Experiments on Peeling

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I. INTRODUCTION

On the assumption that stripping force is determined by the geometry of the joint and the rheological properties of adherend and adhesive and is not influenced by any adhesion forces acting across the adherend–adhesive interfaces, an equation:

$$W_0 = 0.3799 w\sigma (E/E_1)^{1/4}\delta^{3/4}y_0^{1/4} \qquad (1)$$

was derived.[1] In it, W_c is the minimum peeling force (acting perpendicularly to the adhesive film), w the width of the ribbon being stripped off, σ the tensile strength of the adhesive; E and E_1 are the moduli of elasticity of ribbon and adhesive, respectively, δ is the thickness of the ribbon; and y_0 is the initial thickness of the adhesive film.

Equation (1) is only a first approximation, as it assumes both the ribbon and the adhesive to be Hookean solids, and also because it disregards stress concentrations at various boundaries of the "glue line." In this respect, eq. (1) is on the same level of precision as are the assumptions that the stresses in butt joints are pure tensile, and those in lap joints are pure shear stresses. A more thorough theoretical treatment of butt and lap joints was inaugurated several years ago (see, e.g., references 2–5); in this paper an attempt is made to achieve, mainly by experiments, an analogous advance in our understanding of the peeling procedure.

II. THEORY OF PEELING THROUGH NON-HOOKEAN ADHESIVES

Figure 1 indicates the system to which eq. (1) and the following equations are supposed to be applicable. R is a flexible ribbon, P a rigid plate, W force acting normally to P, y_0 the initial thickness of the adhesive film, and $(y + y_0)$ the thickness of the extended adhesive layer which, of course, varies with the distance x from the point of application of force.

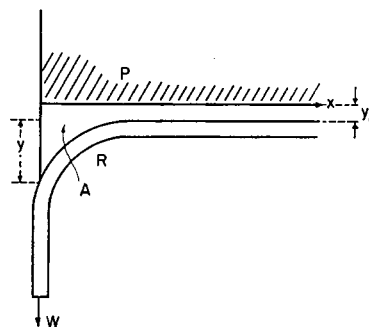If the ribbon is a Hookean solid and if force φ

Fig. 1. Scheme of peeling: (P) rigid plate; (R) ribbon; (A) adhesive; (W) external load.

acts on every rectangle ($w \times dx$ cm.²) of it, then, from the theory of beams on elastic foundation,[6,7]

$$d^4y/dx^4 = -12\varphi/E\delta^3 w dx \qquad (2)$$

If the adhesive is also a Hookean solid, $\varphi = (y/y_0) E_1 w dx$, and integration and introduction of suitable boundary conditions result in eq. (1). Integration is feasible also when stress is proportional to (strain)^n, n being different from unity. Let e_1 be the factor of proportionality; then

$$\text{stress} = e_1(y/y_0)^n \qquad (3)$$

and force is given by

$$\varphi = e_1(y/y_0)^n w dx \qquad (4)$$

Thus,

$$d^4y/dx^4 = (12e_1/E\delta^3 y_0{}^n)y^n \qquad (5)$$

This differential equation is satisfied, for instance, by setting

$$y = Ax^m \qquad (6)$$

$$A = \left[\frac{3(1-n)^4}{(3+n)(2+2n)(1+3n)} \cdot \frac{e_1}{E\delta^3 y_0{}^n}\right]^{1/(1-n)} \qquad (6a)$$

$$m = 4/(1-n) \qquad (6b)$$

The external load W, as long as it is too small to cause peeling, must be equal to the integral of φ extended over the affected length, l, of the ribbon. Thus:

$$W = \int_0^l e_1(y/y_0)^n w\,dx$$
$$= (1 - n/3n + 1)(e_1 w A^n/y_0^n)l^{(3n+1)/(1-n)} \quad (7)$$

At rupture,

$$W_0 = (1 - n/3n + 1)(e_1 w A^n/y_0^n)l_0^{(3n+1)/(1-n)} \quad (8)$$

if l_0 is that length of the ribbon which departs from its initial position just as peeling starts. As determination of l_c is impossible, we introduce the maximum extension Y of the adhesive film, that is, the value of y at the point of application of load W_0. From eq. (6), $Y = Al_0^{4/(1-n)}$; and from eq. (3), $Y^n = \sigma y_c^n e_1$, σ being again the maximum tensile stress possible in the adhesive. Thus,

$$l_c^{4/(1-n)} = Y/A = (\sigma^{1/n}/e_1^{1/n})(y_c/A) \quad (9)$$

and, with eq. (8),

$$W_0 = (1 - n/3n + 1)(w\sigma^{(3n+1)/4n}y_0^{(1-n)/4}/$$
$$e_1^{(1-n)/4n}A^{(1-n)/4}) \quad (10)$$

Introducing A from eq. (6a), we obtain

$$W_0 = [(3 + n)(2 + 2n)/3(1 + 3n)^3]^{1/4}$$
$$(w\sigma^{(3n+1)/4n}E^{1/4}\delta^{3/4}y_0^{1/4}/e_1^{1/4n}) \quad (11)$$

If we set $n = 1$,

$$W_0 = 0.537 w\sigma(E/e_1)^{1/4}\delta^{3/4}y_0^{1/4} \quad (12)$$

thus, the wrong equation (12) differs from the correct equation (1) only by its numerical constant; eq. (12) cannot be correct because m in eqs. (6b) and (6) would be infinitely great if n were equal to unity.

The value of n is less than one, probably for all customary adhesives. According to eq. (11), W_0 increases, then, more rapidly than proportional to σ; for instance, when $n = 0.5$,

$$W_0 = 0.688(w\sigma^{1.25}E^{0.25}\delta^{0.75}y_0^{0.25}/e_1^{0.5}) \quad (13)$$

Equations (3) to (13) are reproduced here because, presumably, such experimental conditions can be devised that the approximations on which the theory is based are valid, but these approximations proved to be invalid in the experiments described in this paper.

III. EXPERIMENTAL PROCEDURE

The experimental set-up is represented in Figure 1. The rigid plates were of glass (Gold Seal Micro Slides), $7.6 \times 5.0 \times 0.12$–$0.13$ cm. The ribbons were of aluminum (donated by Aluminum Co. of America and Reynolds Metal Co.) and, in a few instances, of nickel A. The two types of aluminum used most were (a) 1235 alloy, 0 temper, dry annealed, 0.0076 cm. thick, and (b) 1145 alloy, H-19 temper, 0.0025 cm. thick. Two different materials (glass and metal) were employed to facilitate detection of deleterious impurities in the adhesive; if this does not adhere well to one solid and adheres to the other, the weak boundary layer is likely to have its origin in the first solid, but if the adhesive falls off both glass and metal, the cause of non-adherence should be looked for in the adhesive itself. Having glass as one of the adherends offered also the advantage of being able to inspect the joint before breaking and to eliminate those containing large bubbles or similar imperfections.

Polyethylenes were the chief adhesives utilized. To avoid formation of weak boundary layers, they were dissolved in boiling toluene or cyclohexane and precipitated with acetone.[8,9] A series of polyethylenes with graded mechanical properties was obtained by mixing a polyethylene wax (Epolene N donated by Eastman Chemical Products, Inc.) with a low-pressure, high-molecular polyethylene (Marlex 50), in the ratios 1:0, 1:0.67, 1:1.5, and 1:3; pure Marlex 50 discolored too easily at the temperatures needed to make the extensive joints used. The mixing, as a rule, was achieved by dissolving both ingredients in boiling toluene and vaporizing the solvent.

The preparation of joints generally comprised the following steps. (1) The aluminum ribbons were rubbed (100 strokes) with a cloth saturated with a suspension of Alpha Polishing Alumina No. 2 in distilled water and rinsed with distilled water. After superficial drying in air they were rubbed with tissue paper saturated with distilled acetone and degreased in the vapor of distilled toluene. Apparently, contaminations from untreated parts of the ribbon did not diffuse onto the treated surface in significant amounts, as the stripping force was independent of whether one or both sides were polished, washed, and degreased as above. A more prolonged polishing apparently had no effect on the minimum peeling stress. Each ribbon was discarded after just one test.

(2) The glass slides were degreased in toluene vapor. In many instances, a slide could be used for
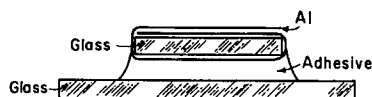
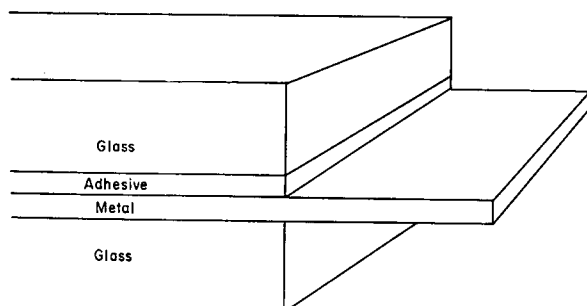Fig. 2. An intermediate stage in making adhesive joints.
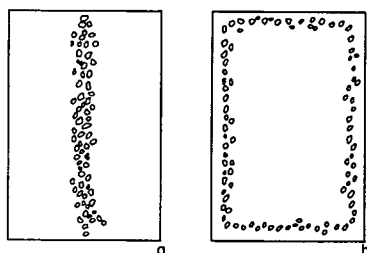

Fig. 3. A method of making adhesive joints.

Fig. 4. Two methods of distributing the adhesive on the adherend.

several consecutive experiments. Between these, the remaining polyethylene was carefully scraped off with a razor blade and extracted with acetone and boiling toluene. As this treatment did not remove the last traces of the adhesive, a more drastic procedure was resorted to whenever the adhesive was changed; in these instances the slides were boiled in a solution of chromic acid and sulfuric acid before being washed with water and acetone and degreased.

(3) In order to give a degree of dimensional stability to the foils which are so easily deformed, the aluminum ribbons were tightly wrapped around microscope slides, 7.5 × 2.5 × 0.1 cm., and joints were made as indicated in Figure 2. As nickel foils could not be wound in this manner, rectangles of, e.g., 10.0 × 5.0 × 0.005 cm., were flattened on a steel plate alleged to be flat within 0.00025 cm., supported on one face by a glass slide, and glued along the opposite face to a similar slide (see Fig. 3). Later, also aluminum was glued in this fashion.

(4) The adhesive was applied as a powder, as thin slides, or as a gel in toluene, either to both glass and metal or to one of the adherends; none of these

variations had a detectable effect on the peeling force. In all instances, the adhesive was heated on the adherend until so much attachment developed that further handling was possible without losing any of the material. The temperature during this operation was not determined but it presumably corresponded to the lower half of the melting range.

The details of the initial distribution of the adhesive proved important. In a deliberate comparison on a system of Epolene/Marlex (1:0.67), hard aluminum foil (0.0025 cm. thick), and glass, the powder was spread once along the axis of the slide (see Fig. 4a) and, in a parallel experiment, around it (Fig. 4b). Further treatment was identical in both instances. After melting and solidification, polyethylene showed no large bubbles in the arrangement in Fig. 4a, while the adhesive layer of Figure 4b contained bubbles up to 0.8 cm. in length. The stripping force for 2-cm. wide ribbons was 538 and 725 g.-wt. for the first, and 333 and 396 g.-wt. for the second arrangement; obviously, bubbles entrapped by a polyethylene corral can lower the resistance to stripping almost to one-half.

In the most frequent procedure, six sandwiches of the types illustrated in Figures 2 and 3 were interleaved with sheets of a thin aluminum foil (to prevent spreading of glue from one sandwich to another), placed one on top of the other, and wrapped in another sheet of foil (to prevent sliding when the adhesive is liquid). A thermocouple (sometimes two thermocouples) was stuck in the middle of the pile, and the pile was heated in an oven. The oven used in earlier experiments was large and could not be evacuated; when nitrogen was passed through it, the concentration of oxygen remaining in the oven was presumably still quite considerable. The newer oven was home-made, its inside dimensions were 19.0 × 16.5 × 6.5 cm.; although it was not truly airtight, it could be emptied easily with a vacuum pump and filled with pre-purified nitrogen. Nitrogen atmosphere was necessary to depress or suppress discoloration of polyethylene when heated above 180 or 200°C. The excellent adherence of polyethylenes to solids, attained in nitrogen, is another argument against the hypothesis that oxygenated or similar polar groups are required for adhesion.

In a few preliminary experiments it was observed that, when in each sandwich the metal was above the glass, the peeling force was smaller than when glass was above metal; the latter arrangement was used in the majority of experiments reported.

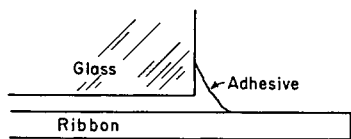The "pile of sandwiches" was rapidly (e.g., in 15

Fig. 5. Accumulation of the adhesive at the front edge.

minutes) heated to the appropriate temperature, kept near this temperature for, for instance, 30 min., and then permitted to cool with the oven; this cooling period lasted at least one hour. The maximum temperature and the duration of heating were found for each adhesive by trials.

A weight was placed on the pile before heating and kept on until the end of the cooling period. It varied between 240 and 4800 g., corresponding to pressures between 1.3×10^4 and 25×10^4 dynes/cm.². No definite effect of the pressure could be detected because higher pressure meant thinner adhesive films, and these two parameters could not be separated.

Whenever a factor other than the nature of the adhesive was varied, the different joints were assembled in one pile and heated and cooled together. This was true in the experiment on the importance of adhesive distribution, in the comparison of different ribbons, and so on.

After cooling, the foils were cut to size, the plates placed in a horizontal position (checked with a precision level), and weights applied to their outstanding length (see Figs. 1 and 3) until slow peeling (of the order of 0.1 cm./sec.) was noticed. If the movement was seen to accelerate, some weight was taken off; when the movement stopped, an additional load was put on. Thus, several values for the stripping force were found in each test, and a mean value could be calculated.

Two types of values were not counted. Usually, a part of the adhesive was squeezed out of the clearance and on the projecting length of the rib-

bon, forming a bond between the latter and the 5.0×0.12 cm. side of the glass plate (see Fig. 5). Breaking this bond usually required a force several times as great as that needed for the further stripping; often the ribbon broke rather than the adhesive outside the clearance. Thus, either the excess of the adhesive was removed before measurement or the force needed to rupture it was disregarded. The other cause of excessive values is pictured in Figure 6. The adhesive, instead of breaking at the knee of the ribbon, formed a "curtain" which sometimes extended for several millimeters. The minimum force for rupturing the curtain was, e.g., twice or three times as great as the regular stripping force.

Dead weight loading was preferred to stressing in a machine because it guarantees a 90° angle of peel; a complicated aligning device would be called for to achieve the constant stripping angle of 90° in a tensile test machine.

The stress–strain curves of the adhesive employed were determined on films made in a manner almost identical with that of making the joints. A glass slide was covered with a sheet of thin aluminum foil, a calculated amount of adhesive was spread along the central line of the sheet, another piece of aluminum foil was placed on top of the adhesive layer, and a second glass slide put on top of the "sandwich." Such a five-layer composite body was substituted for a joint in a pile described above, and the pile was heated and cooled together; in some instances, the pile comprised more than one five-layer body and fewer than five joints. Thus, the thermal history of the adhesive film was identical with that of the adhesive joints. Only the subsequent operations were different: the aluminum foil was dissolved in a sodium hydroxide solution, the liberated film of the adhesive was rinsed with distilled water, dried in air at room temperature, and strained by known forces.

IV. EXPERIMENTAL RESULTS

1. The Magnitude of the Peeling Force

The force required for stripping is considerably smaller than that calculated from eqs. (1) or (13). This is illustrated in Table I. Its first column lists the polyethylenes used; the second column contains the average value of peeling force W_0 determined on aluminum ribbons 1.0 cm. wide and 0.0076 cm. thick; the third column shows the mean thickness of the "glue line," and in the fourth and fifth columns the values of W_0 calculated from eqs.
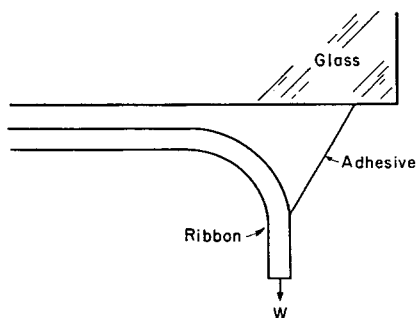

Fig. 6. An "irregular" mode of rupture in the adhesive.

TABLE I
Stripping Force (for 1 cm. Width) of Aluminum-Polyethylene-Glass Joints

| Polyethylenes | W_0, dynes | y_0, cm. | W_0 calculated, dynes | |
			From eq. (1)	From eq. (13)
Epolene	1.0×10^5	0.009	5.79×10^5	—
Epolene + Marlex (1:0.67)	2.5×10^5	0.011	7.49×10^5	—
Epolene + Marlex (1:1.5)	4.2×10^5	0.010	7.32×10^5	14.3×10^5
Epolene + Marlex (1:3)	7.4×10^5	0.019	—	27.2×10^5

(1) and (13), respectively, are given. In computing the averages of the experimental W_0 values, the low results due to visible bubbles and similar defects were included on equal footing with the others, and the analogous data were used when computing the breaking stress σ which appears in the above equations. If the results obtained on defective adhesive films are disregarded, the magnitude of both the experimental W_0 and the stress σ would be raised by 20 to 40%, but the ratio of the theoretical to the experimental W_0 would not be markedly altered.

The following values, derived from our measurements, were used to calculate the theoretical values of W_c. It may be repeated here that they were determined on thin cast foils (usually between 0.01 and 0.02 cm. thick) and thus may be very different from the corresponding mechanical constants found on large specimens or on extruded foils. The values obtained were for Epolene, $\sigma = 5.0 \times 10^7$ baryes and $E_1 = 3.2 \times 10^9$ baryes; for Epolene + Marlex (1:0.67), $\sigma = 4.7 \times 10^7$ and $E_1 = 1.09 \times 10^9$ baryes; and for Epolene + Marlex (1:1.5), $\sigma = 4.6 \times 10^7$ baryes and stress $= 1.84 \times 10^8$ (strain)$^{0.5}$. The actual stress–strain curve was not a parabola, but the error introduced by using an approximate equation is not serious enough to affect our conclusions. For calculating W_0 according to eq. (1), the value of E_1 was assumed to be 10^9 baryes; the modulus of elasticity of aluminum is given in tables as 7×10^{11} baryes. Values obtained for Epolene + Marlex (1:3) were $\sigma = 0.7 \times 10^7$ baryes and stress $= 2.0 \times 10^8$ (strain)$^{0.5}$. This equation also is a crude approximation only.

2. Effect of the Ribbon

The effect of the ribbon thickness δ was smaller than expected and depended on the adhesive. Soft aluminum ribbons of 0.0076 and 0.0025 cm. thickness were compared. Both eqs. (1) and (13) predict the peeling force to be 2.3 times as great for the thick as for the thin foil. The experimental results are summarized in Table II.

The absolute values of W_0 are different from those of Table I because in the latter case the values were taken from experiments in which adhesive joints and free adhesive foils were constituents of one pile of "sandwiches," while in the experiments on which Table II is based each pile comprised three thick and three thin ribbons. Thus, the heating regime and some other details were different for Table I and Table II.

TABLE II
Peeling Strength (for 1 cm. Width) and Ribbon Thickness

	Epolene + Marlex (1:0.67)	Epolene + Marlex (1:1.5)	Epolene + Marlex (1:3)
	Mean W_0, dynes, $\times 10^5$		
$\delta = 0.0025$ cm.	2.55	4.2	9.3
$\delta = 0.0076$ cm.	4.5	4.4	8.0
	Mean y_0, cm.		
$\delta = 0.0025$ cm.	0.006	0.012	0.021
$\delta = 0.0076$ cm.	0.006	0.011	0.020
	Number of samples		
$\delta = 0.0025$ cm.	3	15	6
$\delta = 0.0076$ cm.	3	15	6
	Extreme values of W_0, dynes, $\times 10^5$		
$\delta = 0.0025$	2.1–3.2	2.4–5.9	5.2–13.3
$\delta = 0.0076$	2.8–6.3	2.1–5.8	5.1–12.4

It is seen that W_0 increased with δ in the instance of the 1:0.67 mixture only, and also this increase was smaller than expected. When the ratio of Epolene to Marlex decreased, the effect of δ on W_0 also decreased and apparently even became negative.

The temper (or degree of annealing) of the aluminum foil should not affect the peeling force because the modulus of elasticity E of the metal is independent of the temper. In reality, the W_0 values for a very hard foil (temper designation −H19) were 1.5–1.8 times as great as for a very soft foil of an identical thickness (0.0025 cm.), at least for the 1:3 mixture of Epolene and Marlex. The experimental data were: (1) w = 0.5 cm.; hard Al: $y_0 =$

0.015 cm., W_0 in three experiments: 7.35, 5.39, and 5.88 $\times$ 10⁵ baryes; soft Al: $y_0 = 0.013$ cm., W_0 in three experiments: 3.43, 2.75, and 4.02 $\times$ 10⁵ baryes; (2) $w = 1.5$ cm.; hard Al: $y_0 = 0.015$ cm, W_0 in three tests: 15.7, 11.5, and 7.45 $\times$ 10⁵ baryes; soft Al: $y_0 = 0.013$ cm., W_0 in two tests: 6.67 and 7.16 $\times$ 10⁵ baryes. The highest of the above values for $w = 0.5$ cm. is almost equal to the W_0 for $w = 1$ cm. listed in Table I; thus, the force required to peel a hard ribbon is nearer to the theoretical force than that needed for a soft foil.

When a soft A nickel strip, 0.0053 cm. thick, was compared with a soft aluminum strip, 0.0076 cm. thick, identical peeling forces were expected because the modulus of elasticity of "A" nickel (containing 0.6% of material other than nickel and cobalt) is given in tables as 21×10^{11} baryes, and $(21 \times 10^{11})^{1/4}(0.0053)^{3/4}$ is by only 3% greater than $(7 \times 10^{11})^{1/4}(0.0076)^{3/4}$. In reality, the nickel joints appeared a little stronger than aluminum joints, perhaps because soft nickel has a much higher yield strength than soft aluminum and in this respect is comparable to hard aluminum. The experimental data were for Epolene + Marlex (1:3), $w = 1.0$ cm; (1) nickel: $y_0 = 0.019$ cm., W_0: 6.13, 5.58, and 6.33 $\times$ 10⁵ baryes; (2) aluminum; $y_0 = 0.020$ cm., W_0: 5.28, 4.60, and 5.66 $\times$ 10⁵ baryes.

The length of the ribbon still attached to the glass was varied between 0.6 and 7.5 cm., but no significant difference was detected.

V. EXPERIMENTS ON STRESS CONCENTRATION

As the experimental values of the stripping force were consistently lower than those predicted, the possibility of weak boundary layers was considered. A more thorough polishing, washing, and degreasing of aluminum had no effect on W_0. No significant difference was noticed also between the values of W_0 observed on fresh glass plates and on glass plates from which not all polyethylene was removed. That our polyethylenes contained no material giving rise to weak boundary layers was shown by earlier experiments.[9]

The amount of polyethylene remaining on the adherends after the rupture was estimated visually, and all joints were divided in five classes, namely: (I) almost everything on glass; (II) more on glass than on metal; (III) comparable amounts; (IV) less on glass than on metal; and (V) almost nothing on glass. As an example, 21 stripping experiments with the 1:3 mixture of Epolene and Marlex, aluminum, and glass gave eight cases in group I, two in group II, seven in group III, three in group

IV, and one in group V. When the adhesive film was slightly undercut (for instance, for the depth of 0.1 cm.) at the polymer-glass interface so that peeling started nearer to glass than to aluminum, the results were, for group I, II, III, IV, and V, 9, 11, 24, 15, and 4 cases, respectively. It seems impossible to account for these data by the existence of a weak boundary layer.

As pointed out in an earlier paper,[10] a deformation analogous to those occurring in butt joints must take place in peeling. Let the Cartesian coordinate x be plotted parallel to the longest dimension of the ribbon, and z parallel to the width of the latter. When a force acting in the direction $+y$ is applied, the adhesive expands toward $+y$, but it must contract along the $-z$ and $+z$ directions, as indicated in Figure 7.
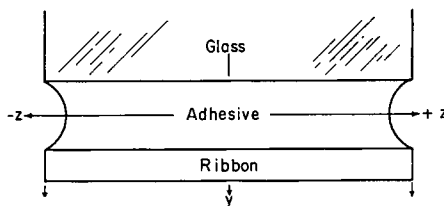

Fig. 7. Stress concentrations during peeling.

Because of these deformations, there will be a stress concentration near the right-hand and the left-hand ends of the adhesive film of Figure 7. The dependence of the stress concentration factor on z is unknown but, as a crude approximation, we may consider the width w of the adhesive film as consisting of two parts; one part, of w_1 centimeters, is the sum of the two deformed regions indicated in Figure 7, and along the rest (i.e., $w - w_1$) the stress is practically uniform. According to Saint-Venant's principle, w_1 must be of the order of the initial thickness of the adhesive film (i.e., y_0). Let f_1 be the mean value of force/width acting along stretch w_1, and f_0, the analogous ratio in $(w - w_1)$; naturally, f_0 must be smaller than f_1. Thus, $W_0 = f_1 w_1 + f_0(w - w_1)$ or

$$W_0 = f_0 w + (f_1 - f_0)w_1 \qquad (14)$$

meaning that W_0 should be an (approximately) linear function of w instead of being simply proportional to w as postulated in eqs. (1) and (13). Figure 8 confirms the expectation for the instance of the 1:3 mixture of Epolene + Marlex, aluminum (0.0076 cm. thick), and glass; the thickness of the adhesive was 0.019–0.029, mean 0.024 cm. The
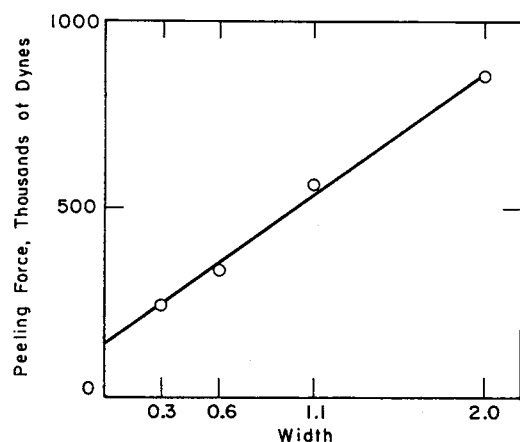
Fig. 8. Peeling force as a function of width (in cm.) for a flexible adherend.

3.7 × 10⁴ and 6.6 × 10⁴, six ribbons of 1.1 cm. gave 6.2×10^4 to 13.1×10^4; and six ribbons of 2.0 cm., 11.4×10^4 to 20.5×10^4 dynes. Heating temperature was 126°. The thickness of the glue line varied between 0.0029 and 0.0050 cm. The mean values of W_0 and the ratio W_0/w are gathered in Table III.

TABLE III
Ribbon Width (w) and Peeling Force (W_0) for Epolene N

w, cm.	W_0, dynes ($\times 10^5$)	W_0/w, dynes ($\times 10^4$/cm.)
0.3	0.24	8.0
0.6	0.51	8.5
1.1	0.99	9.0
2.0	1.70	8.5

experimental points are averages of all readings (24 to 30 per point) made on six specimens (thus, the average number of readings per specimen was 4.5) because this method of averaging gives a slightly better straight line than the means of six tests. Because of the wide spread of individual values, usual in adhesion measurements, the position of the line is not very definite, but it still appears worthwhile to test whether the value of f_1w_1 derived from the graph can have the meaning attributed to it. As f_1w_1 is about 1.5×10^5 dynes and as w_1 must be about 0.03 cm., f_1 comes out equal to 50×10^5 dynes/cm., i.e., not very different from the theoretical value listed in Table I, namely, 27×10^5 dynes/cm. The f_0 in eq. (14) is about 3.5×10^5 dynes/cm., i.e., really smaller than f_1.

The ratio W_0/w should be smaller the greater w, according to eq. (14). This has been observed many times on the Epolene + Marlex (1:3) mixture; unfortunately, only two or three widths usually were compared so that a graph analogous to Figure 8 would not be convincing. For example, the values listed for hard and soft Al in Section IV give W_0/w equal to 1241 and 680 dynes/cm. for $w = 0.5$ cm., and 770 and 461 dynes/cm. for $w = 1.5$ cm.

The ratio W_0/w decreased when w increased also in the instance of the Epolene + Marlex (1:1.5) mixture. Thus, for four strips 0.5 cm. wide, it was 5.0×10^5 to 6.8×10^5 dynes/cm., and for four strips 1.0 cm. wide, 3.2×10^5 to 5.3×10^5 dynes/cm.

However, W_0 was proportional to w, that is, W_0/w was constant, in the instance of Epolene alone. Four ribbons (Al, 0.0076 cm.) of 0.03 cm. gave W_0 ranging from 1.7×10^4 to 3.3×10^4 dynes; five ribbons of 0.06 cm. afforded values between

The striking difference between Epolene and the 1:3 mixture was not likely to be caused by any chemical difference; however, the first material is brittle and the second highly flexible, and this dissimilarity was suspected to be responsible for the behavior observed. To test the hypothesis, another brittle substance of unlike composition was selected, a polyvinyl acetate of molecular weight 12,000 (according to the trade literature), designated as Gelva V7 and donated by Shawinigan Resins Corporation. Joints with Gelva were prepared in the same manner as the Epolene joints; the oven temperature was about 135°. Results are listed in Table IV. Four aluminum ribbons, 0.0076 cm. thick, were used for every width; the thickness of the adhesive film was 0.016–0.020 cm. It is seen that W_0/w is constant within the limits of the experimental error. Thus, the proportionality between W_0 and w seems to be a property of brittle adhesives.

TABLE IV
Ribbon Width w and Peeling Force W_0 for Gelva V7

w, cm.	Range of W_0 values, dynes ($\times 10^5$)	Mean W_0, dynes ($\times 10^5$)	W_0/w, dynes ($\times 10^5$/cm.)
0.3	0.29–0.50	0.40	1.33
0.6	0.70–1.01	0.80	1.33
1.1	1.51–1.94	1.70	1.55
2.0	2.63–3.13	2.92	1.46

Peeling of Epolene and Gelva joints differed from that of Epolene–Marlex joints also in another respect. When the joints with brittle adhesives were peeled, no visible film remained on the metal ribbon. It was attempted to measure the thickness

of the invisible film with a micrometer gage and, in the instance of polyethylene wax, values of a few microns usually were obtained. As nothing was known about the existence of weak boundary layers on Gelva, a special experiment was performed. A piece of aluminum foil, 0.0076 cm. thick, was glued to a glass slide with adhesionable Marlex 50, a calculated amount of Gelva placed on the foil, covered with another piece of the foil, and heated (to 134°). Both aluminum pieces were cut from the same roll and treated in identical manner, but in every instance practically the whole polyvinyl acetate remained on the foil stiffened with glass after stripping while nothing was visible on the flexible ribbon. Obviously, the act of bending the ribbon weakened the adhesive near the bend.

This effect can be understood if the bend is looked at more closely. The stress in the ribbon (considered as a Hookean solid) at the boundary with the adhesive is $\delta E/2R$, R being the radius of curvature of the ribbon. At the bend, this R was near 0.1 cm. in our experiments. Taking $\delta = 0.0076$ cm. and $E = 7 \times 10^{11}$ dynes/cm.^{2} (for aluminum), we obtain stresses of about $2.7 \times 10^{10}/$dynes/cm.^{2}. Plastic deformation of soft aluminum foil starts (according to tables) at about 4×10^8 dynes/cm.^{2}; thus, the metal must flow near the adhesive. This will not affect the adhesive as long as it also can be plastically deformed. However, when the polymer is brittle, the effect must be analogous to that observed when a rigid coat is applied to a rubber membrane and the membrane is stretched; the brittle coating cracks.

The existence of the phenomenon postulated was shown in the following series of experiments. Joints were made as usual but, before peeling, the ribbon was stretched (usually for two minutes) as shown in Figure 9; the angle α was different from zero to
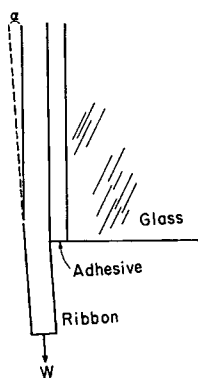

Fig. 9. Test for detecting damage to the brittle adhesive caused by an extension of the adherend.

make sure that no peeling component was present in the stretching force; the values of α used are estimated as between 1 and 5°. The stretching force usually was 3.8×10^6 dynes/cm. width; as aluminum ribbons 0.0076 cm. thick were employed, the stretching stress was 5×10^8 dynes/cm.^{2}, that is, slightly greater than the yield stress. Greater stresses could not be applied as the ribbon broke at the bend. After the stretching, the ribbon was peeled by a force about one-quarter that needed to strip an unstretched ribbon, and the length determined which was separated by this small force; this was believed to be equal to the length of adhesive film damaged by the plastic deformation of aluminum. This length was 0.3, 0.4, 0.5, 0.2, 0.4, 0.3, 0.2, 0.2, 0.3, and 0.2 cm. for Gelva at $y_0 = 0.02$ cm., and 0.3, 0.3, 0.5, 0.5, 0.4, 0.3, 0.3, 0.6, 0.4, 0.1, 0.4, 0.7, and 0.6 cm. for Epolene at $y_0 = 0.01$–0.03 cm, but a weak region of about 0.2 cm. was detected in only one of nine strips tested when an Epolene + Marlex (1:3) mixture was the adhesive at y_0 about 0.02 cm.

Thus, brittle adhesives break near the ribbon because they cannot follow the plastic elongation of the metal. Flexible adhesives can, but their deformation at the two extremities of the width of the ribbon causes stress concentration in these regions and thus simulates a low strength.

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Synopsis

The force required to peel an aluminum (or nickel) ribbon glued to a rigid glass plate with a polyethylene or poly-

vinyl acetate was determined. It proved to be smaller than expected (from a theory, partly new) both for adhesives which were almost Hookean solids and for those whose stress–strain curve could approximately be represented by stress = const. (strain)$^{0.5}$. In the latter group, the difference between the theory and the experiment was due to stress concentrations at the right-hand and the left-hand edges of the adhesive layer; these stress concentrations formed because, when a pull was applied, the adhesive contracted in the directions perpendicular to that of pull. As a consequence of this effect, the peeling force W_0 was not proportional to the width w of the ribbon; sometimes, a linear relation $W_0 = aw + b$ seemed to be valid. On the other hand, near-Hookean adhesives of a low total elongation required a peeling force proportional to w; the stress concentration dangerous for these materials occurred at the boundary between the adhesive and the bent ribbon at the sharp bend. The damage to the brittle adhesive caused by the ribbon deformation could be reproduced without bending, namely by extension of the ribbon beyond its yield strength.

Résumé

On a déterminé la force requise pour ôter un ruban d'aluminium (ou de nickel) collé à une plaque de verre rigide avec du polyéthylène ou de l'acétate de polyvinyle. On a prouvé que cette force est plus petite que prévue (d'après une théorie, en partie nouvelle) pour les adhésifs qui sont à peu près des solides obéissant à la loi de Hooke et pour ceux dont la courbe de force de tension-élongation pourrait être approximativement représentée par force = constante (tension interne)$^{0.5}$. Dans le dernier groupe, la différence entre la théorie et l'expérience provient des accumulations de tension aux extrémités droite et gauche de la couche adhesive; ces accumulations de tension proviennent de ce que, quand on applique une traction, l'adhésif se contracte dans des directions perpendiculaires à cette traction. En conséquence de cet effet, la force de traction W_0 n'est pas proportionnelle à la largeur w du ruban; parfois, une relation linéaire $W_0 = aw + b$ semble être valable. D'autre part, des adhésifs, obéissant à peu près à la loi de Hooke et de faible élongation totale, nécessitent une force de traction proportionnelle à w; l'accumulation de tension critique se situe à la frontière entre l'adhésif et le ruban plié à l'endroit de courbure forte. Le dégat causé à l'adhésif fragile, par le déformation du ruban, pourrait être reproduit sans pliage, par exemple, par extension du ruban au-dessus de sa capacité de rendement.

Zusammenfassung

Es wurde die Kraft bestimmt, die notwendig war, um ein Aluminium- oder Nickelband, das auf eine starre Glasplatte mit Polyäthylen oder Polyvinylacetate geklebt war, abzuziehen. Sie erwies sich als kleiner, als (nach einer zum Teil neuen Theorie) sowohl für Klebstoffe, die nahezu Hookesche Festkörper sind, als auch für solche, deren Spannungs-Dehnungskurve näherungsweise durch Spannung = const. (Dehnung)$^{0.5}$ dargestellt werden kann, zu erwarten war. Bei der letzteren Gruppe wurde der Unterschied zwischen Theorie und Experiment durch eine Spannungskonzentration an der rechts- und linksseitigen Kante der Klebstoffschichte verursacht; diese Spannungskonzentration entstand dadurch, dass bei Anwendung eines Zuges, das Klebemittel sich senkrecht zur Zugrichtung kontrahierte. Als Folge dieses Effektes war die Abziehkraft W_0 der Breite w des Bandes nicht proportional; in manchen Fällen schien eine lineare Beziehung von der Form $W_0 = aw + b$ zuzutreffen. Andrerseits war die bei fast-Hookeschen Klebestoffen bei niedriger totaler Elongation anzuwendende Abziehkraft zu w proportional; die für diese Stoffe gefährliche Spannungsanhäufung trat an der Grenze zwischen dem Klebstoff und dem gegobenen Band bei der scharfen Krümmung auf. Die Schädigung des spröden Klebstoffes, die durch die Deformation des Bandes verursacht wurde, konnte auch ohne Verbiegung reproduziert werden, indem nämlich das Band über seine Fleissgrenze hinaus gedehnt wurde.