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[54] **PRESSURE-SENSITIVE ELECTROSTATIC IMAGING LABELS**

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Related U.S. Application Data

[63] Continuation-in-part of Ser. No. 113,762, Feb. 8, 1971, abandoned.

[52] **U.S. Cl.** ..... **96/1.5**; 96/1 R; 96/1.8; 117/201; 117/215; 117/218; 117/76 P; 161/406; 283/18; 283/21

[51] **Int. Cl.** **G03g 5/02**; B32b 7/06; G09f 3/00

[58] **Field of Search** ..... 96/1.5, 1.8; 117/201, 215, 117/218, 230, 76 P; 161/406; 283/18, 21

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[57] **ABSTRACT**

There is provided pressure-sensitive electrostatic imaging labels and label stock consisting of a laminate of an electrostatic imaging face material, an adhesive layer and a release coated liner containing an electroconductive agent. The composite laminates have electrical characteristics which permit excellent reproduction of images using photocopying systems conventional to the electrostatic imaging face material used.

22 Claims, No Drawings

PRESSURE-SENSITIVE ELECTROSTATIC IMAGING LABELS

BACKGROUND OF THE INVENTION

This is a Continuation-in-Part of my application Ser. No. 113,762 filed Feb. 8, 1971 now abandoned.

Two of the principal methods presently used in the office photocopy field are the xerographic method and an electrophotographic method typically known as the "Electrofax" process.

In the former, a deposited copy formed by a toner is transferred from a selenium coated drum to conventional paper stock.

In the latter, a charged or latent image is created on the surface of an electrophotographic base stock and a toner directly applied to create the desired image.

Example of an electrophotographic stock is a composite layer of an electroconductive material, normally zinc oxide, a highly resistive resin binder for the zinc oxide, a barrier layer, a carrier, normally paper, and second conductive resin layer. This stock is also known to the art as electrostatic paper.

At least the photoconductive material normally possesses a high degree of electrical resistivity. A surface charge of ionic oxygen is applied by a corona discharge. The charged surface is then exposed to a back lighted image. That portion of the surface which is exposed to the light becomes conductive and the charge dissipates away in proportion to the amount of light reaching the surface. This leaves a residual charge having a pattern corresponding to the matter to be reproduced on or near the surface layer. When a toner is applied, it is attracted to the charged image and by one of several known mechanisms secured to the charge portion of the surface.

A similar process occurs when a dielectric or electrographic stock is used. A photo-sensitive cell is disposed between the material to be copied and the electrographic stock. A surface charge corresponding to the image is created on the surface by light exitation and the image created by toner deposition.

In order to obtain a clean copy free of ghosts, specks, shadows and the like, the electrostatic imaging stock are constructed to have certain electrical characteristics. Photoconductive stock, for instance, should have surface resistivity of between about 10^7 and about 10^{10} ohms to permit charge dissipation along the surface.

The requirements for photoconductive stock performance and the copy mechanisms involved have been well published, as, for instance, in "*Recent Developments in Sensitized Paper Technology*", Diamond, Tappi, Vol. 50, No. 12, December 1967, pages 67A-76A; "*Some Considerations in Paper Substrate Selection for Electrophotographic Coating*", Diamond et al., Tappi, Vol. 48, No. 10, October 1965, pages 96A–98A; "*Performance Structure Relationships of Electroconductive Polymers*", Hoover et al., Tappi, Vol. 51, No. 12, December 1968, pages 552–558; "*Base-Paper-Barrier Properties Relating to the Electrofax Process*", Uhl, Tappi, Vol. 48, No. 12, December 1965, pages 77A–82A; as well as many others.

Independent of the several factors which must be considered in the construction of electrostatic imaging copy paper in use, it has been believed that to yield good images, the copy paper must be maintained in intimate and direct contact with a ground to provide the sink for charge dissipation.

SUMMARY OF THE INVENTION

It has now been found that conventional electrostatic imaging stock can be employed as the face material in the construction of pressure-sensitive adhesive labels and label stock if the release liner of the label or label stock is treated with an electroconductive agent, preferably an electroconductive polymer.

The composite structure comprising an electrostatic imaging face material, a pressure-sensitive adhesive interlayer and a conductive release liner, the conductivity of the release liner being due to the presence of the electroconductive substance and enhanced by the pressure of moisture. The resultant constructions have been found to have surface and volume resistivities consonant with the operating parameters of the copy equipment designed for the electrostatic imaging face material. As a consequence, excellent copies can be made without equipment modification.

It is most surprising and unexpected that placing between a conventional electrostatic imaging paper and ground a layer of an adhesive, a release lacquer and the liner treated with an electroconductive agent will not impede the functionability of the electrostatic imaging stock. This is most unexpected since, of at least the adhesive and the lacquer layers, may be of high resistivity.

For optimum performance, the release liner should contain an electroconductive agent in an amount of from about 0.6 grams per square meter per mil or more. The adhesive layer should be thin, generally in the order of about 0.25 to about 2.0 mils, preferably, from abouot 0.4 to about 0.7 mil. For conventional styrene-butadiene polymer based adhesives this corresponds to an adhesive weight of from about 10 to about 35 grams per square meter. It is preferred to employ an adhesive weight of from about 10 to about 15 grams per square meter. For some applications the lacquer for the release liner should be formulated to provide a 90° release value of 250 grams at 300 in./min. using a Tag and Label Manufacturers Incorporated tester.

To prevent curl and to aid in the condition of a charge, the total construction should also be manufactured to be in equilibrium with an atmosphere having relative humidity between about 20 and 80 percent, preferably, about 40 to about 60 percent. This corresponds to a paper moisture content of from about 2 to about 12 percent by weight, preferably, from about 5 to about 9 percent by weight.

DESCRIPTION

According to the present invention there is provided electrostatic imaging labels and label stock which may be used in conventional copy processes for direct image reproduction without modifications of the copy system or the nature of the electrostatic imaging copy paper.

The electrostatic imaging labels and label stocks provided in accordance with the practice of this invention comprise, in general, a conventional electrostatic imaging face material, a pressure-sensitive adhesive interlayer and a release liner containing an electroconductive agent, preferably an electroconductive polymer. The composite laminates have surfaces and volume resistivities consonant with the electrical performance requirements for the face material and provide thereby clean image reproduction free of shadows, ghosts, specks and the like.

By an electrostatic imaging face material, there is meant a construction having either photoconductive properties, such as provided by zinc oxide or other photoconductive materials, or dielectric properties. Both of the constructions are adapted to receive and hold a surface charge for toner application. The preferred construction for the photoconductive face material is a laminate comprising an outer photoconductive layer of zinc oxide, a highly resistive resin binder for the zinc oxide, a barrier layer, a paper base and an inner layer of an electroconductive resin. The preferred base material for a dielectric construction is paper stock to which there is applied one or more electroconductive layers. Polymeric stock such as polyolefins, polyesters and the like may be also used.

By a pressure-sensitive adhesive, there is meant an adhesive which has sufficient tack to be applied to a substrate without solvent or heat activation. Illustrative, but no wise limiting of the pressure-sensitive adhesives which may be employed there may be mentioned conventional acrylic based and styrene-butadiene based adhesives which are formulated by the additions of tackifiers, fillers, antioxidants and the like, to the base polymer. Styrene-butadiene based polymers are preferred.

By a release liner, there is meant a paper coated with a release substance or lacquer which permits the liner to be peeled free of the face material and adhesive without disrupting the surface of the adhesive. The preferred release lacquer formulations contain silicon release agents, such as polymethylsiloxane, as well as additives which modify the force required to separate the liner from the adhesive.

By electroconductive substance, there is meant an inorganic salt, an organic salt or a conductive polymer such as a polyelectrolyte which may be incorporated into the body of, or onto, the surface of the release liner. Illustrative, but no wise limiting of the electroconductive substances which may be used, there may be mentioned water soluble vinylbenzene quaternary ammonium polymers as described in U.S. Pat. No. 3,011,918 incorporated herein by reference, water soluble polycationic conductive polymers and the like. Water soluble polycationic polymers are preferred.

In the construction of the electrostatic imaging label stock of this invention the greatest impediment to current or charge flow appears to be the adhesive layer. Optimum performance, however, will be obtained for adhesive layers having a thickness from about 0.25 to about 2.0 mils, preferably about 0.4 to about 0.7 mil which corresponds, for a styrene-butadiene based adhesive, to an adhesive weight of from about 10 to about 35 grams per square meter, preferably from about 10 to about 13 grams per square meter.

Where thicker adhesive layers are desired and/or where the adhesive has an unusually high resistivity, there may be incorporated in the adhesive a compatible electroconductive agent, or conductors such as metal powders, graphite and the like, to lower the effective resistivity of the adhesive.

Where a polymethylsiloxane based lacquer is used as the release coating for the release liner the lacquer does not appear to present a major impediment to current flow. As will be shown, the surface resistivity of the lacquer layer is not materially greater than the surface resistivity of a backing material which has been treated with an electroconductive agent.

Bond strength of the release liner to the adhesive may be varied widely. Where the end product, however, is to be cut into labels and the label stock passed over sharp edges as is conventional to many copy operations, the lacquer should be formulated to provide a 90° release value of at least about 250 grams or more at a feed rate of 300 inches per minute, using the Tag and Label Manufacturers Incorporated tester. Peel values of this level or more will prevent dispensing of the labels during printing operation while readily permitting separation of the release liner from the adhesive when it is desired to apply the label to the substrate.

As indicated, the release liner is treated with electroconductive material to reduce its resistivity to a level sufficiently low to permit a composite label construction to have a net resistivity sufficiently low to permit good copy reproduction. For an electrostatic imaging stock having a photoconductive face material a typical surface resistivity will be from about 10^7 to about 10^{10} ohm while the composite laminate will have a volume resistivity of about 10^{13} ohm-cm. The electroconductive agent may be applied to one or both surfaces of the release liner. This will depend, in part, on the thickness of the liner, its permeability and the conductivity of the electroconductive material.

The preferred release liner base for the label construction of this invention is what is known in the art as 35 lb. kraft paper, this being the weight of 500 sheets measuring 25 inches × 38 inches. The application of a polycationic conductive polymer to one side of this release liner paper stock to a preferred level of 1.2 grams per square meter or more results in a release liner having excellent electrical characteristics. Although not bound by theory, this high conductivity may be achieved because of migration of the conductive polymer into the body of the release liner.

Where the release liner is thick, or of low permeability, it may be necessary to apply the electroconductive material to both surfaces to achieve a degree of conductivity sufficient to mimic the electrical characteristics of the face material.

The electroconductive agent may be applied to the release liner prior to or following construction of the label or label stock. Prior application is, however, preferred for manufacturing purposes.

Moisture control is an important factor in the construction of the electroconductive labels or label stock of this invention. The composite construction should be equilibrium with an atmosphere having a relative humidity of from about 20% to about 80%, preferably from about 40% to about 60%. This corresponds to a paper moisture content for each paper based layer of about 2 to about 12 percent, preferably, from about 5 to about 9 percent by weight. The presence of moisture in the construction enhances conductivity and prevents curl.

The electrical characteristics of the pressure-sensitive electrostatic imaging labels and label stock of this invention are such that excellent copies, free of ghosts, specks and shadows may be attained on the face materials when used in copy apparatus designed for such face materials. While slightly higher surface voltages may have to be applied to charge the surface, the voltage required has been found to be fully within the normal range of ajustment of such apparatus. In addition, once the required voltage is reached, the composite construction will begin to leak charge and the volt-

age will not continue to increase. If ordinary release liners were employed the applied voltage will continue to increase with time.

In use, the label stock can be cut to the surface of the release liner to provide a plurality of contiguous labels on a carrier (the release liner) without impairing surface conductivity. In addition, a matrix may be cut leaving a plurality of non-contiguous, discrete labels attached to the release liner carrier without materially effecting either surface or volume resistivity.

The following are examples of the construction of a pressure sensitive electrostatic imaging label and their performance characteristics.

EXAMPLE 1

A photoconductive paper containing a zinc oxide as the photoconductive material and manufactured by Summit Office Products for use in an Apeco Office Copier was employed as a face material. There was applied to the back of the face material a styrene-butadiene pressure-sensitive based adhesive to a level of 12 grams per square meter. There was used as the release liner a 35 lb. kraft paper coated on the release side with a polymethylsiloxane based release lacquer and on the opposite side with a polycationic conductive polymer known as Calgon 261 and manufactured by the Calgon Corporation. The polycationic conductive polymer was applied to a level of 0.75 lbs. per ream of paper as a 10% solution in methanol.

The resultant label stock was cut into 8½ inches × 11 inches sheets. One series of the sheets (Group A) was cut to the release liner to provide a pluraltiy of labels which butted lengthwise but which were separated by an ⅛ inch between rows due to removal of the matrix.

A second group (Group B) was cut to release liner so that the labels abutted lengthwise and crosswise on the release backing sheet.

Table I illustrates the surface resistivities of the coated side and the release liner as a function of relative humidity.

TABLE I

	RESISTIVITY (ohms) 20% (R.H.)	40% (R.H.)	55% (R.H.)	90% (R.H.)
GROUP A Electrophotographic				
Face Side	1×10^{10}	7×10^{9}	9×10^{9}	6×10^{8}
Release Liner	9×10^{7}	5×10^{7}	6×10^{7}	2×10^{7}
GROUP B Electrophotographic				
Face Side	8×10^{9}	5×10^{9}	5×10^{9}	2×10^{8}
Release Liner	6×10^{7}	3×10^{7}	6.5×10^{7}	1×10^{7}

Labels were removed from two of the sheets and surface resistivity measured at a relative humidity of 40%. The average surface resistivity of the side of the release liner treated with the conductive polymer was 2.8×10^{7} ohms. The average resistivity of the lacquered side of the release liner was 6×10^{8} ohms.

TABLE II

	CURL 10% (R.H.)	40% (R.H.)	55% (R.H.)	90% (R.H.)
Group A — Face Side Up	1" face	flat	flat	2" face
Group A — Face Side Down	"	"	"	"
Group B — Face Side Up	"	"	"	3" face
Group B — Face Side Down	"	"	"	"

Machine exposure setting was then determined on an Apeco Copier and compared to a control which was the electroconductive paper used in the construction of the face material. The results are shown in Table III.

TABLE III

APECO COPIER	Machine Setting
Group A — Correct Exposure	7
— Exposure Latitude	6–9
Group B — Correct Exposure	7
— Exposure Latitude	6–9
Control — Correct Exposure	4
— Exposure Latitude	3–8

EXAMPLE 2

A label stock constructed from a 3 mil photoconductive paper, an 0.5 mil styrene-butadiene resin based pressure-sensitive adhesive and a 2 mil 35 lb. kraft paper having a polymethylsiloxane based lacquer coat on one surface and the opposed surface coated with Calgon 291 to a level of 0.75 lb. per ream.

The test method employed was ASTM D-257-66. There was used a Keithley Model 610C and a Keithley Model 6105 Resistivity Adaptor of measuring volume resistivity. Ambient conditions varied from a relative humidity of 47% at 72°F to 46% at 75°F. The applied voltage in each instance was 162 volts.

The electrical characteristics of the label construction in comparison to its component parts are shown in Table IV.

TABLE IV

	Surface Resistivity in Ohms Dark Exposed	Light Exposed
Face Material	1.33×10^{10}	0.72×10^{9}
Release Liner	1.01×10^{10}	0.97×10^{10}
Label Construction	1.15×10^{10}	1.17×10^{10}
	Volume Resistivity in Ohm-Cm Dark Exposed	Light Exposed
Face Material	1.68×10^{12}	0.51×10^{12}
Release Liner	1.48×10^{12}	1.8×10^{12}
Label Construction	1.13×10^{13}	0.73×10^{13}

In the surface resistivity test measurements were made on the zinc oxide surface of the face material and label construction and on the polymer treated surface of the release liner. The surface resistivity was determined by the expression

$$\text{ohms} = \frac{53.36 \times \text{volts}}{\text{amperes}}$$

Volume resistivity was determined by the expression

$$\text{ohm—cm} = \frac{22.9 \times \text{volts}}{\text{amperes} \times \text{thickness in cm}}$$

When moistened the release liner had a lower resistivity than the face material.

EXAMPLE 3

A label constructed using a dielectric paper face material was tested in copy apparatus adapted for the face material alone. Excellent copies were made.

What is claimed is:

1. A multi-ply electrostatic imaging construction for pressure-sensitive labels and label stock comprising a composite laminate of:

a. an electrostatic imaging face material comprised of an electrostatic imaging surface and a support for the electrostatic imaging surface;

b. a pressure-sensitive adhesive interlayer of a thickness of from about 0.25 to about 2.0 mils in adhesive contact with said support; and

c. a release liner comprising a release coating in contact with said pressure-sensitive adhesive interlayer and a paper base containing at least one electroconductive agent, the paper in said composite laminate having moisture content of from about 2 to about 12 percent by weight and said composite laminate having, within said range of moisture content, a volume resistivity, in ohm-cm, greater than the volume resistivity of the electrostatic imaging face material.

2. An electrostatic imaging construction as claimed in claim **1** in which the electrostatic imaging surface of face material is a photoconductive imaging surface and the laminate has a photoconductive imaging surface resistivity of about 10^7 to about 10^{10} ohms and a composite laminate volume resistivity of about 10^{13} ohms-cm within the range of moisture content.

3. An electrostatic imaging construction as claimed in claim **2** in which the paper has a moisture content of from about 5 to about 9 percent by weight.

4. An electrostatic imaging label construction as claimed in claim **1** in which the electroconductive agent is selected from the group consisting of polymers of water soluble vinylbenzene quaternary ammonium and water soluble polycationic conductive polymers.

5. An electrostatic imaging label construction as claimed in claim **2** in which the electroconductive agent is selected from the group consisting of polymers of water soluble vinylbenzene quaternary ammonium and water soluble polycationic conductive polymers.

6. An electrostatic imaging label construction as claimed in claim **3** in which the electroconductive agent is selected from the group consisting of polymers of water soluble vinylbenzene quaternary ammonium and water soluble polycationic conductive polymers.

7. An electrostatic imaging construction as claimed in claim **1** in which the face material is electrographic.

8. An electrostatic imaging label construction as claimed in claim **4** in which the electroconductive agent content of said release liner is about 1.2 grams per square meter.

9. A multi-ply electrostatic imaging construction for pressure-sensitive labels and label stock comprising a composite laminate of:

a. an electrostatic imaging face material comprised of an electrostatic imaging surface and a support for the electrostatic imaging surface;

b. a pressure-sensitive adhesive interlayer having a thickness of from about 0.25 to about 2.0 mils in adhesive contact with said support;

c. a release liner comprising a release coating in contact with said pressure-sensitive adhesive interlayer and a paper base containing at least 1.2 grams per square meter of at least one electroconductive agent selected from the group consisting of water soluble vinylbenzene quaternary ammonium and water soluble polycationic conductive polymers, the paper in said composite laminate having a moisture content of from about 2 to about 12 percent by weight and said composite laminate, within said range of moisture content, having a volume resistivity, in ohm-cm, greater than the volume resistivity of the electrostatic imaging face material.

10. An electrostatic imaging label construction as claimed in claim **9** in which the pressure-sensitive adhesive layer has a thickness of from about 0.4 to about 0.7 mil.

11. An electrostatic imaging label construction as claimed in claim **9** in which the paper has a moisture content of from about 5 to about 9 percent by weight.

12. An electrostatic construction as claimed in claim **10** in which the paper has a moisture content of from about 5 to about 9 percent by weight.

13. An electrostatic imaging label construction as claimed in claim **1** in which the pressure-sensitive adhesive interlayer has a thickness of from 0.4 to about 0.7 mil.

14. A multi-ply electrostatic imaging construction for pressure-sensitive labels and label stock comprising a composite laminate of:

a. an electrostatic imaging face material comprised of an electrostatic imaging surface and a support for the electrostatic imaging surface;

b. a pressure-sensitive adhesive interlayer of a thickness of from about 0.25 to about 2.0 mils in adhesive contact with said support; and

c. a release liner comprising a release coating in contact with said pressure-sensitive adhesive interlayer and a paper base containing at least one electroconductive agent, the paper in said composite laminate having moisture content of from about 2 to about 12 percent by weight, said composite laminate having a volume resistivity of about 10^{13} ohm-cm within said range of moisture content.

15. A multi-ply electrostatic imaging construction for pressure-sensitive labels and label stock comprising a composite laminate of:

a. an electrostatic imaging face material comprised of an electrostatic imaging surface and a support for the electrostatic imaging surface;

b. a pressure-sensitive adhesive interlayer having a thickness of from about 0.25 to about 2.0 mils in adhesive contact with said support;

c. a release liner comprising a release coating in contact with said pressure-sensitive adhesive interlayer and a paper base containing at least 1.2 grams per square meter of at least one electroconductive agent selected from the group consisting of water soluble vinylbenzene quaternary ammonium and water soluble polycationic conductive polymers, the paper in said composite laminate having a

moisture content of from about 2 to about 12 percent by weight, said composite laminate having a volume resistivity of about 10^{13} ohm-cm within said range of moisture content.

16. An electrostatic imaging construction as claimed in claim **15** in which the electrostatic imaging surface of face material is a photoconductive imaging surface and the laminate has a photoconductive imaging surface resistivity of about 10^{7} to about 10^{10} ohms.

17. An construction as claimed in claim **16** in which the paper has a moisture content of from about 5 to about 9 percent by weight.

18. An construction as claimed in claim **15** in which the electroconductive agent is selected from the group consisting of polymers of water soluble vinylbenzene quaternary ammonium and water soluble polycationic conductive polymers.

19. An construction as claimed in claim **16** in which the electroconductive agent is selected from the group consisting of polymers of water soluble vinylbenzene quaternary ammonium and water soluble polycationic conductive polymers.

20. An construction as claimed in claim **17** in which the electroconductive agent is selected from the group consisting of polymers of water soluble vinylbenzene quaternary ammonium and water soluble polycationic conductive polymers.

21. An construction as claimed in claim **17** in which the face material is electrographic.

22. An electrostatic imaging construction as claimed in claim **18** in which the electroconductive agent content of said release liner is about 1.2 grams per square meter.

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UNITED STATES PATENT AND TRADEMARK OFFICE

CERTIFICATE OF CORRECTION

PATENT NO. : 3,907,557
DATED : September 23, 1975
INVENTOR(S) : William J. Bolduc, Jr.

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

Column 1, line 55, "96A" should read --94A--.

Column 2, lines 13-14, "pressure" should read --presence--

line 32, "abouot" should read --about--

line 42, "condition" should read --conduction--.

Signed and Sealed this

tenth Day of *February 1976*

[SEAL]

Attest:

RUTH C. MASON
Attesting Officer

C. MARSHALL DANN
Commissioner of Patents and Trademarks