50 (2002) 892–829.
[26] T. Diehl, On using a penalty-based cohesive-zone finite element approach, Part II: Inelastic peeling of an epoxy-bonded aluminium strip, Int. J. Adhes. Adhes 28 (2008) 255–265.
[27] Ph. Martiny, F. Lani, A.J. Kinloch, T. Pardoef, Numerical analysis of the energy contributions in peel tests: a steady-state multilevel finite element approach, Int. J. Adhes. Adhes 28 (2008) 222–236.
[28] Y. Wei, J.W. Hutchinson, Interface strength, work of adhesion and plasticity in the peel test, Recent Adv. Fract. Mech. (1998) 315–333.
[29] H. Hadavinia, L. Kawashita, A.J. Kinloch, D.R. Moore, J.G. Williams, A numerical analysis of the elastic–plastic peel test, Eng. Fract. Mech. 73 (2006) 2324–2335.
[30] R.A. Sauer, Enriched contact finite elements for stable peelings computations, Int. J. Numer. Methods Eng 87 (2011) 593–616.
[31] D. Taylor, The theory of critical distances, Eng. Fract. Mech 75 (2008) 1695–1705.
[32] N. Guli, R. Wang, A.N. Sinclair, J.K. Spelt, A calibrated finite element model of adhesive peeling, Int. J. Adhes. Adhes 23 (2003) 199–206.
[33] M. Zhou, Y. Tian, N. Pesika, H. Zeng, J. Wang, Y. Meng, S. Wen, The extended peel zone model: effect of peelling velocity, J. Adhes. 87 (11) (2011) 1045–1058.

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[34] P. Rahulkumar, A. Jagota, S.J. Bennison, S. Saigal, Cohesive element modeling of viscoelastic fracture: application to peel testing of polymers, Int. J. Solids Struct. 37 (13) (2000) 1873–1897.
[35] J. Du, D.D. Lindeman, D.J. Yarusso, Modeling the peel performance of pressure-sensitive adhesives, J. Adhes. 80 (7) (2004) 601–612.
[36] Abaqus Version 6.13. Hibbitt Karlsson and Sorensen Inc, Providence, USA.
[37] S. Marzi, O. Hesebeck, M. Brede, F. Kleiner, A rate-dependent cohesive zone model for adhesively bonded joints loaded in mode I, J. Adhes. Sci. Technol. 23 (6) (2009) 881–898.
[38] J.G. Williams, Stress Analysis of Polymers, 2nd ed., John Wiley & Sons, 1980.
[39] M.A.P. Mohammed, E. Tarleton, M.N. Charalambides, J.G. Williams, Mechanical characterization and micromechanical modeling of bread dough, J. Rheol. 57 (2013) 249.
[40] S. Tantideeravit, M.N. Charalambides, D.S. Balint, C.R.T. Young, Prediction of delamination in multilayer artist paints under low amplitude fatigue loading, Eng. Fract. Mech 112–113 (2013) 41–57.
[41] S.M. Goh, M.N. Charalambides, J.G. Williams, Determination of the constitutive constants of non-linear viscoelastic materials, Mech. Time-Depend. Mater 8 (2004) 255–268.
[42] A.J. Kinloch, C.C. Lau, J.G. Williams, The peeling of flexible laminates, Int. J. Fract 66 (1994) 45–70.
[43] I. Georgiou, H. Hadavinia, A. Ivankovic, A.J. Kinloch, V. Tropsa, J.G. Williams, Cohesive zone models and the plastically deforming peel test, J. Adhes. 79 (2003) 239–265.
[44] J.A. Williams, J.J. Kauzlarich, Application of the bulk properties of an acrylic pressure-sensitive adhesive to peeling, J. Adhes. Sci. Technol 21 (2007) 515–529.
[45] J.A. Williams, J.J. Kauzlarich, Mandrel peeling of a flexible laminate with a pressure-sensitive adhesive: comparison of experiment with numerical analysis, Strain 47 (5) (2011) 439–448.