ANATOMY OF THE DEFORMATION OF PRESSURE SENSITIVE ADHESIVES FROM RIGID SUBSTRATES

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Relevance

Bonding. Holding. Debonding. Three mechanistic steps that define the service life of any pressure sensitive adhesive. But, do we really know how pressure sensitive adhesives (PSAs) deform? Can we anticipate how bulk and surface properties of PSAs and substrates will impact their deformation? Is there an individual physical property that allows designing pressure sensitive adhesives? And most importantly for the industrial researcher, can we really predict how physical properties and deformation will ultimately define performance?

Background

The extent of the deformation of any pressure sensitive adhesive (PSA) during debonding, which is independent if the failure occurs under tensile, shear or peel modes, has been widely recognized as a critical factor determining its capacity to instantly bond, hold a load or resist debonding. In the case of pressure sensitive adhesive tack, defined by Carl A. Dahlquist as "the ability of a material to form a bond instantaneously upon contact with a solid surface such that a substantial force is required to effect separation [1]", the property is related to a bonding step but it is traditionally quantified from a debonding process. Tack values depend on both the bulk modulus of the PSA and the degree of contact with the substrate, but it is ultimately the extent and shape of the volume being deformed that defines its magnitude. In the case of peel strength, it was also Dahlquist [2] reporting on Paul Stedry's efforts to reach a theoretical understanding of the mechanics of peeling of PSAs via stop action photographs, who pointed out that the extent of the deformation was a function of peel rates and that the evident differences in peel profiles were a result of the debonding strengths. Later, Zosel [3] reported on the role played by degree of chain entanglement to define fibrillation processes, debonding profiles and ultimately tack values. Work by Newby, Choudhury and Brown [4] demonstrated that standard 3M Scotch® Tape debonds from fluorinated substrates with a higher force compared with substrates with higher surface free energy due to interfacial slippage and by the creation of different fingering instabilities observed at the peel front. With the advent of advanced visualization techniques and instrumentation, Creton et al. [5], Shull et al. [6], and Nakamura et al. [7] have successfully demonstrated that both extent and shape of the deformation define the strength of the interface between a viscoelastic material and a substrate. Very recently, Ciccotti et al. [8] have started postulating theoretical models to help provide a measurement of the local strain response of the debonding region coupled to a cohesive zone modeling to describe what happens when PSAs deform from rigid substrates during steady-state peel experiments.

Deformation of pressure sensitive adhesive tapes. An industrial perspective.

In an attempt to decouple the three mechanistic steps of the lifetime of a pressure sensitive adhesive, namely bonding, holding, and debonding, it is necessary to acknowledge that soft, viscoelastic materials not only debond in intrinsically complex geometries but also that there is no individual bulk or surface property of the material responsible for the features observed during debonding of tape. In this report, a combination of high speed imaging with high strain bulk rheology and substrate energetics was used to identify the features of the deformation. Additionally, statistically significant relationships between bulk properties of the PSA (mainly adjusted by molecular characteristics and the rate of deformation) and surface properties of rigid substrates are discussed. All this because the extent, shape and strength of the deformation and debonding of a PSA are critical for applications of industrial relevance, such as pressure sensitive adhesion to materials with different surface free energies. The main factors affecting the deformation of a pressure sensitive adhesive tape are stiffness of backing film, angle of peel test, PSA modulus, PSA/substrate interfacial energy, substrate/environment interfacial energy, PSA/environment interfacial energy and peel rate. The features identified to be sensitive to these main factors were kinetic peak of the peel force, average peel force, fibril take-off angle, fibril take-off length and fibril center-to-center distance [Figure 1].

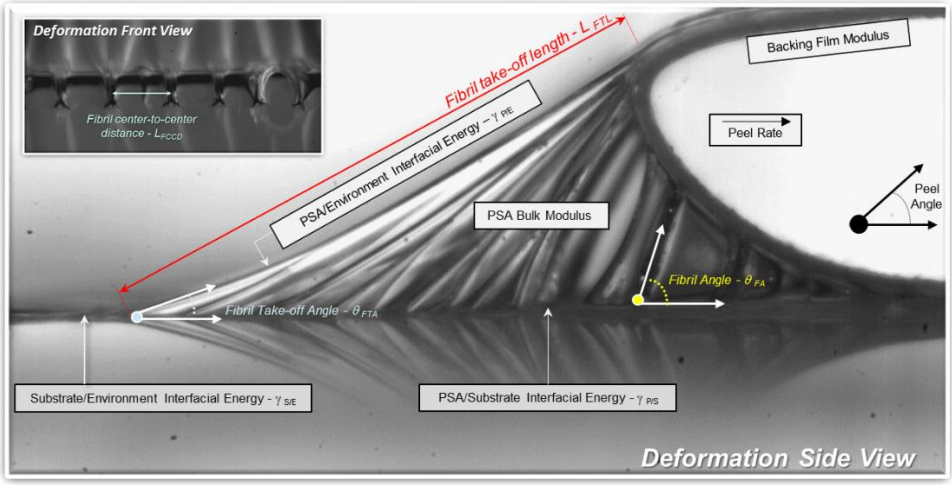

Figure 1. Factors influencing shape and extent of deformation of a pressure sensitive adhesive tape during a peel test (boxes). Relevant features measured during debonding profile experiments (colored).

Experimental

The deformation characteristics of tackified, acrylic pressure sensitive tapes composed of a poly(2-ethylhexyl acrylate)-based PSAs (75 ± 5 micrometers thick) supported on polyethylene terephthalate (PET) backing film (50 micrometers thick) are reported here. The level of crosslinking within the PSA was varied to obtain samples with high-strain modulus of 600, 1800 and 3000 kPa for “3M PSA Low X”, “3M PSA Mid X” and “3M PSA High X”, respectively. High-strain modulus was determined using a free-standing multilayer stack of each of the transfer PSAs, unsupported by any backing with a thickness of 600 micrometers. High-strain modulus was determined from the slope of the linear region typically in the stress-strain curve before the yield or fracture point. For adhesives used in this study, that region was between 10 and 20 units of engineering strain. The Elongational Viscosity Fixture (EVF) of an Ares G2 Rheometer (from TA) at room temperature with a deformation rate (Hencky

Strain) of 1 s^{-1} at room temperature (25° C) was used for this series. 0.5 inch wide x 4 inch long strips of tape were applied following ASTM D3330 to precleaned panels of stainless steel (reported surface free energy @ 20 °C = 79 mJ/m^2), polycarbonate (reported surface free energy @ 20 °C= 54 mJ/m^2) or polypropylene (reported surface free energy @ 20 °C = 29 mJ/m^2). Debonding profiles were obtained in a TA.XTplus Texture Analyzer (from Stable Micro Systems) in a 90° peel testing mode.
A series of experiments were designed to examine the effects and interactions of three key variables:

1. The adhesive high strain modulus (as dictated by its crosslinking level)
2. The peel rate
3. The substrate surface free energy

The experimental responses measured were various characteristics of the debonding shape as defined above, as well as the peel force, both the kinetic peak and the steady state value.

Results and Discussion

High speed imaging in combination with high strain rheology was successfully used to study the deformation of pressure sensitive adhesives. By imaging the peel front of model copolymers of poly (2-EHA)-based PSAs having different levels of crosslinking, relationships between the responses of peel kinetic peak (ozf/0.5 in), average peel force (ozf/0.5 in), fibril take-off angle (degrees) affected by high-strain modulus (kPa), substrate surface free energy (mJ/m2) and peel rate (in/min) were constructed. Selected response surfaces are shown in Figure 2. Only statistically significant terms ($p \leq 0.05$) were used within each of the response surfaces.

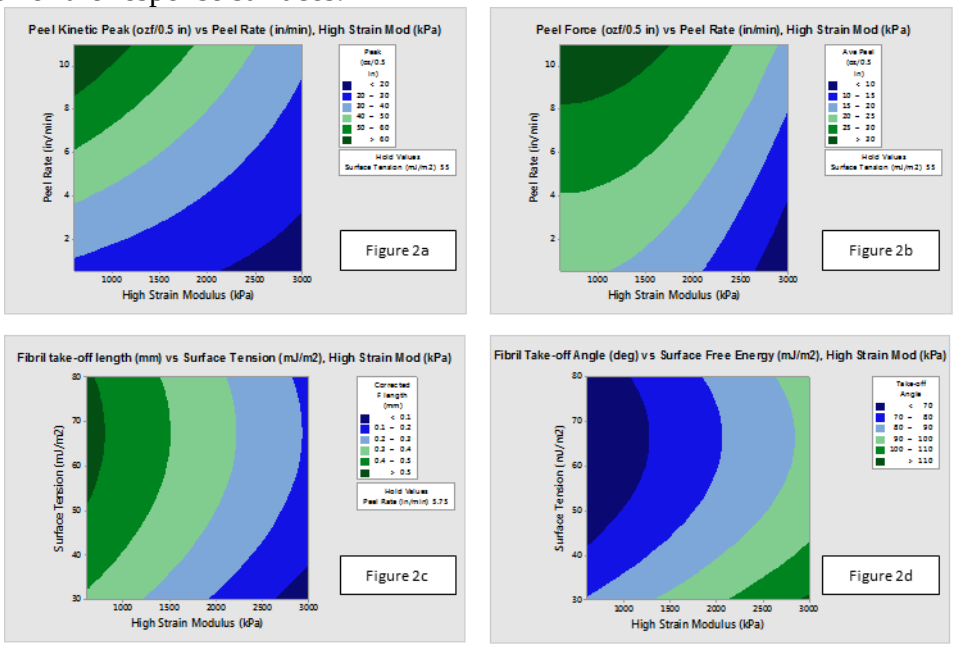

Figure 2. Selected response surfaces for a) peel kinetic peak (ozf/0.5 in), b) average peel force (ozf/0.5 in), c) fibril take-off angle (degrees) and d) fibril take-off length (mm).

- Both kinetic peak of peel and average peel force were sensitive to variations in high strain modulus, substrate surface free energy and peel rates (Figure 2a and 2b), as it will be expected for a viscoelastic material being deformed from surfaces with different levels of interaction. It is important to highlight that peel forces (both peak and average) correlated to intrinsic bulk properties of the pressure sensitive adhesives when subject to large deformations.

- Fibrils take-off length was also sensitive to all three factors: high strain modulus, substrate surface free energy and peel rates (Figure 2c). This result was also expected as the extent of the deformation will be strongly influenced by the viscoelastic nature of a highly deforming material.
- In contrast, fibril take-off angle, was insensitive to peel rates, at least for the peel rate levels presented in this report, and depended only on bulk properties and substrate surface energetics (Figure 2d). This was the feature of the PSA deformation most heavily affected by substrate energetics.
- Center-to-center fibril distance was perhaps the most convoluted property of all when imaging the deformation of PSAs and generalizations about its dependence on either PSAs or surface energetics of the substrates will be reserved for further experimental analyzes.

Conclusions

The effect of PSA's high-strain modulus, substrate surface free energy and peel rate on the deformation of the peel front of a poly(2-ethylhexyl acrylate) PSAs were investigated. With the help of high-speed imaging and the quantification of peel force profiles during debonding, evidence is presented that both substrate surface energetics as well as bulk properties intrinsic to large deformation of viscoelastic materials must be contemplated when investigating PSAs debonding phenomena. Among all the properties screened, fibril take-off angle is the most sensitive and useful to quantify the effect that substrate surface energetics play during the deformation of a PSA from a rigid substrate.

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