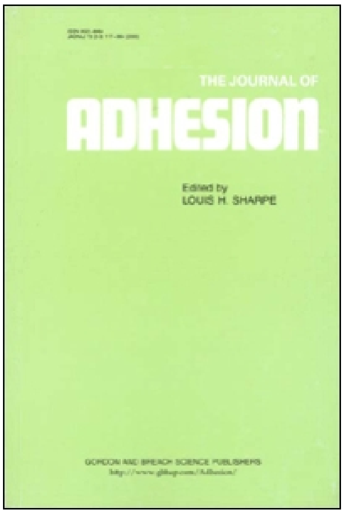

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Rheology of Peeling

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Rheology of Peeling

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ABSTRACT

In this paper, the peeling of an elastic adherend from a rigid substrate is analyzed rheologically. The theoretical relation between peel strength and peeling rate is introduced by the stress distribution over the interlayer at the steady state of peeling and by the assumed mechanical properties of the interlayer. Application of the theoretical relation to the experimental data gives us information about the mechanical properties of the interlayer. It may be concluded that in peeling the mechanical properties of the interlayer can be defined and they are related to the mechanical properties of both materials forming the interlayer.

INTRODUCTION

THE PEELING TEST has usually been employed as a convenient method for determining the strength of an adhesive. The phenomenon of peeling is, however, very complicated, depending upon the physical properties of adhesive and adherend. Though much work has been done to throw light on the phenomenon of peeling, [1-5] attention has generally been paid only to the deformation of the adhesive and the phenomenon was explained in terms of the properties of the adhesive. The systems chosen before generally consisted of a rigid substrate, soft adhesive and flexible adherend or of two identical flexible adherends holding a soft adhesive layer between them. The mechanical approach was usually for the one dimensional case. The use of these treatments would seem to make it very difficult to analyze the effect of the adherend and to determine the mechanical properties of the interlayer.

MECHANICAL ANALYSIS OF PEELING

To study the mechanical properties of the interlayer, the peeling of an elastic adherend from rigid substrate is chosen as a typical case. In this

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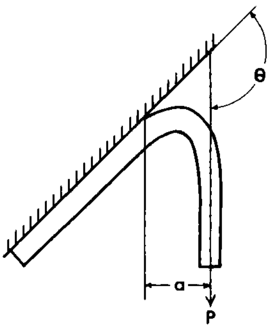

Figure 1. Schematic diagram of adherend in steady state peeling.

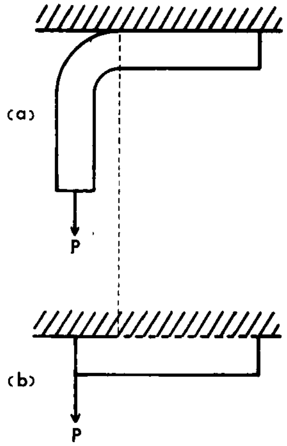

Figure 2. Adherend in steady state peeling and its model for theoretical analysis.

case, the previous mechanical analysis could not be applicable. Therefore, we must introduce another technique than has been used before.

At first, we paid attention to the shape of the adherend in the peeled part, which is shown in Fig. 1. In a state of steady peeling, the value of *a*, the horizontal distance between the failure point and the axis of the peeling load can be expressed as

$$a^2 = \frac{2EI_z}{P}(1 - \cos\theta) \tag{1}$$

where E and I_z denote Young's modulus and second moment of area of adherend, respectively, P, the peeling load and θ, the peeling angle. The value of a has a great effect on peeling.

Then we tried to determine the stress distribution over the interlayer where fracture occurs. Several analytical methods were tested but it seemed difficult to analyze the problem in this way. We, therefore, chose a numerical method.

To calculate the stress distribution, it is more convenient to assume a simple arrangement of the region in which the stress is to be analyzed. Therefore, we chose the model shown in Fig. 2(b) for the stress calculation instead of the one shown in (a). It was confirmed by photoelastic measurement that there is little difference in stress distribution over the interlayer between these two cases, when an equal moment and load were chosen in each case. The normal stress distributions, particularly, were almost identical.

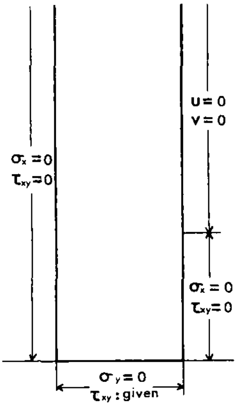

Figure 3. Boundary conditions for calculation of Airy stress function.

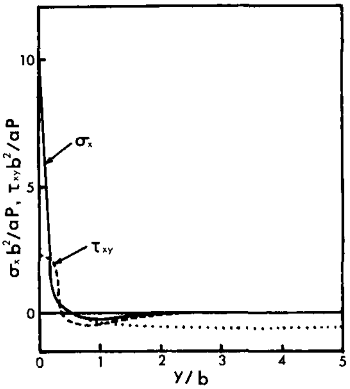

Figure 4. Stress distribution over the interlayer and its approximated value (dotted line).

Boundary conditions for the adherend when the peeling angle is 90° are chosen as shown in Fig. 3. It is assumed that there is no displacement on the interlayer because the opposing material along the interlayer is rigid. Parabolically distributed shear stress is assumed on the edge of the region.

Under these boundary conditions, the Airy stress function is solved [6] numerically by means of a relaxation method and the stress distribution is calculated. These calculations were done in two dimensions. The stress distribution for the case of $a = b$, where b is the thickness of the adherend, is shown in Fig. 4. The normal tensile stress is very large at the failure point. It decreases suddenly with increase of the distance from the failure point, reaches zero at x_0 and, in turn, changes into compressive stress. These stress distributions change with varying values of a, θ, b, E, and P.

The tangential stress, shown by the broken line in Fig. 4, is considerably smaller than the normal stress and we ignored it. Compressive stress seems to have no or little effect on peeling. Therefore, the actual stress distribution may be replaced by the one shown by the dotted line.

Thus, the peeling stress is approximately given as

$$\sigma = A_e{}^{-kx} - C \tag{2}$$

where A, C and k are constants and x denotes the distance from the failure point along the interlayer. From this equation, we can express the peeling stress at any point on the interlayer as a function of time, t. If peeling progresses at a constant rate, v, and t is defined to be 0 when the stress becomes zero, the stress at any point is defined by

$$\sigma = C(e^{kvt} - 1) \qquad (3)$$

Here, an assumption is made that the mechanical properties of the interlayer can be described in terms of a three element viscoelastic model: a Kelvin element in series with a spring as shown in Fig. 5. When the stress defined by equation (3) is applied on the model, the deformation is calculated as

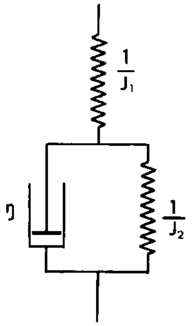

Figure 5. Three element model for the mechanical properties of the interlayer.

$$D(t) = \int_0^t ckv \exp(kvt') \left[J_0 + J_1 \left\{ 1 - \exp\left(-\frac{t-t'}{\tau} \right) \right\} \right] dt'$$

$$= CJ_0\{\exp(kvt) - 1\} - CJ_1\left\{ 1 - \frac{1}{kvt+1}\exp(kvt) - \frac{kvt}{kvt+1}\exp\left(-\frac{t}{\tau}\right) \right\} \qquad (4)$$

where $\tau (=1/J_1\eta)$ denotes the relaxation time of the model.

The work done on the model can be calculated as

$$D(t) = \int_0^t D\frac{d\sigma}{dt}dt = \int_0^t Dckv \exp(kvt)dt$$

$$= C^2\left[J_0 \left\{ \frac{1}{2}\exp(2kvt) - \exp(kvt) \right\} \right.$$

$$-J_1\left\{ \exp(kvt) - \frac{\exp(2kvt)}{2(kv\tau+1)} - \frac{(kv\tau)^2}{(kv\tau)^2-1}\exp\left(kvt\frac{t}{\tau}\right) \right\}$$

$$\left. + \frac{1}{2}J_0 + J_1\left\{ 1 - \frac{1}{2(kv\tau+1)} - \frac{(kv\tau)^2}{(kv\tau)^2-1} \right\} \right] \qquad (5)$$

The assumption that failure occurs at some constant value of the work done gives us

$$P = \frac{1}{A_0 + A_1\left[\dfrac{1 - 2\alpha k\tau v}{k\tau v + 1} + \dfrac{2(k\tau v)^2}{(k\tau v)^2 - 1}\left\{\alpha(1+\alpha)e^{-\frac{x_0}{\tau v}} - \alpha^2\right\}\right]} \qquad (6)$$

where A_0 and A_1 are constants and $\alpha = C/(A - C)$. In this way, the relation between peel strength and peeling rate is obtained.

EXPERIMENTAL DATA AND APPLICATION OF THE THEORY

Polyisobutylene (PIB) was used as the elastic adherend, and various materials (glass, polyvinyl chloride (PVC) and so on) are used as substrates for our experiment.

PIB (Vistanex MML-80) was dissolved in chloroform, then the solution was poured over the substrate and the solvent was evaporated. The resultant film thus obtained was cut into strips two centimeters wide with a razor blade.

This specimen was held at a constant temperature. After the temperature of the specimen had attained a fixed value, a constant load was placed on the free end of the PIB film and the peeling rate was measured on the surface of the substrate directly.

Experimental peeling data for the PIB and glass system at various temperatures, and the theoretical curves fitted to the data, are shown in Fig. 6. The agreement of the data with the theoretical relation is fairly good at temperatures lower than 20°C, but not so good at temperatures higher than 50°C. Similar data are also obtained for many systems of PIB and other sub-

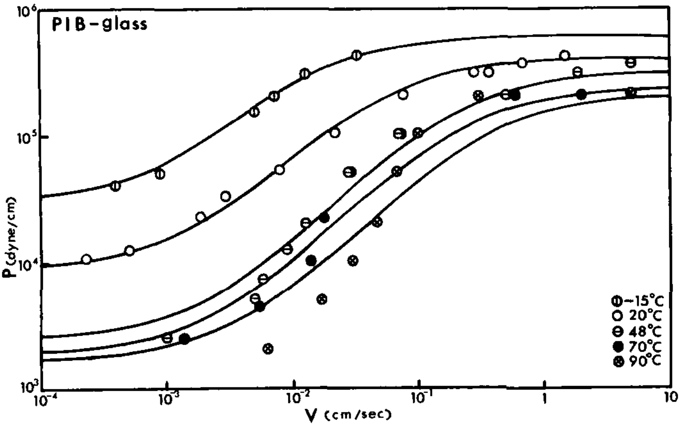

Figure 6. Peeling data for PIB-glass system at various temperature and theoretical peeling curve fitted to the data.

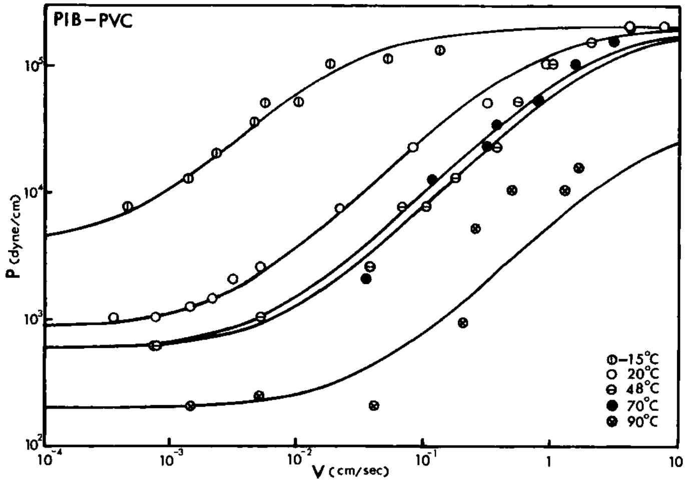

Figure 7. Peeling data for PIB-PVC system at various temperature and theoretical peeling curve fitted to the data.

strates. These curves are referred to as "peeling curves." Among these substrates are chromium plated steel sheet, PVC, polytetrafluoroethylene (PTFE) and polyethylene (PE).

In Fig. 7, curves for the PIB and PVC system are shown. These peeling curves can be characterized by three essential parameters: $P_o(=1/(A_0+A_1))$, the peeling strength extrapolated to zero peeling rate; $P_\infty(=1/A_0)$, the peeling strength at infinite peeling rate; and τ, the relaxation time of the three element model which describes the mechanical properties of the interlayer.

TEMPERATURE DEPENDENCE OF PEELING

The temperature dependence of P_0 and P_∞ are shown in Figs. 8 and 9. These parameters usually decrease with increasing temperature. Their temperature dependence is similar to the temperature dependence of surface energy given by Berry. [7, 8]

The temperature dependence of τ is very interesting. The value of τ is originally the value which denotes the relaxation time of the interlayer where separation occurs during peeling. It is, therefore, important to analyze the temperature dependence of τ in order to study the mechanical properties of the interface on fracture.

As a whole, τ shows a temperature dependence similar to that of the shift factor for PIB as shown in Fig. 10. Nevertheless, there seem to be some exceptions. For the PIB-PVC system the curve clearly shows an abrupt change at a temperature between 70°C and 90°C. This change may be related to the glass transition of PVC. A similar but smaller change is also

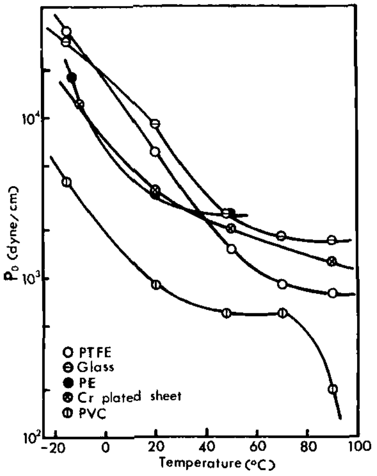

Figure 8. Temperature dependence of P_0 for PIB-various adherend systems.

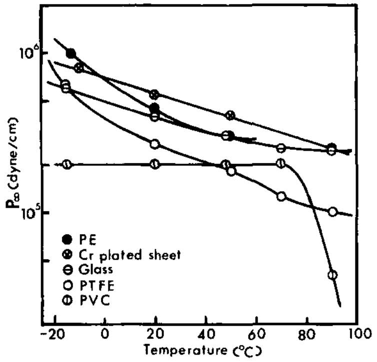

Figure 9. Temperature dependence of P_∞ for PIB-various adherend systems.

seen at a temperature near 30°C for PIB-PTFE system. From the data of McCrum [9] and Illers & Jenckel, [10] we can find a peak of tan δ for PTFE at the temperature at which the anomalous behavior occurs. It may be suggested that τ shows properties characteristic of some composite system of adherend and substrate.

To compare the temperature dependence of τ with that of a_T, we first examined whether the WLF equation was applicable to the temperature dependence of τ. The original WLF equation is as follows:

$$\log a_T = \frac{-C_1(T - T_s)}{C_2 + T - T_s}$$

where C_1 and C_2 are constants and T_s denotes the reference temperature which is about 50°C higher than glass transition temperature. It can be rearranged as

$$\log a_T = -C_1 + \frac{C_1 C_2}{T - T'}, \text{ where } T' = T_s - C_2$$

The value of log τ is plotted against $1/(T\text{-}T')$, changing T'. And when the plots form a straight line, the value of T' is chosen as an appropriate value. The slope of this line gives the value of C_1C_2. Thus we can obtain the two relations for three constants; C_1, C_2 and T_s.

When one of the three constants is defined to be original value, the other two values can be calculated by these two relations. The values of C_1 and C_2 are given as 8.86 and 101.6 respectively in the original WLF equation. Nevertheless, they are treated as constants the values of which are undecided. The value of T_s for PIB is used as the original value of T_s. There values are listed in Table I.

As a whole, the value of C_1C_2 is smaller than original value, therefore it is impossible to apply the WLF equation to the temperature dependence of τ. The value of T_s is also smaller than the one for PIB. However, although it is impossible to apply the WLF equation to it, a WLF type equation is applicable to the temperature dependence of τ.

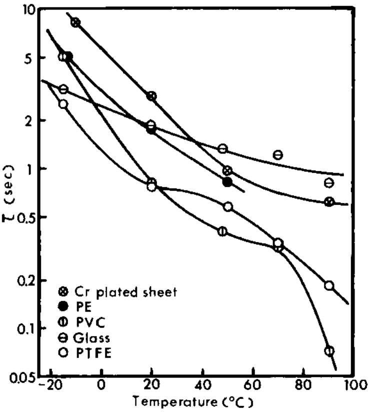

Figure 10. Temperature dependence of τ for PIB-various adherend systems.

EFFECT OF PLASTICIZER

The effect of plasticizer in the adherend has already been reported by Hata. [11] He has made clear that the temperature-plasticizer content superposition principle is applicable to peeling. Our data for PIB and various material systems suggested that peeling was not only affected by the adherend but by the substrate. Therefore, we studied the effect on peeling of plasticizer in the substrate.

In this case, the specimen for peeling was obtained as follows. PVC (plasticized or unplasticized) film was at first fixed onto a glass plate with adhesive tape. PIB film which had previously been prepared on a glass plate was placed on the PVC film, then heated at 55°C about 5 hours under a pressure of 100g/cm^2. After that the peeling load was applied at the edge of PIB film which was peeled at a 90° angle. The peeling curves were obtained at various temperatures, from which the temperature dependence of the three essential parameters can be estimated.

As was described before, the temperature dependence of τ is most important for understanding the mechanical or rheological properties of the interlayer. As shown in Fig. 11, an increase in plasticizer content has an effect similar to an increase in temperature. This suggests that the plasticizer

Table I.

	T' $(=T_s-C_2)$	C_1C_2	$T_s = 240°K$		$C_1 = 8.86$		$C_2 = 101.6$	
			C_1	C_2	$T_s(°K)$	C_2	$T_s(°K)$	C_1
PIB—glass	0	429.2	1.79	240	48.4	48.4	101.6	4.22
PIB—PE	0	829.3	3.45	240	93.6	93.6	101.6	8.16
PIB—Cr	178	364.4	5.88	62	219.2	41.1	279.6	3.59
PIB—PVC	213	82.0	3.04	27	222.3	9.3	314.6	0.81

content-temperature superposition is applicable to this case. Nevertheless, these curves cannot be perfectly superposed by shifting along the abscissa. These data also suggest that the temperature dependence of τ is related to the mechanical properties of the substrate.

EXCHANGE OF SUBSTRATE AND FLEXIBLE MEMBER

If we exchange the substrate for a flexible member, it seems that information about the interlayer can be obtained, particularly, whether the mechanical properties of the interlayer are changed by the reversal of substrate and flexible member. Accordingly, a PIB and Aluminum (Al) foil system was studied. PIB film was formed on the Al foil and one of them was fixed onto a glass plate with adhesive tape. The peeling load was applied at an end of the other film and the peeling rate was measured. The thickness of the PIB film was 0.03cm and of the Al foil 0.0015 cm.

We had standard peeling curves which had been calculated for the case of 0.03cm thick adherends. Therefore, we used these standard curves for the estimation of the three parameters. When the thickness of adherend is not equal to 0.03cm, the value of τ obtained from the standard curves is not correct. It is denoted by τ_0. The value of τ_0 must be modified by the thickness of adherend, but the modification is not simple. For a rough approximation, τ_0 must be multiplied by the ratio of the thickness to 0.03cm. Correct modification is very difficult, because the change of stress distribution with thickness of adherend is very complicated. The correct value of τ seems to be a little smaller than the rough approximation.

Therefore, we used τ_0 as it is. The temperature dependence of τ_0 for the two cases is shown in Fig. 12. It seems that the difference between the two cases is not large. There may be many reasons why the absolute value of τ is not the same for each case. One of the principal reasons is that when the PIB film is used as the substrate, the interlayer is not in the fixed state. Although it is not clear that the absolute value of τ remains unchanged in an exchange of substrate and adherend, the temperature dependence of τ is quite similar in these two cases. Thus, the belief that the mechanical properties of the interlayer can be defined is supported. Further, the suggestion that the mechanical or rheological properties of the interlayer are determined by those of both materials forming the interlayer is also supported.

CONCLUSION

As mentioned above, the analysis of peeling gives much interesting information. There are, however, many problems which have not been solved. One of them is the effect of the peeling conditions on peeling. A study of this will be given in a future paper.

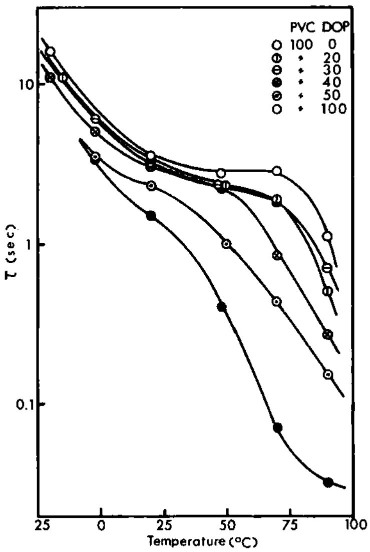

Figure 11. Temperature dependence of τ for PIB-plasticized PVC systems.

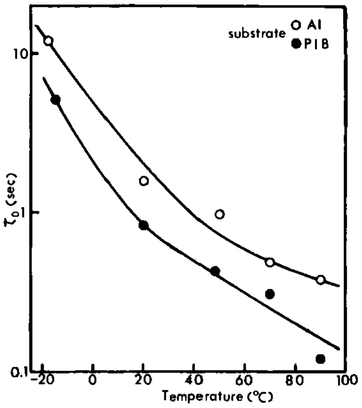

Figure 12. Temperature dependence of τ_0 for PIB-Al system when PIB is used as adherend and as substrate.

It may be concluded, on the basis of our data and our interpretation of it, that, in peeling, the mechanical or rheological properties of the interlayer can be defined and that its properties are related to the mechanical or rheological properties of both materials forming the interlayer.

To understand why the interlayer shows such behavior, we will have to analyze the peeled surface by microscopic or other special techniques. Such studies are proceeding.

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