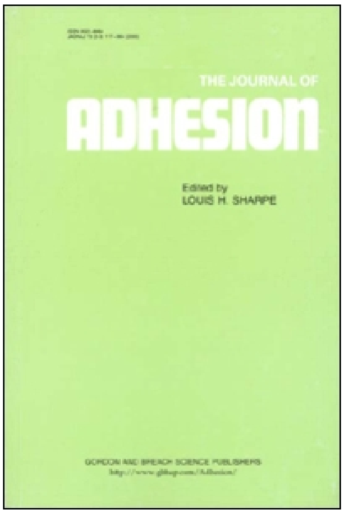

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Mechanisms of Adhesive Failure†

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In the peeling test of adhesive tapes as well as in other experiments for adhesive failure, the transition of failure modes from cohesive to interfacial has been observed by several workers in the process of increasing rate or decreasing temperature. It is accompanied by an abrupt change of adhesive strength. These facts cannot be explained by the failure mechanism based on a weak boundary layer. (It would be willful to assume two kinds of weak boundary layers). In this paper, the phenomena above referred to and the dependence of adhesive strength on rate, temperature, thickness, and some physical properties of adhesives are attempted to be explained rheologically. The author has proposed a simple model theory to interpret the so-called failure envelope of T. L. Smith, where viscoelastic substances were represented by Maxwell elements connected in parallel and appropriate criteria for failure were introduced to an element, which was considered a weak point in the substance (*Zairyo* (*Materials*) 17, 322 1968). In addition to this treatment for cohesive failure, the following new criterion is introduced to the same model; that is, interfacial failure occurs when the elastic work of deformation of the whole system reaches a critical value. The formulae obtained represent the observed behavior at least qualitatively. Other dependence of adhesive strength on the variables aforementioned and the mutual reduction between them are also discussed.

INTRODUCTION

In the peeling test of adhesive tapes, as well as in other experiments for adhesive failure, the transition of failure modes from cohesive to interfacial has been observed by several workers in the process of increasing rate of separation or decreasing temperature. It is accompanied by an abrupt change of adhesive strength. As an example, results reported by Fukuzawa

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et al.[1] for the 180° peeling of adhesive tapes of different thicknesses are shown in Figure 1, where Bakelite plates were used as an adherend. In the figure the differences of failure modes, observed by the naked eye, are also shown by different indicators, cohesive failure by dots, interfacial failure by circles, and mixed failure of both modes by half circles. The fact that the thicker the adhesive layer, the larger the peeling strength becomes, is expected from all theories of peeling. Cohesive failure becomes predominant for thicker tapes, whereas interfacial failure does so for thinner ones. For the intermediate thickness (0.07 mm and 0.11 mm), the peeling strength varies abruptly at a certain rate of peeling, where the failure mode also transforms from cohesive to interfacial.

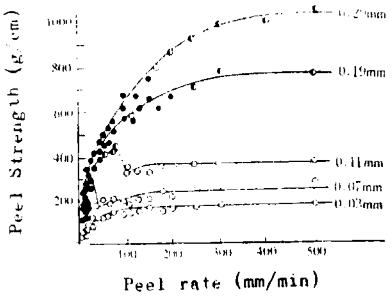

FIGURE 1 Dependences of peel strength on peel rate and thickness of adhesive tapes.

Similar results have been obtained by Kaelble[2], who confirmed the interfacial failure by measurements of contact angle and the critical surface tension of the adherend, which gave equal values before and after the peeling test. Nakao[3] has observed similar behaviours for shear tests of adhesion. These facts, particularly the discontinuous change of adhesive strength, cannot be explained by the failure mechanism based on a weak boundary layer, as proposed by Bikerman[4]. It would be willful to assume that two kinds of weak boundary layers exist. As well as the facts here described, there are many results correlating adhesive strength to surface chemical functions as discussed in another paper persented in this symposium by the author, which suggests a possibility of interfacial failure even for properly wetted adhints.

In this paper, an attempt is made to interpret the facts shown in Figure 1 and to show that the variability of failure modes is essentially of rheological character, depending on rate of separation, temperature, thickness of adhesive layer, and its physical properties.

DEPENDENCE OF ADHESIVE STRENGTH ON RATE OF SEPARATION, TEMPERATURE, AND OTHER FACTORS

In order to visualize the author's idea, at first, the simplest model of viscoelasticity, composed of three elements, is considered for the case of shear adhesion test. In Figure 2 the surface force is represented by a spring, the modulus of which is G_1, and the mechanical behaviours of adhesives by a Voigt model, its modulus and viscosity coefficient being G_2 and η respectively, and the adherend is assumed to be rigid enough to neglect its deformation. Then, using the notations in Figure 2,

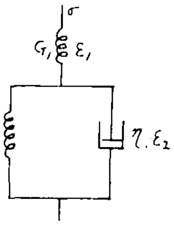

FIGURE 2 Three elements model.

$$\sigma = \varepsilon_1 G_1 = \varepsilon_2 G_2 + \eta\, \mathrm{d}\varepsilon_2/\mathrm{d}t \tag{1}$$

Assuming these relations to be valid up to failure, the following criteria for failure are introduced; failure occurs either at the interface or in the bulk phase, according to whether the strain of the surface spring ε_1 reaches a critical value ε_{1_c}, or the strain of the adhesive layer ε_2 reaches ε_{2c}. Thus,

interfacial failure $\sigma_b{}^S = \varepsilon_{1c} G_1$ (2)

or

cohesive failure $\sigma_b{}^B = \varepsilon_{2c} G_2 + \eta(\mathrm{d}\varepsilon_2/\mathrm{d}t)_{\varepsilon 2 = \varepsilon 2c}$ (3)

where $\sigma_b{}^S$ and $\sigma_b{}^B$ represent the stresses at break for the two cases. The problem of which mode of failure will occur is dependent on the strain rate. Because of the viscosity term in the bulk phase, its deformation is hindered at higher rates and the condition of interfacial failure $\varepsilon_1 = \varepsilon_{1_c}$ will be realized faster than $\varepsilon_2 = \varepsilon_{2_c}$, whereas, at lower rates, the condition of cohesive failure $\varepsilon_2 = \varepsilon_{2_c}$ will first be realized. In this treatment, however, $\sigma_b{}^S$ has no rate dependence, which is improved in the following section. As for $\sigma_b{}^B$, its dependence on various factors is formulated as follows.

We now consider a shear test with constant speed of machine, v, and an adhesive layer of thickness, h. Let the strain at break be $\varepsilon_b (= \varepsilon_1 + \varepsilon_{2c})$ and the time at break be t_b. Then $v = \varepsilon_b h / t_b$, and

$$(\mathrm{d}\varepsilon_2/\mathrm{d}t)_{\varepsilon 2 = \varepsilon 2c} \simeq \varepsilon_{2c}/t_b = \varepsilon_{2c} v / \varepsilon_b h \tag{4}$$

Combining Eqs. (1) at the condition of $\varepsilon_2 = \varepsilon_{2c}$, (3) and (4), and eliminating $(\mathrm{d}\varepsilon_2/\mathrm{d}t)_{\varepsilon_2=\varepsilon_{2c}}$ and ε_1, we obtain

$$\sigma_b{}^B = 1/2\{\varepsilon_{2c}(G_2 - G_1) + \sqrt{\varepsilon_{2c}{}^2(G_1 + G_2)^2 + 4\varepsilon_{2c}G_1\eta v/h}\} \qquad (5)$$

This equation expresses, at least qualitatively, experimental facts for shear adhesive strength such that it grows larger with the increase of machine speed, modulus and viscosity coefficient of adhesive, and with the decrease of thickness[5]. The increase of $\sigma_b{}^B$ with these factors also means that the possibility of cohesive failure decreases, instead, the possibility of interfacial failure will relatively increase. The strengthening of surface layers by the methods of CASING and TCR developed by Schonhorn and his co-workers[6,7] gives rise to the increase of the modulus G_2, and in the extreme case of strengthening, interfacial failure may be expected to occur.

The transition of failure modes due to the change of temperature can be also discussed with the use of Eq. (5). As the viscosity coefficient η decreases with the increase in temperature, ε_2 easily reaches the critical value ε_{2c} and $\sigma_b{}^B$ becomes smaller at higher temperature. In other words, a state is soon reached in which cohesive failure occurs. On the contrary, with the decrease in temperature, interfacial failure becomes more easy. The facts demonstrating this effect of temperature on the failure mode are seen in Figure 3, again by Fukuzawa. Similar results have been obtained by Kaelble[2] who further investigated successfully the time-temperature superposition.

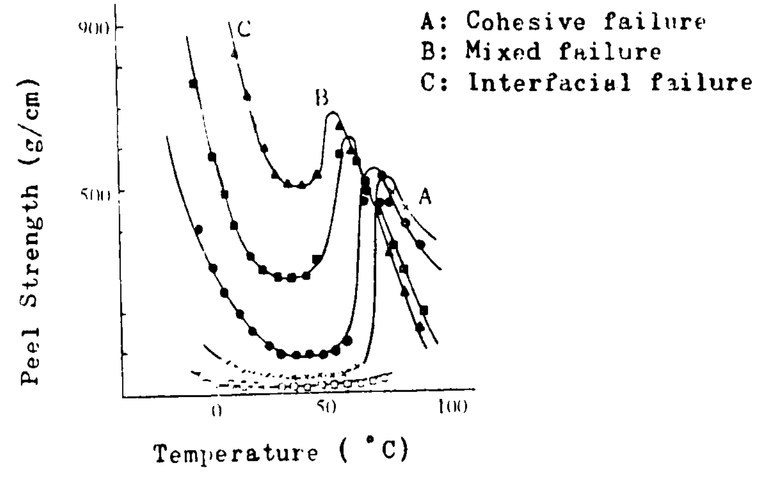

FIGURE 3 Temperature dependence of peel strength of adhesive tapes of different contents of tackifier resin.

□: (10%), ×: (30%), ●: (40%), ■: (50%), ▲: (60%).

Although the above equations were described for the case of shear adhesion, if the shear modulus G and the viscosity coefficient η are converted to Young's modulus E and the tensile viscosity coefficient η_t, the equations are valid for a tensile test. Even in the case of peeling, as far as its essential mechanism is based on the tensile deformation of the adhesive layer, it is allowable to discuss the behaviour of peeling strength with these equations, qualitatively, but with certain necessary limitations.

ADHESIVE STRENGTH AND FAILURE MODE, TAKING FAILURE ENVELOPE INTO CONSIDERATION

In the treatment described above, adhesive strength for interfacial failure remains constant against rate of separation, though in fact, there are not a few results showing the dependence of adhesive strength on rate and other factors, even in the case of interfacial failure, as shown in Figure 1. Considering the viscoelastic deformation up to failure at the interface, such dependence must be taken into consideration. Further, in this section, it is attempted to explain the discontinuous change of both adhesive strength and failure mode shown in Figure 1, and the behaviours of adhesive failure, similar to T. L. Smith's failure envelope[8] for cohesive failure.

Attempts theoretically to explain the failure envelope have been made by several workers[9–11]. The author's theory is based on the following model. (1) Failure in viscoelastic bodies starts at a weak point, then the residual part breaks down carrying the whole load. (2) The failure at the weak point may occur either by scission or slipping of chain molecules. The simplest model representing these concepts is two Maxellw models (system 1 and 2) connected in parallel, as shown in Figure 4, where system 1 designated by subscript 1 is regarded as the weak point.

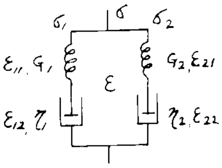

FIGURE 4 Four element model.

Criteria for failure are introduced to system 1 as follows: failure occurs either when strain of the spring ε_{11} reaches a critical value ε_{11c} (case A), or strain of the dashpot ε_{12} reaches a critical value ε_{12c} (case B). The problem which occurs depends on strain rate, temperature and other factors. The circumstances are the same as discussed in the preceding section, that is, at a larger strain rate, the spring is stretched faster than the dashpot and the condition $\varepsilon_{11} = \varepsilon_{11c}$ will first be realized, and at a smaller rate, the condition $\varepsilon_{12} = \varepsilon_{12c}$ will occur. For the experiment with constant rate $R(= d\varepsilon/dt = \varepsilon/t)$, the results are given as

$$\sigma_b(R) = G_1\tau_1 R\{1 - \exp(-\varepsilon_b(R)/\tau_1 R)\} + G_2\tau_2 R\{1 - \exp(-\varepsilon_b(R)/\tau_2 R)\} \quad (6)$$

(A) at a larger strain rate $\varepsilon_b(R) = \tau_1 R\varepsilon_{11c}/(\tau_1 R - \varepsilon_{11c})$ (7)

(B) at a smaller strain rate $\varepsilon_b(R) = 1/2(\varepsilon_{12c} + \sqrt{\varepsilon_{12c}^2 + 4\tau_1 R\varepsilon_{12c}})$ (8)

where $\tau_1 = \eta_1/G_1$ and $\tau_2 = \eta_2/G_2$ are relaxation times of two Maxwell elements. In the case of (A), $\varepsilon_b(R)$ can be exactly calculated as $\varepsilon_b(R) = -\tau_1 \ln(1 - \varepsilon_{11c}/\tau_1 R)$, instead of Eq. (7). However, $\varepsilon_b(R)$ corresponding to Eq. (8) cannot be obtained in an analytical form, therefore an approximation of $d\varepsilon_{12}/dt \simeq \varepsilon_{12}/t_b$ is used for the calculation of both $\varepsilon_b(R)$'s. Figure 5 shows the two parts of the failure envelope (open circle) on the stress-strain map with the parameter of strain rate, and Figure 6 shows the dependence of $\varepsilon_b(R)$ on strain rate. $\varepsilon_b(R)$ reaches a maximum value at the point of intersection of the two curves. Putting Eq. (7) equal to Eq. (8), we obtain

$$\varepsilon_{b\max} = (\varepsilon_{12c}/\varepsilon_{11c})\tau_1 R_{\max} \quad (9)$$

or

$$t_{b\max} = (\varepsilon_{12c}/\varepsilon_{11c})\tau_1 \quad (10)$$

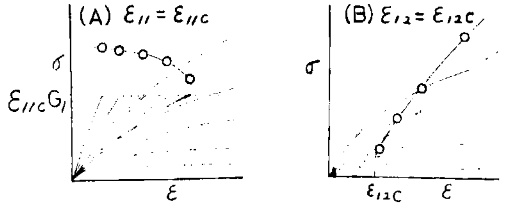

FIGURE 5 Explanatory diagrams of the model theory for the failure envelope (–O–). Solid line: System 1, Dashed line: System 2. (A) A case of large strain rate, (B) A case of small strain rate.

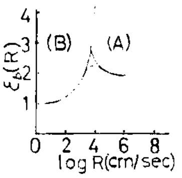

FIGURE 6 Dependence of the breaking strain $\varepsilon_b(R)$ on strain rate R by Eqs. (7) and (8). ($\tau_1 = 10^{-3}$ sec, $\varepsilon_{11c} = 2, \varepsilon_{12c} = 1$).

Effects of temperature on $\sigma_b(R)$ and $\varepsilon_b(R)$ can be discussed through the dependence of τ_1 and τ_2 on temperature. As seen in Eqs. (6)–(8), τ_1 and τ_2 have influences upon $\sigma_b(R)$ and $\varepsilon_b(R)$ in quite the same manner as R. Effects of thickness, h, of adhesive layer can be also deduced, converting R to v/h, where v is the speed of test machine. Similar treatment based on the generalized Maxwell model is also possible[11], which is omitted here because the purpose of this paper is to make clear the principal idea.

The above treatment is concerned with cohesive failure in viscoelastic bodies. The problem in this paper is how to introduce interfacial failure into the model. Here we consider that the interfacial force is essentially elastic, and its energy at failure is equal to the stored energy or the elastic work of deformation of bulk phase up to the interfacial failure. Therefore we can express the interfacial failure by introducing a critical value W_c into the elastic work of the bulk system. In the case of this model represented by Figure 4, it is

$$1/2\varepsilon_{11}{}^2G_1 + 1/2\varepsilon_{21}{}^2G_2 = W_c \tag{10}$$

Using again the approximations $\mathrm{d}\varepsilon_{12}/\mathrm{d}t \simeq \varepsilon_{12}/t_b = \varepsilon_{12}R/\varepsilon_b(R)$ and $\mathrm{d}\varepsilon_{22}/\mathrm{d}t = \varepsilon_{22}R/\varepsilon_b(R)$ and eliminating ε_{11} and ε_{21} from Eq. (10), we obtain

$$\left(\frac{\varepsilon_b(R)\tau_1R}{\varepsilon_b(R) + \tau_1R}\right)^2G_1 + \left(\frac{\varepsilon_b(R)\tau_2R}{\varepsilon_b(R) + \tau_2R}\right)^2G_2 = 2W_c \tag{11}$$

Putting $\varepsilon_b(R)$ calculated from this equation into Eq. (6), the rate dependent $\sigma_b{}^S(R)$ for interfacial failure can be obtained. $\sigma_b{}^S(R)$ is an S-type increasing function of R, as shown in Figure 7, starting from $\sigma_{b0}{}^S = (\tau_1G_1 + \tau_2G_2) \times \sqrt{2W_c(\tau_1{}^2G_1 + \tau_2{}^2G_2)}$ at $R_0 = \sqrt{2W_c(\tau_1{}^2G_1 + \tau_2{}^2G_2)}$ and converging to $\sigma_{b\infty}{}^S = \sqrt{2W_c(G_1 + G_2)}$ at $R \rightarrow \infty$, while $\varepsilon_b(R)$ is a decreasing function toward $\varepsilon_{b\infty} = \sqrt{2W_c(G_1 + G_2)}$. In Figure 7, $\sigma_b{}^B$ according to Eq. (6) combined

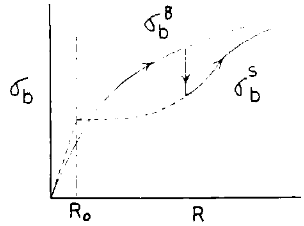

FIGURE 7 Dependence of adhesive strength and failure mode on strain rate. $\sigma_b{}^B$: Cohesive failure, $\sigma_b{}^S$: Interfacial failure.

with (8) is also shown. Generally speaking, in the region of smaller strain rates, cohesional failure arising from the viscous flow term occurs predominantly, while in the region of larger strain rates, interfacial failure arising from the elastic term occurs successively. The problem of where the transition between them occurs is dependent on the critical values given to the two failure modes. As a suggestion, it may be said that the transition occurs at $R_{\max}$ corresponding to the maximum $\varepsilon_b(R)$.

MUTUAL REDUCTION BETWEEN RATE, TEMPERATURE, AND THICKNESS

Here, the possibility of mutual reduction between rate, temperature, and thickness of adhesive layer for adhesive strength is shown, without specifying the failure mode. The experimental facts for peeling have been given by the author[12], Kaelble[2,13], and Nonaka[14], and they proved that rate-temperature superposition according to the WLF equation was established. The superposition following Arrhenius' equation has also been reported by Koizumi *et al.*[15] and Nakao[16].

It is assumed, as is usually done for thermally simple viscoelasticity, that when temperature varies from T_0 to T, the relaxation time τ_0 and the modulus G_0 become τ and G according to the following relations:

$$\tau = a_T\tau_0, \qquad G = (\rho T/\rho_0 T_0)G_0 \tag{12}$$

where a_T is a shift factor, ρ and ρ_0 are density at each temperature. As $\rho T/\rho_0 T_0 \simeq 1$, here, for simplicity, the change of modulus with temperature is not considered. Let us again consider a shear test which is represented by the model described in the preceding section, and put the relation $\tau = a_T\tau_0$ into the Eqs. (6), (7), (8) and/or (11) and convert R to v/h, then the following relationships are easily obtained:

$$\left.\begin{aligned}\sigma_b(T, v, h) &= \sigma_b(T_0, a_T v, h)\\ \varepsilon_b(T, v, h) &= \varepsilon_b(T_0, a_T v, h)\end{aligned}\right\} \tag{13}$$

$$\left.\begin{aligned}\sigma_b(T, v, h) &= \sigma_b(T_0, v, h/a_T)\\ \varepsilon_b(T, v, h) &= \varepsilon_b(T_0, v, h/a_T)\end{aligned}\right\} \tag{14}$$

Eq. (13) expresses the rate-temperature reduction for adhesive strength and failure strain, while Eq. (14) similarly expresses the thickness-temperature reduction. It is noteworthy that the change of temperature can be reduced by the change of thickness, in addition with the same shift factor for the rate-temperature reduction a_T. That is to say, a curve of adhesive strength vs. thickness of adhesive layer at temperature T_0 can be superposed by dividing the thickness h by a_T to the curve at temperature T. When the thickness varies from h_0 to h as $h = a_h h_0$, similarly we obtain

$$\left.\begin{aligned}\sigma_b(T, v, h) &= \sigma_b(T, v/a_h, h_0)\\ \varepsilon_d(T, v, h) &= \varepsilon_b(T, v/a_h, h_0)\end{aligned}\right\} \tag{15}$$

This is the thickness-rate reduction, which is a matter of common knowledge to research workers.

These mutual reductions between rate, temperature, and thickness are not restricted to such models or equations as considered in this paper, but they are deduced from every equation in which the term τR or $\tau v/h$ is included as a combined form.

CONCLUSIONS

Formulations and discussions described here seem to be merely based on assumptions. However, in order to understand the transition behaviour of the failure mode, it is necessarily required to assume two kinds of failure mechanisms, one of which should be an interfacial failure. The present theory is based on this fact and criteria for failure were introduced as rigorously as possible within the limitation of simplicity of the model.

As shown in the third section, viscoelastic deformation occurs even in interfacial failure, in which adhesive strength increases with increasing rate of separation. For understanding this behaviour it is not necessary to consider an electrical double layer in the interface as done by Derjaguin, or a diffusion layer as by Voyutskii.

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