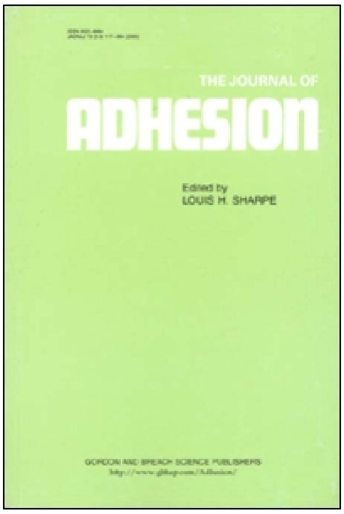

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Effect of Peel Load on Stringiness Phenomena and Peel Speed of Pressure-Sensitive Adhesive Tape*

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The factors governing interfacial separation in lightly cross-linked polymer adhesives at low pulling rates as demonstrated by their stringiness phenomenon are investigated.

Cohesive failure and adhesive/substrate interfacial separation of uncross-linked polymer adhesives have been adequately explained. However, in lightly cross-linked polymer adhesives, where cohesive failure cannot occur because there is no viscous flow, there are two regions of interfacial separation at low rate and this phemonenon cannot be readily explained by present viscoelastic theories.

Investigation of the stringiness phenomenon of peeling pressure-sensitive adhesive tapes at constant loads shows that two peeling speeds exist for any peeling load up to the vicinity of 200 g/25 mm. Also it is clear that stringiness structure differs greatly at each peeling speed. The stringiness phenomenon of each of these two regions is analyzed using Miyagi's observation apparatus. These two measurements are then reversed and a comparison shows that the two peeling speeds correspond to each steady peeling region.

This field of investigation, when added to the present viscoelastic property studies, should lead to a new peeling adhesive theory which, in turn, may lead to the development of new high peel force pressure-sensitive adhesives.

KEY WORDS Pressure-sensitive adhesive tape; stringiness phenomena of PSA; stringiness pattern; peel force vs peel rate; interfacial separation; cross-linked natural rubber adhesive.

1 INTRODUCTION

Peeling behavior is one of the most important properties of pressure-sensitive adhesive tapes, and various theoretical analyses have been investigated on the distribution of adhesion stress during peeling.[1–5] Satas *et al.* have studied the effects of 90° and 180° peeling of various backings.[6]

The peel force *vs.* velocity spectrum of pressure-sensitive adhesive tape has

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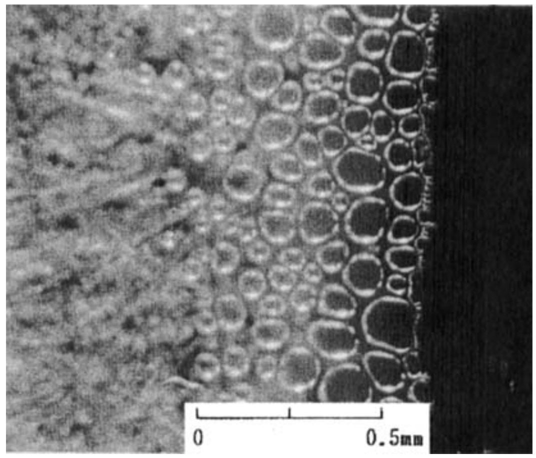

PHOTO 1 Photomicrograph of a honeycombed stringiness pattern of a pressure-sensitive adhesive on porous backing.

been studied and analyzed from the point of view of time dependence of adhesive viscoelastic property by Gent *et al.* and Aubrey *et al.*[7,8]

From their investigations in uncross-linked polymer adhesives, it is clear that there are three regions of steady peeling and two regions of oscillatory peel force (stick-slip peeling). In the middle pulling-rate region, the locus of separation is at the adhesive/substrate interface, and in the lower rate region, separation occurs within the bulk of the adhesive ("cohesive" separation). Between these regions, the separation is stick-slip peeling. In cross-linked polymer adhesives, where viscous flow cannot occur, cohesive failure is absent from the velocity spectrum, and interfacial separation then extends throughout all lower rates.

However, some cross-linked polymer adhesives, such as the natural rubber/tackifier mixtures used in commercial PSA tape, have two regions of steady, adhesive/substrate interfacial separation in the two rate regions mentioned above, and in each region, the peel force shows a dependence on pulling

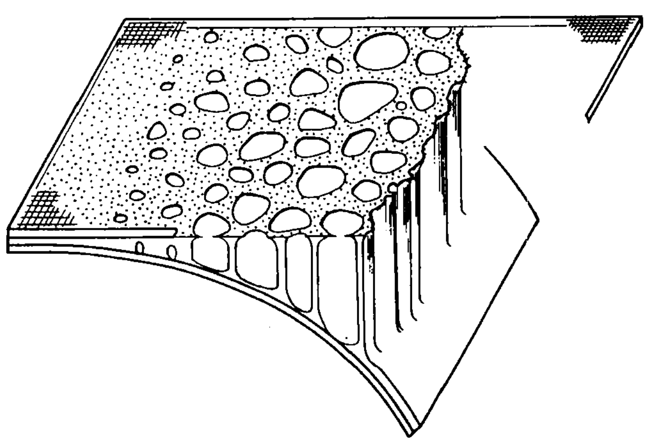

FIGURE 1 Illustration of a honeycombed stringiness pattern of a pressure-sensitive adhesive on porous backing.

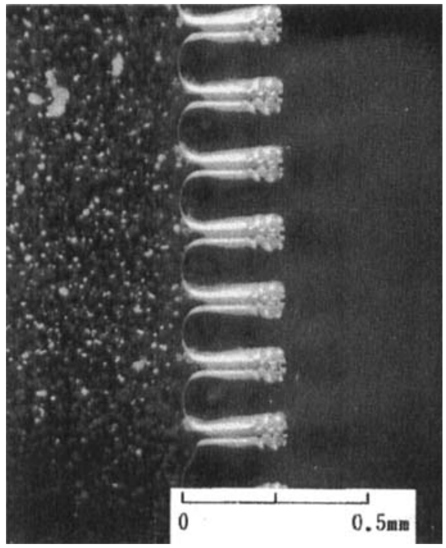

PHOTO 2 Photomicrograph of a sawtooth stringiness pattern of a pressure-sensitive adhesive on nonporous backing.

rate. This phenomenon cannot be explained from the viscoelastic properties of the polymer adhesive.

In the present author's previous papers, using Miyagi's peeling phenomenon observation apparatus, the separation of adhesive tape from an adherend (transparent substrate) was observed and the effects of various backing materials on stringiness patterns were investigated.[9,10]

The studies showed that, at a relatively low peeling speed (10 mm/min or less), the stringiness patterns could be broadly classified into two types depending on the type of backing material used: a porous backing produces a honeycombed stringiness pattern (Photo 1 and Figure 1) while a nonporous backing produces a sawtooth stringiness pattern (Photo 2 and Figure 2).

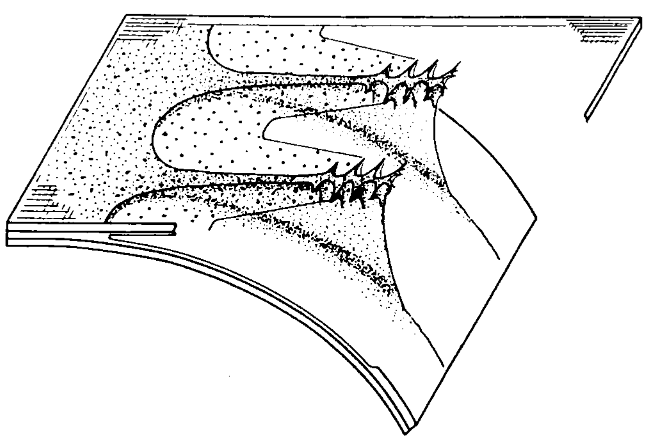

FIGURE 2 Illustration of a sawtooth stringiness pattern of a pressure-sensitive adhesive on nonporous backing.

In these photos, the peeling direction is from right to left. The peeling area is in the center of the photo. The region to the right of the peeling area has already separated from the glass substrate, and the region to the left of the peeling area is still attached to the glass substrate.

In this paper, first, using uncross-linked and cross-linked natural rubber adhesives, a 90° peel force at constant pulling rates is measured. Next, a 90° peel speed at constant peel loads is measured using Miyagi's apparatus. The difference between these 90° peeling phenomena is investigated. Finally, the stringiness phenomenon of peeling pressure-sensitive adhesive tapes is investigated. These procedures were utilized because constant pulling rates cannot be photographed. The following sections will also make clear why two steady adhesive/substrate interfacial separation regions exist.

2 EXPERIMENTAL METHOD AND SPECIMENS

2.1 Peeling Phenomenon Observation Apparatus

A schematic diagram of the observation apparatus is shown in Figure 3.[11] The adhesive tape to be tested was attached to the bottom surface of a Pyrex glass plate substrate. Peeling weight L was applied to one end of the adhesive tape, through a pulley, to peel it off. Peeling angle θ can be set to any angle by changing the position of the pulley. The peeling angle is 90° when a pulley is not used.

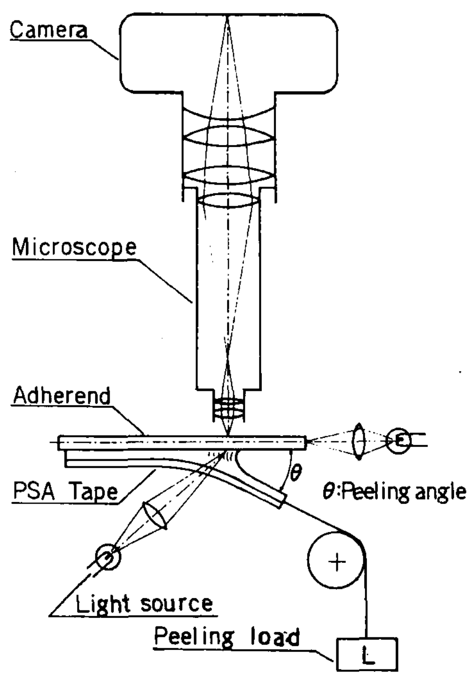

FIGURE 3 Schematic diagram of the observation apparatus.

When a small lens-focused light is shone on the peeling area of the adhesive tape, random light reflection occurs at the stringiness area of the adhesive. Thus, peeling behavior and the stringiness phenomenon can be observed with an optical microscope. A peel speed was calculated by measuring the time required for 2 mm peel-off at a constant peel load.

2.2 Specimens

A 40 μm-thick, biaxially-oriented polypropylene (OPP) film was used as a backing. Its modulus of longitudinal rigidity at 23°C was 230 kg/mm^2 and in the transverse direction was 300 kg/mm^2.

The uncross-linked adhesive used for this investigation was a mixture of 100 parts by weight of natural rubber, 60 parts by weight of aliphatic hydrocarbon resin, and 30 parts by weight of terpene resin. To prepare the cross-linked adhesive, this mixture was cross-linked by heating at 150°C for 2 minutes with 3 parts by weight of isocyanate cross-linking agent.

The modulus of the adhesives were measured under the following conditions.

Tensile tester:	Instron-type tensile tester (CRE type)
Distance between the jaws:	10 mm/min
Pulling rate:	1.0 mm/min
Atmosphere:	23 ± 2°C, 55 ± 10% r.h.

The modulus of the uncross-linked adhesive was 0.8 kg/cm^2 and that of the cross-linked one was 2.0 kg/cm^2.

Two kinds of test specimens were made by coating an OPP film backing with a 30 μm-thick adhesive. One specimen was uncross-linked and the other was cross-linked.

2.3 Observation Conditions

These PSA tapes (each 25 mm wide) were attached to an ultrasonically-cleaned glass plate substrate by rolling a rubber roller weighing 2.0 kg over the tape twice (once in each direction). They were then left standing for approximately 24 hours after which they were attached to the aforementioned apparatus and the peeling phenomenon was observed.

Peeling weights varied from 40 to 400 g/25 mm, the temperature was 23 ± 2°C and the humidity was 55 ± 10% r.h. Photos were taken when the stringiness phenomenon stabilized.

3 RESULTS AND DISCUSSION

3.1 90° Peeling at Constant Pulling Rates

The analysis of the 90° peel force curve *vs.* various pulling rates, using uncross-linked and cross-linked natural rubber as the adhesives, is shown in Figure 4.

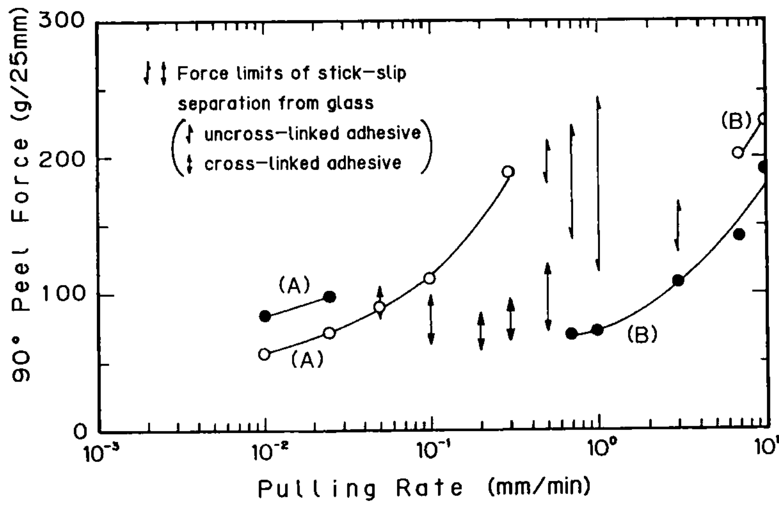

FIGURE 4 90° peel force curve *vs.* various constant pulling rates, using uncross-linked (○) and cross-linked (●) rubber as the adhesives.

It is clear that the uncross-linked adhesive showed three different types of peeling phenomena depending on the pulling rate. In a low pulling-rate range up to 0.3 mm/min. (region A), separation occurred within the bulk of the adhesive and the peel force showed a strong dependence on pulling rate. At higher rates, the locus of separation was at the adhesive/substrate interface (region B). Between regions A and B, the separation was stick-slip peeling. These results coincided with the results of Gent and Aubrey.[7,8]

At very low pulling rates, as the cohesive force of the uncross-linked adhesive is weaker than the adhesive/substrate interfacial strength, separation was caused by cohesive failure. However, at a higher pulling rate, because the cohesive force with its elastic modulus increased, the separation occurred at the adhesive/substrate interface.

In contrast, at very low pulling rates, the separation of the cross-linked adhesive occurred at the adhesive/substrate interface. Peel force showed a weak dependence on pulling rate. In the middle pulling-rate region of 0.025 to 0.7 mm/min., however, a stick-slip phenomenon occurred suggesting a transition state. When the pulling rate was raised further, the adhesive again showed its steady adhesive/substrate interfacial separation. Thus, the cross-linked adhesive was found to have two pulling-rate ranges in which steady interfacial separation occurred.

As Aubrey[12] stated, if the peel force of adhesive/substrate interfacial separation increases uniformly with the elastic modulus of the adhesive, it should increase in a simple pattern as the pulling rate increases; instead, we found that the peeling force divided into two pulling-rate ranges (see Figure 4). This phenomenon cannot be readily explained by any present viscoelastic theories.

A careful comparison of uncross-linked and cross-linked adhesive peel force revealed that the cross-linked adhesive developed the same type A/B region patterns as is normally found in uncross-linked adhesives. In other words, it is apparent that the cross-linked adhesive had a higher cohesive force causing its interfacial separation from the substrate.

Further analysis of the peel force of each adhesive, in each pulling-rate region, showed that the cross-linked adhesive had a higher adhesive strength than the uncross-linked one in region A, insofar as its higher cohesive force resulted in its interfacial separation, but a lower adhesive strength in region B because of its lower degree of elongation and viscous flow.

3.2 90° Peeling at Constant Peel Loads

3.2.1 *Measurement of peel speed* By using Miyagi's apparatus and applying constant peel loads at an angle of 90°, peel speeds and stringiness phenomena of uncross-linked and cross-linked adhesive were investigated. The peel speeds which were measured are shown in Figure 5. The peel speed curves show two different peel speeds under each peel load in the range of 50 to 200 g/25 mm.

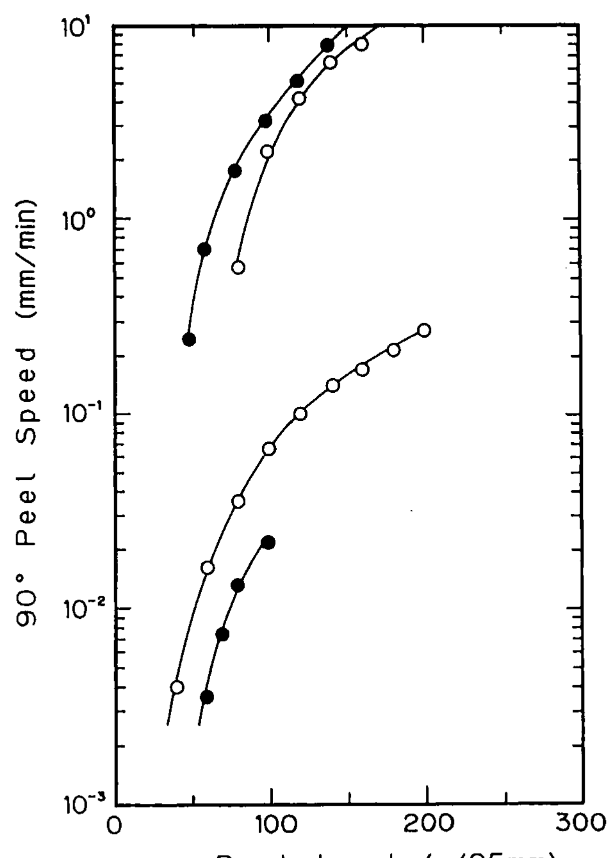

FIGURE 5 90° peel speed curve *vs.* various constant peel loads, using uncross-linked (○) and cross-linked (●) rubber as the adhesives.

Inversing and overlaying Figure 5 on Figure 4 shows that the two peel speeds correspond to the steady pulling-rate regions A and B. Here, the uncross-linked adhesive undergoes cohesive failure in region A and shows a steady adhesive/substrate interfacial separation in region B. On the other hand, the cross-linked adhesive undergoes steady adhesive/substrate interfacial separation in both regions A and B. These results coincide with the results of the peeling test at constant pulling rates. Here again, conventional viscoelastic theories are useless for explaining the presence of two different peel speeds at which steady adhesive/substrate interfacial separation occurs under the same peel loads.

3.2.2 *Stringiness phenomena* The stringiness patterns of the uncross-linked adhesive in region A (cohesive failure) and in region B (interfacial separation) are shown in Photos 3 and 4, respectively. In Photo 3, the stringiness, showing cohesive failure of the adhesive, appears as greatly elongated sawtooth forms. Further, the photo clearly shows that some adhesive still remains attached to the substrate. The adhesive remaining on the substrate and the webs extending to the leading edge, though not shown in Photo 3, make it impossible to determine the stringiness width. The stringiness patterns of the interfacial separation shown in Photo 4 are simple, regular sawtooth forms, having small webs near the bottom of each sawtooth. Each tooth has a rounded tip without any small projection suggesting that no concentration of stress occurred.

The stringiness pattern of the cross-linked adhesive in regions A and B (interfacial separation in both of the cases) are shown in Photos 5 and 6, respectively. Figures 6 and 7 show stringiness pattern illustrations derived from Photos 5 and 6, respectively.[9,10] These patterns are entirely different from each other, even though both of them represent steady adhesive/substrate interfacial separation.

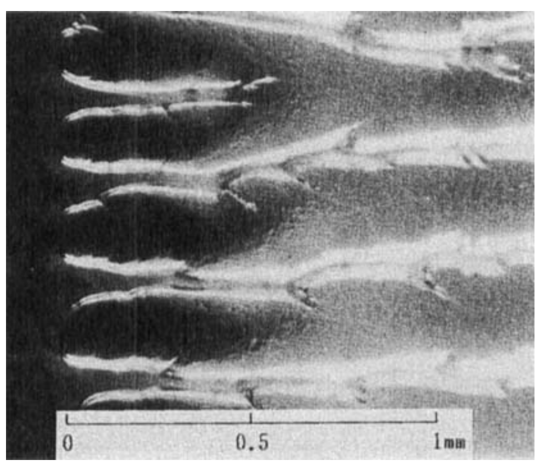

PHOTO 3 Photomicrograph of the stringiness pattern of the uncross-linked natural rubber adhesive at a peel load of 100 g/25 mm in region A.

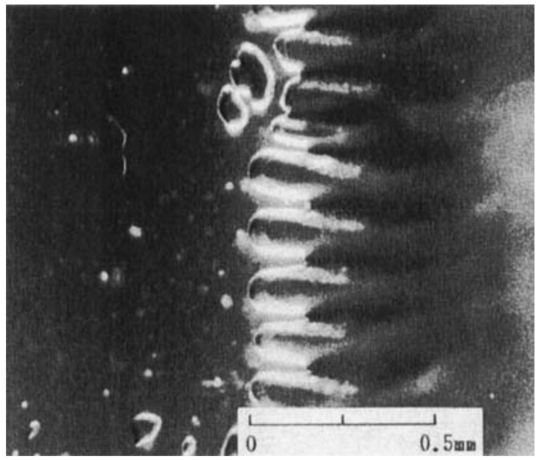

PHOTO 4 Photomicrograph of the stringiness pattern of the uncross-linked natural rubber adhesive at a peel load of 100 g/25 mm in region B.

The pattern shown in Photo 6 is the same as that shown in Photo 4. It follows, therefore, that the behavior of the interfacial separation of both adhesives is the same in region B. On the other hand, the pattern shown in Photo 5 is entirely different from not only that shown in Photo 6, but also from that shown in Photo 3. The stringiness pattern in region A has a sawtooth stringiness with webs extending to the leading edge of the peeling area. Here it can be seen that adhesive is detaching cleanly from the substrate. This interfacial separation form

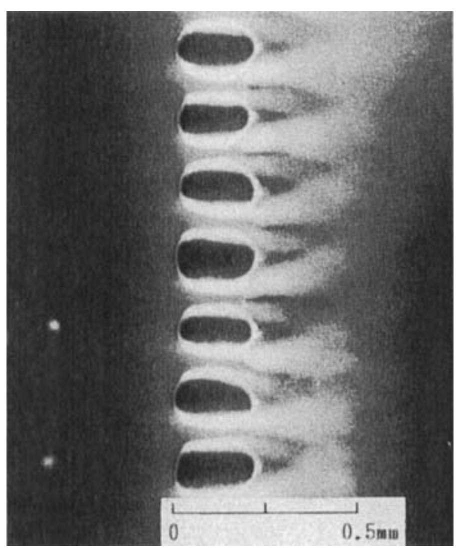

PHOTO 5 Photomicrograph of the stringiness pattern of the cross-linked natural rubber adhesive at a peel load of 100 g/25 mm in region A.

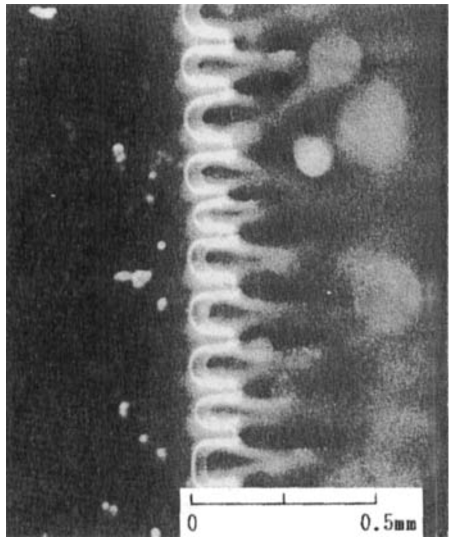

PHOTO 6 Photomicrograph of the stringiness pattern of the cross-linked natural rubber adhesive at a peel load of 100 g/25 mm in region B.

can be considered as a variation of the stringiness pattern shown in Photo 3 of the uncross-linked adhesive which resulted from cohesive failure. It is apparent that the higher cohesive force of the cross-linked adhesive results in interfacial separation instead of cohesive failure. Also, at the right-hand edge of stringiness structure shown in Photo 5, many small projections can be seen which indicate that the concentration of stress occurred in that area. The number of these stress concentration points makes a difference of at least one peel speed figure between regions A and B.

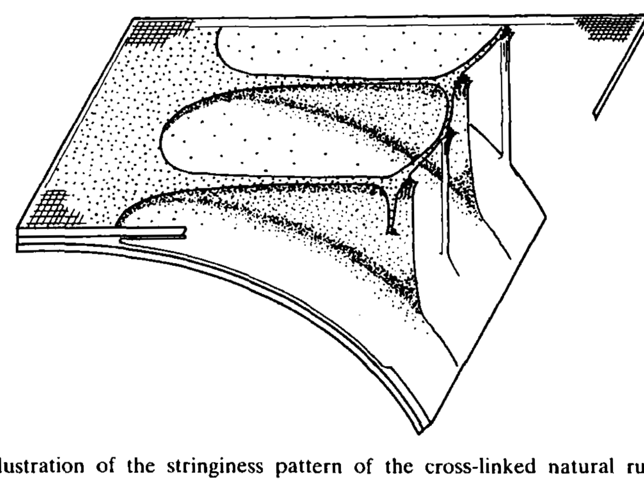

FIGURE 6 Illustration of the stringiness pattern of the cross-linked natural rubber adhesive in region A.

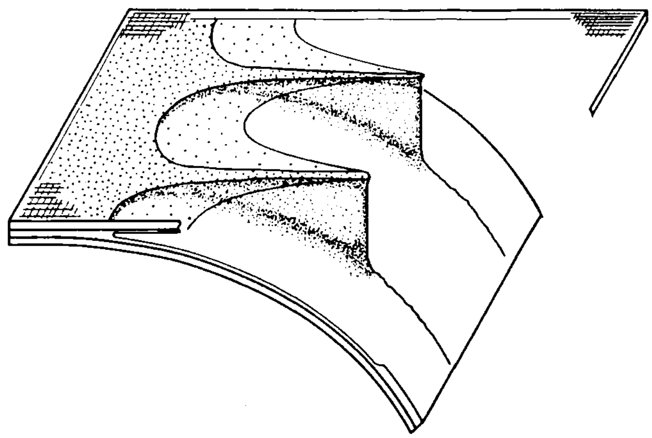

FIGURE 7 Illustration of the stringiness pattern of the cross-linked natural rubber adhesive in region B.

4 CONCLUSION

As seen in the previous sections, present viscoelastic theories could not account for the two regions of interfacial separation in lightly cross-linked, pressure-sensitive adhesive. Investigation of the stringiness phenomenon of peeling pressure-sensitive adhesive tapes at constant peeling loads shows that, there are two peeling speed regions where steady interfacial separation occurs. These two regions correspond to the steady pulling-rate regions mentioned above. The existence of these two regions of interfacial separation can be explained by the differences in their stringiness patterns (Figure 8).

The existence of two steady peeling regions at low pulling rates is not due to the variation in the viscoelastic state of the adhesive caused by peeling speed but, rather, to the changes in the stringiness pattern concentrated in the pulling force area.

The existence of two regions of steady interfacial separation in cross-linked pressure-sensitive adhesive, which could not be explained by using present peel adhesion theories based on the viscoelastic properties of high polymer when measured at constant pulling rates, can now be explained by the investigation of the stringiness phenomenon. However, as this paper concentrated only on the effect of peeling loads, further research into polymer structure, adhesive thickness, effect of substrates, etc., will be necessary before this stringiness phenomenon is completely understood. This field of investigation, when added to the present viscoelastic property studies should lead to a new peeling adhesive theory which, in turn, may lead to the development of high peel force pressure-sensitive adhesives.

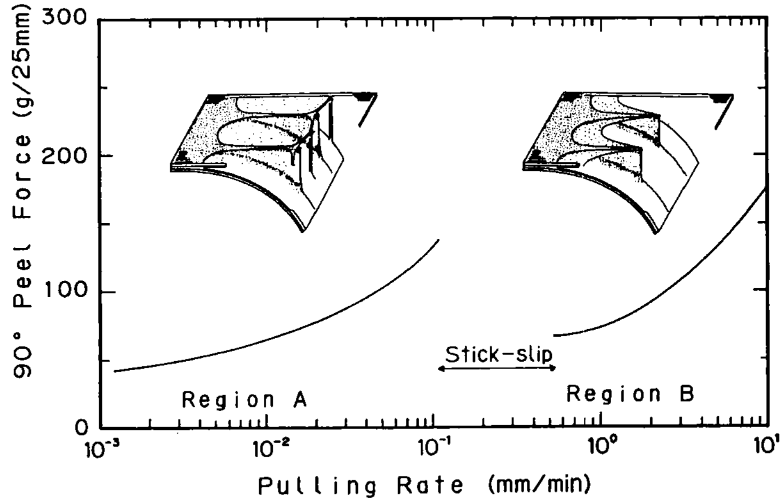

FIGURE 8 90° peel force curves and stringiness patterns in two pulling rate regions of a cross-linked pressure-sensitive adhesive.

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