Journal of Non-Newtonian Fluid Mechanics 000 (2016) 1–10

Contents lists available at ScienceDirect

Journal of Non-Newtonian Fluid Mechanics

journal homepage: www.elsevier.com/locate/jnnfm

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

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ARTICLE INFO

Article history:
Received 28 October 2015
Revised 11 January 2016
Accepted 25 January 2016
Available online xxx

Keywords:
Pressure-sensitive adhesive
Peeling
Finite element
Viscoelastic
Cohesive zones
Tack

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 poker-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.

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.

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http://dx.doi.org/10.1016/j.jnnfm.2016.01.016

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 peeling 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], xfem [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 font. 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.

Table 1
The Prony series parameters.

Van der Waals	$g_{0.1}$	g_1	g_{10}	g_{100}	g_{1000}
	0.740	0.044	0.099	0.046	0.038

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 poker-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-hardening region of the stress–strain curve to be expressed analytically through:

$$\sigma = \begin{cases} E\varepsilon & (\varepsilon \le \varepsilon_y) \\ \sigma_y \left(\dfrac{\varepsilon}{\varepsilon_y} \right)^n & (\varepsilon > \varepsilon_y) \end{cases} \quad (1)$$

where is the ε_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-