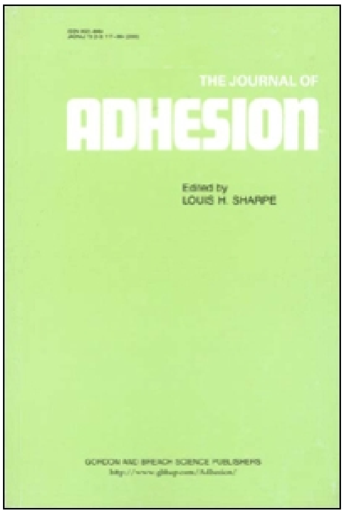

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Yoshiaki Urahama[a]; Kentaro Yamamoto[b]
[a] Central Research Laboratory, Nitto Electric Industrial Co., Ltd., Ibaraki, Osaka, Japan [b] Laboratory of Applied Metrology, Department of Precision Engineering, Faculty of Engineering, Meiji University 1-1-1, Kawasaki, Kanagawa, Japan

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Evaluation of Adhesive Properties of PSA Tape by Rolling Adhesive Moment Tester

YOSHIAKI URAHAMA

Central Research Laboratory, Nitto Electric Industrial Co., Ltd. 1–2, 1–chome, Shimohozumi, Ibaraki, Osaka 567, Japan

KENTARO YAMAMOTO

Laboratory of Applied Metrology, Department of Precision Engineering, Faculty of Engineering, Meiji University 1–1–1, Higashimita, Tama-ku, Kawasaki, Kanagawa 214, Japan

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The rolling adhesive moment of pressure-sensitive adhesives is studied over a wide range of rolling rates, giving the velocity spectrum of the rolling adhesive moment. To determine the significance of these velocity spectra, their correlation to various pressure-sensitive adhesive properties has been examined.

As a result, the following have been made clear:

(1) The time-temperature superposition principle is applicable to the correlation coefficient between the rolling adhesive moment and the 180° peel force. In other words, the effect of temperature on the 180° peel force over a wide temperature range from −20°C to 40°C can be predicted from the profile of the velocity spectrum of the rolling adhesive moment,

(2) The correlation coefficients to mechanical properties of the adhesives prove that the mechanical properties of the acrylic adhesives and those of the rubber adhesives each have different effects on adhesion.

KEY WORDS Test methods; mechanical properties; rolling ball test; time-temperature superposition; rubber PSA: acrylic PSA.

1 INTRODUCTION

Pressure-sensitive adhesive tapes (PSA tapes) are used in various applications such as electrical insulation, surface protection, nameplate adhesives, medical care, and household uses. Usually, they are used for the purpose of long term holding or securing in position in each field. Accordingly, the adhesion of the tape should be evaluated over the time schedule suitable for each application purpose and service condition. The actual measurement, however, is commonly made over a shorter time range than the actual servcie time due to the limitations of the testing apparatus or a measuring time limit. Moreover, in order to enhance the reproducibility of the test data, the tests are conducted at one point under the strictly limited, specific conditions typically regarding the shape of test specimens, temperature and pull rate.

However, if the adhesion was measured over the time expanded to the length of 3 orders to obtain data in a time spectrum, the properties of the tape could be safely predicted for a long time range corresponding to the actual application conditions through extrapolation from the actual measurement into the shorter or longer time area. Eventually, the total characteristics of a PSA tape may be perceived with the aid of one spectrum.[1]

The test method for adhesion, such as 180° peel force, also includes another problem in that measurement is carried out in the presence of the backing. In the case of double-coated tapes, a test specimen is lined with polyester film or similar for measurement.[2] Conventional studies made concerning the effect of the backing on adhesion,[3,4] prove that the adhesion is greatly affected by thickness and Young's modulus of both the adhesive and the backing.[5,6,7]

For such reasons, it is difficult to define the original characteristics of PSA tape from the measurement of the velocity spectrum of adhesion because the effect of the backing on adhesion depends on the peeling rate. Also, "stick-slip" occurs in a special rate range which seems to have a close relation not only to the viscoelastic properties of the adhesive but also to the Young's modulus and elongation of the backing.

Therefore, in order to analyze the original characteristics of the pressure-sensitive adhesive, it is necessary to evaluate the properties of the adhesive alone, and then to evaluate them using a PSA tape to include the effect of the backing.

For these purposes, the rolling friction or rolling adhesive moment of the pressure-sensitive adhesive or a PSA tape is determined over a wide range of rolling rates by rolling a ball or cylinder in contact with the adhesive surface.[8,9,10]

The rolling adhesive moment of a rubber or acrylic pressure-sensitive adhesive transfer tape over a wide range of rolling rates was measured by Miyagi's rolling ball method,[9] and the difference in the velocity spectra of the rolling adhesive moment for different adhesive compositions was investigated. This paper reports the significance of the velocity spectrum of the rolling adhesive moment and the peculiarity of each rate range achieved through the study of the correlations between the data at each rolling rate tested, and the 180° peel force at various measuring temperatures and the mechanical properties of pressure-sensitive adhesives.

2 EXPERIMENTAL

2.1 Rolling adhesive moment tester of the rotary drum type

Figure 1 is a sketch of the rolling adhesive moment tester. The ball for measuring the rolling adhesive moment is adjusted to be neutral (0) in weight with the aid of the helical spring fixed just past the

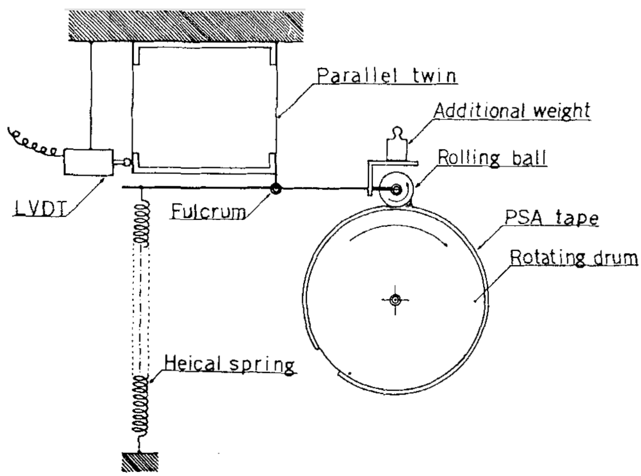

FIGURE 1 Scheme of the rolling adhesive moment tester.

fulcrum. The ball can then be loaded as required by adding weights. The PSA tape is applied to a drum with the surface to be measured facing out, and the drum is rotated at a constant rate. The ball loaded with an added weight is placed on the adhesive face. The ball is rotated on the adhesive by rotating the drum. At this time, the rolling adhesive moment of the ball generated by adhesion is converted into the displcement of parallel twins and detected by an LVDT (linear variable differential transformer).

This study of the rolling adhesive moment is conducted by rolling a steel ball as used in ball bearings, 24/32" in diameter, to which a weight of 28.2 gf (equivalent to its own weight) is added, at a temperature of $23 \pm 2°C$ and at a rate ranging form 50 to 50,000 mm/min.

2.2 180° peel force

The 180° peel force is measured in accordance with PSTC-1, Peel Adhesion for Single Coated Tapes, 180° Angles.

In addition to this standard measurement, measurements at temperatures of 0, 10, 20, 40, 60, 80 and 100°C were made for examination of the temperature dependence.

2.3 Mechanical properties of pressure-sensitive adhesives

The mechanical properties of pressure-sensitive adhesives were measured under the following conditions.

Tensile tester:	Instron-type tensile tester (CRE type)
Distance between the jaws:	10 mm
Pulling rate:	50 mm/min
Atmosphere:	$23 \pm 2°C$, $50 \pm 5\%$ Rh

2.4 Glass transition temperature of pressure-sensitive adhesives

Glass transition temperatures of pressure-sensitive adhesives were calculated by second-order regression of the peak curve of its loss modulus (G″) temperature sprectrum. Loss modulus of adhesives were measured at 1 Hz by using a Viscoelastic Spectrometer (VES-S Model, Iwamotoseisakusho).

2.5 Test specimens

As shown in Table I, using 6 kinds of rubber pressure-sensitive adhesives and 17 kinds of acrylic pressure-sensitive adhesives, 23 kinds in total, transfer-type adhesives of 50 μm in thickness were prepared as test specimens for measuring rolling adhesive moment. Test specimens for 180° peel force were then prepared by applying a backing of 25 μm-thick polyester film to those transfer type adhesives of 50 μm thickness.

TABLE I
Types of PSA studied

Specimen no.	Polymer	System	Tackfier
1	Natural rubber	Solution	Hydrocarbon resin
2	Natural rubber	Solution	Hydrocarbon resin
3	Elstomer	Solution	Hydrocarbon resin
4	Elastomer	Solution	Hydrocarbon resin
5	Elastomer	Solution	Rosin
6	Elastomer	Solution	Terpene resin
7	Acrylate	Solution	—
8	Acrylate	Water-borne	Terpene resin
9	Acrylate	Solution	Xylene resin
10	Acrylate	Solution	Xylene resin
11	Acrylate	Solution	Xylene resin
12	Acrylate	Solution	—
13	Acrylate	Solution	—
14	Acrylate	Solution	—
15	Acrylate	Water-borne	—
16	Acrylate	Solution	—
17	Acrylate	Solution	—
18	Acrylate	Solution	—
19	Acrylate	Solution	—
20[a]	Acrylate	Solution	—
21[a]	Acrylate	Water-borne	—
22[a]	Acrylate	Water-borne	Rosin
23	Acrylate	Solution	—

[a] Adhesive included glass chips.

3 RESULTS AND DISCUSSION

3.1 Results

Table II shows the results of the tests for rolling adhesive moment.

Table III shows the results of the 180° peel force tests of pressure-sensitive adhesive tapes and Table IV shows the mechanical properties and *Tg* of pressure-sensitive adhesives.

3.2 Analysis method

Figure 2 shows the results of the tests for rolling adhesive moment using one kind each of the acrylic adhesives and rubber adhesives. The abscissa indicates the rolling rate and the ordinate indicates the

TABLE II
Rolling adhesive moment [*gf* · mm]

Specimen no.	50	100	300	500	1000	2500	7000	10,000	30,000	50,000
1	240	221	281	267	279	275	309	309	300	316
2	318	342	394	463	462	540	543	509	448	388
3	184	197	170	156	211	232	249	239	203	191
4	433	440	586	715	740	796	701	609	354	273
5	185	140	104	87	83	22	1	1	0	0
6	88	90	85	85	70	58	1	0	0	0
7	238	226	205	191	190	178	236	202	210	205
8	619	727	828	835	890	663	490	442	312	264
9	317	359	364	359	346	285	271	211	74	47
10	144	167	215	262	269	300	297	291	232	184
11	422	431	440	433	452	412	234	186	83	58
12	196	221	238	264	300	269	207	271	177	173
13	204	206	218	236	239	188	80	52	30	28
14	47	50	56	72	84	79	43	40	0	0
15	431	392	342	286	266	148	70	47	0	0
16	181	152	152	114	57	38	20	15	11	22
17	343	457	314	276	114	57	36	30	14	22
18	391	362	276	276	210	105	38	32	14	14
19	314	295	295	286	229	76	50	38	19	10
20	105	162	171	171	171	191	162	143	114	95
21	162	181	229	219	200	171	171	171	114	95
22	562	676	705	733	829	857	562	448	210	76
23	238	238	191	181	152	86	67	48	19	13

TABLE III
180° Peel force at various temperature [gf/25 mm]

Specimen No.	Temperature [°C] 0	10	20	40	60	80	110°C
1	1920	1350	920	570	450	730 c	1000 c
2	2200	1570	1130	720	720 c	730 c	400 c
3	1770	1330	950	630	680 c	1490 c	780 c
4	3370	2720	1930	1300	830	370	10
5	1370	1280	1120	870	820 c	1150 c	480 c
6	2020	1420	1020 c	720 c	530 c	90 c	20
7	1520	1200	980	920	1220 c	1250 c	730 c
8	2250	2180	4480 c	2620 c	1550 c	870 c	670 c
9	2530	2000	1700	1520	1380	1230 c	1120 c
10	1600	1370	1130	980	920	700	580 c
11	3050	2500	2080	1680	3330 c	2730 c	1370 c
12	1650	1270	1020	970	900	980	1130 c
13	1600	1430	1180	1220	900	870	1050
14	1000	1080	750	570	500	770	1580 c
15	1650	1500	1330	1770	1950	830	720
16	430	1770	1470	1300	1130	950	900
17	550	1380	1380	1350	1130	970	1150
18	550	1420	1250	1280	1170	1070	1400
19	620	1820	1450	1400	1220	1070	1080
20	820	720	580	450	350	320 c	380 c
21	920	820	700	580	620	900	1000
22	3120	3070	2230	1580 c	1600 c	930	620
23	2050	1680	1280	1320	1180	1020	980

c: cohesive failure.

rolling adhesive moment calculted by multiplying rolling friction by the radius of the ball.

As shown in Figure 2, the rolling adhesive moment of the acrylic adhesive sharply decreases at rolling rates over 10,000 mm/min. Results in Figure 2 for the rubber adhesive sample indicate relatively less rate dependence of the rolling adhesive moment.

In this way, the rolling adhesive moment for all the specimens was determined at rolling rates ranging from 50 to 50,000 min/min.

Next, the method for analyzing these test results is discussed below. The analysis is made by regarding each set of data obtained through testing all of the adhesive specimens with a particular test method and conditon as one group, and by finding the coefficients of correlation between those groups of data; not by investigating the time and temperature dependence of the properties of each

TABLE IV
Mechanical properties and Tg

Specimen no.	Young's Modulus [Kg/cm]	Tensile Compliance [cm/Kg]	Tensile Strength [Kg/cm]	Elongation [%]	Tg [°C]
1	2.0	0.50	1.3	1160	−37.2
2	1.2	0.83	0.7	1500	−39.5
3	2.2	0.45	0.7	1540	−42.9
4	2.6	0.38	19.1	1620	−33.7
5	5.5	0.18	2.7	1800	−29.0
6	2.4	0.42	1.8	1510	−24.9
7	0.7	1.43	0.3	4580	−58.8
8	1.1	0.91	0.85	2150	−33.7
9	1.4	0.71	5.9	2290	−21.1
10	1.6	0.63	11.5	1500	−31.9
11	1.2	0.83	1	7300	−21.3
12	2.0	0.50	11.6	2030	−36.7
13	2.7	0.37	3.5	910	−37.0
14	4.4	0.23	7.6	1100	−12.7
15	3.2	0.31	12.7	930	−17.2
16	2.9	0.34	7.1	850	−27.1
17	4.0	0.25	6.2	1020	−22.8
18	3.4	0.29	4.5	1140	−22.1
19	3.5	0.29	5.1	920	−28.5
20	74.5	0.01	12.8	230	−47.9
21	48.9	0.02	6.1	500	−48.1
22	45.1	0.02	2.6	230	−20.1
23	2.7	0.37	5.9	960	−32.1

adhesive. For example, the scatter diagram for the rolling adhesive moment at a rolling rate of 50 mm/min and the 180° peel force at a temperature of 40°C is presented in Figure 3. The correlation coefficient is 0.83. Figure 4 presents the scatter diagram for the rolling adhesive moment at a rolling rate of 1,000 mm/min and the 180° peel force at 40°C. The correlation coefficient is 0.60. In Figure 5, at a rolling rate of 30,000 mm/min, the correlation coefficient is −0.02 and plotted data were scattered.

Thus, it is to be proved that the 180° peel force and the rolling adhesive moment correlations depend on the rolling rate by seeking high correlation coefficients, at various rolling rates.

Adopting this method, the significance of each velocity in the velocity spectrum of the rolling adhesive moment as shown in Figure 2 is analyzed through examination of the correlation between

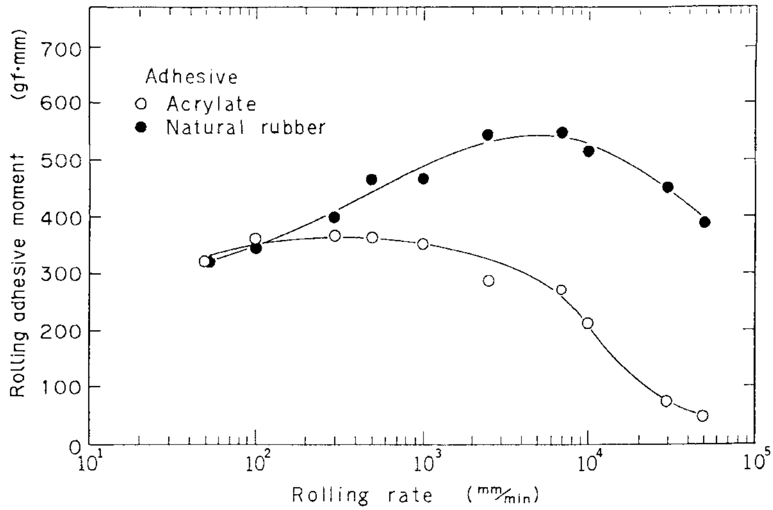

FIGURE 2 Rolling adhesive moment velocity spectrum.

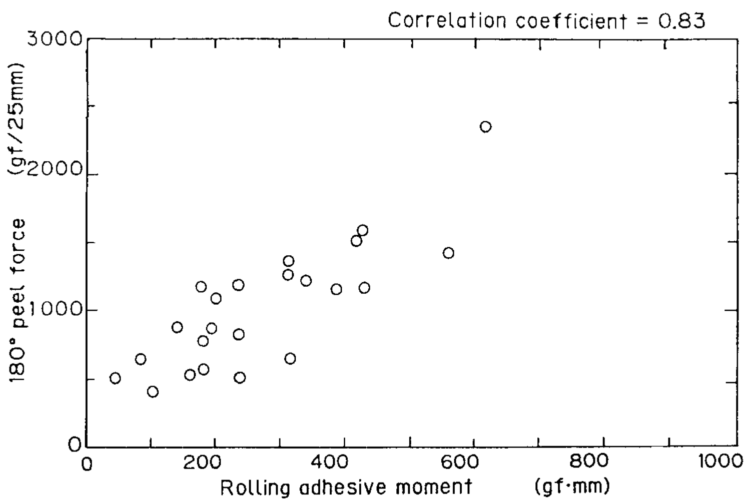

FIGURE 3 Scatter diagram for rolling adhesive moment at a rate of 50mm/min and 180° peel force at a temprature of 40°C.

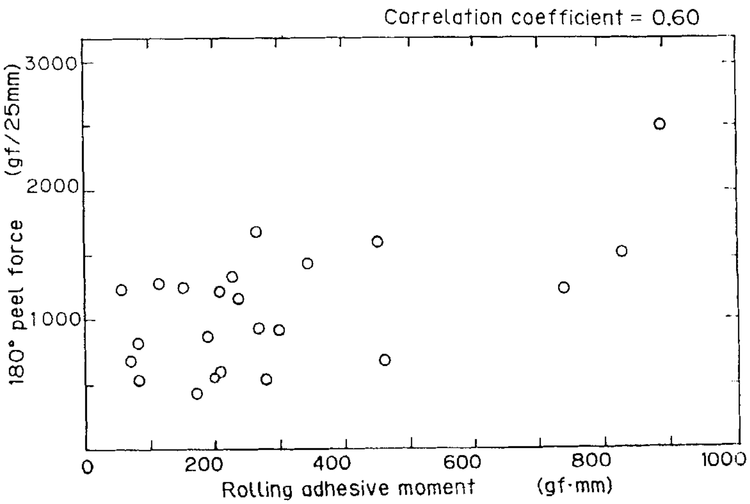

FIGURE 4 Scatter diagram for rolling adhesive moment at a rate of 1,000 mm/min and 180° peel force at a temperature of 40°C.

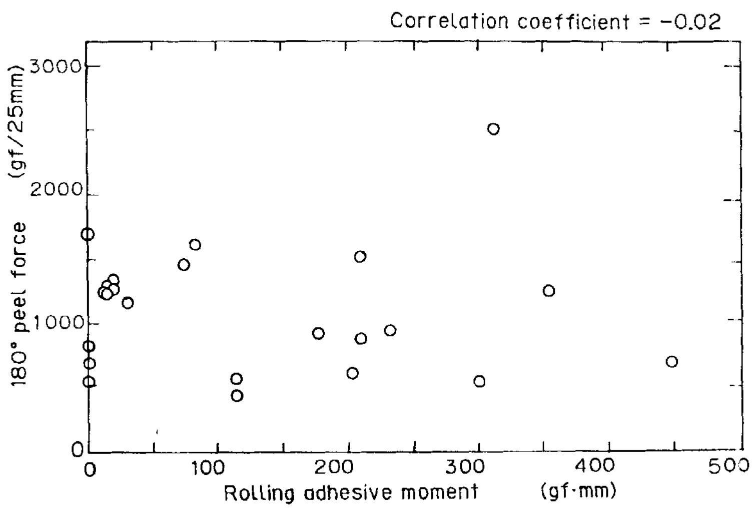

FIGURE 5 Scatter diagram for rolling adhesive moment at a rate of 30,000 mm/min and 180° peel force at a temperature of 40°C.

the group of rolling adhesive moment data at each rolling rate and
1) 180° peel force at various temperatures, and
2) mechanical properties of the adhesives.

3.3 Correlation to 180° peel force at various temperatures

The correlation coefficients between 180° peel force at set temperatures of 0, 10, 20, 40, 60, 80, and 100°C, and the rolling adhesive moment at each rolling rate were obtained and are shown in Figure 6 as the rate dependence of the coefficients. The coefficients are plotted at each temperature for the 180° peel test. The abscissa indicates the rolling rate and the ordinate shows the coefficient of correlation at each rolling rate.

As is apparent from Figure 6, the rolling adhesive moment at high rolling rates (over a short testing time) is closely correlated with the 180° peel force at low temperatures while the rolling adhesive moment at low rates (over a long testing time) is closely correlated with the 180° peel force at high temperatures.

Accordingly, it follows that the time-temperature superpositon

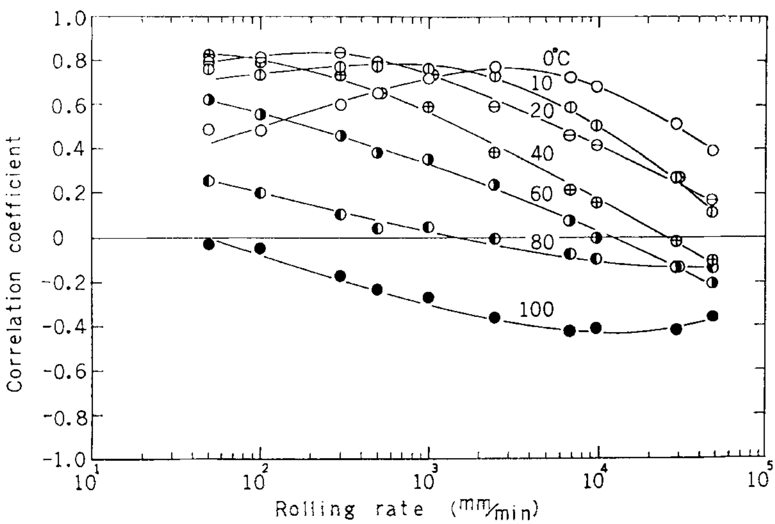

FIGURE 6 Effect of rolling rate on coefficient of correlation between rolling adhesive moment and 180° peel force at various temperatures.

principle usually applicable to the mechanical properties of amorphous high polymers such as rubber will also be applicable to the adhesion of pressure-sensitive adhesives.

Figure 7 presents the master curve of the correlation coefficient drawn by shifting the coefficient at each temperature shown in Figure 6 with reference to the 20°C curve and superpositioning them. The shift factor in the process is shown in Figure 8.

In Figure 8, the shift factor is presented by a broken line, which is obtained from the Williams, Landel, and Ferry time-temperature superposition principle, where the shift factor a_T is given by Eq. (1).[11]

$$\text{Log}\, a_T = \frac{-C_1(T - Tg)}{C_2 + T - Tg} \tag{1}$$

where C_1 and C_2 are constants. For simplicity, the glass transition temperature Tg of the adhesives is employed here as a reference temperature, so that C_1, C_2 take the values 17.4 and 51.6°C. And we employ as Tg -32°C; that is, the average glass transition temperature of all 23 kinds of pressure-sensitive adhesives used for the test which are shown in Table IV.

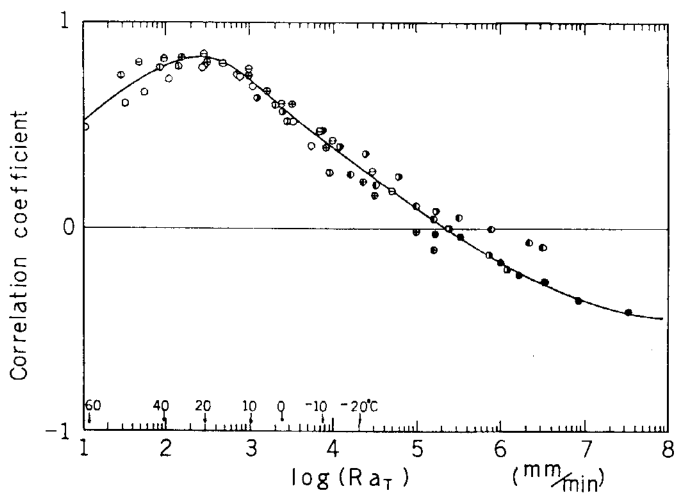

FIGURE 7 Master curve for correlation coefficient *versus* rolling rate Ra_T reduced to 20°C.

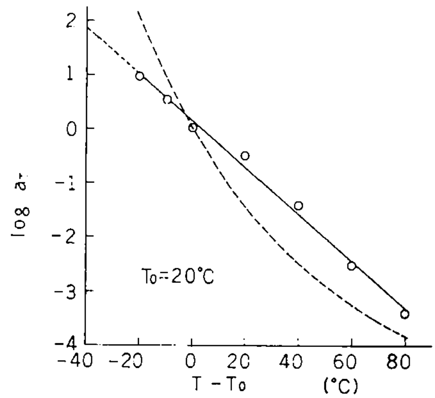

FIGURE 8 Temperature dependence of the shift factor a_T.

Assuming that the shift factor is of an Arrhenius type, we calculated that ΔH was 21 Kcal/mol.

The adhesion is a composite of interfacial chemical behavior where the pressure-sensitive adhesive face wet the adherend, and viscoelastic behavior where the peel force is distributed by deformation of the pressure-sensitive adhesive bulk. Here, as for the viscoelastic properties, the time-temperature superposition principle is applicable to those properties because the adhesive is an amorphous high polymer. As shown in Figure 7 and mentioned before, the principle is applicable to 180° peel force in terms of the rolling adhesive moment; consequently, it follows that the viscoelastic properties of the adhesive make a large contribution to adhesion.

On the basis of this shift factor, the rolling rate equivalent to each set temperature for the 180° peel force at a pulling rate of 300 mm/min was obtained and is shown in Figure 7. Thus, it is found that the rolling rate of 50,000 mm/min corresponds to −20°C or below in 180° peeling.

According to the results given above, it can be said that the pressure-sensitive adhesive properties (180° peel force) over a wide temperature range from −20°C to 40°C are available from the velocity spectrum of the rolling adhesive moment at normal temperature over rolling rates ranging from 50 to 50,000 mm/min.

3.3 Correlation with mechanical properties

Specimens with sample No. 20, 21 and 22 were omitted from this analysis, because they contained glass chips in their adhesives.

Figure 9 represents the correlation between the rolling rate and the Young's modulus, tensile compliance, tensile strength, and elongation of the pressure-sensitive adhesive.

In Figure 9, the rolling adhesive moment shows a positive correlation to the tensile compliance which becomes higher as the rolling rate increases, and a low positive correlation to the elongation and tensile strength. On the other hand, the rolling adhesive moment shows a negative high correlation to Young's modulus as the rolling rate increases. As a result, it follows that the modulus of the adhesive makes some contribution to the rolling adhesive moment and adhesion at high velocity (low temperatures).

The correlation, however, is low from a general view point. Therefore, the test data of the acrylic adhesives and the rubber adhesives are separately analyzed. The correlations for the acrylic adhesives are illustrated in Figure 10.

The elongation and tensile strength, as per the test results of all the adhesives, show low positive correlation to the rolling adhesive

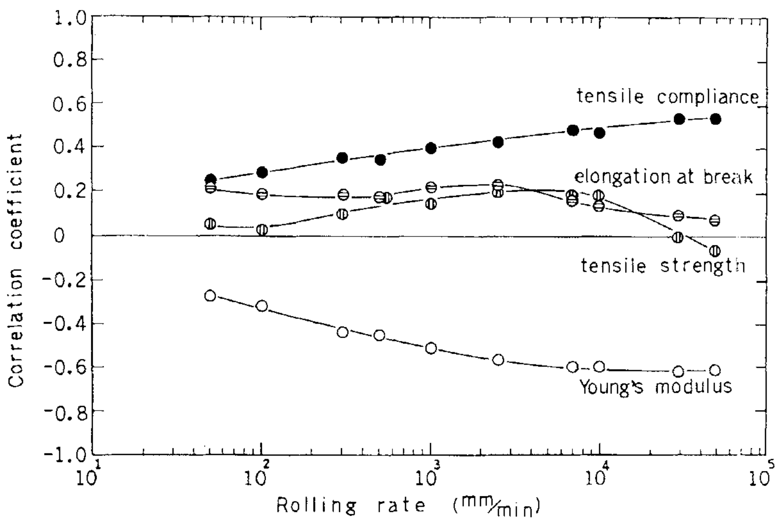

FIGURE 9 Effect of rolling rate on coefficient of correlation between rolling adhesive moment and mechanical properties of all adhesives.

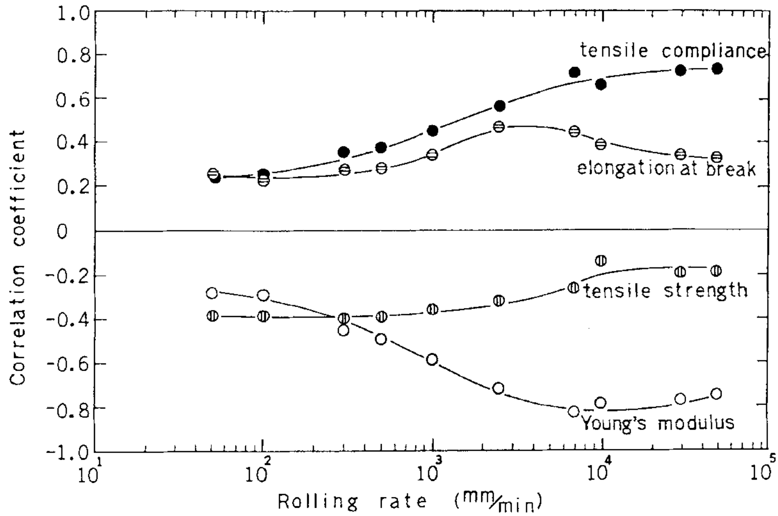

FIGURE 10 Effect of rolling rate on coefficient of correlation between rolling adhesive moment and mechanical properties of adhesives.

moment but, in the case of the acrylic adhesive only, the tensile strength shows negative correlation, and Young's modulus shows a quite high negative correlation of -0.8 at high rolling rates. The softness and good elongation of the adhesive displays positive correlation to the rolling adhesive moment; consequently, from the fact that the modulus and tensile strength show negative correlation, this conformability makes a great contribution to the rolling adhesive moment at high velocity (low temperatures).

Simply for reference, the correlation coefficient for the rubber adhesive is shown in Figure 11, in spite of an insufficient number of specimens and lack of reliability.

As for the peel force of a pressure-sensitive adhesive, many theoretical and experimental formulas are submitted on the basis of the analysis of its mechanical behavior. Most of them are presented as follows.

$$W_0 \propto \sigma \cdot (1/Ea)^{1/4} \tag{2}$$

where W_0 is the peeling force, σ is the tensile strength of the50 adhesive, and Ea is the modulus of the adhesive. That is, that the

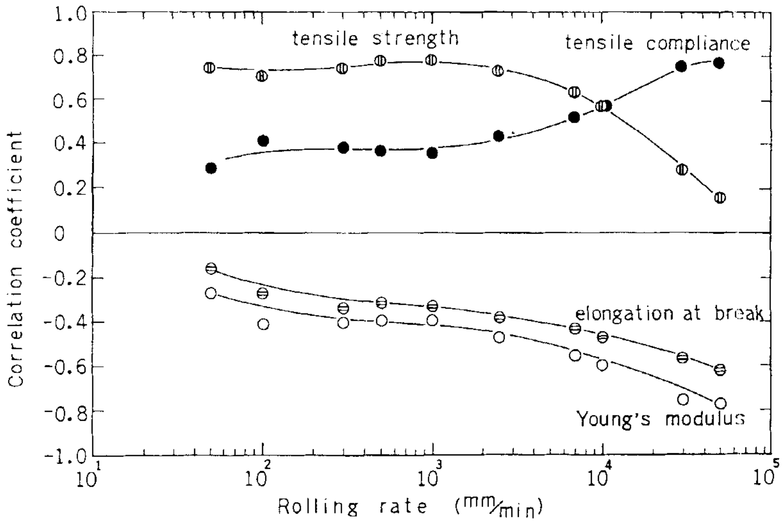

FIGURE 11 Effect of rolling rate on coefficient of correlation between rolling adhesive moment and mechanical properties of acrylic adhesives.

peel force is proportional to the tensile strength of the adhesive, while it is inversely proportional to 1/4 power of the Young's modulus.

From Figure 11, for the rubber adhesives, there is a relatively high positive correlation between the rolling adhesive moment and the tensile strength, and a low negative correlation between the rolling adhesive moment and Young's modulus, which agrees with the formula (2).

On the other hand, in the data for acrylic adhesives in Figure 8, the rolling adhesive moment shows negative correlation to the tensile strength, which does not agree with the formula (2).

As can be seen from our discussion, the rubber adhesives and the acrylic adhesives have different adhesion effects from the mechanical point of view.

4 CONCLUSION

Using 23 different kinds of transfer type pressure-sensitive adhesive tapes, the rolling adhesive moment is determined by the rolling ball method over rolling rates ranging from 50 to 50,000 mm/min. By studying the correlation between the rolling adhesive moments obtained and the results of property evaluation, *e.g.* 180° peel force at various temperatures or the mechanical properties of the adhesive, the significance of the velocity spectrum of the rolling adhesive moment is analyzed.

As a result, the following have been made clear:

1. The time-temperature superposition principle is applicable to the correlation coefficient between the rolling adhesive moment and the 180° peel force. In other words, it follows that the rolling rate axis indicates the measuring temperature for 180° peel force. Furthermore, the effect of temperature on the 180° peel force over a wide temperature range from -20°C to 40°C can be predicted from the profile of the velocity spectrum of the rolling adhesive moment.

2. The correlation coefficients between the rolling adhesive moment and the mechanical properties of the adhesive prove that the mechanical properties of the acrylic adhesive and those of the rubber adhesive each have different effects on adhesion.

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