Theory of Tack of Pressure Sensitive Adhesive. I

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Synopsis

If the tack of a pressure-sensitive adhesive is closely related to the rolling motion of a ball on the material, it is more scientific to express tack in terms of the rolling friction coefficient, which depends on the physical properties of the materials, and not on any trivial conditions of measurements. It is shown that the rolling friction coefficient of a pressure-sensitive adhesive can experimentally be determined from the pulling cylinder method much more easily than the rolling ball method and that we can theoretically calculate the rolling friction coefficient by making some assumptions, concerning deformation and failure of a pressure-sensitive adhesive.

INTRODUCTION

Tack is one of the most important properties of a pressure-sensitive adhesive, and it is measured mainly by two kinds of methods; one is the probe tack test and the other the rolling ball tack test. Here, the physical problems concerning the latter are discussed.

It is believed that rolling motion of a ball on a pressure-sensitive adhesive reflects tackiness of the adhesive because the motion must be closely related to bonding and unbonding processes which occur simultaneously at the surface of contact. Therefore, the rolling ball tests have been used in many countries for a very long time.

In the Dow method of measuring the rolling ball tack, balls are rolled under a standardized condition on an inclined surface on which a sample of a pressure-sensitive adhesive is placed, and the diameter of the largest ball which stops within some region is determined. And in case of the PSTC-6 method, a ball of a certain diameter rolls down on an inclined path, and it goes on a horizontal surface of pressure-sensitive adhesive. Here the tack of the adhesive is expressed by rollout distance.

These ways of expressing tack are useful in some practical cases, but the physical meaning of the values is not necessarily clear. For scientific purposes, we had better develop a method, by which tack of pressure sensitive adhesive is expressed with significant physical meanings. Fundamentally, the rolling motion of a ball must be described by a set of equations of motion, where the rolling friction coefficient is involved. If the rolling motion of a ball really reflects tackiness of a pressure-sensitive adhesive, we have to take into account the rolling friction coefficient of it. The rolling friction coefficient is independent of such factors as the angle of inclination and leading distance, but is dependent on the physical properties of the materials. Therefore, it would be reasonable to think that tack may be expressed in terms of this fundamental quantity in order to treat the problem on scientific bases.

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In previous report,[1] equations of motion of a rolling ball are solved, and methods to evaluate rolling friction coefficient of pressure-sensitive adhesive from experimental data are shown. It became clear that we have to perform a rather elaborate analysis according to some complicated equations, because the velocity of a ball changes at every moment and at the same time the rolling friction coefficient varies as a function of velocity.

Then, in this report, it will be shown that rolling friction coefficients of pressure-sensitive adhesives can be determined by the pulling cylinder method much more easily than by the rolling ball method. If a force to pull a cylinder on a pressure-sensitive adhesive at a constant velocity is measured, one can calculate its rolling friction coefficient without any elaborate analysis. Measurements can be made under a well-defined condition, and no assumption is needed concerning the dependence of the rolling friction coefficient on velocity. In addition, it is shown that the rolling friction coefficient can theoretically be calculated under some assumptions.

It would be worthwhile to accumulate data of tackiness of many pressure-sensitive adhesives which are expressed in terms of rolling friction coefficients in order to promote our understandings on science of adhesion.

ROLLING MOTION OF A CYLINDER ON A PRESSURE-SENSITIVE ADHESIVE

A cylinder of radius R, length b, and weight Mg is pulled by a force P along an inclined surface with an angle of α, upon which a sample of pressure sensitive adhesive is placed as schematically shown in Figure 1. Equations of motion[2] are

$$M\ddot{x} = P - F - Mg\sin\alpha \tag{1}$$

$$M\ddot{y} = 0 = N - Mg\cos\alpha \tag{2}$$

$$I\ddot{\varphi} = RF - fN \tag{3}$$

$$x = R\varphi + \text{const} \tag{4}$$

where F is the static frictional force, N is the normal force, φ and I are the angle of rotation and moment of inertia, respectively, of a cylinder, and f is the rolling friction coefficient of the adhesive. From these equations, we get

$$(MR + I/R)\,\ddot{x} = RP - RMg\sin\alpha - fMg\cos\alpha \tag{5}$$

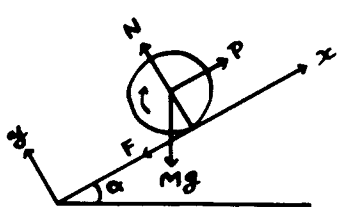

Fig. 1. Rolling cylinder.

If a cylinder is pulled at a constant velocity, then $\ddot{x} = 0$. Therefore,

$$P = Mg \sin \alpha + (f/R)Mg \cos \alpha \tag{6}$$

In case where $\alpha = 0$,

$$P = (f/R)Mg \tag{7}$$

or

$$f = PR/Mg \tag{8}$$

It is very easy for us to measure the force required to pull a cylinder on a pressure-sensitive adhesive at a constant velocity, and then the value of f is calculated without difficulty. There is another advantage that R and Mg can easily be varied independently in the case of a cylinder.

There are no literature values of f obtained by the rolling cylinder method as far as sticky materials are concerned. However, we can find some data, which are related to the values of f of the materials. Bull, Martin, and Vale[3] measured "tack" of a particular pressure-sensitive adhesive by means of a rotary drum technique. Tack in their study is directly proportional to f, and it is shown that f decreases as velocity of rotation increases in some range. On the other hand, Watanabe and Amari[4] studied on tack of printing ink by rolling cylinder method. They expressed their data in terms of tack energy density Dv instead of f. It is reasonable to think that these two quantities are somewhat proportional to each other. It is shown that Dv (or f) becomes very low when the velocity of the rolling cylinder becomes extremely low for the very soft materials. Then, if we plot the values of f against logarithm of velocity for a viscoelastic material over a very wide range of velocity, it would be possible to imagine that some curve is obtained, which increases from a relatively low value to a certain maximum and then decreases as velocity becomes higher.

THEORETICAL CALCULATIONS OF ROLLING FRICTION COEFFICIENTS

Now, what is the physical meaning of rolling friction coefficient? If stresses generated under a rolling cylinder are not symmetrical, there occurs rolling friction. It is quite natural to think that the rolling friction coefficient f of a viscoelastic material generally consists of two terms:

$$f = f_c + f_a \tag{9}$$

where f_c represents rolling friction caused by compressive deformation of the substrate material and f_a that caused by adhesion or extensional deformation of the substrate, which are schematically shown in Figure 2.

Theoretical and experimental studies concerning f_c have been developed by many researchers such as Eldredge and Tabor,[5] Tabor,[6] Flom and Bueche,[7] May, Morris, and Atack,[8] Flom,[9] and Minato and Takemura,[10] but

Fig. 2. Rolling friction caused by adhesion (f_a) and that caused by compressive deformation of the substrate (f_c).

few authors have studied f_a. In the case of a pressure-sensitive adhesive, f_a must be much larger than f_c, and therefore, we have only to develop theoretical calculations of f_a here.

When a cylinder is pulled on a pressure-sensitive adhesive in one direction, the adhesive will elongate and pull the cylinder from the backside, disturbing its rotation, as shown in Figure 3. If compressive deformation of the adhesive is neglected, strain of the adhesive ϵ at θ is expressed as

$$\epsilon = (R/h)(1 - \cos\theta) \tag{10}$$

and rate of strain $\dot{\epsilon}$ is

$$\dot{\epsilon} = (R\dot{\theta}/h)\sin\theta = (v/h)\sin\theta \tag{11}$$

where h and v are original thickness of the adhesive layer and velocity of the cylinder, respectively. If we adopt a mechanical model, stress σ generated by elongation of the adhesive can be expressed as a function of θ. Then, the moment m_a caused by the extended part of the adhesive can be calculated as follows:

$$dm_a = R\,\sigma\,bR\,d\theta\cos\theta\sin\theta \tag{12}$$

$$m_a = R^2 b\int_0^{\theta_b}\sigma(\theta)\cos\theta\sin\theta\,d\theta \tag{13}$$

where θ_b is the angle where failure is steadily proceeding. This moment m_a is set to be equal to f_aMg, according to the definition of the rolling friction coefficient. Therefore, a general expression for f_a is

$$f_a = \frac{R^2 b}{mg}\int_0^{\theta_b}\sigma(\theta)\cos\theta\sin\theta\,d\theta \tag{14}$$

Now, results of the calculations for some cases are given. It is shown that curves of f_a vs. log v are different if we choose different viscoelastic model and different failure criterion.

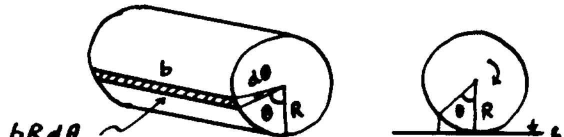

Fig. 3. Rolling cylinder.

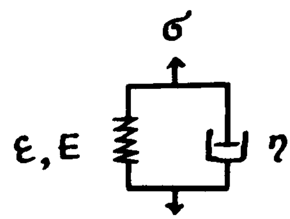

Fig. 4. A single Voigt element: σ = stress; ϵ = strain; E = modulus of a spring; η = viscosity of a dashpot.

Model I: A Single Voigt Element

If we assume that mechanical behaviors of the pressure-sensitive adhesive can be described by a single Voigt element, as Hata[11] did in his theory of peeling, stress σ is given as a function of θ like this:

$$\sigma = (ER/h)(1 - \cos\theta) + (\eta v/h)\sin\theta \tag{15}$$

Figure 4 shows the model. Substituting Eq. (15) into Eq. (14), we get

$$\begin{aligned} f_a = {} & (ER^3b/Mgh)\left(\frac{1}{2}\cos^2\theta_b - \cos\theta_b + \frac{1}{2}\right) \\ & + (\eta R^2bv/Mgh)\left(\frac{1}{2}\theta_b - \frac{1}{4}\sin 2\theta_b\right) \end{aligned} \tag{16}$$

θ_b can be determined if we adopt some failure criterion.

Failure Criterion (A): $\sigma = \sigma_c$

In case where a failure criterion is $\sigma = \sigma_c$, which means that failure occurs when stress becomes a certain critical value σ_c, the value of θ_b is then determined by substituting σ_c into eq. (15) and solving it numerically for θ, and f_a is calculated according to eq. (16). Curves of f_a vs. log v are shown in Figure 5. f_a decreases antisigmoidally as v increases in this case.

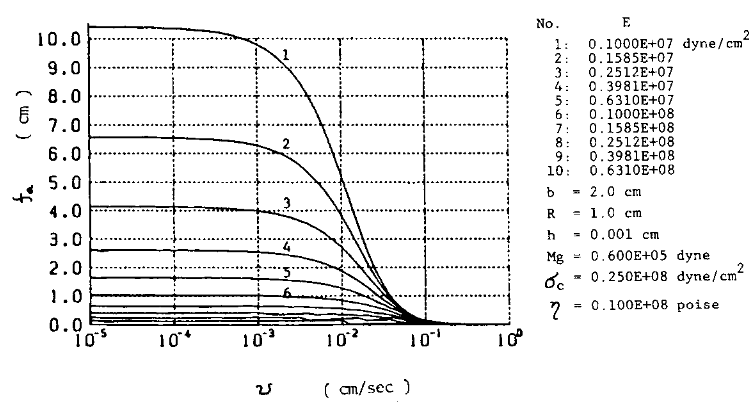

Fig. 5. Curves of f_a vs. log v for a single Voigt element. Failure criterion is $\sigma = \sigma_c$. Values of the parameters used for the calculations are also given in the figure.

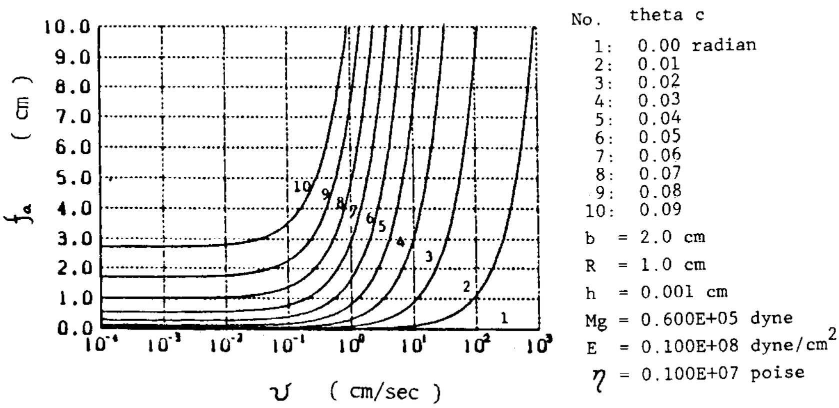

Fig. 6. Curves of f_a vs. log v for a single Voigt element. Failure criterion is $\epsilon = \epsilon_c$.

Failure Criterion (B): $\epsilon = \epsilon_c$ or $\theta = \theta_c$

If we adopt this failure criterion, which means that failure occurs when strain becomes a certain critical value ϵ_c, f_a can be calculated according to eq. (16), assuming θ_b is a constant (θ_c). f_a increases as v increases, as shown in Figure 6.

Model II: A Single Maxwell Element

If we assume that mechanical behaviors of the pressure-sensitive adhesive can be described by a single Maxwell element, as Fukuzawa[12] did in his theory of peeling, the following equations hold:

$$\sigma = E\epsilon_1 = \eta\, \dot{\epsilon}_2 \tag{17}$$

$$\dot{\epsilon}_1 + \dot{\epsilon}_2 = \dot{\epsilon} \tag{18}$$

From these equations, we get the differential equation

$$\frac{d\sigma}{d\theta} + \frac{ER}{v\eta}\sigma = \frac{ER}{h}\sin\theta \tag{19}$$

and the solution is

$$\sigma = \frac{ER}{h}\cdot\frac{1}{E^2R^2/v^2\eta^2 + 1}\cdot\left(\frac{ER}{v\eta}\sin\theta - \cos\theta + e^{-(ER/v\eta)\theta}\right) \tag{20}$$

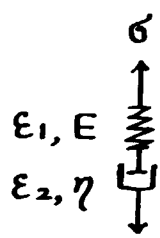

Fig. 7. A single Maxwell element. σ = stress; ϵ_1 = strain of a spring; ϵ_2 = strain of a dashpot; E = modulus of a spring; η = viscosity of a dashpot.

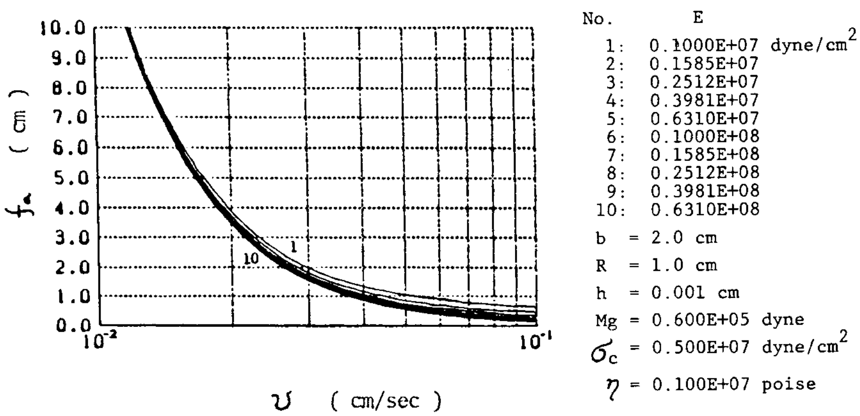

Fig. 8. Curves of f_a vs. log v for a single Maxwell element. Failure criterion is $\sigma = \sigma_c$.

Then, the rolling friction coefficient is calculated according to eqs. (20) and (14):

$$f_a = \frac{ER^3b}{Mgh} \cdot \frac{1}{E^2R^2/v^2\eta^2 + 1} \cdot \left\{ \frac{ER}{3v\eta} \sin^3 \theta_b + \frac{1}{3} (\cos^3 \theta_b - 1) + \frac{1}{2} \cdot \frac{1}{E^2R^2/v^2\eta^2 + 4} \cdot \left[2 - e^{-(ER/v\eta)\theta_b} \left(\frac{ER}{v\eta} \sin 2\theta_b + 2 \cos 2\theta_b \right) \right] \right\} \tag{21}$$

Failure Criterion (A): $\sigma = \sigma_c$

Equation (20) is numerically solved for $\theta(= \theta_b)$ and the value of f_a is calculated according to eq. (21). Figure 8 shows that f_a decreases very sharply as v increases.

Failure Criterion (B): $\epsilon = \epsilon_c$ or $\theta = \theta_c$

The value of f_a is calculated according to eq. (21), assuming θ_b is a constant. f_a increases sigmoidally as v increases in this case, as shown in Figure 9.

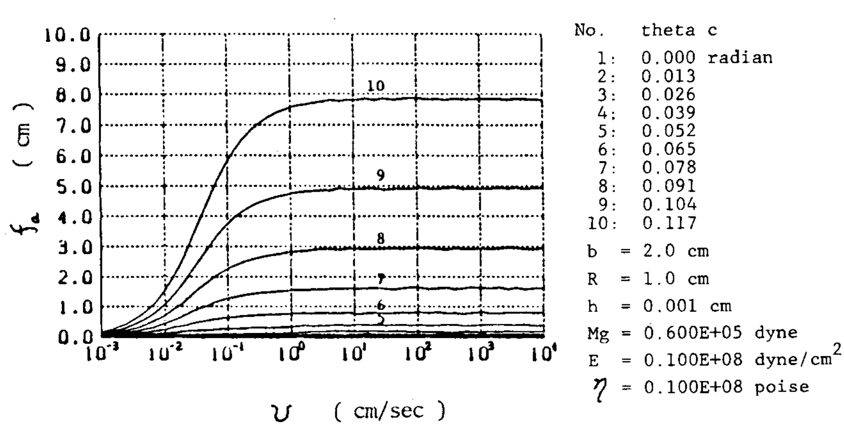

Fig. 9. Curves of f_a vs. log v for a single Maxwell element. Failure criterion is $\epsilon = \epsilon_c$.

Model III: Two Maxwell Elements in Parallel Connection

Hata[13,14] has shown that failure envelopes of viscoelastic materials (adhesives) can be reproduced if we choose a simple viscoelastic model as shown in Figure 10, and assume some failure criteria. His theory can be made use of for the calculation of f_a. Stresses of the model are expressed by the following equation:

$$\sigma = \sum_{i=1}^{2} \frac{E_i R}{R} \cdot \frac{1}{E_i^2 R^2/v^2\eta_i^2 + 1} \cdot \left(\frac{E_i R}{v\eta_i} \sin\theta - \cos\theta + e^{-(E_i R/v\eta_i)\theta} \right) \tag{22}$$

f_a is obtained from eqs. (22) and (14):

$$\begin{aligned} f_a = \sum_{i=1}^{2} \frac{E_i R^3 b}{Mgh} \cdot \frac{1}{\dfrac{E_i^2 R^2}{v^2\eta_i^2} + 1} \cdot &\left\{ \frac{E_i R}{3v\eta_i} \sin^3\theta_b + \frac{1}{3}(\cos^3\theta_b - 1) \right. \\ &\left. + \frac{1}{2} \cdot \frac{1}{E_i^2 R^2/v^2\eta_i^2 + 4} \cdot \left[2 - e^{-(E_i R/v\eta_i)\theta_b} \left(\frac{E_i R}{v\eta_i} \sin 2\theta_b + 2\cos 2\theta_b \right) \right] \right\} \end{aligned} \tag{23}$$

Failure Criterion (A): $\epsilon_{11} = \epsilon_{11c}$

In the region where the rate of strain is very high, failure occurs when strain of the spring in the weak point (element 1) reaches a critical value ϵ_{11c}. ϵ_{11} of the model is expressed as a function of θ; then

$$\epsilon_{11} = \frac{\sigma_1}{E_1} = \frac{R}{h} \cdot \frac{1}{E_1^2 R^2/v^2\eta_1^2 + 1} \cdot \left(\frac{E_1 R}{v\eta_1} \sin\theta - \cos\theta + e^{-(E_1 R/v\eta_1)\theta} \right) = \epsilon_{11c} \tag{24}$$

Failure Criterion (B): $\epsilon_{12} = \epsilon_{12c}$

In the region where the rate of strain is very low, failure occurs when strain of the dashpot in the weak point reaches a critical value ϵ_{12c}.

$$\dot{\epsilon}_{12} = \frac{d\epsilon_{12}}{d\theta} \cdot \frac{d\theta}{dt} = \frac{v}{R} \cdot \frac{d\epsilon_{12}}{d\theta} = \frac{\sigma_1}{\eta_1} \tag{25}$$

$$\begin{aligned} \epsilon_{12} = \frac{E_1 R^2}{hv\eta_1} \cdot \frac{1}{E_1^2 R^2/v^2\eta_1^2 + 1} \cdot &\left[\frac{E_1 R}{v\eta_1}(1 - \cos\theta) \right. \\ &\left. - \sin\theta - \frac{v\eta_1}{E_1 R}\left(e^{-(E_1 R/v\eta_1)\theta} - 1\right) \right] = \epsilon_{12c} \end{aligned} \tag{26}$$

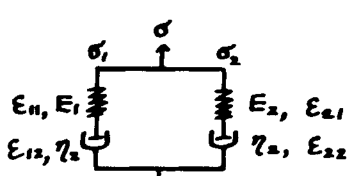

Fig. 10. Two Maxwell elements in parallel connection: σ, σ_i = stress; ϵ_{i1} = strain of a spring; ϵ_{i2} = strain of a dashpot; E_i = modulus of a spring; η_i = viscosity of a dashpot.

Failure Criterion (C): $W = W_c$

In the case where adhesion is concerned, interfacial failure occurs when stored energy in the springs of the model reaches a critical value W_c:

$$W = \sum_{i=1}^{2} \frac{1}{2} \epsilon_{i1}^2 E_i = \sum_{i=1}^{2} \frac{1}{2} \cdot \frac{\sigma_i^2}{E_i} = W_c \tag{27}$$

Some appropriate values are given as the critical values, ϵ_{11c}, ϵ_{12c}, and W_c, and eqs. (24), (26), and (27) are solved for θ, and values of f_a are calculated according to eq. (23). An example of the above calculations is shown in Figure 11. Shapes of the curves are different if different critical values are given. It is shown that f_a vs. log v curves changes drastically as the failure mode changes from dashpot failure (or ductile failure) to spring failure (or brittle failure), and then to interfacial failure.

Model IV: Multiple Maxwell Elements in Parallel Connection

Hata[15] generalized his theory of fracture, using the multiple Maxwell model, and obtained substantially the same conclusion. The theory of f_a can also be developed for the same model:

$$\sigma = \sum_{i=1}^{n} \frac{E_i R}{h} \cdot \frac{1}{E_i^2 R^2/v^2\eta_i^2 + 1} \cdot \left(\frac{E_i R}{v\eta_i} \sin\theta - \cos\theta + e^{-(E_i R/v\eta_i)\theta}\right) \tag{28}$$

$$f_a = \sum_{i=1}^{n} \frac{E_i R^3 b}{Mgh} \cdot \frac{1}{E_i^2 R^2/v^2\eta_i^2 + 1} \cdot \left\{\frac{E_i R}{3v\eta_i} \sin^3\theta_b + \frac{1}{3}\left(\cos^3\theta_b - 1\right) + \frac{1}{2} \cdot \frac{1}{E_i^2 R^2/v^2\eta_i^2 + 4} \cdot \left[2 - e^{-(E_i R/v\eta_i)\theta_b}\left(\frac{E_i R}{v\eta_i} \sin 2\theta_b + 2\cos 2\theta_b\right)\right]\right\} \tag{29}$$

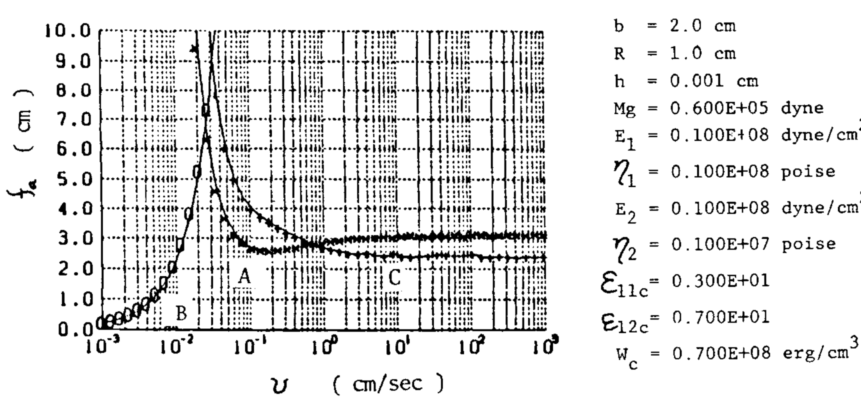

Fig. 11. Curves of f_a vs. long v for two Maxwell elements in parallel connection. Failure criteria are: (A) $\epsilon_{11} = \epsilon_{11c}$ (spring failure); (B) $\epsilon_{12} = \epsilon_{12c}$ (dashpot failure); (C) $W = W_c$ (interfacial failure).

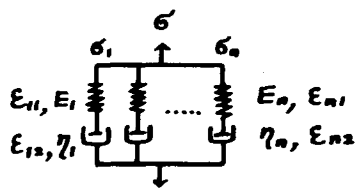

Fig. 12. Multiple Maxwell elements in parallel connection: σ, σ_i = stress; ϵ_{i1} = strain of a spring; ϵ_{i2} = strain of a dashpot; E_i = modulus of a spring; η_i = viscosity of a dashpot.

Failure Criterion (A): $\epsilon_{kl} = \epsilon_{klc}$

Failure occurs when strain of the spring of the kth element reaches a critical value ϵ_{klc}.

$$\epsilon_{kl} = \frac{R}{h}\cdot\frac{1}{\dfrac{E_k^2R^2}{v^2\eta_k^2}+1}\cdot\left(\frac{E_kR}{v_{\eta_k}}\sin\theta - \cos\theta + e^{-(E_kR/v\eta_k)\theta}\right) = \epsilon_{klc} \tag{30}$$

Failure Criterion (B): $\epsilon_{l2} = \epsilon_{l2c}$

Failure occurs when strain of the dashpot of the lth element reaches a critical value ϵ_{l2c}:

$$\epsilon_{l2} = \frac{E_lR^2}{hv\eta_l}\cdot\frac{1}{E_l^2R^2/v^2\eta_l^2+1}\cdot\left[\frac{E_lR}{v\eta_l}(1-\cos\theta) - \sin\theta - \frac{v\eta_l}{E_lR}\left(e^{-(E_lR/v\eta_l)\theta} - 1\right)\right] = \epsilon_{l2c} \tag{31}$$

Failure Criterion (C): $W = W_c$

Failure occurs when stored energy in the springs of the model reaches a critical value W_c:

$$W = \sum_{i=1}^{n}\frac{1}{2}\epsilon_{i1}^2E_i = \sum_{i=1}^{n}\frac{1}{2}\cdot\frac{\sigma_i^2}{E_i} = W_c \tag{32}$$

Relaxation spectra of the model are tentatively assumed as shown in Figure 13. Again, some appropriate values are given as ϵ_{k1c}, ϵ_{l2c}, and W_c,

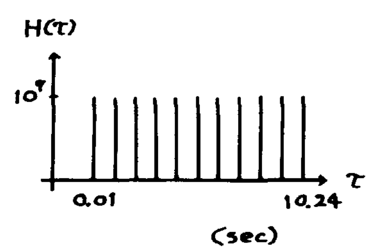

Fig. 13. Tentative relaxation spectra.

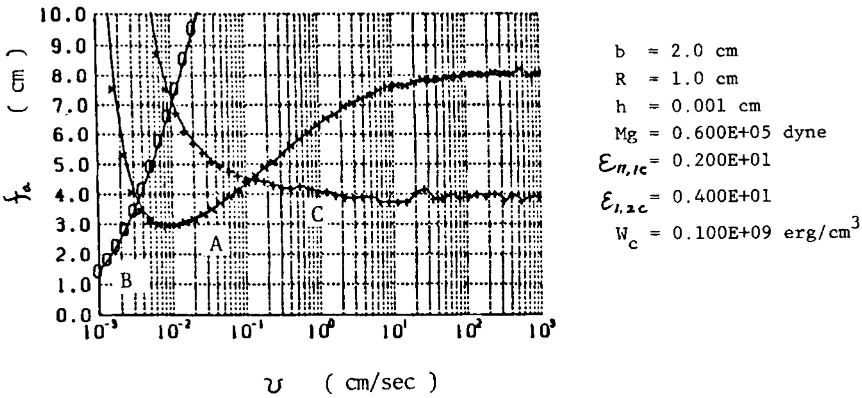

Fig. 14. Curves of f_a vs. long v for multiple Maxwell elements in parallel connection. Failure criteria are: (A) $\epsilon_{II,I} = \epsilon_{11,1c}$ (spring failure); (B) $\epsilon_{I,2} = \epsilon_{1,2c}$ (dashpot failure); (C) $W = W_c$ (interfacial failure).

and eqs. (30)–(32) are solved for θ, and f_a is calculated according to eq. (29). Figure 14 shows that similar curves are obtained for this model, too.

CONCLUSION

The rolling friction coefficient is a fundamental quantity to describe rolling motion of a ball or a cylinder on pressure sensitive adhesives, and therefore, tack must be expressed in terms of the rolling friction coefficient. The rolling friction coefficient is determined from the pulling cylinder experiments much more easily than the rolling ball experiments. The rolling friction coefficients can theoretically be calculated if we make some assumptions concerning deformation and failure of the adhesive. Curves of the rolling friction coefficient vs. the logarithm of velocity are different if we choose different mechanical model and failure criteria. It would be reasonable to expect that a curve of f vs. log v for a pressure-sensitive adhesive generally has a peak or peaks, and that f becomes low when velocity becomes extremely high or extremely low. Therefore, we will have to choose either model III or model IV if we want to interpret variations of f in very wide range of velocity.

Anyway, it is recommended that researchers of pressure-sensitive adhesives accumulate data on tack, which are expressed in terms of f, instead of some empirical parameters. If we prepare a series of pressure-sensitive adhesives having various viscoelastic properties, and obtain systematic data on f of them as a function of velocity, we can compare the experimental and theoretical curves, and these analyses will help us understand in detail the mechanism of tack or adhesion in general.

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