

Failure Mechanisms in Peeling of Pressure-Sensitive Adhesive Tape

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Synopsis

Studies on the peel behavior of pressure-sensitive tape comprising a polyester backing and polyacrylate adhesive have shown that, in peeling from a plane glass surface, three fundamentally different modes of peeling may be distinguished, depending upon the rate of pulling. At low rates, deformation by flow of the adhesive appears to determine the peel behavior and the peel force is strongly rate dependent. At high rates, little or no viscous deformation of the adhesive occurs and the peel force is independent of rate. At intermediate pulling rates, cyclical instability of mode of failure involving alternate storage and dissipation of elastic energy in the backing, results in the phenomenon of "slip-stick" peeling, in which failure is jerky and regular. Results have been obtained which show how the pulling rates at which transitions from one mode of peel to another occur, and the peel force values for a given type of failure, depend upon such factors as molecular weight of adhesive, thickness of adhesive, thickness of backing film, and angle of peeling.

INTRODUCTION

Resistance to peeling is a most important property of pressure-sensitive adhesive tape. Many studies have been made of the variation of peeling resistance with changes in tape design and composition (type and thickness of both the flexible "backing" strip and the adhesive) and testing conditions (nature of the rigid "substrate" from which the tape is peeled, angle of application of the force of peel, temperature, rate of peel, etc.). The most usual type of test has been to measure the force required to peel a tape of unit width, at constant rate and angle, from a plane substrate of metal or glass.

Under these conditions, commercial tapes fail most commonly by separating cleanly from the substrate, i.e., they show an "adhesive" mode of separation at the adhesive/substrate interface. An "adhesive" separation may alternatively occur, however, at the adhesive/backing interface. A further possibility is "cohesive" separation, located within the bulk of the adhesive.

Apart from these possibilities of "adhesive" or "cohesive" locations of separation, however, there are indications that the mechanism of failure can vary in other fundamental ways. For example, with tapes showing adhesive failure, the peel strengths may increase with peel rate,¹⁻⁷ may be

rate independent,⁷ or may decrease with rate.⁷ Where cohesive failure occurs, the measured force of peel at a given rate may either be more or less steady or alternatively may undergo regular oscillations of large amplitude,^{4,7} a phenomenon known as "slip-stick" peeling. Gardon observed "slip-stick" to occur both with wholly adhesive and with wholly cohesive modes of separation.⁷

The present study, besides confirming that "slip-stick" peeling may occur with wholly adhesive separation, shows its occurrence also with cohesive and adhesive separation alternating in step with the oscillation of peel force.

The present work was begun in an attempt to identify, characterize, and investigate the mechanism of each of the modes of failure which could occur. Thus it is necessary first to explore the extent to which existing theories of peel are applicable to the different types of failure. Furthermore, where existing theories are found inadequate or inapplicable, attempts will be made to suggest appropriate modifications or to devise new theories which will survive experimental test. Thus, it is hoped to arrive at a satisfactory understanding of the mechanism of peel under all conditions.

METHOD OF APPROACH

For the purpose of this investigation, it was decided to standardize on a form of tape that would be simple and reproducible and reasonably representative of the wide variety available.

A polyester film was chosen as the backing, having high chemical purity (no plasticisers or other additives), good resistance to solvents, heat, and moisture, and ready availability in a wide variety of thicknesses. Experiments were confined mainly to adhesives based on poly(*n*-butyl acrylate), which has the advantages of chemical purity and availability in different molecular weights, although it may not completely simulate some of the sophisticated acrylate copolymers and their compounds used in industrial adhesives. The thicknesses of backing and adhesive, except when deliberately varied, were chosen so as to be reasonably similar to those used industrially.

The tapes were tested by peeling from clean glass plates, at an angle of 90°, at room temperature. This method was chosen after preliminary tests had indicated that much more reproducible results were obtained using chromic acid-cleaned glass rather than the more conventional standard stainless steel finish.⁸ Each tape was generally tested at a variety of discrete rates of pulling, and it is intended in future work to vary the temperature also.

Details of the tape preparation, composition, and testing are given later in the experimental section, which need not be read before the discussion below.

RESULTS AND DISCUSSION

Effect of Pulling Rate on Mode of Failure and Peel Strength for a Poly-*(n*-butyl Acrylate)-Coated Polyester Film

The dependence of peel strength upon the rate of jaw separation (pulling rate) when a poly(*n*-butyl acrylate)-coated polyester film is peeled at a 90° angle from glass using an Instron tensile testing instrument can be seen from Figure 1.

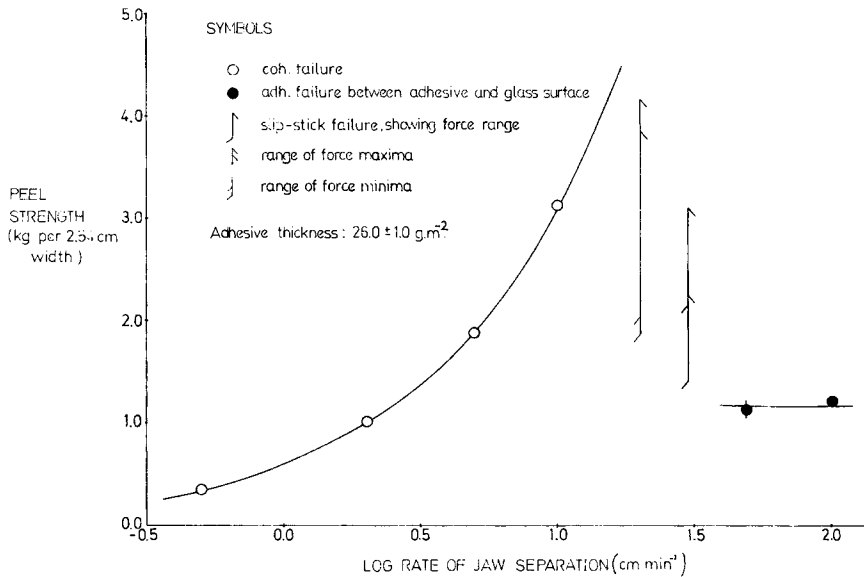


Fig. 1. Effect of pulling rate on peel strength and mode of failure for poly(*n*-butyl acrylate) adhesive on polyester backing.

The curve is seen to comprise three regions, distinguished by the occurrence, at intermediate pulling rates, of regular oscillations of the peel force, whereas at slower and faster pulling rates these regular oscillations are absent.

For the convenience of discussion these regions are represented diagrammatically in Figure 2.

With results in the first region, at the lowest rates, the adhesive showed "legging" (i.e., filaments of adhesive were formed between backing and substrate) to a considerable extent during peel. Also in this region it was always found after peeling that some adhesive remained on the backing film and some remained on the glass, i.e., the failure was "cohesive" within the adhesive. The adhesive surface after the separation was rough. The way in which the peel force increases with rate in this region resembles the way in which the shear stress increases with shear rate in the steady-state flow of viscoelastic polymers in the soft state. This indicates that in the

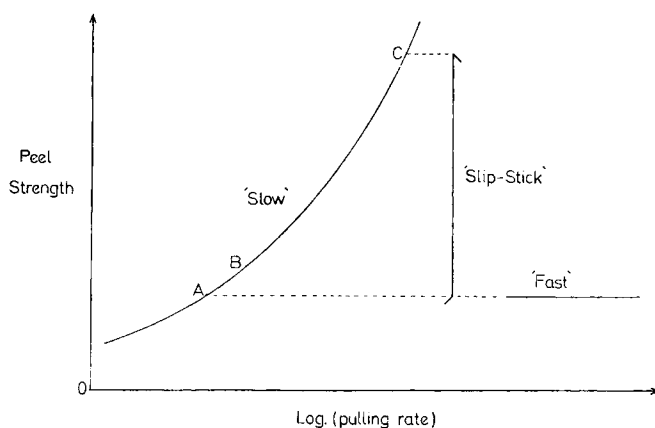


Fig. 2. Diagrammatic illustration of the dependence of peel strength and mode of failure on rate of pulling (poly(*n*-butyl acrylate)-coated polyester film peeled from glass).

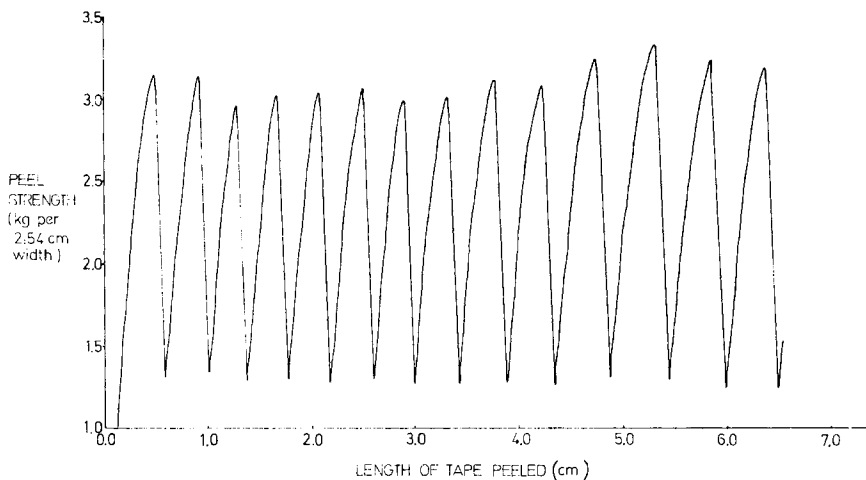


Fig. 3. Copy of an Instron chart showing "slip-stick" peeling. Poly(*n*-butyl acrylate) adhesive (26.5 gm^{-2}) on polyester backing. Pulling rate: $10.0 \text{ cm. min.}^{-1}$

slow region the main process involved in the separation is viscous flow of the adhesive, a view also held by Voyutskii⁴ and Bright.² Results reported in later sections give further support to the conclusion that viscous flow is the predominating process leading to failure in this region.

At the highest pulling rates ("fast" region), the failure is wholly "adhesive" and the force appears to be more or less independent of the pulling rate. Although only two of the data points in Figure 1 are relevant, more values have been obtained in the experiments described later. These agree, within experimental error, with the postulate that the peel force is independent of rate in this region. During peeling in this region, no "legging" can be seen to occur at the line of separation. The adhesive surface after

peeling is smooth, glossy, and apparently undisturbed. These observations, together with other results reported later, suggest that viscous flow does not occur in this region and that the behavior of the adhesive is almost elastic. This would account for the independence of peel load on pulling rate.

In the region of intermediate pulling rates, the mode of failure is "slip-stick," that is, a regular, jerky peel in which the observed peel force oscillates between quite well-defined limits. An autographic recording obtained in this region is shown in Figure 3.

In this region, it was seen from the peeled film and glass that "cohesive" separation within the adhesive alternated with "adhesive" separation at the adhesive/glass interface. The alternations of force were in step with the alternations of mode of failure, and furthermore the rising and falling parts of the autographic trace corresponded with "cohesive" and "adhesive" separations, respectively.

These and other observations (some of which appear later in this paper) have led us to an explanation of the mechanism of slip-stick peeling, which in outline is as follows. Assume that the rate of pulling is steady in the intermediate range giving slip-stick peeling. Imagine that the force of peel has momentarily the value attained in "fast" peeling, although at this instant the mechanism of peel is the "slow" one. This situation would be represented by the point A in Figure 2, if the tape was being pulled at the rate shown by A. But the tape is being pulled much faster than this, and so the force of peel will increase rapidly. At a later instant the force will pass through the value represented by the point B (Fig. 2), where the amount of elastic energy stored in the unattached backing will appreciably exceed the value for "fast" peeling at a steady rate. This excess of elastic energy would be available to sustain briefly a rate of peel greater than the rate of pulling, if the force of peel were to fall below that of the point B. Such a fall in force (to the level of A) would occur if the mechanism of peel were to change over to the "fast" one. Thus the situation would become increasingly unstable until, at some point such as C in Figure 2, the mechanism of peel does change to the "fast" one. Then fast peeling will occur briefly, at the expense of the stored elastic energy. This peeling will be at a rate much faster than the steady rate of pulling and will continue until the excess elastic energy is dissipated and the force has fallen to the level of A. The steady rate of pulling is insufficient to sustain peel with the "fast" mechanism, so that the mechanism must then revert to the "slow" one. This completes the cycle of a slip-stick oscillation.

Effect of Pulling Rate on Mode of Failure and Peel Strength for Other Tapes

The question which immediately arises on consideration of the above observations is whether or not the mode of peeling of all adhesive tapes will similarly pass (at suitable rates and/or temperatures) from a region of

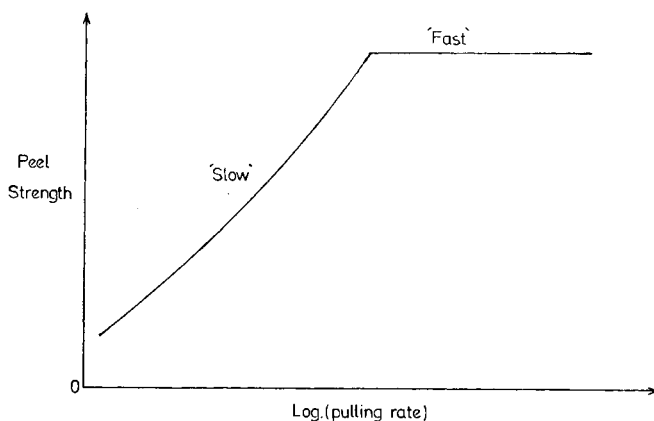


Fig. 4. Diagrammatic illustration of the dependence of peel strength and mode of failure on rate of pulling, as reported by Gardon (acrylate-coated cellophane film in T-peel).⁷

cohesive failure involving flow in the adhesive, through a slip-stick region, to a region of rate-independent adhesive failure.

Some results reported by Gardon⁷ indicate that all three modes do not always occur. Using an acrylate adhesive, he also found at low rates a cohesive failure with rate-dependent peel force, and at high rates an adhesive failure with rate-independent peel force; but, in contrast, there appeared to be no intervening region of slip-stick failure. However, the absence of the intervening region would be expected on the basis of our postulated mechanism for slip-stick peeling as given above. This absence would be a consequence of another feature of Gardon's results, namely that the peel force for adhesive separation (i.e., with fast peeling) was *not* lower than the highest value for cohesive separation. Thus, the generalized diagram in his case would be of the form shown in Figure 4. It is clear that this form of curve does not provide the conditions for instability (i.e., higher peel force at low rates of peel than at high rates) that are required according to our proposed mechanism for slip-stick peel.

Further experiments we have made suggest that the slow "leggy" region is not necessarily characterized by a cohesive type of separation. Thus, using a vinyl acetate-2-ethylhexyl acrylate copolymer as the adhesive, we have found that, at the lowest pulling rates, separation is cohesive and "leggy." However, with an increase in rate, the mode of failure becomes adhesive at the adhesive/glass interface although still "leggy." This region is then succeeded by slip-stick failure of a wholly adhesive type, and, at still higher rates, by smooth, "nonleggy" adhesive separation from the glass (Fig. 5). The behavior of a natural rubber-based adhesive is also shown in Figure 5. In this case, failure is wholly adhesive at the adhesive/glass interface at all but the very slowest of the rates used. At the low rates, the separation is "leggy" in nature and the peel force gradually increases with pulling rate. At higher rates, slip-stick failure was observed, followed at the

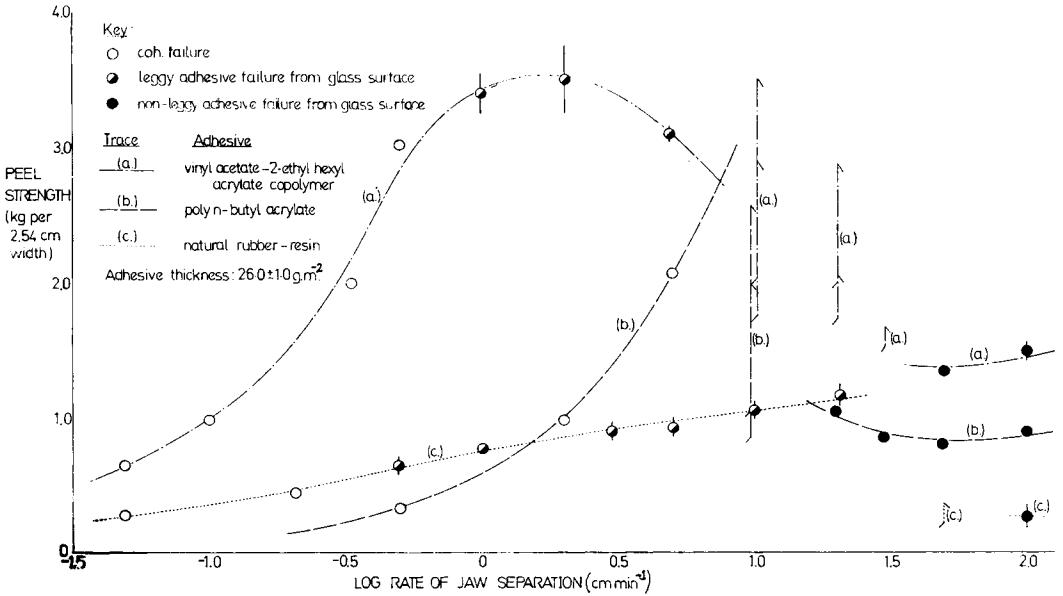


Fig. 5. Effect of pulling rate on peel strength and mode of failure for different adhesives on polyester backing.

fastest rate by a smooth “nonleggy” adhesive failure at low peel force. Indeed, further evidence of the occurrence of this succession of types of behavior is seen in the unwind characteristics of some commercial tapes, which progress from a steady but “leggy” adhesive peel at low rates through a slip-stick region (characterized by a noisy unwind and with the unwound tape showing clearly visible transverse striations of disturbed adhesive) to a smooth nonleggy adhesive peel at the highest rates of unwind. The adhesive after a low-rate leggy peel always appears to have an irregular disturbed surface, whereas after a high-rate nonleggy peel the adhesive surface appears smooth, glossy, and undisturbed.

It therefore appears that the three principal kinds of peel behavior of pressure-sensitive adhesive tapes that should be distinguished have the following characteristics:

(1) At slower rates the peel force is steady and increases with rate. The separation is either cohesive (and “leggy”) or “leggy” adhesive and in either case leaves a rough surface on the peeled adhesive.

(2) At intermediate rates there is a region of slip-stick failure with large regular oscillation of force. The separation may be wholly cohesive, adhesive alternating with cohesive, or wholly adhesive. The peel will be noisy if the slip-stick oscillation occurs at audiofrequencies. Where slip-stick separation is wholly adhesive, it is believed that “leggy” failure alternates with “nonleggy” failure, as shown by corresponding striations seen on the peeled adhesive.

(3) At higher rates the peel force is steady and independent of rate. The separation is adhesive and "nonleggy," and leaves a smooth surface on the peeled adhesive.

Of course, it is to be expected that the rates at which the transitions occur will vary enormously with different tapes and test conditions and may fall outside the accessible range of rates, so that a given tape may well appear to give only one of these types of failure.

Effect of Varying Molecular Weight of Adhesive Polymer

While keeping other factors constant, the relationship between peel strength and pulling rate has been examined using a range of poly(butyl acrylate) homopolymer adhesives of different molecular weights (as measured by dilute solution viscometry). Results are shown in Figure 6.

In the "slow" peeling region (with steady force and cohesive separation) the peel strength increases with molecular weight, as would be expected if viscous flow of the adhesive is the controlling factor.

The rate at which the transition to slip-stick occurs increases with decreasing molecular weight, although with the two lowest molecular weight adhesives this transition did not occur, even at the highest crosshead speed of 100 cm min^{-1} obtainable with the Instron tester. Presumably, however, these samples would also exhibit slip-stick and smooth adhesive failures at higher rates than used here.

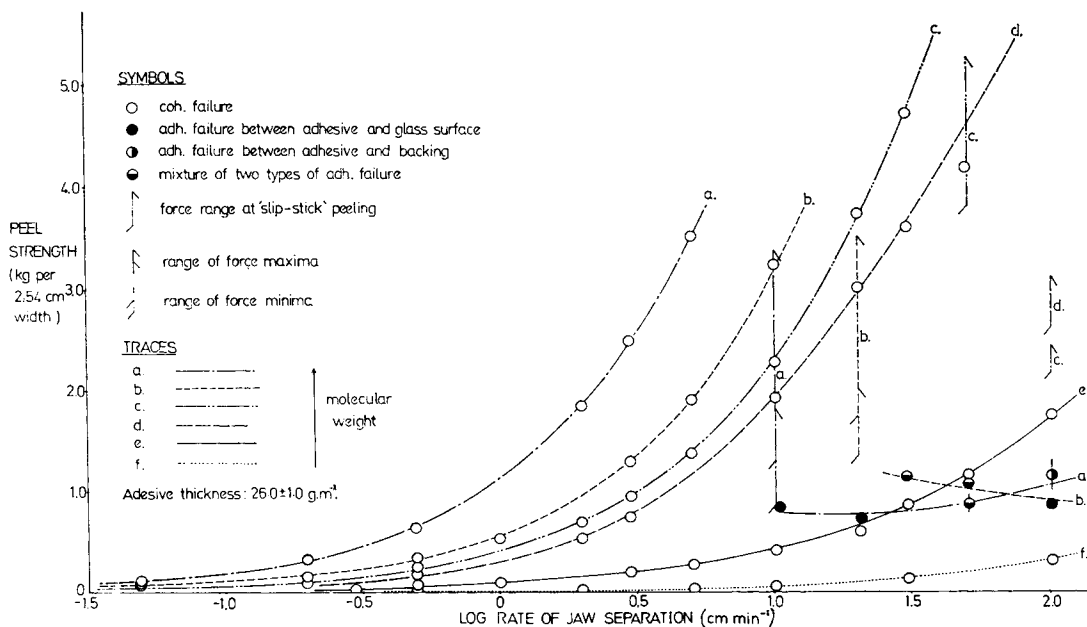


Fig. 6. Effect of molecular weight on peel strength at various rates of jaw separation for poly(*n*-butyl acrylate) adhesives.

In the "fast" peeling region (with steady force and smooth adhesive failure), the amount of data available is small but gives no indication of appreciable dependence of peel strength on molecular weight. Such an independence of the molecular weight of the adhesive would be further evidence that here there is no flow of the adhesive and that the adhesive deforms high-elastically and almost completely reversibly, i.e., it behaves as if on the "high elastic plateau" of its viscoelastic response curve.

On this basis the shift of the two transitions to higher rates with decrease of molecular weight would be seen to arise solely from the shift of the curve for the slow region to higher rates with decrease of molecular weight.

Effect of Varying the Thickness of Adhesive

Figure 7 illustrates the effect of varying the thickness of adhesive while keeping other factors constant.

It is clear that, in the region of steady cohesive failure (at the lowest rates), there is little effect of adhesive thickness on peel strength. This result, as well as those given earlier for this region, could be accounted for by the following explanation, which we have not yet subjected to additional test. Assume that all the work of peel is used in causing flow of the adhesive (neglecting the work stored elastically in the peeled backing). At a given instant, the region in which serious amounts of flow are occurring would be expected to be very restricted and to be confined to the immediate

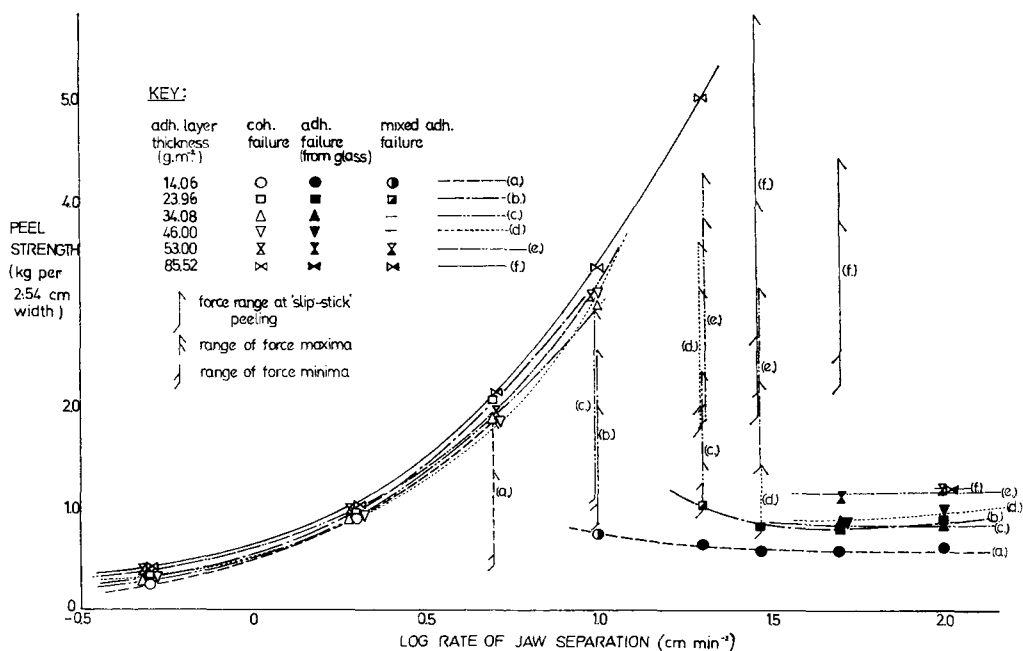


Fig. 7. Effect of adhesive thickness on peel strength at various rates of jaw separation for poly(*n*-butyl acrylate).

neighborhood of the line of separation at the head of the advancing front of air. Then it could well be that the amount of flow required for separation (i.e., the amount having occurred along the track of this neighborhood, per unit width and unit length peeled) is constant, independent of rate of peel or thickness of adhesive. Thus the peel force would increase with rate of peel and molecular weight of adhesive but not change with thickness of adhesive. If this is the correct explanation, it appears that some additional factor may enter over a very wide range of thicknesses of adhesive, because Gardon⁷ found an increase of peel force with adhesive thickness from 1.2 μ to 258 μ .

The most noticeable effect of increasing the adhesive thickness is to cause the transition to slip-stick peel behavior to occur at higher pulling rates. It also appears to increase the mean force in the slip-stick region.

In the region of steady adhesive separation at the fastest rates, the peel strength increased with adhesive thickness. This was also observed by Gardon⁷ and was attributed to a reduction in localized stress concentration due to an increase in the area of adhesive under stress.

Thus the effect on the slip-stick region of increase in thickness of adhesive, in increasing both the pulling rate and the mean force, could be attributed solely to the effect on the "fast" region of thickness in increasing the peel force. If the transition to slip-stick peeling requires a given degree of instability, as indicated by the ratio of force in "slow" peel to force in "fast" peel, then an increasing thickness would require higher speeds of "slow" peel so as to reach a higher steady peel force for the transition to slip-stick, and the mean forces in the slip-stick region would also increase.

Effect of Varying Thickness of Backing Film

The effect of variation in thickness of the polyester support on the relationship of peel strength to pulling rate is shown in Figure 8. The adhesive thickness was kept as constant as possible and all the other test conditions were nominally identical.

It can be seen that, for the thinner films, the familiar form of relationship is observed between peel strength and pulling rate, showing the three types of failure. However, for the thicker films the slip-stick region has been replaced by a peeling process in which there is large irregular fluctuation in peel strength, accompanied by apparently random simultaneous differences in the mode of separation across the width of tape. (Cohesive failure, and adhesive failure from either interface, occurred apparently at random over the area of peeled tape.)

In the "slow" region, where the failure is cohesive, the peel strength is unaffected by thickness of backing in the range 0.015 to 0.052 mm, but then decreases with thickness up to 0.126 mm. The reason for this is not clear at the moment.

At higher pulling rates, where adhesive failure occurs there does not appear to be any systematic relationship between either peel strength or location of separation and thickness of backing film. This region is complicated by the occurrence of adhesive separation from both substrate and

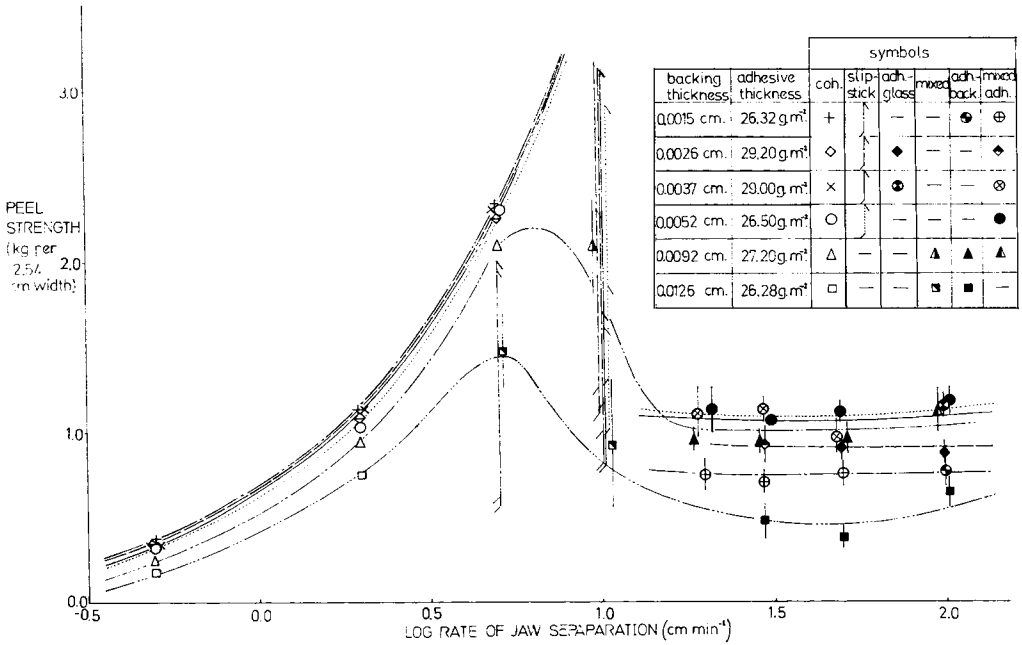


Fig. 8. Effect of backing thickness on peel strength at different pulling rates for poly(*n*-butyl acrylate) adhesive on polyester backing.

backing, but nevertheless the peel force values appear to be approximately independent of pulling rate.

It will be possible to give an explanation for the absence of slip-stick behavior with the thicker films in a later paper in which the slip-stick mechanism is to be more fully analyzed.

Effect of Variation of Peel Angle

Figure 9 shows how the peel angle affects the peel strength, and affects the mechanism of failure, at two pulling rates.

At the lower pulling rate (10 cm min⁻¹), leggy cohesive separation occurred at angles of 60° and 70°, but at higher angles, up to 150°, the failure was slip-stick, with alternating cohesive and adhesive separation. A line joining the slip-stick force maxima with the peel strengths for cohesive failure at 60° and 70° is a curve very similar in shape to those obtained by Kaelble.^{6,9} We are currently investigating the proposition that the recorded slip-stick force minima may have no real significance but are largely determined by the limited speed of response of the recorder.

At the higher pulling rate (30 cm min⁻¹), slip-stick failure with alternating cohesive and adhesive separation occurred at angles up to 80°. At 90° and above, smooth “nonleggy” adhesive separation occurred, giving a curve with a less obvious minimum than in the case of cohesive separation.

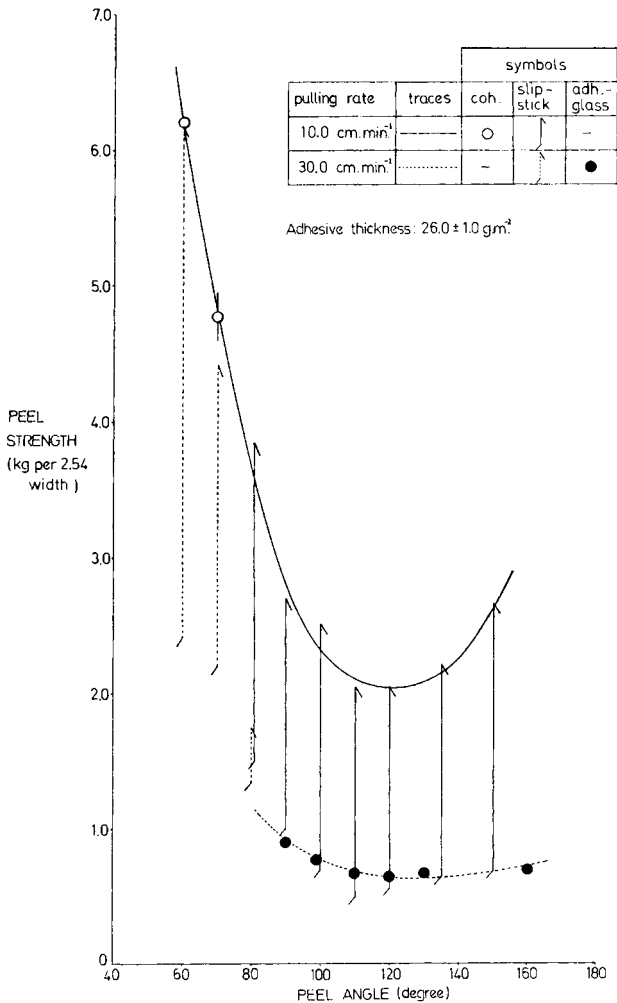


Fig. 9. Effect of peel angle on peel strength and mode of failure for poly(*n*-butyl acrylate) adhesive on polyester backing.

It appears, therefore, that the effect of reducing the peel angle from about 90° to lower angles is to cause the mode of failure to move from the slip-stick to the "leggy" cohesive region at the lower rate, and from the "non-leggy" adhesive to the slip-stick region at the higher pulling rate. Thus, both of the transitions move to higher pulling rates as the angle decreases. The reason for this is not, as yet, clear.

CONCLUSION

Three main modes of failure have been identified in the peeling of pressure-sensitive tapes from a glass substrate.

As the pulling rate was increased, the failure at first was by a "leggy" separation (either adhesive or cohesive in location) and in which the peel force increased with rate. Then the failure passed through a slip-stick region involving either wholly adhesive or regularly alternating cohesive with adhesive separation. Finally, at the highest rates, failure was by a "nonleggy," wholly adhesive separation, in which the peel force was rate independent. The essential characteristics of these three modes have been inferred by considering also the results of other authors.

Measurements have also been made of the way that the peel strengths in these regions, and the pulling rates for the transitions from one mode of failure to another, are affected by the adhesive polymer molecular weight, the thickness of adhesive, the thickness of backing film, and the angle of peel.

On the basis of these results, outline theories have been proposed for the mechanisms of the peel processes which account for many of the observed features but which are currently undergoing further test and refinement.

EXPERIMENTAL

Materials

Unless otherwise specified, Melinex type O uncoated polyester film of 0.001 in. thickness (I.C.I. Ltd.) was used as the backing film. This film is biaxially oriented and is tough, transparent, and flexible, with good dimensional stability and resistance to heat, solvents, and moisture. It is believed to contain no plasticizer or other additives. At an elongation rate of 100%/min, this film has a Young's modulus of about 40000 kg/cm², its yield strength is about 1150 kg/cm², and it yields at about 3% extension.

The adhesive was poly(*n*-butyl acrylate) obtained in the form of its 25% w/w acetone-benzene solution (Acronal 4L) supplied by BASF (United Kingdom) Limited. For experiments designed to investigate the effects of molecular weight of adhesive, grades of poly(*n*-butyl acrylate) of different degrees of polymerization were used (Acronal 4L as a 50% w/w ethyl acetate solution, and Acronal 4F which is solvent free). The Acronal 4L (acetone-benzene solution) was fractionated further by precipitation from acetone into methanol in order to obtain a still higher molecular weight fraction. The average molecular weights of different grades of poly(*n*-butyl acrylate) were estimated by dilute solution viscometry. The *K* and α values used for the Mark-Houwink equation for poly(*n*-butyl acrylate) with acetone as solvent at 30°C were 6.85 and 0.75, respectively.¹⁰

Infrared spectra of the various molecular weight grades of polymer were identical and consistent with a reference spectrum of pure poly(*n*-butyl acrylate). The alcohol obtained by saponification of each polymer was shown by gas-liquid chromatography to be *n*-butanol, with negligible impurities.

Preparation of Tapes

The adhesive was coated on to the polyester film using a laboratory hand-spreading device with which the film was slowly (at about 3.0 meters per min) drawn through a nip between a smooth curved surface and a "doctor blade." The variation of thickness of adhesive along the direction of spreading was estimated to be less than $\pm 2.5\%$, whereas that along the width of the film was estimated to be less than $\pm 1\%$. Such a small variation in adhesive thickness was considered to be negligible, a view supported by experimental results obtained on the effect of adhesive thickness (Discussion, "Effect of Varying the Thickness of Adhesive").

After spreading, the film was allowed to dry on a flat surface for 20 min, then transferred to an aerated but dust-free box to condition for at least 72 hr in the dark before testing. It was found that the peel strength always became constant and reproducible after 48 hr. One-inch wide tapes were cut from the film with the help of a specially made template and support. The edges and the first 4 in. of the coated film were discarded because it was found that the variation of adhesive thickness there was highest.

The thickness of adhesive was found by measuring the loss of weight when the adhesive was removed. A 5×5 cm² portion of the coated film, cut with the aid of a square template, was accurately weighed. The adhesive was removed by swelling in methanol, then by peeling or rubbing off. The adhesive-free film was finally washed with acetone and dried to constant weight.

Peel Adhesion Test

The peel test was done on an Instron tester with a specially designed jig (details to be published) which ensured that the angle of peel remained constant during any single test. The crosshead speed of the Instron tester ranges from 0.05 to 100 cm min⁻¹. The Instron tester was calibrated against a standard load prior to each test.

The glass plates were cleaned, after removal, of any excess adhesive by a methanol wash, and by immersion in a chromic acid bath for at least 24 hr before use. Immediately before use, the glass plates were rinsed with distilled water, then with acetone, and finally dried at 60°C.

The tape to be tested was applied to the glass plate by a rubber hand roller 2.5 cm in diameter so as to avoid the inclusion of air bubbles. It was found that changing the pressure of application of the tape on to the glass surface had no effect on the resulting peel strength, providing that no air bubbles were visible.

The peel test was carried out within 2.5 min after the application of the tape to the glass surface. A freshly cleaned glass plate was used for every test and the peel angle was kept at 90° except where deliberately varied.

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