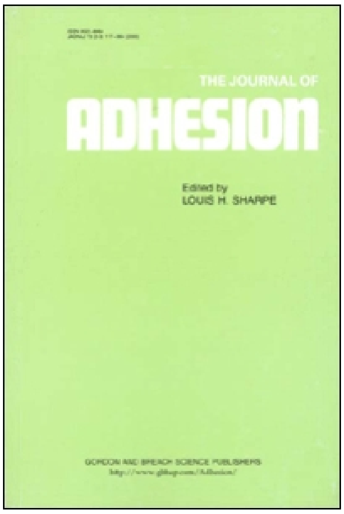

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Hideaki Suzuki[a]; Kosuke Hamada[a]; Yukiko Suzuki[a]
[a] Research and Development Division, New Oji Paper Co Ltd, Hyogo, Japan

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Physical Considerations in the Development of Pressure-Sensitive Adhesive Sheets*

HIDEAKI SUZUKI, KOSUKE HAMADA and YUKIKO SUZUKI**

New Oji Paper Co., Ltd., Research and Development Division, Amagasaki Center
4-3-1 Jokoji, Amagasaki-shi, Hyogo 660, Japan

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A pressure-sensitive adhesive sheet is a special kind of paper used in non-impact printers which use a heating process to apply toner to paper. As a result, it needs special characteristics that general pressure-sensitive adhesive paper for labels do not require.

One of these characteristics is that the edge of the folded paper used in non-impact printers must not incline after printing. This was done by making the degree of orientation of the fibers in the face stocks and the release liners low.

The other characteristics are that adhesive must not ooze out from the edges during the slitting or guillotining process and that the labels must not come off of the release liner by themselves during the printing process. Ooze characteristics were found to be related to the adhesive coat weight. An adhesive paper with both a high peel strength and lower adhesive coat weight was developed by studying the dynamic viscoelastic properties of adhesives and release layers. The storage modulus of the release layer concerned with the release force was also found to be related to the self-peeling tendency of the labels.

These points were considered during the development of pressure-sensitive adhesive paper used in non-impact printers which use a heating process to apply toner to paper.

KEY WORDS: Paper for non-impact printers; oozing property of adhesive; dynamic viscoelastic properties of adhesives and release layers; self-peeling tendency; fiber orientation of paper.

INTRODUCTION

A pressure-sensitive adhesive sheet is generally constructed to hold a pressure-sensitive adhesive between a face stock and a release liner. Consequently, a pressure-sensitive adhesive sheet has multiple layers, and the components making up the layers are in turn made from multiple components. Therefore, if the various layers and components are given different functions, useful pressure-sensitive adhesive sheets can be created.

Pressure-sensitive adhesive paper used in non-impact printers which use a heating process to apply toner to paper was developed. This kind of pressure-sensitive adhesive paper needs the following three characteristics in particular.

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**Corresponding author.

First, the edge of folded paper used in non-impact printers must not incline after printing. If the edge of folded paper inclines sharply after printing, (Fig. 1), the folded adhesive paper pulls toward the inclining direction and causes the printer to jam. Paper has stress which is caused by many conditions during the papermaking process; the fiber orientation of the paper is also decided during the procedure.

The stress and fiber orientation of paper does not change during papermaking. Furthermore, their appearance does not change unless the paper is treated by heating or moisturizing. If the paper is treated by heating or moisturizing, the stress releases from the paper and the paper changes its shape or size based on the fiber orientation of the paper. As a result, the folded paper tends to incline.

The fiber orientation of different kinds of paper was studied using microwave by attenuation based on the interaction between polarized microwaves and dipole moments of fibers in a paper sheet.[1] To make the edge of folded paper vertical, it is necessary that the orientation angle of fibers (θ) be low and that the degree of orientation of fibers be parallel to the machine direction (Fig. 2)[2], or that the degree

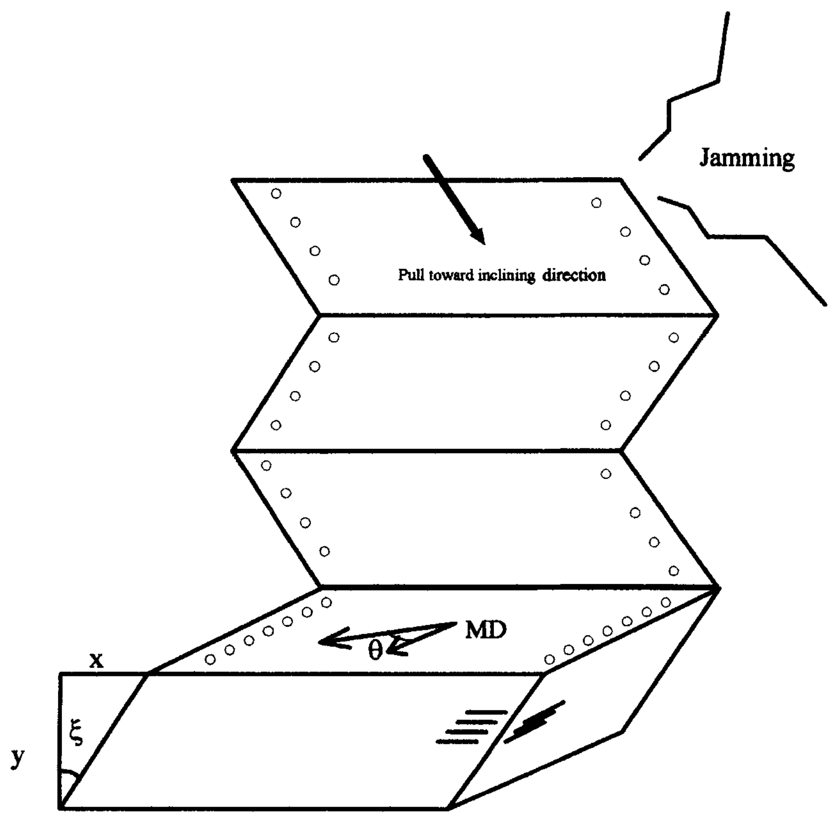


FIGURE 1 Inclination of folded papeer after printing, $\tan\xi = x/y$ (degree of inclination of the folded paper), y = thickness, x = displacement, θ; orientation angle of fibers measured from the machine direction (MD).

of orientation of fibers in a sheet be low (Fig. 3). This inclining-edge problem was solved by making the degree of orientation of fibers low in the face stock and the release liner.[3]

Second, adhesive must not ooze out from the edge during the slitting or guillotining process. Slitting pressure-sensitive adhesive paper is similar to cutting it with a pair of scissors. Scissor blades exert pressure on pressure-sensitive adhesive paper

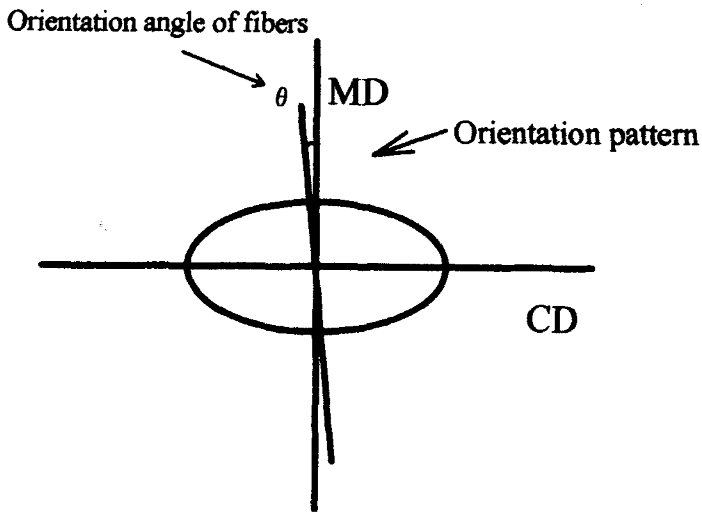


FIGURE 2 The degree of orientation of fibers parallel to MD.

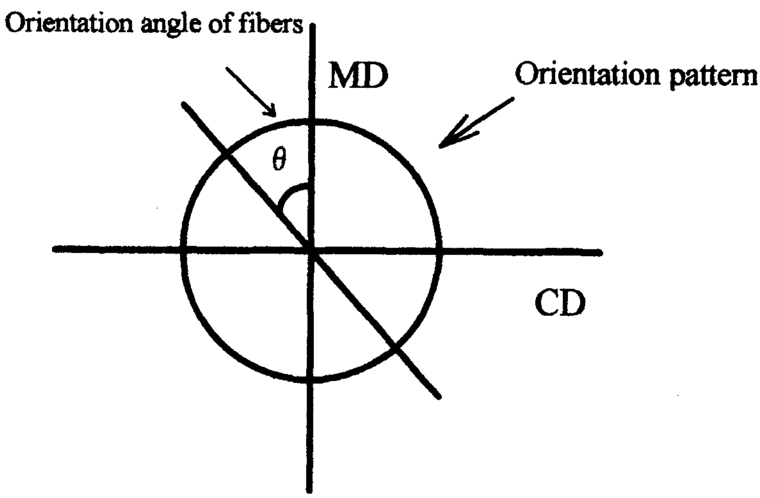


FIGURE 3 The low degree of orientation of fibers.

during cutting. In the same way, a slitting blade exerts pressure on pressure-sensitive adhesive paper during slitting. A guillotine blade also exerts pressure on a pile of pressure-sensitive adhesive papers during guillotining. In the case of guillotining, pressure is exerted on the pile of pressure-sensitive adhesive paper by not only the guillotine blade, but also by the clamp. Once the adhesive oozes out from the edges, it remains on the edge of the adhesive paper and sticks to parts of the printer, preventing the adhesive paper from being used for printing. It was found that the oozing-adhesive level became lower as the adhesive-coat weight decreased. However, a lower adhesive-coat weight leads to poorer peel strength. Therefore, the dynamic viscoelastic properties of adhesives were studied in order to develop an adhesive paper with both high peel strength and low adhesive-coat weight.

The third characteristic is that the labels must not come off of the release liner by themselves during the printing process. If the labels come off of during printing, they stick to parts of the printer and stop the printing process. It is believed that two factors cause the labels to come off of the release liner by themselves during printing. One is that after one label comes off of the liner during printing, it causes the others to follow. The other is that the labels have already started to come off of the liner before printing has even started. In either case, this problem can be solved by having a high release force, which is related to the crosslinking level of the release layer[4]. It was found that this premature releasing of the labels was related to the dynamic viscoelastic property of the adhesive as well as that of the release layer.

EXPERIMENTAL PROCEDURE

Printing Property

The printing property of whether or not a non-impact printer would jam during printing was evaluated using a MELCOM 8270, which is made by Mitsubishi Electric Corp.

Fiber Orientation of Paper

The fiber orientation of the paper was measured using a MOA-2001, made by New Oji Paper Co.Ltd. The principle for determining the fiber orientation of paper is based on the interaction between the dipole moments of molecules constituting a sheet of paper. This interaction depends on the direction in which the dipole moments differ for anisotropic sheet material. A 100 × 100 mm test sheet in a sample holder is placed between a pair of rectangular waveguides constituting a cavity resonator system. A polarized microwave is used to irradiate vertically a sheet that is rotated around the central axis normal to the sheet plane. The transmitted mcirowave intensities are detected by a diode detector and are then digitized for each rotating angle. A batch of 360 data points includes the measuring frequency, the maximum and minimum values of transmitted microwave intensities, the ratio of these two values, the orientation angle, the ratio of the transmitted microwave intensity in the cross direction to that in the machine direction, and the orientation patterns.

Oozing-property Evaluation

120 pressure-sensitive adhesive sheets were stacked in a pile, and the end of the pile was cut with a hand guillotine (Fig. 4). The amount of adhesive oozing out from the edge of the pile being cut was then evaluated. The oozing-property was evaluated by touching the edge while looking through a 45 power microscope. Pictures were also taken of the blade of the hand guillotine that was used to cut the pile of pressure-sensitive adhesive sheets.

Dynamic Viscoelastic Properties

Adhesive The dynamic viscoelastic properties were measured using M-500 parallel plates made by Rheology Co. Ltd. Kyoto, Japan. Measurements from −60 °C to 100 °C were taken at 5 °C intervals. The data for the master curves were obtained according to the time-temperature superposition principle using a reference temperature of 20 °C. The measured adhesive's thickness was about 1.5 mm.

Release agent Solvent-type silicone release agents were used and were turned into silicone layers with a thickness of about 2.0 mm. The dynamic viscoelastic properties were measured using a DVE-V4 pull-type tool made by Rheology Co. Ltd. The master curves of the release layers were obtained in the same way as those for the adhesive.

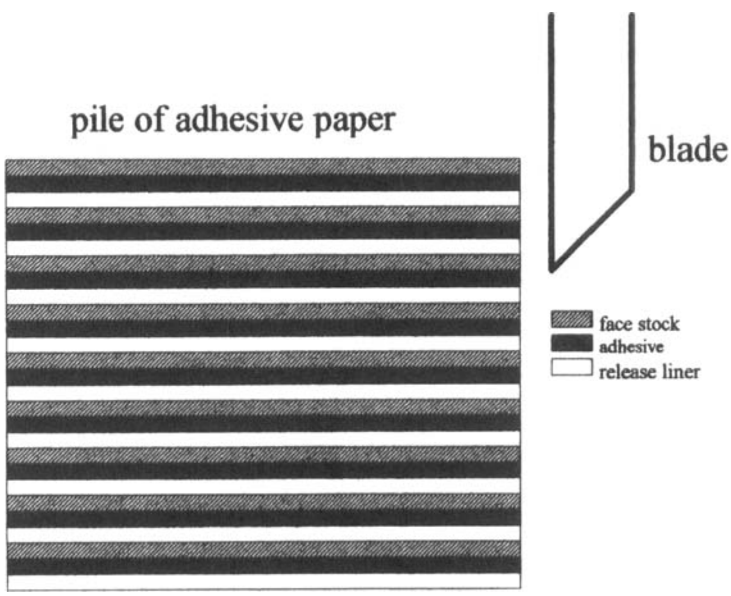


FIGURE 4 Schema of cutting with a hand guillotine.

Label Self Peel-off Evaluation

Labels were made from a pressure-sensitive adhesive paper by using a cutting-die and removing the waste. They were then attached to a release liner an rolled onto a pipe whose diameter was similar to that of a roll in the non-impact printer (Fig. 5). Several hours later the edges were checked for lifting.

RESULTS AND DISCUSSION

Fiber Orientation of Paper

Figures 6 and 7 show the orientation pattern, the measuring frequency, and the maximum, minimum, cross direction and machine direction values of the transmitted microwave intensities. Also shown are the orientation angle and the ratio of the transmitted microwave intensity in the cross direction to that in the machine direction for the face stock, release liner, and the pressure-sensitive adhesive paper with the same face stock and release liner. The MOR value is a ratio of the maximum to the minimum values of the transmitted microwave intensities. If the MOR value is 1, the orientation pattern is a circle; this means that the fibers in the paper have no orientation. Figure 6 shows the data from the adhesive folded paper that inclined after printing. The orientation angles of the face stock, the release liner and the pressure-sensitive adhesive paper were 16°, 17°, and 15°, respectively. These orientation patterns were distortional. The degree of inclination ($\tan\xi = x/y$, see Fig. 1) of this adhesive folded paper was 0.700. Figure 7 shows the data from the pressure-sensitive adhesive paper that did not incline very much after printing. The orientation angles of the face stock, the release liner and the pressure-sensitive adhesive paper were 3°, −48°, and −26°, respectively; the orientation patterns were similar to circles. The degree of inclination ($\tan\xi$) of this folded adhesive paper was 0.087. This pressure-sensitive adhesive paper did not disturb the printing process. This means that the orientation angle of fibers is not always required to be low and that

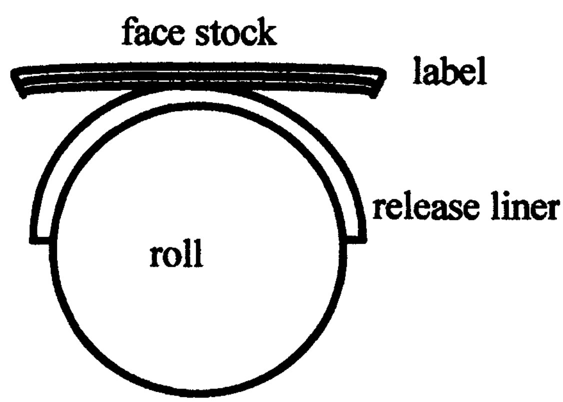
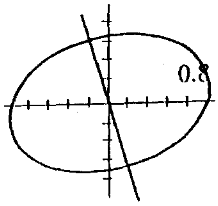
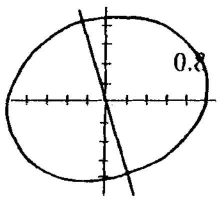


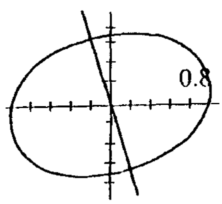
FIGURE 5 Schema of label coming off the release liner.



NAME = Face stock
F =3.980862 GHz
MAX =0.792
MIN =0.535
ANGLE =16 °
MOR =1.463

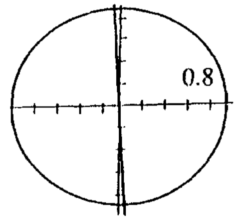


NAME = Release liner
F =3.980761 GHz
MAX =0.797
MIN =0.653
ANGLE =17 °
MOR =1.208

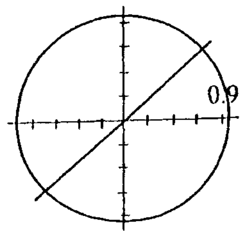


NAME = Pressure-sensitive adhesive paper
F =3.977133 GHz
MAX =0.797
MIN =0.558
ANGLE =15 °
MOR =1.424

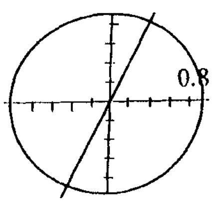
FIGURE 6 The fiber orientation of inclined paper.



NAME = Face stock
F =3.981086 GHz
MAX =0.797
MIN =0.715
ANGLE =3 °
MOR =1.105



NAME = Release liner
F =3.980950 GHz
MAX =0.809
MIN =0.782
ANGLE =-48 °
MOR =1.019



NAME = Pressure-sensitive adhesive paper
F =3.977684 GHz
MAX =0.800
MIN =0.751
ANGLE =-26 °
MOR =1.059

FIGURE 7 The fiber orientation of non-inclined paper.

the orientation degree of fibers is not always required to be parallel to the machine direction to prevent pressure-sensitive adhesive paper from inclining. The limit of the degree of inclination would appear to be 0.364. If the degree of inclination of the folded adhesive paper is under 0.364, then the adhesive paper should not disturb the printing process.

Oozing-property and Label Self Peel-off Property

Table I gives a summary of the ooze properties of different adhesives. P-22 is the permanent type of adhesive that is currently being used, while P-15 is the adhesive that has been developed for used in non-impact printers.

Adhesive paper with varying adhesive coat weights of 12 g/m^2, 15 g/m^2, 19 g/m^2 were made. As can be seen from Table I, when the coat weight was reduced, the amount of oozing also decreased. However, when the coat weight is reduced, the peel strength generally weakens. Table II shows the peel strength at every coat weight for each adhesive. P-15 had almost the same peel strength at a coat weight level of 12 g/m^2 as did P-22 at a coat weight level of 15 g/m^2. Figures 8 and 9 show the master curves of P-22 and P-15. The storage modulus of P-15 tended to be higher than that of P-22, P-22 except in high and low frequency areas; the loss modulus of P-15 also tended to be higher than that of P-22, except in high frequency areas. Frequencies were chosen based on Eq. (1), which corresponds to the velocities of the measured peel strength, printing, the guillotining and slitting processes, and the velocity at which the labels came off.

$$\text{Test frequency} = \frac{\text{Separation speed}}{\text{Adhesive thickness}} (\text{Hz}) \tag{1}$$

It was assumed that the printing frequency was the same as the frequency of the guillotining and slitting processes. Table III shows the storage and loss moduli of the adhesives and the release layer at the frequencies corresponding to the cases

TABLE I
Ooze property

Adhesive-coat weight	P-22	P-15
12 g/m^2	Excellent	Excellent
15 g/m^2	Fair	Fair
19 g/m^2	Poor	Poor

TABLE II
Peel strength (N/25mm)

Adhesive-coat weight	P-22	P-15
12 g/m^2	4.22	6.37
15 g/m^2	6.57	7.75
19 g/m^2	7.45	8.14

adherend is polyethylene

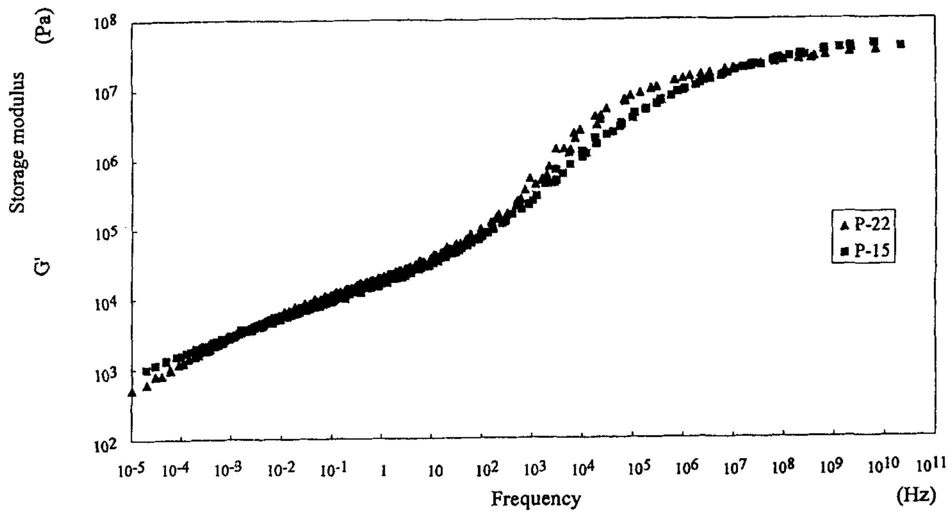


FIGURE 8 Master curves of storage modulus.

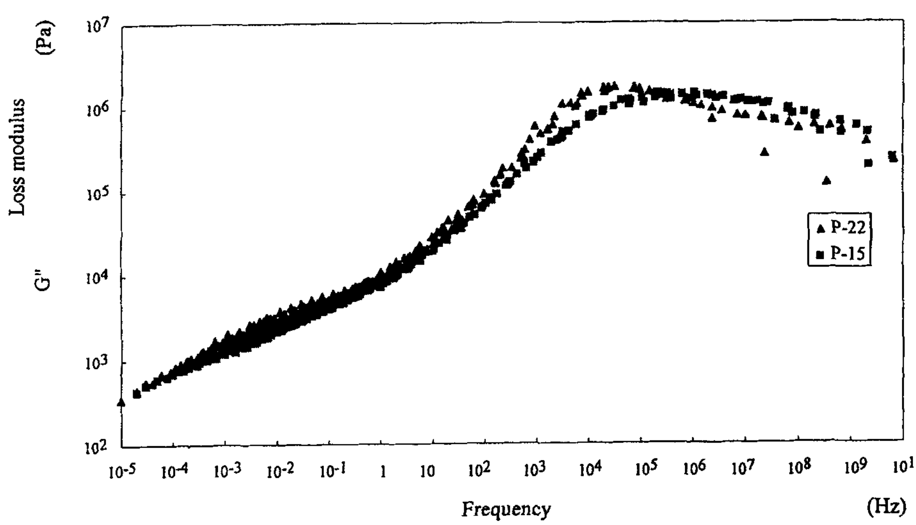


FIGURE 9 Master curves of loss modulus.

	P-22		P-15		Release Agent A	Release Agent B
(Pa)	G'	G''	G'	G''	G'	G'
Peel freq. 10^2 Hz	2.2E + 5	2.5E + 5	1.4E + 5	1.4E + 5	—	—
Print freq. 10^5 Hz	1.1E + 7	1.3E + 6	6.3E + 6	1.3E + 6	2.7E + 5	4.8E + 5
Peel off freq. 10^{-4} Hz (Before starting ro print)	8.2E + 3	3.9E + 3	7.2E + 3	3.1E + 3	3.1E + 5	5.7E + 5

mentioned above. Lower storage and loss moduli corresponded to a higher peel strength[6]. The 100 Hz results, which corresponded to the velocity of the measured peel strength, were focused on. P-15 has a lower storage modulus and a lower loss modulus than does P-22 at 100 Hz. Therefore, P-15 clearly has a higher peel strength than P-22 when the adhesive coat weights are the same.

The dynamic viscoelastic properties of adhesives were studied in order to determine the optimum oozing-property. The relationship between the oozing-property and the storage modulus of other adhesives was investigated at 100 KHz, which corresponds to the frequency of the guillotining and slitting processes. A higher storage modulus corresponds to a better oozing-property. The most effective method to improve the oozing--property was to reduce the adhesive-coat weight without decreasing the peel strength. The lower the values of the storage and loss moduli were, the easier it was to increase the peel strength. However, if the issue of reducing the adhesive coat weight is not considered, then a higher value of the storage modulus corresponds to a better oozing-property.

How the release coefficient of friction affects the oozing-property was also examined. Though this report does not go into detail on the relationship between the release coefficient of friction and the oozing-property, it is considered that a higher coefficient of friction corresponds to a better oozing-property. When pressure is applied by a slitting blade or a guillotine blade and a clamp, the adhesive around the slitting blade or the guillotine blade and the clamp moves around on the release liner and then oozes out from the edge of the adhesive paper after cutting.

Next, the problem of the labels coming off of the release liner by themselves one after another during printing is examined. The printing speed is assumed to be 100 KHz. As can be seen from the Table III, the storage modulus of the P-22 is higher than that of the P-15 at 100 KHz. It can be assumed that the P-15 adheres more strongly to the release liner than does the P-22. Table III also shows the storage moduli of the release agents. The crosslinking level of release agent B is higher than that of release agent A. Release agent B has a higher release force than release agent A, and the storage modulus of release agent B is higher than that of release agent A at 100 KHz. Therefore, the corsslinking level of the release layer is high and the storage modulus of the release layer is also high at 100 KHz. Though it is not evident how closely the crosslinking level relates to the value of the release force, it is generally clear that a higher crosslinking level corresponds to a smaller dependence on the peeling speed. The dependence on the peeling speed means that a higher peeling speed equates to a higher release force. In other words, when the release force is plotted against the peeling speed for various croslinking levels of release agents, as the crosslinking level increases, the inclination decreases. The release force is considered to be large enough to prevent the labels from peeling off of the release liner during printing. It is affected by the adhesive type, *i.e.*, solvent type or emulsion type, acrylic type or NBR type, and so on. This report does not examine the adhesive type in detail. However, at a minimum, it can be concluded that the printing speed determines how large the release force must be.

Finally, the problem of labels coming off of the release liner by themselves before the printing process starts is examined. This phenomenon occurs at very low velocities because it is only the stiffness of the paper that causes the labels to peel back

TABLE IV
Self-peeling tendency of the labels

	Release Agent A	Release Agent B
Print freq. 10^5 Hz	comes off	does not come off
Peel off freq. 10^{-4} Hz (Before starting to print)	comes off	does not come of

from the release liner. Specifically, it occurs at a printing velocity of 10^{-4} Hz. The storage modulus of the P-22 was higher than that of the P-15 at 10^{-4} Hz. It was also assumed that the P-15 adheres more strongly to the release liner than the P-22 at 10^{-4} Hz. Release agent B was assumed to have a high release force at 10^{-4} Hz because it had a high crosslinking level, which means that it has a high release force at the very low speeds given above. Table IV compares the self-peeling tendency of the labels when the two release agents are used. This result supports our assumption.

CONCLUSION

It was found that pressure-sensitive adhesive paper used in non-impact printers presented three problems. First, the edge of the folded adhesive paper used in non-impact printers must not incline after printing. Second, adhesive must not ooze out from the edges during the slitting or guillotining processes. Third, the labels must not come off of the release liner by themselves during printing. The first problem was solved by changing the orientation degree of the fibers, while the other two were solved by studying the dynamic viscoelastic properties of adhesives and release layers.

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