Theory of Tack of Pressure-Sensitive Adhesive. II

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Synopsis

When a cylinder is pulled on a pressure-sensitive adhesive, the bonding and debonding processes proceed simultaneously within the surface of contact. In the previous paper, the theory of rolling friction coefficient of pressure-sensitive adhesives was proposed for the case where bonding occurs instantaneously. The theory is modified in this paper so as to include both the time-dependent bonding process and debonding process at the same time. Effect of fiber-forming on rolling friction is also examined. It is ascertained that the modified theory reproduces quite well the experimentally observed features of the curves of rolling friction coefficient against velocity.

INTRODUCTION

The tack of pressure-sensitive adhesives is often measured by means of some standardized rolling ball methods in the practical cases,[1] but the physical meaning of the measured values is not studied enough. The author[2-5] has pointed out that, in order to understand the tack on scientific bases, we had better express it by the rolling friction coefficient f, which depends upon the physical properties of the materials, and not upon any trivial experimental parameters such as the leading distance, angle of inclination of the surface, etc. It is shown that the rolling motion of a ball on pressure-sensitive adhesives can satisfactorily be described by a simple mechanical theory, in which f of the material is expressed as $f = \varphi_0 + \varphi_1 v$, with φ_0 and φ_1 being constants. In other words, values of φ_0 and φ_1 can be determined by the least squares method, although we have to carry out some numerical calculations. Then, the authors[5,6] have proposed the pulling cylinder method, according to which f of the material is given by the following simplest equation:

$$f = PR/\mathrm{Mg} \tag{1}$$

where P is the force to pull a cylinder of radius R and weight Mg at a constant velocity v. If the cylinder is pulled at different v, the dependence of f upon v can experimentally be clarified. Plot of f against $\log v$ for a viscoelastic material generally gives such a curve which increases from a very low value to a certain maximum and then decreases as v becomes larger. The curve shifts toward higher velocity region as the mechanical relaxation time of the adhesive decreases or thickness of the adhesive layer increases. It is experimentally ascertained[3] that f obtained from rolling ball experiments

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corresponds to that from pulling cylinder experiments in the velocity range from 10 to 100 cm/s.

Then, in order to interpret these characteristics, one of the authors[7] has developed in the previous paper a theory in which f is expressed in terms of viscoelastic parameters of the adhesives. However, only the debonding process was treated in the theory, which means that the bonding process was implicitly assumed to proceed instantaneously. Actually the bonding process is known to be time-dependent, and this factor must be taken into consideration in the theory of rolling friction of sticky materials. In addition, it is well known that some fiberlike structures of the adhesive are formed in the debonding process within the deforming zone, and this may also be an important factor.

In this paper, the authors try to modify the previously proposed theory so as to include these effects.

THEORETICAL CALCULATIONS OF ROLLING FRICTION COEFFICIENTS

If we want to treat the rolling friction of a viscoelastic material in a very strict sense, we have to think that f consists of two terms:

$$f = f_c + f_a \tag{2}$$

where f_c represents rolling friction caused by compressive deformation of the substrate and f_a that caused by adhesion or extensional deformation of the substrate. But here again, we assume that f_a is much larger than f_c in the case of pressure-sensitive adhesives. It can experimentally be shown[8] that f is inversely proportional to the weight (Mg) of the cylinder, provided that it is not extremely heavy. This means that we do not have to take sinking of a cylinder into account when we think of a light touch of a material on a pressure-sensitive adhesive.

Suppose a cylinder is pulled on a pressure-sensitive adhesive in one direction. The adhesive will elongate as a result of adhesion, and disturb rotation of the cylinder as schematically shown in Figure 1. Then, strain ϵ and rate of strain $\dot{\epsilon}$ of the adhesive are expressed as a function of angle θ of the cylinder:

$$\epsilon = (R/h)(1 - \cos\theta) \tag{3}$$

$$\dot{\epsilon} = (v/h)\sin\theta \tag{4}$$

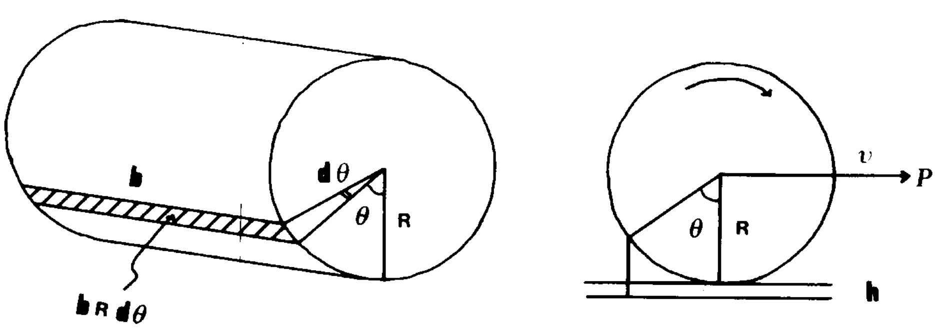

Fig. 1. Rolling cylinder.

where h and v are original thickness of the adhesive layer and velocity of the cylinder, respectively.

If we adopt a proper mechanical model, stress σ generated by elongation of the adhesive can be expressed as a function of θ. A surface element $bR\,d\theta$ is pulled by the force $\sigma bR\,d\theta$ downward (perpendicularly toward the base for small θ), and therefore the moment dm_a caused by extension of the adhesive can be calculated by integrating it from $\theta = 0$ to $\theta = \theta_b$:

$$m_a = \int_0^{\theta_b} R\sigma(\theta) bR\,d\theta \cos\theta \sin\theta$$

$$= R^2 b \int_0^{\theta_b} \sigma(\theta) \cos\theta \sin\theta\, d\theta \tag{5}$$

where θ_b is the angle where failure is steadily proceeding on the surface of the cylinder. Then, the rolling friction coefficient f_a is

$$f_a = (m_a/\mathrm{Mg}) = (R^2 b/\mathrm{Mg}) \int_0^{\theta_b} \sigma(\theta) \cos\theta \sin\theta\, d\theta \tag{6}$$

This is the general expression of f_a, which was derived in the previous paper.[7]

Consideration of the Time-Dependent Bonding Process

It is well known that the true bonding area increases gradually as a function of contact time t. Bright[9] and Kanamaru[10] assumed that the rate of increase of the bonding area A is proportional to the still unbonded part $(A_\infty - A)$, namely,

$$dA/dt = k(A_\infty - A) \tag{7}$$

where A_∞ is the area at $t = \infty$, which is equal to the nominal bonding area. Then,

$$A/A_\infty = 1 - e^{-kt} \tag{8}$$

Therefore, the area of the surface element in Figure 1, $bR\,d\theta$, must be replaced by $bR\,d\theta\,(1 - e^{-kt})$ in deriving the equation for f_a. As the cylinder rolls at a constant velocity,

$$R\theta = vt \qquad \text{or} \qquad t = R\theta/v \tag{9}$$

and then a general expression of f_a for this case is

$$f_a = (R^2 b/\mathrm{Mg}) \int_0^{\theta_b} \sigma(\theta)(1 - e^{-kR\theta/v}) \sin\theta \cos\theta\, d\theta \tag{10}$$

One of the mechanical models adopted in the previous paper is the two Maxwell elements in parallel connection, which is shown in Figure 2. Hata[11,12] has shown that the rheological features of adhesive strength in general can

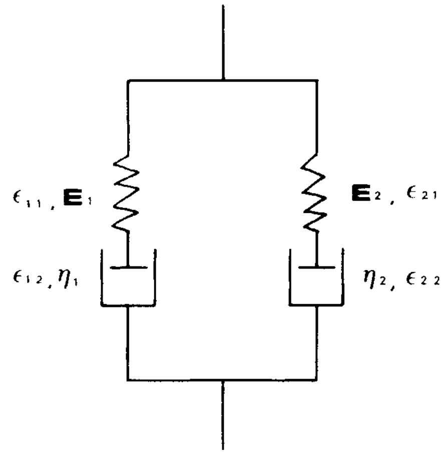

Fig. 2. Two Maxwell elements in parallel connection.

approximately be explained according to this model, if we assume the following failure criteria:

A. In the region where rate of strain is very high, cohesive failure occurs when strain ϵ_{11} of the spring in the weak point (element 1) reaches a critical value ϵ_{11c}.
B. In the region where rate of strain is very low, cohesive failure occurs when strain ϵ_{12} of the dashpot in the weak point reaches a critical value ϵ_{12c}.
C. Interfacial failure occurs when stored energy W in the springs of the model reaches a critical value W_c.

Mathematical expression for $\sigma(\theta)$, $\epsilon_{11} = \epsilon_{11c}$, $\epsilon_{12} = \epsilon_{12c}$, and $W = W_c$ are as follows:

$$\sigma(\theta) = \sum_{i=1}^{2} \frac{E_i R}{h} \frac{1}{E_i^2 R^2/v^2\eta_i^2 + 1} \cdot \left(\frac{E_i R}{v\eta_i} \sin\theta - \cos\theta + e^{-(E_i R/v\eta_i)\theta} \right) \tag{11}$$

$$\epsilon_{11} = \frac{R}{h} \cdot \frac{1}{E_1^2 R^2/v^2\eta_1^2 + 1} \cdot \left(\frac{E_1 R}{v\eta_1} \sin\theta - \cos\theta + e^{-(E_1 R/v\eta_1)\theta} \right) = \epsilon_{11c} \tag{12}$$

$$\epsilon_{12} = \frac{E_1 R^2}{hv\eta_1} \cdot \frac{1}{E_1^2 R^2/v^2\eta_1^2 + 1} \cdot \left[\frac{E_1 R}{v\eta_1}(1 - \cos\theta) - \sin\theta - \frac{v\eta_1}{E_1 R}\left(e^{-(E_1 R/v\eta_1)\theta} - 1\right) \right] = \epsilon_{12c} \tag{13}$$

$$W = \sum_{i=1}^{2} \frac{1}{2} \cdot \frac{\sigma_i^2}{E_i} + W_c \tag{14}$$

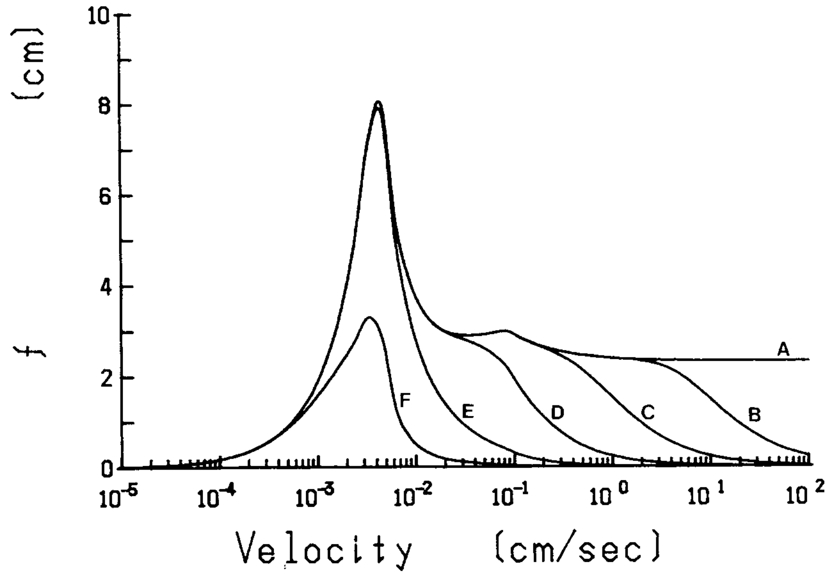

Fig. 3. Curves of f against v calculated according to eq. (10). Values of the parameters are as follows; $b = 2.0$ cm; $R = 1.0$ cm; $Mg = 6.0 \times 10^4$ dyn; $h = 0.001$ cm; $E_1 = 10^{10}$ dyn/cm^2; $E_2 = 10^{10}$ dyn/cm^2; $\eta_1 = 10^{10}$ P; $\eta_2 = 10^9$ P; $\epsilon_{11c} = 0.1$; $\epsilon_{12c} = 0.3$; $W_c = 7.0 \times 10^7$ erg/cm^3; $k = \infty$ (curve A), 1000 (curve B), 100 (curve C), 10 (curve D), 1 (curve E), and 0.1 (curve F).

Some appropriate values are given for the parameters of the mechanical model, for the critical values in the failure criteria and for the rate constant of the bonding process, and values of f_a are calculated according to eq. (10), where θ_b is determined by solving eqs. (12), (13) and (14) for θ. An example of these calculations is illustrated in Figure 3. It is clearly shown that the bonding process has a remarkable influence upon the shape of the curve of f_a vs. log v in the higher velocity side. The curve goes down drastically at some velocity which is approximately equal to $v = k/10$ in this case, and f_a is almost zero at higher velocity region (higher than $v = k$). This means that if velocity of pulling the cylinder on pressure-sensitive adhesives is extremely high, debonding occurs before the efficient contact of the two materials is realized. If k is large enough, which means that the bonding process proceeds instantaneously, the resultant curve is the same as that obtained according to the previously proposed theory.

Consideration of Fiber Forming

It has been well known that fiberlike structures are formed in the deforming zone of the adhesive when a pressure-sensitive tape or a label is peeled off from a solid. Recently, Urahama[13] observed the shapes and sizes of the fibers which are formed between a backing material and a transparent substrate during the peeling process which proceeds at relatively low velocity, by means of a microscope equipped with a camera. He pointed out the fact that the fibers have very complicated shapes, which depend strongly upon the type of

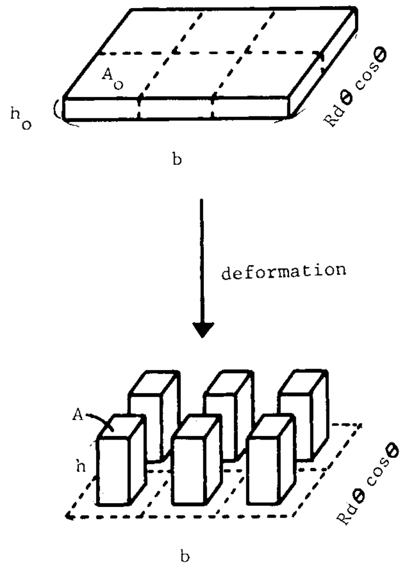

Fig. 4. Simplified model of fiber forming.

backing materials (porous or nonporous), rather than upon the type of adhesives (elastomer or acrylics).

Similar phenomena must occur when a cylinder rolls on a pressure-sensitive adhesive. Here in this paper, we will simplify the phenomena of fiber forming like this: The adhesive layer is *a priori* divided into small elements, as schematically shown in Figure 4, and as each element is elongated in one direction, it contracts in the other two directions without any change in its volume, which means that the effective bonding area is decreasing. Then,

$$h_0 A_0 = hA \tag{15}$$

$$\frac{A}{A_0} = \frac{h_0}{h} = \frac{1}{\epsilon + 1} \tag{16}$$

where A is the effective bonding area and A_0 is that before deformation. Therefore, the area of the surface element in Figure 1 should be multiplied by a factor of $(\epsilon + 1)^{-1}$ in deriving the equation for f_a. If we consider the time-dependent bonding process and the fiber-forming process at the same time, a general expression of f_a must be expressed as follows:

$$f_a = \frac{R^2 b}{\mathrm{Mg}} \int_0^{\theta_b} \sigma(\theta)(1 - e^{-kR\theta/v}) \cdot \frac{1}{(R/h)(1 - \cos\theta) + 1} \cdot \sin\theta \cos\theta \, d\theta \tag{17}$$

Figure 5 shows an example of the curve of f_a vs. log v, which is calculated according to eq. (17). Here again, the mechanical model shown in Figure 2 is

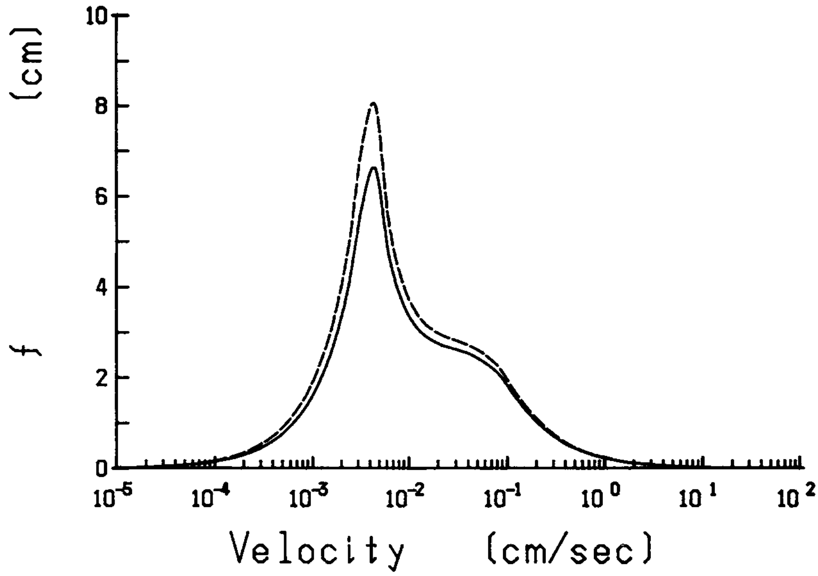

Fig. 5. A curve of f against v calculated according to eq. (17) for $k = 10$. Values of the parameters are the same as those given in Figure 3. Curve D in Figure 3 is also shown by a dotted line for comparison.

adopted, and therefore the mathematical expressions for $\sigma(\theta)$, $\epsilon_{11} = \epsilon_{11c}$, $\epsilon_{12} = \epsilon_{12c}$, and $W = W_c$ must be (11), (12), (13), and (14), respectively. Values of the parameters are the same as those given in Figure 3. We can compare the two curves (Figs. 3 and 5) for the same values of the parameters. The major effect of fiber forming upon the curve seems to suppress the absolute value of f_a.

CONCLUSION

It has been pointed out that the tack of pressure-sensitive adhesives is closely related to the rolling friction coefficient of the materials, and in the previous paper the author has proposed a theory by which f_a is calculated for the case where a cylinder is pulled on an adhesive at a constant velocity. However, only the debonding process was considered there. In this paper, the theory is modified so as to include the bonding process which is time-dependent, and also the fiber forming in a very simplified manner.

The bonding process has a remarkable effect on the shape of the curve of f_a vs. log v, especially in the higher velocity region, and the fiber forming has an effect that the absolute value of f_a is somewhat suppressed for the same values of the parameters. The general features of the experimentally observed curves[5] of f_a vs. log v for some pressure-sensitive adhesives can be reproduced by this modified theory much better than the previous one.

References

1. J. Johnston, *Adhesives Age*, **26**, 34 (1983).
2. F. Urushizaki, H. Yamaguchi, and H. Mizumachi, *J. Adhesion Soc. Jpn.*, **20**, 295 (1984).
3. H. Mizumachi and T. Saito, *J. Adhesion*, **20**, 83 (1986).

4. H. Mizumachi, *J. Adhesion Soc. Jpn.*, **20**, 522 (1984).
5. H. Mizumachi and Y. Hatano, *J. Adhesion*, **21**, 251 (1987).
6. H. Mizumachi, *Mater. Technol.* (*Jpn.*), **2**, 6 (1984).
7. H. Mizumachi, *J. Appl. Polym. Sci.*, **30**, 2675 (1985).
8. T. Tsukatani, Y. Hatano, B. Tomita, and H. Mizumachi, Proc. 36th Annual Meeting, Japan Wood Res. Soc. 1986, p. 147.
9. W. M. Bright, in *Adhesion and Adhesives*, J. E. Rutzler, Jr. and R. L. Sovage, Eds., Wiley, New York, 1954, p. 130.
10. K. Kanamaru, *Kolloid Z.*, **192**, 51 (1963).
11. T. Hata, *Zairyo*, **17**, 322 (1968).
12. T. Hata, *J. Adhesion Soc. Jpn.*, **8**, 64 (1972).
13. Y. Urahama, *J. Adhesion Soc. Jpn.*, **23**, 171 (1987).

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