ELECTRIFICATION DUE TO PEELING OF PRESSURE-SENSITIVE ADHESIVE PAPER
I. ANOMALOUS ELECTRIFICATION

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ABSTRACT

Electrification of pressure-sensitive adhesive paper due to peeling was studied by measuring the electric charge and the surface potential with use of toner for electrophotography and a surface potentialmeter, respectively. It was found that the electric charge on the surface of the release-coating after completion of peeling was negative in a sign S near the starting portion from which the release-coating was peeled, while it was positive in a sign E near the end portion. The surface potentials of both the peeled release-coating and the unpeeled face-stock in region E increased with increasing lable-length. The surface potential of the peeled release-coating had a maximum at a peeling rate of about 0.6 cm/sec while that of the unpeeled face-stock increased monotonically as the adhesive paper was peeled. These findings suggest that the anomalous electrification should be due to electric polarization induced by peeling.

1. INTRODUCTION

Several studies on peeling of an adhesive from an adherend have been carried out to understand the peeling strength of the adhesive on the basis of its rheological and electrical properties. In one of the previous studies, an attempt was made to interpret the peeling strength in terms of the geometrical and rheological properties of a pair of adhesive and adherend under viscoelastic deformation. Deryagin investigated the electrification phenomenon during peeling, and concluded that the peeling strength could be related to electrostatic forces between charges induced on the stripped interfaces. Hata et al. also undertook a similar study, but showed no direct correlation between the amount of induced charges and the peeling strength. In these two studies, they used a Farady-cage, by which only the net charges on the whole test specimen could be determined. They implicitly assumed that the charges induced on each side of the peeled adhesive-adherend interface were uniform and the same in sign throughout, while the outer surface opposite to the interface would carry no charges. This assumption, however, has not yet been substantiated. Thus, as far as we know, no plausible explanation has as yet been made for this phenomenon on peeling.

This paper describes the distribution of induced charges on stripped and opposite surfaces of a pair of adhesive and adherend and the interpretation of the peeling strength in terms of the distribution. Since few studies on peeling have dealt with pressure-sensitive adhesive paper, we selected it as a sample.

2. EXPERIMENTAL

Figure 1 shows schematically the experimental equipment used for surface potential measurements on pressure-sensitive adhesive. The test samples which are of 1.0 ~ 7.0 cm in length (L) and 7.5 cm in width consist of face-stock, a pressure-sensitive adhesive, a silicone release-coating on a polyethylene film of 15 μm in thickness, and a protective backing. The thickness and intrinsic surface resistivity of constituents of pressure-sensitive adhesive paper are shown in Table 1. The
Fig. 1. Schematic diagram of the equipment used for peeling pressure-sensitive adhesive paper and measuring the surface potential. Face-stocks are peeled from the release-coating by pulling the long release paper at a constant speed, and are allowed to drop at a corner of the metal-supporter: L, label-length; (S), starting point of peeling; (E), end point of peeling; $V$, surface potential on the peeled release-coating; $V_f$, surface potential on the face-stock; $V_a$, surface potential on the adhesive.

Table 1. Thickness and intrinsic electrical surface resistivity of each face-stock, the adhesive, the silicone coating, and the backing.

<table>
<thead>
<tr>
<th>Material</th>
<th>Thickness/μm</th>
<th>Intrinsic Electrical Surface Resistivity/Ω</th>
</tr>
</thead>
<tbody>
<tr>
<td>Face-Stock</td>
<td>100</td>
<td>$1.0 \times 10^{10}$</td>
</tr>
<tr>
<td>Adhesive</td>
<td>30</td>
<td>$1.0 \times 10^{12}$</td>
</tr>
<tr>
<td>Silicone Coating</td>
<td>1</td>
<td>$6.0 \times 10^{12}$</td>
</tr>
<tr>
<td>Backing</td>
<td>100</td>
<td>$2.0 \times 10^{10}$</td>
</tr>
</tbody>
</table>

The peel force was about 14 grams when the face-stock with 5.0 cm in width was peeled from the release paper (i.e., the protective backing coated with silicone) at an angle of 180° and a rate of 0.35 cm/sec.

The face-stock with a fixed size was peeled from the metal-contact release paper by bending the release paper at a corner of the metal plate with 0.4 cm in thickness, pulling it at a constant speed. The release paper was supplied continuously from a core of the pressure-sensitive adhesive paper placed at the other position. The detached face-stocks were allowed to drop in succession at the corner of metal plate.

A surface potentialmeter (SSV II-40, Kawaguchi Electric Co., Tokyo) was used for surface potential measurements. The peeling rate of a face-stock was 0.33 cm/sec in most cases. The probe of the potentialmeter was held at a distance of 0.3 cm from the surface (see Fig. 1). Potentials $V_f$ on the surface of the peeled release-coating were measured in the order from starting portion (S) to the end portion (E), and potentials $V_a$ on the adhesive and $V_f$ on the face-stock were also determined. For potential $V_p$ on the protective backing, the measurement was not feasible because it is difficult to set the potentialmeter over the protective backing and the metal plate as an opposite electrode on the peeled adhesive under peeling process. Hence, the set-up was so modified that the release paper could be pulled up at an angle of 30° from the metal supporter and the probe was held behind the backing (see Fig. 4).

Intrinsic electrical surface resistivity was measured by use of a room-temperature resistivity chamber (Model VE-30, Kawaguchi Electric Co., Tokyo). Toner for electrophotography was used to examine the distribution of electric charges on the peeled release paper. The half period was determined to be about 48 hours for the silicone release-coating and less than 1 second for the protective backing, from the rate at which the surface potential decays after treatment of corona discharge. The potentials observed at a few seconds later were taken as those measured immediately after peeling, because the half period of the potential decay on the release-coating was about 48 hours. The intrinsic surface resistivities of the adhesive and the release-coating were larger by about two decades than those of the face-stock.
and the protective backing. This fact suggests that virtually no transport of charges between the face-stock and adhesive and also between the protective backing and release-coating occurs. All measurements were made at 20°C and 65% RH.

3. RESULTS

3.1 Distribution of Electric Charge

Figure 2 shows typical results for the distribution of positively charged toner on the surfaces of a release-coating (a) and a protective backing (b) which were peeled immediately before the measurement. The blackened portions due to positively charged toner show the existence of negative charges on the paper. Here, the toner is stuck on the starting portion (1) of the release-coating and on the end portion (4) of the protective backing. From only such an experiment, we can not see whether on the end portion of the release-coating (2) and the starting portion of the protective backing (3) charges are positive or neutral. Thus, in order to see whether positive charges exist, negatively charged toner was also used. We ascertained that the end portion (2) of the release-coating and the starting portion (3) of the protective backing were blackened by the negatively charged toner. From these results we conclude that the portions (2) and (3) are positively charged. These patterns of charge distribution on the peeled release paper almost did not depend on the angle of peeling.

3.2 Surface Potential of Peeled Release-Coating

Figure 3 shows a surface potential pattern of the release-coating measured immediately after the complete peeling of the release paper from left to right. Interestingly, the surface potential is about -50 volts at the starting portion, but is anomalously large and positive (about 150 volts) at the end portion. The potentials at the starting portion and the end portion are expressed by \( V_i \) and \( V_m \), respectively.

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Fig. 2. Distribution of the electric charges on the release paper treated with a liquid developer containing toner. Three labels are peeled from left to right. The blackened parts stuck by toner show the existence of the negative charge. \( L \), label-length of 3.5 cm; (a), surface of the peeled release-coating; (b), surface of the protective backing; 1 and 3, starting portion; 2 and 4, end portion; peeling rate, 0.33 cm/sec.

Fig. 3. Surface potential \( V_r \) on the peeled release-coating; \( V_i \), surface potential at the starting portion; \( V_m \), surface potential at the end portion; (S), starting point of peeling; (E), end point of peeling; peeling rate, 0.33 cm/sec.
The surface potential on the protective backing measured at 20 seconds after peeling is shown in Fig. 4. The pattern is contrasted with that of the release-coating, being positive at the starting portion and negative at the end portion. These potential patterns correspond to those developed by toner.

Figure 5 shows the patterns of the surface potential on the release-coating after completion of peeling at different label-length L. The potential patterns change with L. In Fig. 6, values of the surface potential \( V \) on the release-coating shown in Fig. 5 are plotted against L. The maximum potential \( V_m \) at the end portion of the release-coating increases almost linearly with increasing label-length, while the maximum potential \( V_i \) at the starting portion remains nearly constant. Since the surface potential is related to the surface charge density on the body, the result implies that the charge density at the starting portion of the peeled surface is about the same in this region.

The effective charges \( Q \) which are defined as integrals of surface potential over each length at the starting and the end portions of the unpeeled release-coating are also plotted in Fig. 7 against label-length. The effective charges of the starting and end portions change linearly with L. Of course, the sign of each effective charge is opposite and the difference in the absolute values of the effective charges is very small. That is, the sum of positive and negative charges on the peeled release-coating has a tendency to approach zero.

The patterns of surface potential on the release-coating after completion of peeling at different peeling rates \( R \) are shown in Fig. 8. The patterns change with \( R \). Figure 9 shows the peeling rate dependence of the surface potentials.
Fig. 7. Effective charges $Q$ at the starting and end portions of the release-coating are plotted against label-length $L$: peeling rate, 0.33 cm/sec.

Fig. 8. Patterns of the surface potential on the release-coating after completion of peeling at different peeling rates: label-length, 7.0 cm.

$V_m$ and $V_i$ at the peeled release-coating. $V_m$ has a maximum of about 150 volts at the peeling rate of about 0.6 cm/sec, while $V_i$ at the starting portion is almost constant and about -50 volts. The appearance of the maximum in $V_m$ may be ascribed to the two opposite effects, i.e., the induction of electric polarization at the end portion by charges existed on the peeled surface and the decay of the electric charges accumulated at the end portion. For example, at a peeling rate of 0.33 cm/sec it takes about 21 seconds to peel the whole adhesive paper with 7.0 cm in length. At such a slow rate it is likely that the electric charges induced at the peeling front are transferred toward the unpeeled end portion and decayed before completion of peeling. In this case, the effect of decay should be remarkable. At a high rate of peeling, on the other hand, the induced charges may not be transferred to the end portion, although the charges once accumulated may not appreciably have decayed, before the complete peeling. In this case, the rate of electric induction and transfer due to peeling is predominant. These considerations put a qualitative interpretation on the observed fact that $V_m$ at both low and high rates of peeling is small, while it is the largest at an intermediate rate of peeling.

3.3 Surface Potential of Face-Stock

On peeling, the surface potential $V_f$ at the unpeeled end portion of face-stock increases with increasing label-length $L$ and eventually levels off, as can be seen in Fig. 10. Here, the probe of potentialmeter on the face-stock was set at the inner position of 6 mm from the peeling front. The effective charge $Q$ at the unpeeled end portion of the face-stock also increased linearly with
increasing label-length $L$. The dependences of $V_f$ and $Q$ on $L$ are similar to those observed in the end portion of the release-coating. These suggest close correlations between the potentials and also between the effective charges for the release-coating and the face-stock. The charges on the peeled surface induced by peeling may in turn induce polarization on the opposite surface, i.e., on the face-stock and the protective backing. As the result, the sign of the induced charge at the end portion of the surfaces should become opposite to that at the starting portion. At the same time, the sign of the charge at the end portion of the unpeeled face-stock may also be opposite to that at the end portion of the unpeeled protective backing. The accumulated charges at the end portions of the face-stock and the protective backing induce polarization at the interface between the adhesive and the release-coating, in other words, the charges at the end portion of the face-stock may raise the potential at the end portion of the release-coating.

In Fig. 11, $V_f$ on a face-stock under peeling is plotted against peeled length $L_f$. Here, the probe of potentiometer was set at the end point of the label which moves with a progress of peeling. The potential increases sharply with increasing $L_f$, indicating that the induced electric charges are accumulated more and more at the end of the unpeeled face-stock as the peeling proceeds. Therefore, the electric charge density of the unpeeled region must also increase sharply with increasing peeled length.

3.4 Surface Potential on Adhesive

Since a pressure-sensitive adhesive is sticky, it was difficult to measure surface potentials $V_a$ by continuous scanning of the adhesive surface. Therefore, the adhesive with a face-stock was taken out after peeling and the surface potential on a given point of the adhesive was measured. We found that the potential was positive at the starting portion of the adhesive and negative at the end portion of the adhesive, as opposed to that of the peeled release-coating.

4. DISCUSSION

Electrification due to peeling induces electric charges which are negative at the starting portion and positive at the end portion of the release-coating. This fact is in contradiction to the old concept that the charges induced by peeling are uniform in sign anywhere on the surface. The surface potential $V_i$ at the starting portion of the peeled release-coating is influenced by neither the label-length nor the peeling rate. The amount and sign of the charges induced on the interface between the adhesive and the release-coating depends only on the electrical properties of them.
The surface potential $V_m$ at the end portion of the release-coating and the potential $V_f$ on the face-stock increase with increasing label-length, suggesting the existence of a correlation between these potentials. The dependence of $V_m$ upon peeling rate and that of $V_f$ upon peeled length suggest that electric charges are induced on the face-stock and the protective backing. At a high rate of peeling, the electric charges in the peeled face-stock and protective backing may not be transferred to the unpeeled end portion. On the other hand, at a slow rate of peeling the electric charges induced at the peeling front may be transferred toward the unpeeled end portion and be decayed gradually to the time when the peeling is completed. From such effects for transfer and decay of electric charges a maximum in the surface potential $V_m$ may be observed.

The dependence of potential $V_f$ upon the peeled length $L_f$ suggests that the induced electric charges are accumulated more and more at the end portion of the unpeeled face-stock and protective backing. This suggestion is also supported by the dependences of $V_f$ and $Q$ upon label-length. The levelling off of $V_f$ in Fig. 10 may be ascribed to the fact that the probe of potentialmeter is not set at the end point of label and also to the idea that the extra electric charges beyond the finite electric capacity of the face-stock should disperse in the layer of the face-stock.

Since the face-stock is electrically neutral as the whole, the electric charges in the adhesive induce charges at the peeled portion of the face-stock which then induce electric charges with the opposite sign in the unpeeled portion of the face-stock. This means that a large electric polarization occurs on the face-stock. Similarly, charges on the release-coating should induce electric polarization on the protective backing. The induced polarization in the unpeeled region of the face-stock and the protective backing affects charge in the un-peeled interface between the adhesive and release-coating, resulting in the charge distribution on the release-coating. The surface potential $V_m$ at the end portion of the release-coating as well as the potential $V_f$ on the face-stock may be large.

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粘着紙の剝離带電

I. 異常带電

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粘着紙の剝離帯電現象の詳細を明らかにすることを目的として、粘着紙の各種部位において剝離に起因して生ずる表面電荷および表面帯電分布をそれぞれ電子写真機用トナーおよび表面帯電計を用いて調べた。その結果、剝離紙シリコーン面の前半部は正帯電し、末端部は正で、かつ異常に高い電位が観測されるという特異現象を見出した。また、剝離紙シリコーン面の末端電位およびラベル未剝離部の末端電位は、ラベル長が大きくなるにつれて上昇すること、剝離紙シリコーン面の末端最大電位は、約0.6 cm/secの剝離速度で極大値を持つこと及び剝離が進行するにつれてラベル未剝離部の末端電位は著しく上昇すること等を見出した。これらの結果から、粘着紙の剝離時における特異現象は、剝離に起因して生じた帯電荷が、ラベルおよび剝離紙に電気分極を誘起した結果として説明される。