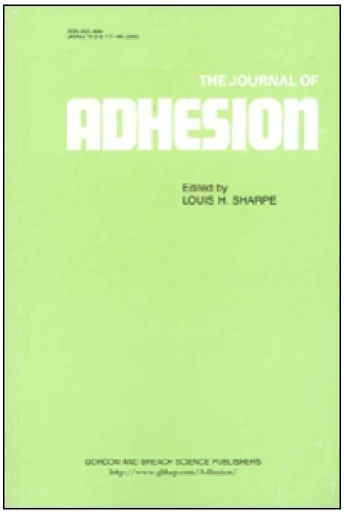

The Journal of Adhesion
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713453635

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To cite this Article Mizumachi, Hiroshi and Hatano, Yasunori(1987) 'Rheological Study on Tack of Pressure-Sensitive Adhesives', The Journal of Adhesion, 21: 3, 251 — 260
To link to this Article: DOI: 10.1080/00218468708074973
URL: http://dx.doi.org/10.1080/00218468708074973

J. Adhesion, 1987, Vol. 21, pp. 251–260
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Rheological Study on Tack of Pressure-Sensitive Adhesives†

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(Received August 26, 1986)

It is pointed out that the tack of pressure-sensitive adhesives should be expressed in terms of rolling friction coefficient f of the adhesives. Values of f were determined by both rolling ball and pulling cylinder experiments, and the dependence of f upon viscoelastic properties and thickness of the adhesives was studied. The experimental results were interpreted by the model theory previously proposed. It is also shown that the tack of pressure-sensitive adhesives by the conventional ball tack tests corresponds to f measured at the velocity ranging from $v \sim 10$ to $v \sim 10^2$ cm/sec.

KEY WORDS Pressure-sensitive adhesives; rolling ball; pulling cylinder; rolling friction coefficient; viscoelastic properties; tack.

INTRODUCTION

The tack of pressure sensitive adhesives is often measured by rolling ball methods.[1] It is reasonable to express the tack in terms of rolling friction coefficient f, because it depends upon the physical properties of the material, and not upon such experimental conditions as the leading distance or the angle of inclination of a surface. It has been shown that f of a pressure-sensitive adhesive can be evaluated by analysing the rolling motion of a ball on it.[2] If we assume that f is

† Presented at the Tenth Annual Meeting of The Adhesion Society, Inc., Williamsburg, Virginia, U.S.A., February 22–27, 1987.

a linear function of velocity v, namely $f = \varphi_0 + \varphi_1 v$, with φ_0 and φ_1 being constants, both the rolling process and rollout distance of a ball on a pressure-sensitive adhesive can satisfactorily be analysed according to a unified theory.[3] However, it has also been pointed out that f of a pressure sensitive adhesive can more easily be determined by a pulling cylinder method, without any assumption about the dependence of f on v.[4,5]

H. Mizumachi[6] proposed a model theory where f is expressed in terms of viscoelastic parameters of the material, by which the dependence of f on v can be calculated in a wide range. In this study, the influence of the viscoelastic properties of several pressure-sensitive adhesives upon f are experimentally examined and the rheological discussions are made on the results.

EXPERIMENTAL

Samples used in this study are shown in Table I which also gives the temperature of the viscoelastic adsorption peak $T(E''_{max})$ at 110 Hz and thickness of the adhesives.

TABLE I
Pressure sensitive adhesives

Sample	$T(E''_{max})$ at 110 Hz (°C)	Thickness (cm)
A1†	−50	4.0×10^{-3}
A2†	−45	4.0×10^{-3}
A3†	−30	4.0×10^{-3}
A4†	0	4.0×10^{-3}
B1‡	−35	2.6×10^{-2}
B2‡	−38	1.8×10^{-3}

† The pressure-sensitive adhesive is mounted directly on a glass plate.

‡ A double-faced pressure-sensitive adhesive tape is adhered on a glass plate, upon which a sample tape is placed with the adhesive surface facing up.

(A) Measurement of *f* by the rolling ball method

The Nichiban tester[1] was employed to measure the rollout distance $(x - x_0)_{st}$ as a function of initial height H of a ball. Measurements were repeated ten times (10 runs) for one condition, and a new specimen was set in the tester at each run. Values of φ_0 and φ_1 were determined by the least squares method, using the following equation;[3]

$$(x - x_0)_{st} = \frac{7R}{5g\varphi_1}\left\{\left(\frac{10}{7}\mathrm{gH}\right)^{1/2} - \frac{\varphi_1}{\varphi_1}\log\left(1 + \frac{\varphi_1}{\varphi_0}\left(\frac{10}{7}\mathrm{gH}\right)^{1/2}\right)\right\}$$

and then f is given by

$$f = \varphi_0 + \varphi_1 v$$

(B) Measurement of *f* by the pulling cylinder method

A cylinder was pulled at a constant velocity on a horizontal surface of pressure-sensitive adhesives, and the resistance P was measured. Actually, the plate carrying the specimen is driven at various speeds, while the cylinder is placed on the specimen and connected to a load cell. Velocity can be varied from 0.001 to 100 cm/sec. A new specimen was set in the tester at each run. Values of f can be calculated by the following equation[6] without any elaborate analysis;

$$f = \frac{PR}{Mg}$$

RESULTS AND DISCUSSIONS

(A) Dependence of *f* upon viscoelastic properties of a pressure-sensitive adhesive

Figure 1 shows some of the typical examples of a plot of rollout distance $(x - x_0)_{st}$ at 20°C *vs.* initial height H of a ball for a few pressure-sensitive adhesives with different $T(E''_{max})$. $T(E''_{max})$ at 110 Hz is higher than T_g by about twenty degrees. The average relaxation time τ at room temperature increases with the increase of $T(E''_{max})$. The shape of the curve in Figure 1 changes systematically with the change of the average relaxation time of the pressure-sensitive adhesives. Values of φ_0 and φ_1, obtained by the least squares method, for each adhesive are given in Table II.

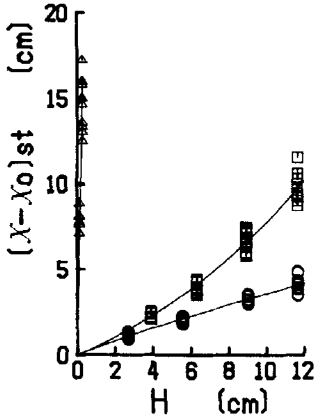

FIGURE 1 Typical examples of a plot of rollout distance $(a-x_0)_{st}$ at 20°C *vs.* initial height H of a ball for some pressure-sensitive adhesives.
○: $A2$, $R = 0.64$ cm, $\varphi_0 = 1.32$, $\varphi_1 = 0.006$
□: $A3$, $R = 0.96$ cm, $\varphi_0 = 2.258$, $\varphi_1 = -0.012$
△: $A4$, $R = 0.159$ cm, $\varphi_0 = 0.00465$, $\varphi_1 = -0.000115$
Theoretical curves are illustrated by the solid lines.

It is interesting that f at room temperature changes from a decreasing function of v (*i.e.*, $\varphi_1 < 0$) to an increasing function (*i.e.*, $\varphi_1 > 0$) as $T(E''_{max})$ decreases.

Then, f of the same pressure-sensitive adhesives were also measured by the pulling cylinder method at various velocities, and the results are illustrated in Figure 2. It is evident that f of a viscoelastic material has a peak at some velocity and that the peak

TABLE II
Values of φ_0 and φ_1 obtained by the least squares method

Sample	R (cm)	φ_0 (cm)	φ_1 (sec)	DEV† (cm^2)
A2	0.96	0.696	0.00066	6.96×10^1
	0.80	0.859	0.00116	3.52×10^1
	0.64	1.32	0.0060	3.14×10^0
A3	0.96	2.26	−0.0120	1.15×10^1
	0.80	2.76	−0.0141	7.31×10^0
	0.64	3.60	−0.0166	1.71×10^0
A4	0.159	0.00465	−0.000115	2.69×10^1

† $DEV = \Sigma (y - y_i)^2$

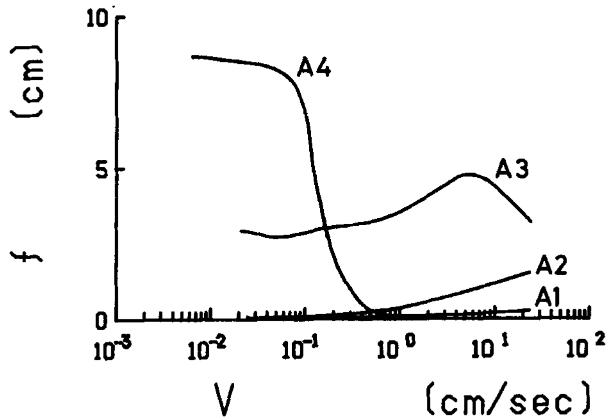

FIGURE 2 Values of f obtained from the pulling cylinder experiments at 20°C plotted against $\log v$ for A1, A2, A3 and A4. $R = 0.5$ cm, $b = 2.5$ cm, $M = 50$ g.

shifts toward the higher velocity region as the average relaxation time τ or T_g decreases, as schematically shown in Figure 7. This is in agreement with the trends of the theoretical curves of f *vs.* $\log v$, calculated according to the model theory.[6] In the previous report,[3] the rolling motion of a ball on some pressure-sensitive adhesives was traced by means of stroboscopic photography, and data on time/position of a ball were analysed according to equations of motion. In every case, all the experimentally observed data fell in the velocity range between approximately 10 and 10^2 cm/sec. Values of φ_0 and φ_1 could be determined from the data, and rollout distance calculated using those values agreed very well with the experimentally measured one. This means that the tack of pressure-sensitive adhesives which is measured by the conventional rolling ball tack tests must be related to f of a relatively narrow range of velocity, namely from $\log v \sim 1$ to $\log v \sim 2$. At room temperature, f of A1 and A2 is an increasing function of v in the above-mentioned velocity range and, on the other hand, f of A3 and A4 is a decreasing function of v in the same range.

It has been made clear that f in the velocity range $10 - 10^2$ cm/sec corresponds well to the rolling motion of a ball in the conventional ball tack tests, and that f of pressure-sensitive adhesives can easily be measured by the pulling cylinder method in a wide range of velocity.

(B) Dependence of *f* upon temperature and thickness of a pressure-sensitive adhesive

Figure 3 and Figure 4 show some examples of $(x - x_0)_{st}$ *vs.* H plots for the two pressure-sensitive adhesives, B1 and B2, at different temperatures. In the case of B1, the curve is almost straight or somewhat convex at 4°C, but it becomes concave at 20°C, while in case of B2, the curve is concave at both temperatures. This means that φ_1 changes from negative to positive as the temperature is raised from 4°C to 20°C in case of B1, but φ_1 is always negative in the same temperature range in case of B2. Values of φ_0 and φ_1 obtained by the least squares method is given in Table III.

Again, f of the same pressure-sensitive adhesives were measured at 4°C and 20°C by the pulling cylinder method. Figure 5 and Figure 6 show f of these adhesives at 4°C and 20°C as a function of velocity. A broad peak is generally observed. In the case of B1, f is a decreasing function of v at 4°C in the velocity range from 10 to

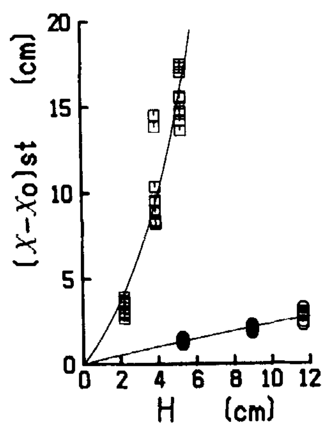

FIGURE 3 Examples of a plot of $(x - x_0)_{st}$ *vs.* H for B1 at different temperatures.
□: 4°C, $R = 0.96$ cm, $\varphi_0 = 0.799$, $\varphi_1 = -0.00746$
○: 20°C, $R = 0.96$ cm, $\varphi_0 = 3.13$, $\varphi_1 = 0.0115$
Theoretical curves are illustrated by the solid lines.

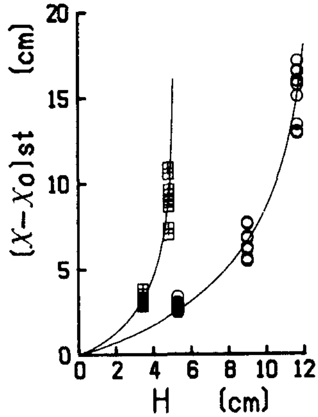

FIGURE 4 Examples of a plot of $(x - x_0)_{st}$ *vs.* H for B2 at different temperatures.
□: 4°C, $R = 0.159$ cm, $\varphi_0 = 0.4488$, $\varphi_1 = -0.005288$
○: 20°C, $R = 0.96$ cm, $\varphi_0 = 3.701$, $\varphi_1 = -0.0279$
Theoretical curves are illustrated by the solid lines.

10^2 cm/sec, but it is almost a constant or a somewhat increasing function at 20°C in the same range, while in the case of B2, f is a decreasing function of v between $v = 10$ and 10^2 cm/sec at both temperatures, which is consistent with the trends observed in the rolling ball experiments.

TABLE III
Values of φ_0 and φ_1 obtained by the least squares method

Sample	Temperature (°C)	R (cm)	φ_0 (cm)	φ_1 (sec)	DEV† (cm^2)
B1	20	0.96	3.13	0.0115	2.20×10^0
		0.80	4.05	0.0002	7.28×10^{-1}
		0.64	0.378	0.0719	7.73×10^{-1}
	4	0.96	0.799	−0.00746	7.78×10^1
		0.80	1.042	−0.00975	5.04×10^1
		0.64	1.152	−0.0100	2.73×10^1
B2	20	0.96	3.701	−0.0279	3.05×10^1
		0.80	3.989	−0.0304	3.67×10^1
		0.64	3.67	−0.0268	1.37×10^1
	4	0.159	0.4488	−0.00529	1.42×10^1

† $DEV = \Sigma (y - y_i)^2$

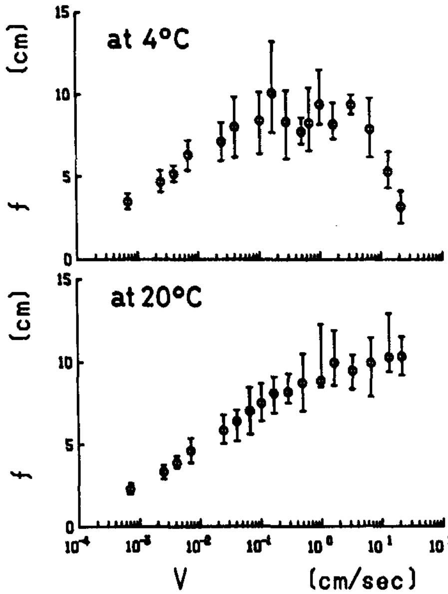

FIGURE 5 Values of f obtained from the pulling cylinder experiments at 4°C and 20°C plotted against log v for B1, $R = 0.5$ cm, $B = 2.5$ cm, $M = 50$ g.

Now, we have to discuss the reason why these differences appear in the two pressure-sensitive adhesives. Viscoelastic properties of the two adhesives are quite similar, but B1 is much thicker than B2. If thickness of the adhesive is different, rate of strain of the adhesive is different at the same velocity of movement of a ball or a cylinder.

It is easily shown by the model theory[6] that a peak of f shifts toward higher velocity region as the adhesive becomes thicker. (Figure 7).

In the case of B1, which is very thick, a peak is then located at very high velocity, and the velocity range of our concern corresponds to the increasing part of the curve of f *vs.* v at 20°C, which corresponds to $\varphi_1 > 0$. But when the temperature is lowered to 4°C, the peak shifts toward the lower velocity region, and then f will become a decreasing function within the above-mentioned velocity range, which corresponds to $\varphi_1 < 0$. On the other hand, the adhesive layer of B2 tape is very thin and in this case a peak of the curve of f *vs.* v is located at very low velocity, and then f is

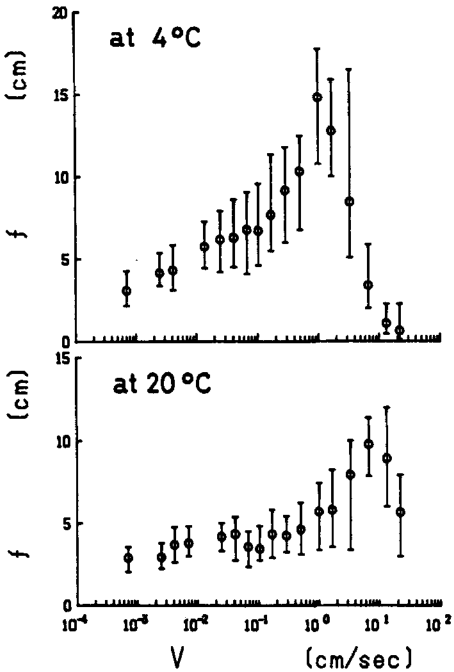

FIGURE 6 Values of f obtained from the pulling cylinder experiments at 4°C and 20°C plotted against log v for B2. $R = 0.5$ cm, $b = 2.5$ cm, $M = 50$ g.

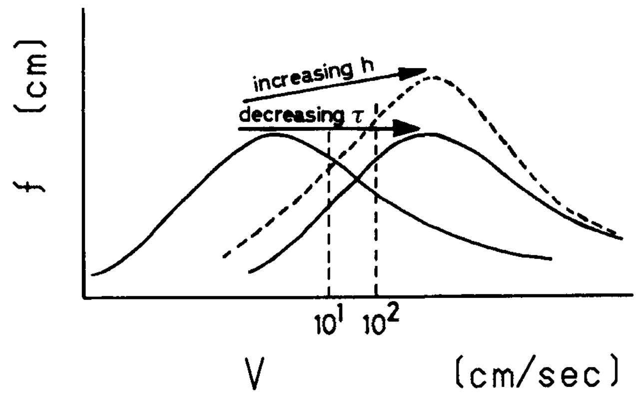

FIGURE 7 Schematic representation of f *vs.* log v curve. Direction of shifts of the curve due to the change of some parameters are shown by the arrows. The curve shifts toward the higher velocity region when the average relaxation time τ decreases (*i.e.*, when temperature is raised for a pressure-sensitive adhesive, or when T_g's of the adhesives are lowered in case the temperature of tests is fixed), or when thickness h of the adhesive layer increases.

decreasing during $10-10^2$ cm/sec at 20°C. When the temperature is lowered, the peak shifts further toward the lower velocity region, and the effective velocity range in the conventional ball tack tests still corresponds to a decreasing part of f, which means that φ_1 is always negative.

CONCLUSIONS

The tack of pressure-sensitive adhesives should be expressed by the rolling friction coefficient f of the adhesives, and it can be estimated by analysing the rolling motion of a ball. By employing the pulling cylinder method, f can be measured more easily.

It has been indicated that f is closely related to the viscoelastic properties of the material. If we plot f of a viscoelastic material against $\log v$ over a very wide range of velocity, a curve having a peak is generally observed, and the curve shifts toward the higher velocity region when the average relaxation time τ is lowered (*i.e.*, when temperature is raised for a pressure-sensitive adhesive, or when T_g's of the adhesives are lowered in case the temperature of measurement is fixed), or when thickness of the adhesive becomes larger.

It was pointed out that f obtained from rolling ball experiments corresponds to that from pulling cylinder experiments in the velocity range of $10-10^2$ cm/sec. The pulling cylinder method is a good way to measure f of pressure-sensitive adhesives over a very wide range of velocity under a well-defined condition.

Acknowledgement

Part of the work was supported by a Grant-in-Aid for Co-operative Research of The Ministry of Education, Science and Culture of Japan. The authors wish to thank Dr. T. Saito of Nichiban Co. Ltd. for providing samples.

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