Detachment of stretched viscoelastic fibrils

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Abstract. New experimental results are presented about the final stage of failure of soft viscoelastic adhesives. A microscopic view of the detachment of the adhesive shows that after cavity growth and expansion, well adhered soft adhesives form a network of fibrils connected to expanded contacting feet which fail via a sliding mechanism, sensitive to interfacial shear stresses rather than by a fracture mechanism as sometimes suggested in earlier work. A mechanical model of this stretching and sliding failure phenomenon is presented which treats the fibril as a nonlinear elastic or viscoelastic rod and the foot as an elastic layer subject to a friction force proportional to the local displacement rate. The force on the stretched rod drives the sliding of the foot against the substrate. The main experimental parameter controlling the failure strain and stress during the sliding process is identified by the model as the normalized probe pull speed, which also depends on the magnitude of the friction and PSA modulus. In addition, the material properties, viscoelasticity and finite extensibility of the polymer chains, are shown to have an important effect on both the details of the sliding process and the ultimate failure strain and stress.

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1 Introduction

Pressure-sensitive adhesives (PSAs) are a class of soft materials commonly exploited on the adherent sides of transparent tape, sticky notes, and other consumer products \cite{1}. A typical PSA is composed of a blend of weakly cross-linked polymers, having both elastic and dissipative aspects. The distinctive feature of PSAs is their ability to conform to unsmooth surface topographies, by merit of their softness, and then to relax the elastic stresses induced by such surface deformations, by merit of their viscoelasticity. This concurrent attainment of a large surface area of contact and low storage of elastic energy near the interface is what makes PSAs desirable materials for temporary, low force adhesion based on physical surface interaction forces.

Although this mode of arriving at a large contact area is crucial to PSA functioning, their actual performance depends most strongly on mechanical behavior during peel off. The separation of a PSA layer from a substrate has been much studied, both experimentally and with analytic and numerical models \cite{2–5}.

The geometry of a thin layer or film of a PSA is ubiquitous in both applications and academic studies, and the most common adhesion experiments on such films are peel, shear, and tack tests (Fig. 1).

In all of these experiments, the PSA is placed under some tension, and if the adhesion to the substrate is strong enough, a large hydrostatic tension in the film leads to cavity formation and growth \cite{6}. The clearest demonstration of this is given by the tack test (see Fig. 1c), where a rigid flat cylindrical punch is brought into contact with
a thin layer of PSA. After intimate contact is established, the punch is retracted. During retraction, cavities typically nucleate at defects along the interface and expand into the film to increase the compliance of the layer in the directions both parallel and perpendicular to the applied force. Once the cavities grow to have size on the same order as the film thickness, they cannot expand further without encountering neighboring cavities. However, the walls between cavities continue to deform and eventually form a web of thin fibril-like structures that can extend to very large stretch ratios in the direction of the applied force [7, 8]. Eventually, the fibrils pull off completely from the substrate as the stretch and stress on them mounts. A similar sequence of cavity formation, growth, fibrillation, and ultimate failure is seen in the peel tests [9], although this experiment is less amenable to both precise microscopic observation and subsequent quantitative analysis.

Previous theoretical studies have provided a good basis for understanding the initial homogeneous deformation, which requires the calculation of the compliance of the confined film [10]. Others have determined the criterion which governs when defects grow into cavities due to an elastic instability [11, 12] or crack growth [12, 13]; as well as the details of cavity growth in a finite sample [3, 14, 15]. What has not been carefully studied, and the focus of this paper, is the final stage of stretching and pull-off of the fibrillar structures which develop after the cavities reach their maximum size.

The paper is organized as follows: first, we present new experimental observations that illuminate the mechanism by which the fibrils detach from the substrate. Next, we develop a model of this process and use a numerical method to solve the governing equations. We present as results the maximum strain in the fibril at pull off, as a function of the normalized pull speed and material behavior, as well as the stress in the fibril. These results are then discussed in the context of experimental observations, and we evaluate its usefulness and limitations.

2 Experimental observations

Tack experiments (Fig. 1c) were performed on a standard industrial PSA to observe in particular the final stage of fibrillar detachment. (See the Materials and Methods section for more details.) Although quantitative details of the mechanisms differ from one PSA to the other these stages are qualitatively similar for a variety of families of PSA [8, 16–18]. A microscope was used to zoom in on a single cavity at random, and the growth of the cavity and subsequent loss of contact between the PSA and the probe surface was filmed. A sequence of images from the film is shown in Figure 2. Note that after nucleation, the cavity grows quickly and uniformly in a self-similar spherical shape (images 1-3). The images shown correspond to the projection of the cavity through the glass slide. Because the cavity has a curved surface separating the inner vacuum from the surrounding PSA, its edges refract and disperse light and appear black in these reflection images.

Eventually, the top of the cavity approaches the glass slide and must deform from its spherical shape. The top of the cavity becomes flattened, allowing light to pass so that visualization of the lower contact surface is possible (images 3-12, Fig. 2). What becomes apparent is that, except for a circular central region (white regions in the images of Fig. 2), the PSA remains in contact with the substrate. That is, the contact line, which moves with the cavity during its initial self-similar expansion, arrests while the cavity continues to grow (images 4-6).

Later, as the cavity’s edges approach the edges of neighboring cavities, the walls between the cavities begin to become stretched in the tensile direction. The elastic force supported by these stretched walls is transferred to the contacting “foot” region beneath and interior to the cavity (see insert in Fig. 3 for schematics of contacting foot). As the force rises, eventually the contact line begins to move again, but at a much slower rate. It continues to move steadily toward the walls between cavities as the displacement of the probe is increased at a constant rate (images 7-12). In its wake, it leaves traces of residue on the probe surface. Eventually, when the contact line of the foot approaches the wall edge, the entire wall pulls off in a rapid catastrophic event that leaves no residue.

Since the geometry of the contacting foot is nearly the inverse of the crack geometry (Fig. 3), its failure by crack propagation is unlikely. Rather, the foot appears to slide relative to the probe surface, with resistance to sliding provided by the adhesion between the foot and probe. This shear mode of failure is supported both by the slow motion of the contact line and the presence of residue traces on the probe surface. There are alternative explanations for the details of the debonding mechanism, such as propagation of a shear crack, or deformation similar to a 180° peel near the tip of the foot. While we believe the sliding mechanism is most likely based on the experimental evidence at hand, certainty would require a careful measurement of the interfacial displacements of the foot relative to the substrate, using some sort of tracer particles, for instance. For definiteness, the sliding mechanism is supposed for the remainder of the paper.

In addition to the visual data shown in Figure 2, the total force and displacement of the entire probe was also recorded, giving a measurement for the nominal (engineering) stress and strain (stretch ratio) on the fibril (Fig. 4a). Furthermore, the motion of the foot was measured precisely from sequences of images like those shown in Figure 2 using an image analysis software. The location of the contact line (edge of the foot) as a function of time was measured along a line normal to both the contact line and neighboring wall. Typical results are shown in Figure 4b. Note that the foot edge moves at a nearly constant rate during sliding; however, this sliding speed varied significantly from sample to sample and cavity to cavity, for the same experimental conditions. This indicates that local variations in the stress on the wall are important to determining how fast foot sliding occurs, but that the overall process is similar in each case.

Based on the physical picture of the foot sliding phenomenon developed from the images in Figure 2, we will