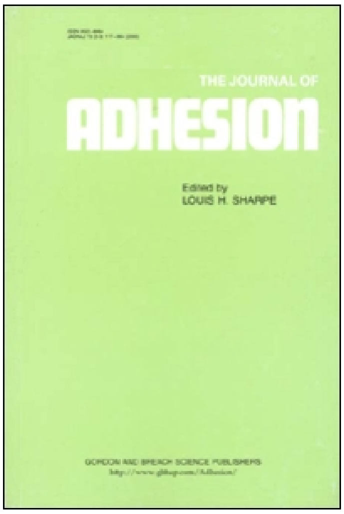

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Autographs from Peeling Fiber Reinforced Pressure Sensitive Adhesives: Correlation with Failure Mechanisms

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Autographs from Peeling Fiber Reinforced Pressure Sensitive Adhesives: Correlation with Failure Mechanisms

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In past studies, we have investigated the nature of the photon emission accompanying the peeling of pressure sensitive adhesive tapes. These studies included producing clear images of the emitted light created by direct attachment/detachment of the adhesive from photographic film (*i.e.*, autographs). Here we present an extension of this work to very slow peeling rates (<0.05 mm/s) in which we examine how the resulting autographs from fiber reinforced tape are influenced by the micro-mechanical behavior of this composite tape during high angle peeling.

KEY WORDS Pressure sensitive adhesives; fiber reinforced tape; peel test; triboluminescence; fracto-emission; photon imaging; autograph.

I INTRODUCTION

When two dissimilar materials are separated, the newly created surfaces are usually electrostatically charged. With the use of suitable probes and an electrometer, we have found that such surfaces carry net charges as well as alternating patches of both positive and negative electrostatic charge. During a fracture or peel process, oppositely charged surfaces form in close proximity, resulting in microdischarges between these surfaces. In some cases, we also find convincing evidence for discharges along the newly created surfaces. These small discharges cause the emission of photons (phE), and radiowaves (RE), as well as a variety of charged and neutral species. [These emissions are generally known as Fracto-Emission (FE)[1–4,6,7]].

Our past investigations of the emission accompanying the peeling of pressure sensitive adhesive tapes from various substrates have included pulse-counting experiments in which the phE, electron, and/or the RE bursts were detected and their size and time correlations determined,[1,2] time resolved spectroscopic analysis of the phE,[3] and direct imaging of the phE by production of autographs on Polaroid® films.[4]

These experiments verified that the phE arose predominantly from highly-localized microdischarges and when peeling took place in air, consisted of discrete molecular nitrogen lines in the blue portion of the spectrum. This N_2 radiation was observed to originate both from the vicinity of the peel and also from the surface of the tape at several mm distances from the peel line,[3] indicating that both discharges across separating surfaces in the peel zone and discharges along the surfaces resulted in gas phase excitation. In addition, it was shown that the emission rate and spatial distributions depended strongly on the conditions of peeling: namely, the substrate, the peel geometry, the peeling rate, and the gas environment.

In this paper, we focus our attention on the peeling of 3M Filament Tape in a sharp-angle peel using the autograph technique.[4] This material exhibited a wide range of instantaneous peel rates (slow-fast peel behavior) due to the mechanical properties of this composite system in a high-angle peel. Furthermore, we wish to illustrate that for this type of system, the autograph technique provides images at very low peel rates (~0.0016 mm/s), and that the observed images are intimately related to the micro-mechanical events accompanying slow, high-angle peel.

II EXPERIMENTAL METHODS

We used 3M No. 893 Filament Tape in this study. This product has an adhesive of natural rubber combined with varying amounts of a tackifying agent which is a terpene-based hydrocarbon resin. More tackifier is used on the face of the adhesive and less in the saturating layer binding the glass filaments to the backing. The back of the polyester tape carrier has been treated with a release coating having a critical surface tension for wetting of approximately 21 dynes/cm [mN/m]. The tape is 2 cm wide.

Peels from the photographic film were performed at either constant applied force or constant strain rate at a peel angle of 150° (dictated by constraints of the film holder). Constant force was supplied by hanging masses between 150–250 grams (a force of 1.5–2.5 N) on the tape's end, shown schematically in Figure 1. For the extremely slow peel rates studied here, small fluctuations in the acceleration of the mass can be neglected. Constant strain rate loading down to 0.0016 mm/s was provided by a motor-driven mechanism. In all cases, only average peel rates were measured. A force transducer was used to measure the instantaneous peel force under condition of constant strain rate. [Note in Fig. 1 the definitions of "longitudinal and transverse directions."]

Autographs [4] were recorded in the dark by peeling filament tape directly from Polaroid Type 107C film (ISO3000). The Polaroid film was developed and printed in a Polaroid camera back in the usual manner. Microscopic examination of the substrates and of the tape showed no evidence of cohesive failure.

To observe visually the slow-rate peeling process, we replace the film with a PMMA substrate and used a stereoscopic microscope at magnifications in the range of 15 to 120X. With a transparent substrate the peeling could be viewed

Schematic of Experimental Arrangement

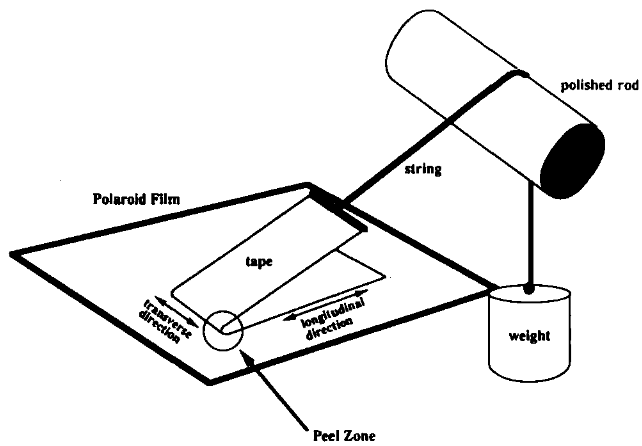

FIGURE 1 Schematic diagram of the experimental arrangement for peeling tape from a substrate under constant applied force conditions.

either through the substrate or from the edge of the tape (on an axis parallel to the peel line).

III RESULTS AND DISCUSSION

Peeling the tape from the surface of Polaroid Type 107C film at low rates produced autographs of the accompanying photon emission. Figure 2 shows an autograph of the light emission as the tape is peeled in a slow then fast sequence where slow peel was at 0.04 mm/s and the fast peel was at ~5 mm/s. The patterns of light intensity show periodic variations in both longitudinal (along the length of the tape and fiber direction) and transverse (normal to the fiber direction and

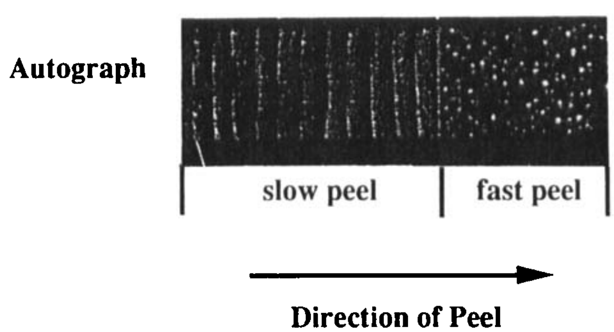

FIGURE 2 Autograph of slow then fast peel of filament tape from Polaroid® Type 107C film. Tape is 2 cm wide. Slow peel at 0.05 mm/s.

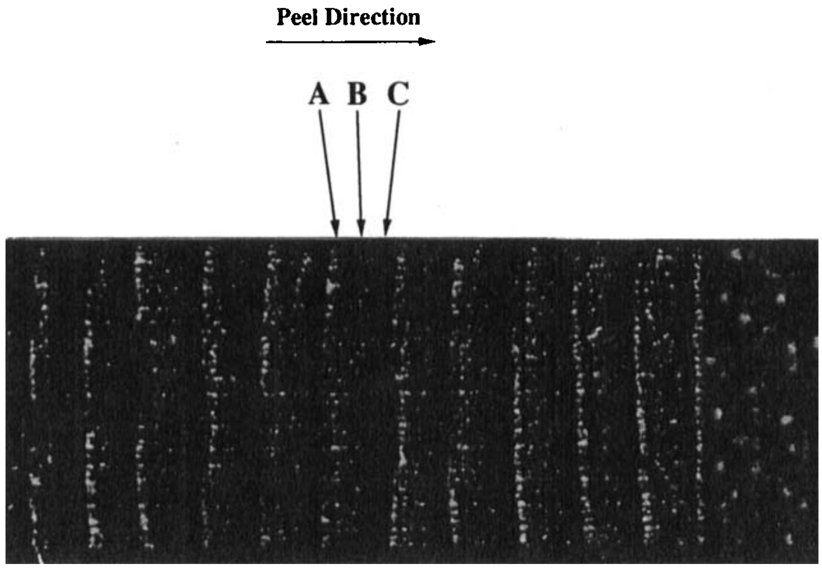

FIGURE 3 Enlargement of Figure 2.

parallel to the peel line—across the width of the tape) directions. In the rapid peel region, we see patterns similar to those reported previously,[4] namely bright spots due to localized, strong discharges with high correlation in position with the position of the fibers.

An enlargement of Figure 2 is shown in Figure 3 where the slow peel region is seen to exhibit many smaller localized discharges. The longitudinal variations in this pattern have an average "wavelength" of 3.8 mm and the transverse variations have spacings the same as distance between fiber bundles (0.54 mm). Following the very dark bands, the longitudinal bands arise showing a sharp increase in emission intensity at their leading edge (at A in Figure 3) followed by a decay in both the intensity and in the average radii of the spots. Those features near the leading edge of the high emission band have a diffuse character, while those in the lower emission region (*e.g.*, B in Figure 3) are sharper and smaller in size. Subsequently (at C in Figure 3) the phE nearly disappears from the autographs. Obviously, the mechanics of this particular sample and peel geometry are strongly influencing the spatial (and time) dependence of the phE.

The tape exhibits transverse creases after peeling at a peel angle of 150°–180° from almost any surface, including the film. This pattern of creases resembles the transverse patterns of light in autographs. Figure 4 is a comparison of an autograph with the corresponding piece of tape after peeling at an average peel rate of 0.008 mm/s. The tape backing was illuminated with a beam of light at a low angle; both tape and autograph are shown at the same magnification. As seen in Figure 4, the creases display a pattern much like the transverse lines of strong emission and the longitudinal lines in the autograph correspond to the spacings of the fiberglass bundles.

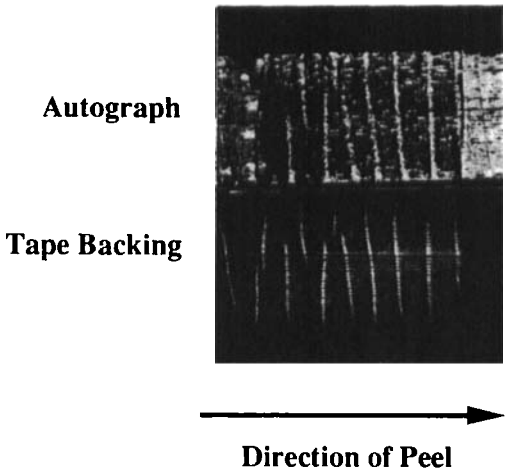

FIGURE 4 Autograph of a slow peel experiment and photograph of creases in the backing of the corresponding piece of tape. Slow peel at 0.008 mm/s.

To investigate the mechanical behavior of this system further, we peeled Filament Tape at constant strain rate from rigid glass or PMMA substrates at a peel angle of nearly 180° simultaneously measuring the force and carefully observing the peel zone. The two kinds of experimental conditions, namely peeling tape from flexible photographic film and peeling it from rigid, smooth glass or PMMA surfaces, might be thought to be incomparable. However, we observed visually that the sequence of mechanical events observed during the peeling from the photographic film (observed in light) occured in a similar fashion when tape was peeled from a rigid substrate.

A typical force *vs* time record is shown in Figure 5a which shows major maxima of ~0.7 N, each followed by a rapid drop to minima of ~0.35 N. We observed in each case that the rapid build-up of force occurred during a period when the peel line appeared to have arrested, whereas the sudden drop in force accompanied the relatively sudden detachment of a creased region of tape. It is important to emphasize that these oscillating forces observed at these low peel rates are unrelated to the more customary stick-slip effects seen at much higher peel rates.

During this particular peel we marked the position of the peel zone on the edge of the tape each time that the peel force exhibited a sudden decrease. These marks allowed us to examine the correlation between the peel force variations and the resulting topography of the tape when photographed under low angle illumination (see Figure 5b). Corresponding features **A–K** are identified in Figures 5a and 5b. At position **A** the experiment was started where the force initially reached a plateau value corresponding to steady, slow peeling. After a slowly accelerating increase, the force reached a maximum then displayed a sharp decrease at **B**. The mark on the tape corresponding to this instant (**B** in Figure 5b) aligned with the creased region of the tape which had detached from the

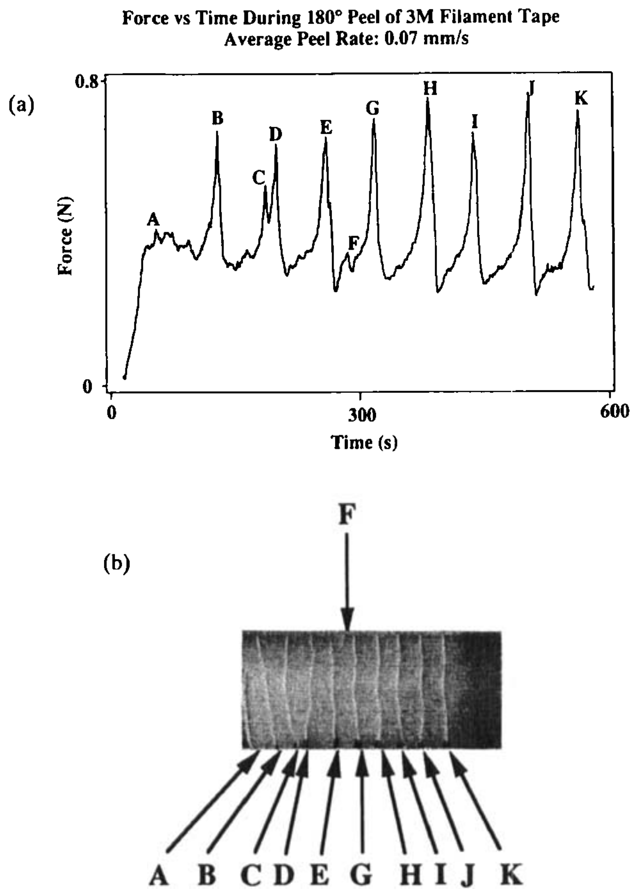

FIGURE 5 (a) Force *vs* time during intermediate rate peeling of filament tape from PMMA (2 cm tape width). (b) Photograph of creases in backing of corresponding piece of tape.

substrate fairly quickly. As the peel continued, the force again increased, followed by a pair of sharp declines at **C** and **D**. In Figure 5b, the corresponding creases were not formed transversely across the tape but in a diagonal manner; crease **C** had not completely detachted before crease **D** began, thus causing the force to show a double maximum.

At point **E** the force again reached a maximum before a sharp drop which was accompanied by fast peel and a crease at **E** in Figure 5b. As the force rises toward **G** it goes through a small minimum at **F**. When we examine the tape in Figure 5b we find a partial crease at **F**. The subsequent sharp force decreases, **H–K**, are typical of the majority of other cycles we measured, namely they represent individual sudden reductions in the force, each accompanied by a single crease fairly well aligned in the transverse direction. All of these variations in the peeling sequence show up readily in the autographs (Figure 2–4).

Kaelble has studied the peeling behavior of pressure sensitive adhesives and

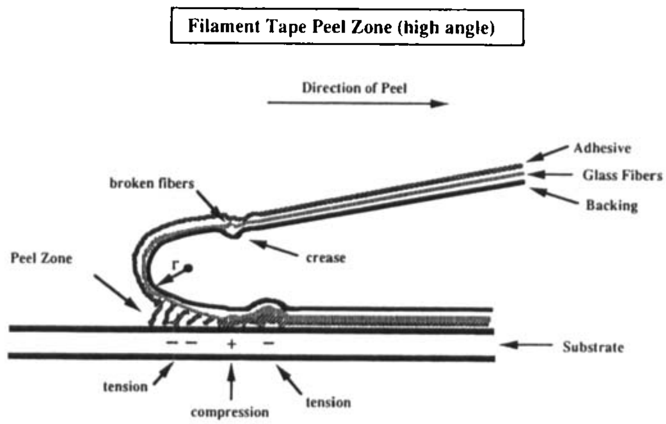

FIGURE 6 Drawing of filament tape profile during peeling from a rigid substrate.

noted that the visco-elastic properties lead to the development of regions of compressive stress(+) as well as tensile stress(−) near the peel line.[5] A sketch of the profile of filament reinforced tape during peeling from a rigid substrate is shown in Figure 6, where the regions of compressive and tensile stress are identified. When the longitudinal edge of the tape is observed during peeling, one sees that the plasticized face adhesive is undergoing longitudinal flow and is squeezed from the region of compressive stress away from the peel line, into a region of tensile stress, about 1 mm ahead of the peel line (see Figure 6). This causes curvature (concave downward in Figure 6) to develop in both the tape backing and in some of the adjacent fibers as they are pushed away from the substrate. The formation of this bulge is assisted by the longitudinal compressive force on the backing due to the 150° loading. For a short distance (approximately 1 mm), as the peel line continues to advance, this bulge moves as a wave in concert with, and ahead of, the peel. This leads to the region of the autograph labeled **B** in Figure 3, one of fairly uniform, low intensity.

In a fairly abrupt fashion, this bulge becomes a permanent crease or fold in the tape backing. In addition, the glass filaments in the strands close to the peel zone are divided such that some of them remain attached to the polyester backing and some remain attached to the substrate. This configuration produces a stiff section in the tape which tends to be peeled as a unit and in essence has a large surface area associated with it. This results in an increase resistance to peeling, thus causing the radius of curvature, (**r** in Fig. 6) to decrease. During this time, the crack front advances very slowly towards the creased region, yielding little evidence of photon emission (region **C**, Figure 3).

When **r** reaches a critical minimum value, a number of the fibers fracture in a brittle fashion. On the average, we measure an $\mathbf{r}_{min}$ of ~30 μm. Accompanying fiber fracture is a sudden change in the forces applied to the creased region of the tape causing it to pull from the substrate fairly quickly, usually propagating transversely from one fiber bundle to the next. This creates the brighter edge of the autograph pattern (region **A** in Figure 3). After this stiff, folded section has peeled from the substrate, the cycle begins again and repeats every 3 to 6 mm along the length of the tape, where the largest spacing occurs at lowest peel rates.

After the peel is completed we can measure that the tape is slightly shortened (0.3%) due the creasing process. Microscopic examination of the tape shows that approximately 20% of the fibers are broken at the leading edge of the crease, precisely where the bend became sharpest. As a further demonstration of this failure mechanism, using a roll of Filament Tape we deliberately delaminated the backing from a single bundle of fibers so that the fibers remained adhering to the release-coated polyester. When this single bundle of fibers was peeled from the polyester in a 150° peel, we observed the same slow-fast peel previously described, with oscillations in the radius of curvature (which are even more extreme than seen in the tape). Similarly, when the single bundle is examined after peeling, extensive fiber breakage is observed at fairly uniform intervals of length (approx. 0.8 mm), roughly 1/3 the interval for the tape.

The origin of the highly localized features on the autographs in the region labled **B**, Figure 3, can be understood in terms of the normal behavior of a viscoelastic adhesive during peeling. Microscopic examination of the tape prior to attachment to the substrate shows that the adhesive face is not planar but follows the contour of the fiber bundles. When the tape is pressed onto any substrate the closest contact is made at these ridges whereas longitudinal bubbles lie in between the ridges and prevent good contact with the substrate. When the tape is subsequently peeled these "lines" of better contact form a locus of interfacial failure which in turn produces aligned spots on the autographs. In contrast, the valleys represent regions of very poor contact, thus yielding no visible film exposure.

For moderately slow peeling the autographs, such as Figure 3, allow us to identify approximately 2 to 4 discharges/mm^2 to tape area peeled in air. This is similar to the areal density of discharges which we counted previously arising from the T-peel configuration for slow peeling[2] of this tape in a nitrogen atmosphere.

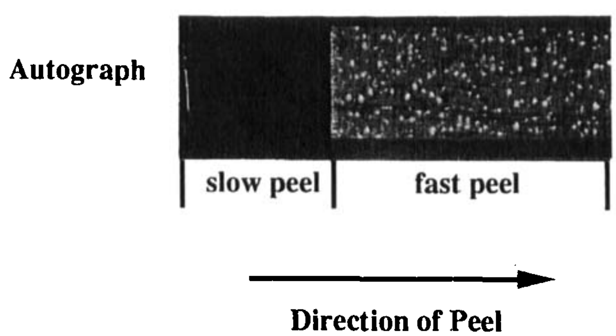

FIGURE 7 Autograph of slow then fast peel of filament tape from Polaroid® Type 107C film. Slow peel was at 0.0015 mm/s.

It is interesting to compare the interval in which the peeling produces little or no light (*e.g.*, region **C**, Figure 3) for the cases of constant applied force *vs* constant strain rate. This region is noticeably wider and darker for constant force than for constant strain rate, reflecting the differences in the mechanical responses of the system to these different types of loading.

We should also note that at constant applied force we recorded emission at an average peel rate of 0.003 mm/s. At a peel rate of 0.0015 mm/s, the resulting autograph shown in Figure 7 appears to be blank. On the original photo, one can see ~20 small discharges. This is consistent with other studies we have carried out involving variations in fracto-emission intensities with crack speed[3,6,7] which we have attributed to the charge separation process and leakage of charge occurring in the crack tip region.

Previously,[2] we learned that the number of photons (as well as the size of the accompanying RE bursts) associated wih the microdischarges increases on exposure to N_2. We suspected that this enhancement would make the autographs more intense and enhance the images at low peel rates. Conversely, we also had discovered that the number of photons created at a microdischarge were quenched in O_2. The gas phase phenomena which explain these results are discussed in Ref. 2. In Figures 8a and 8b we compare the autographs of Filament

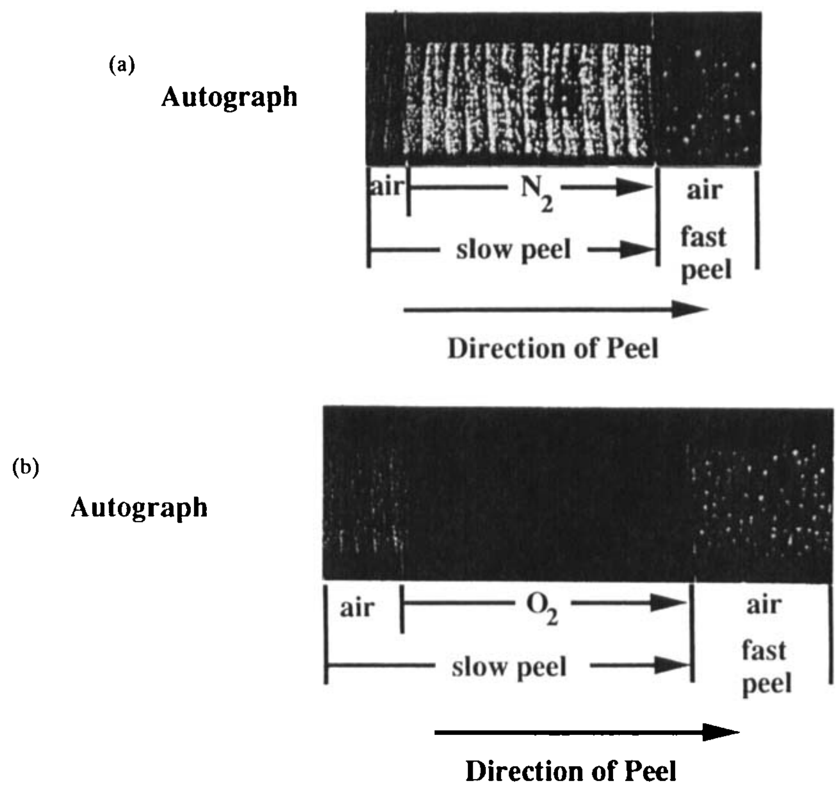

FIGURE 8 Autographs of slow peel (0.033 mm/s.) in air then (a) in N_2 or (b) in O_2 at one atmosphere, followed by fast peel in air.

Tape peeled from film at an average peel rate of 0.033 mm/s in air and when N_2 and O_2 were introduced at one atmosphere; we note the obvious enhancement of light by the N_2 and strong suppression of emission by O_2. Obviously, if one desires to detect extremely slow peel, one could easily introduce N_2 to take advantage of the enhancement.

IV SUMMARY AND CONCLUSION

In summary, we have shown that:

1) phE caused by tape peeling of Filament Tape produced autographs which could be observed at average peel rates approaching 0.0016 mm/s.

2) The bright transverse bands of emission appearing in autographs are the result of periodic events involving viscous flow of adhesive, fiber fracture, and rapid detachment of adhesive from the substrate.

3) At peel rates averaging less than 0.001 mm/s the intermittent nature of the peeling assures that there are periods when the interfacial failure is extremely slow and the corresponding light emission is nearly undetectable.

4) Peeling in the presence of gases other than air has a striking effect on the autographs. Nitrogen supports electrical discharges and causes enhanced light emission. Oxygen quenches discharges and suppresses light emission. These dramatically affect the intensity of the light recorded in autographs, which could be useful for studying very slow failure.

This work illustrates that autographs provide details of the detachment process of a composite tape from a smooth substrate during high-angle peeling. In this type of loading, the fiber-reinforced tape undergoes a sequence of micromechanical steps which strongly influence the instantaneous rate of detachment, which in turn leaves a unique record on the exposed film. The occurrence of this type of failure at such slow peel rates appears to be quite distinct from the usual stick-slip failure seen at very high peel rates. We propose that these and other fracto-emission studies can aid our understanding of the various failure modes in a composite system including filament-reinforced tape and therefore assist in improvements in tape design.

Acknowledgments

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