United States Patent [19]

Hasegawa et al.

[11] **3,787,207**

[45] **Jan. 22, 1974**

[54] **ELECTROPHOTOGRAPHIC PHOTOSENSITIVE PLATE HAVING A POLYIMIDE INTERMEDIATE LAYER**

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[22] Filed: **Dec. 16, 1971**

[21] Appl. No.: **208,708**

[52] **U.S. Cl.** **96/1.5,** 117/218

[51] **Int. Cl.** **G03g 5/00**

[58] **Field of Search** 96/1.5, 1, 1.8; 117/218

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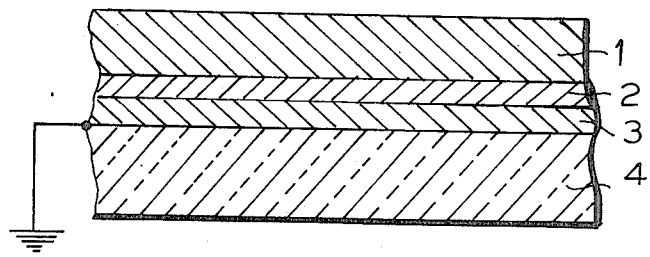
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[57] **ABSTRACT**

This invention provides an improved electrophotographic photosensitive plate with excellent durability and electrophotographic characteristics.

The plate comprises a multilayered electrophotographic photosensitive plate comprising, from the top, a first layer including an organic photoconductive insulating material, a second layer including vitreous selenium, a third layer including a polyimide material and the fourth layer of an electrically conductive backing.

4 Claims, 15 Drawing Figures



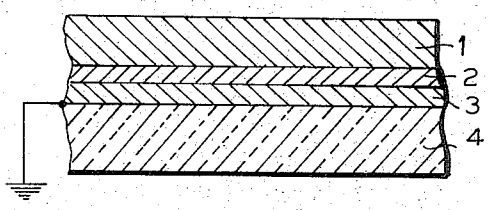


FIG. 1

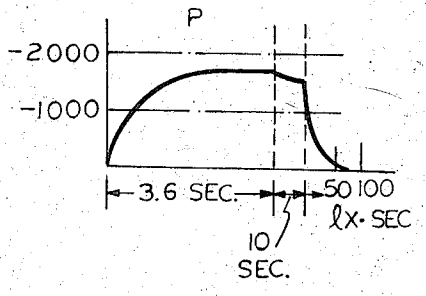


FIG. 5

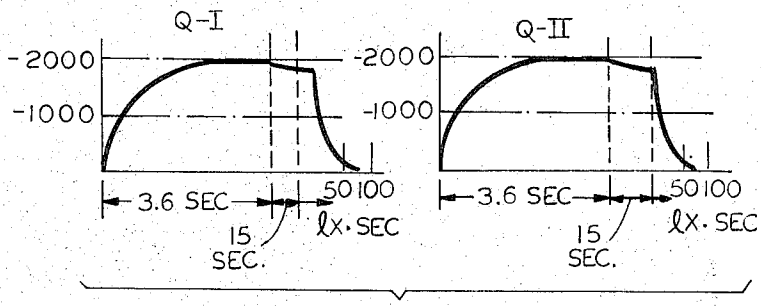


FIG. 6

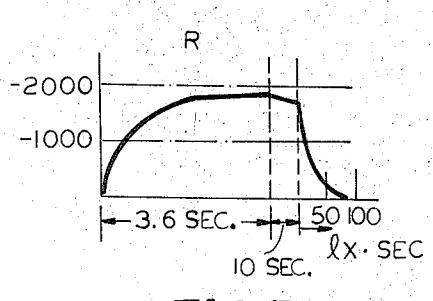


FIG. 7

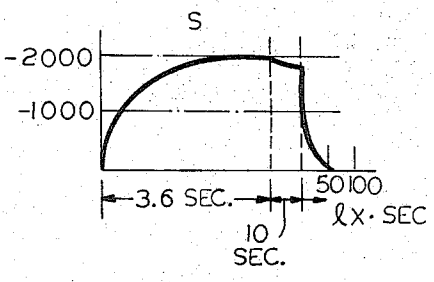


FIG. 8

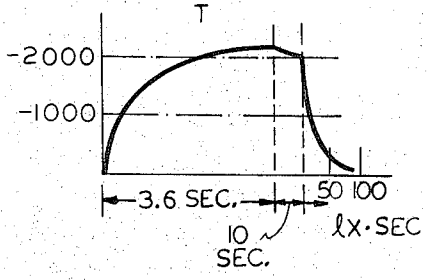


FIG. 9

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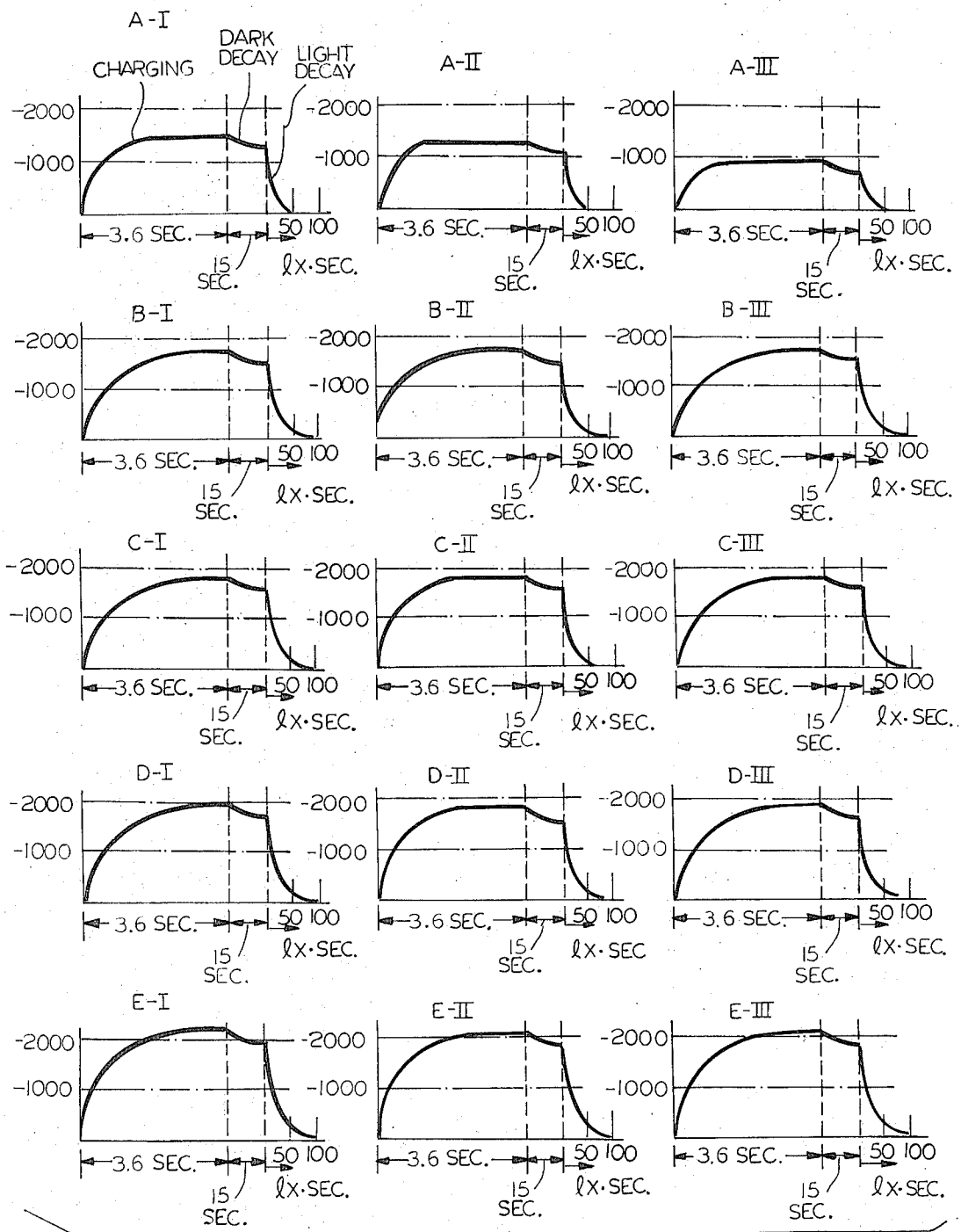


FIG. 2

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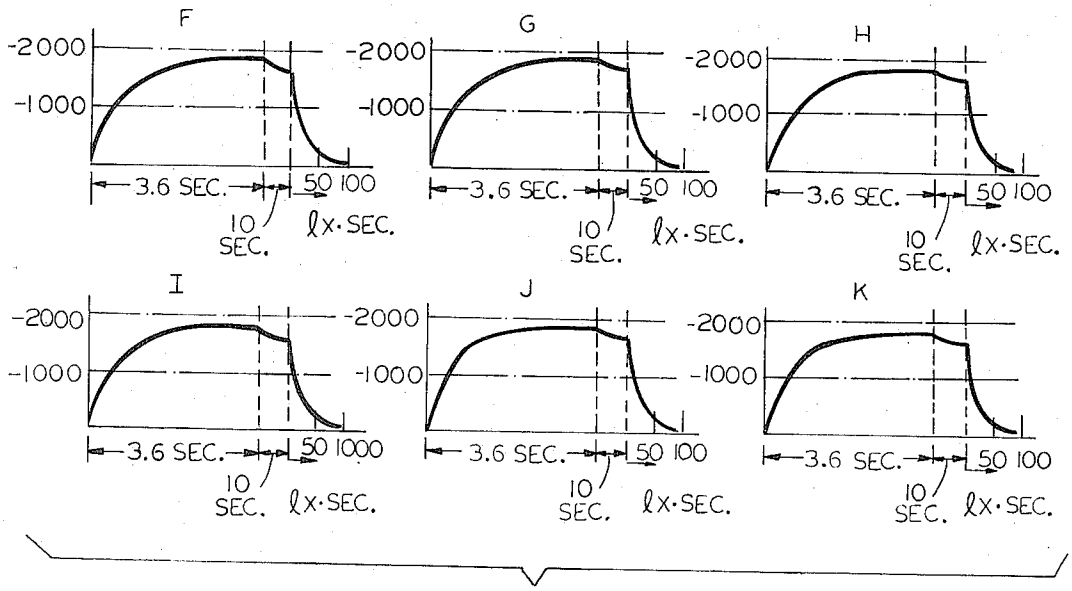


FIG. 3

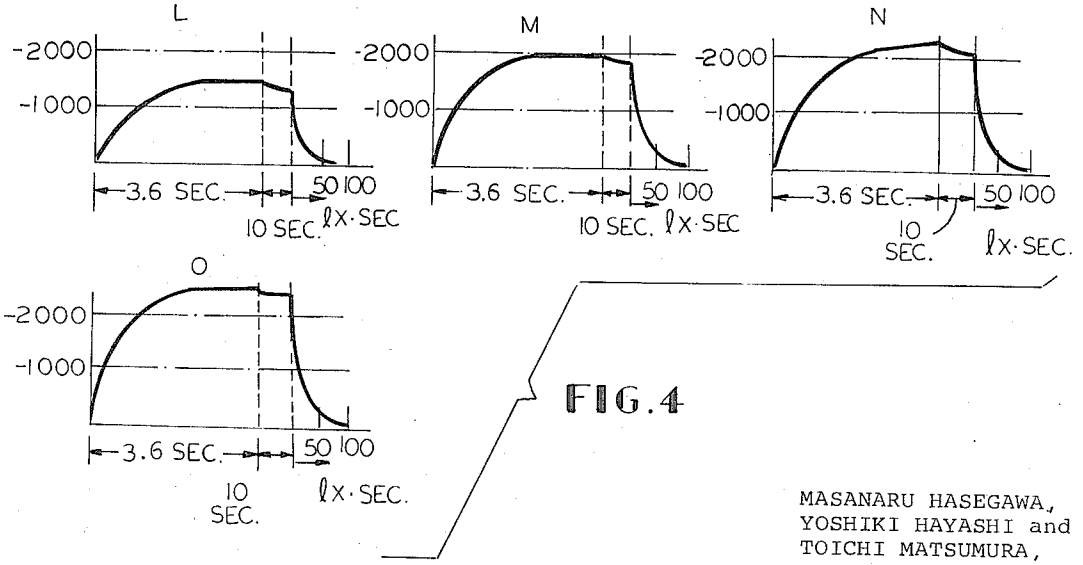


FIG. 4

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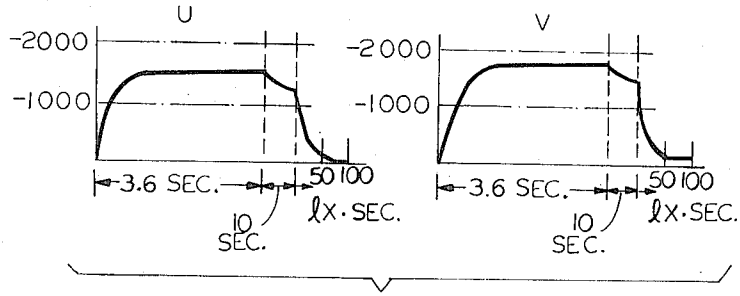


FIG.10

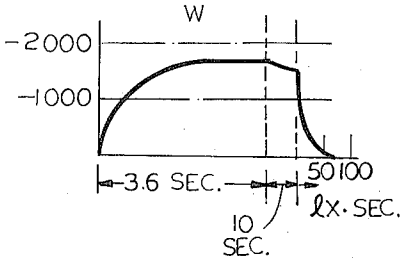


FIG.11

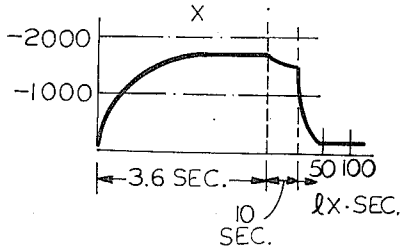


FIG.12

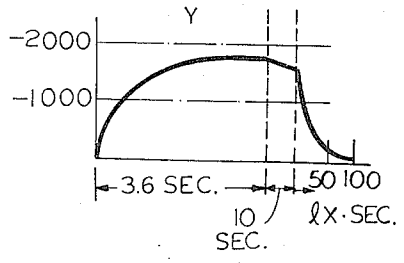


FIG.13

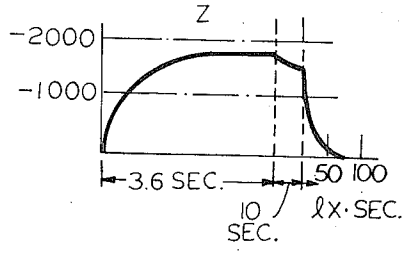


FIG.14

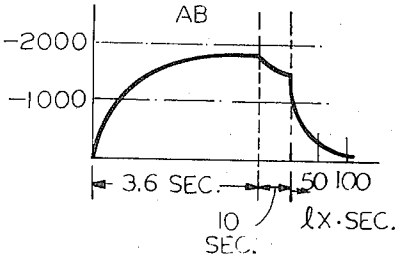


FIG.15

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ELECTROPHOTOGRAPHIC PHOTOSENSITIVE PLATE HAVING A POLYIMIDE INTERMEDIATE LAYER

This invention relates to electrophotography and more particularly an improved electrophotographic photosensitive plate and the material thereof.

It is known that images are formed and are visually developed by miscellaneous electrophotographic processes. The most common process is well known as the Carlson process, which is described in U.S. Pat. No. 2,297,691.

Some other known electrophotographic processes include those disclosed in U.S. Pat. Nos. 2,833,648, 2,982,647 and 3,147,679.

A multilayered electrophotographic photosensitive plate suitable for the above-mentioned electrophotographic processes is described in copending U.S. Pat. application, Ser. No. 889,118, filed on Dec. 30, 1969, now U.S. Pat. No. 3,725,058. Said multilayered electrophotographic photosensitive plate comprises essentially three layers, e.g., a top layer which is a substantially visible ray transmissive layer including an organic photoconductive insulating material, a second layer comprised of a thin vitreous selenium layer and a bottom layer comprised of an electrically conducting backing.

Said multilayered electrophotographic photosensitive plate displays many advantages, such as reusability, high charge acceptability and high photosensitivity in the visible ray region. In addition, it is possible to make a flexible and/or transparent web from this plate.

In the multilayered electrophotographic photosensitive plate of the prior art, the thin vitreous selenium layer is basically vacuum-deposited directly on an electrically conducting backing, such as an aluminum plate and a cuprous-iodide-superposed polyester film. However, when the surface of said conducting backing is very smooth and glassy, the adhesive attraction between said selenium layer and said surface of the electrode is weak and the final product, i.e., the multilayered electrophotographic photosensitive plate may be subject to peeling

Moreover, when the multilayered electrophotographic photosensitive plate of the prior art is repeatedly used in a copying system, such as the toner-transfer type copier described in U.S. Pat. No. 2,357,809 or the charge transfer copier described in U.S. Pat. No. 3,015,304 and 2,825,814, said plate exhibits decreasing charge acceptability or increasingly produces many pinholes with repeated use.

A further disadvantage is that the second layer, including the thin vitreous selenium in the multilayered electrophotographic photosensitive plate of the prior art tends to produce a metal selenide compound when some active metallic materials, such as copper and aluminum, are used as the bottom conductive backing. The metal selenide does not exhibit sufficient activity as a photosensitizing layer in the multilayered electrophotographic photosensitive plate.

Accordingly, it is an object of the present invention to improve the interface adhesiveness between the vitreous selenium layer and the bottom layer of an electrically conducting backing of said multilayered electrophotographic photosensitive plate.

It is a further object of the present invention to provide an improved multilayered electrophotographic photosensitive plate which exhibits unchanged charge acceptability or produces fewer pinholes during repeated operation.

It is a still further object of the present invention to provide novel materials for use in an improved construction of the multilayered electrophotographic photosensitive plate.

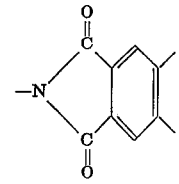
These and other objects of the present invention will be apparent upon consideration of the following detailed description taken together with accompanying drawings, wherein

FIG. 1 is a cross-sectional view of an electrophotographic photosensitive plate according to the present invention,

FIGS. 2 to 15 are graphs showing electrophotographic characteristics of the electrophotographic photosensitive plates used in the test examples described hereinafter.

Referring to FIG. 1, a four-layered electrophotographic photosensitive plate contemplated by the present invention comprises, from the top, a first layer (layer 1) including a transparent organic photoconductive insulating material, a second layer (layer 2) including vitreous selenium and a third layer (layer 3) including a polyimide material and a bottom layer (layer 4) of an electrically conductive backing.

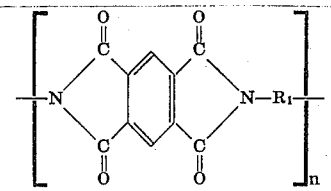
The polyimide material used in said layer 3 in accordance with the present invention is designated as a polymer type organic compound, which has at least one chemical group in recurring unit, said chemical group is represented by the following general formula:



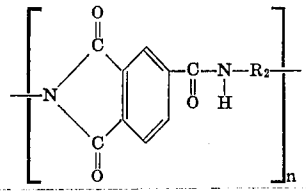
Several polyimide materials including above chemical group are commercially available as heat-durable polymers for use in electrical insulation, adhesive tape base and printed circuit base. Said polyimide materials are marketed in a form of a lacquer, powder and a flexible film.

The polyimide material used in the present invention includes thermoplastic linear polymers and thermosetting crosslinked polymers.

The preferable polyimide materials used in said layer 3 is a polypyromellitimide, which is a product produced by a condensation reaction between pyromellitic dianhydride and a diamine having the following chemical formula:



or a product produced by a condensation reation between trimellitic anhydride and a diamine, having a following chemical formula



wherein:

R_1 and R_2 is selected from hydrogen atom, aromatic group, alkyl group, alkyl-aryl group and diphenyl ether group, and n is an integer.

Here, the two polyimide materials are represented by linear polymer form, however, it is well known by those skilled in the art that the final polyimide material above sometimes exhibits a crosslinked form by heat treatment.

The present invention can be achieved by inserting a layer **3** of above-mentioned polyimide material between the vitreous selenium layer **2** and the conducting base **4** in the three-layered electrophotographic photosensitive plate of the prior art.

The manufacturing process for the novel electrophotographic photosensitive plate including said polyimide layer can be carried out by the following per se well known method.

A preferred conductive base material, such as an aluminum plate, brass plate, steel plate, aluminum vapor deposited on polyester film, or CuI-superposed-polyester film is carefully cleaned with a chemically inert solvent. The cleaned surface is then coated with a solution of a polyimide material by any suitable and available coating method, such as a doctor-blade method, roll coating method, gravure printing method and spray method. If required, said solution can contain a plasticizer, viscosity control material, etc.

The preferable solvent for the polyimide material is such as N-methyl-2-pyrrolidine (d, p.; 202° C), N-N-dimethyl acetamide or their combination which is less volatile than a common organic solvent such as benzene, toluene, methylethyl keton, etc. Therefore, it is preferable to place the coated layer of the polyimide solution in a hot oven of 90°–200° C for about 1 hour in order to make an almost dry and tack-free layer of the polyimide material.

The tack-free polyimide layer is preferably subjected to an aging treatment for improvement of light decay characteristics. The aging treatment is achieved by keeping the tack-free polyimide layer at 100°–150° C for about 10–20 hours.

The operable thickness of the dried polyimide layer is less than 15μ. Use of said polyimide layer thicker than 15μ not only lowers a light decay rate but also increases residual potential. The most preferable thickness range is from 2 to 8μ.

The cured surface is preferably cleaned with an AC corona and then is coated with a vitreous selenium layer by vacuum deposition.

The vacuum-deposited vitreous selenium layer is then coated with an organic photoconductive insulating layer. The vacuum deposition of vitreous selenium and the coating of the organic photoconductive insulating material is carried out in a manner similar to that described in the aforesaid copending U.S. Pat. application Ser. No. 889,118.

The four-layered electrophotographic photosensitive plate of the present invention provides pinhole free images even after the plate is electrophotographically reused 5,000 times or more. The life of the four-layered photosensitive plate of the present invention is longer by about five to 10 times that of the three-layered photosensitive plate.

The interface bonding between the vitreous selenium layer and polyimide layer, and between the polyimide layer and the conducting backing is strong enough to prevent the separation of each of the layers.

In order to improve only the interface bonding and/or the reusability, it is easy for those skilled in the art to insert an insulating layer between said vitreous selenium layer **2** and said conducting backing **4.** However, the insertion of an insulating layer impairs the electrophotographic characteristics (e.g., charging, dark decay, light decay). In fact, when the insulating layer is made of a polymeric material other than a polyimide, such as a polyurethane, epoxy resin, polyvinyl acetate, polyvinyl butyral, polypropylene, chlorinated polypropylene, polystyrene, chloro-sulfonated polyethylene, silicone resin, copolymer of vinyl chloride and vinyl acetate, copolymer of vinylidene chloride and acrylonitrile and copolymer of vinylidene chloride and vinyl chloride, the insertion of the insulating layer results in inferior characteristics with respect to the light decay rate and residual potential in comparison with a three-layered photosensitive plate having no layer **3.** The "residual potential," referred to herein, is designated as a surface potential which persists even when the photosensitive plate is exposed to strong illumination.

A decrease in the light decay rate and/or an increase in residual potential causes the resultant photosensitive plate to have sluggish photosensitive response and to reproduce foggy images, respectively.

The four-layered photosensitive plate, using polyimide material as layer **3,** has never shown any unusual residual potential even when repeatedly used 5,000 times or more.

The useful effect of the polyimide layer in the multi-layered photosensitive plate, according to the present invention is clearly understood by reference to the following examples. However, these examples should not be construed as limitative.

Example 1

A solution of polypyromellitimide (commercially available as a trade name "Pire ML Lacquer," Du Pont de Nemours and Co., U.S.A.), was applied to four cleaned aluminum plates of 1 mm. thickness with a knife-blade in order to form a polypyromellitimide layer having various thicknesses.

Upon the polyimide layer which had been completely dried and cured, the vitreous selenium layers were vacuum-deposited by a conventional method under 1×10^{-4}mmHg with a basket coil type tungsten heater. The selenium metal used was 99.999 percent in purity.

The surface of the vitreous selenium layers were then coated with a solution of an organic photoconductive insulating material comprising 100g of poly-N-vinyl carbazole (commercially available as a trade name "Luvican M-170," BASF, West Germany), 30g of polycarbonate (commercially available under a trade name "Panlite-L," Teijin Chemical Co., Japan), 30g of diphenyl chloride (commercially available under the trade name "Kanechlor," Kenegafuchi

Chemical Co., Japan), 600g of monochlorobenzene and 600g of dichloroethane. The coating was dried so as to complete four four-layered electrophotographic photosensitive plates.

For comparison, a three-layered photosensitive plate having no polyimide layer was also prepared by the same manner as above mentioned.

The thickness of the polyimide layers, the vitreous selenium layers or the organic photoconductive insulating (OPC) layers is listed in Table 1.

TABLE 1

Plate	Polyimide Layer	Se Layer	OPC Layer
A	none	0.2–0.4μ	8μ
B	3μ	"	"
C	5μ	"	"
D	8μ	"	"
E	15μ	"	"

The electrophotographic characteristics (charge acceptance, dark decay and light decay) of the five plates in Table 1 were measured by using a corona discharge device of −6KV and a tungsten lamp of 5 lux in surface illuminance. The test results are shown in FIG. 1, wherein the curves A–I, B–I, C–I, D–I and E–I correspond to electrophotographic characteristics of a virgin plate of A, B, C, D, and E, respectively. Likewise, curves A–II to E–III correspond to electrophotographic characteristics of reused plates A to E, which have previously been subjected to the electrophotographic process cycle (e.g., corona-charging in the dark and illuminating with a tungsten lamp), repeated 1,000 times (II series) and 5,000 times (III series), respectively.

From the curves A–I, A–II and A–III of FIG. 1, the charge acceptability of the three-layered plate A decreases with an increase in the reusing cycle times. In contrast, the four-layered plates B to E do not change with respect to charge acceptability even when reused 5,000 times, as shown in curve B–I to B–III, curve C–I to C–III, curve D–I to D–III and curve E–I to E–III, respectively.

After being subjected to the electrophotographic process cycle repeated 5,000 times, the plates A to E were then examined with an electrophotographic imaging test which is based on Carlson's method using a magnetic brush development technique. The resultant image, which was obtained with a light exposure of about 30 lux·sec. in white area, was satisfactory in sharpness through all the plates A to E. However, only in the plate A, the image was low in density and showed white pinholes numerously on the black area.

The reused plates A to E were also subjected to a TESI imaging test, which is based on the transfer of a latent electrostatic image of negative polarity to a dielectric paper, as described in the French Pat. No. 2,024,150. The transferred image was then developed with a liquid toner containing positively charged particles.

The developed image from plate A was low in density and showed numerous white pinholes on the black area, however, the developed images from plates B to E were high in density and were practically free from pinholes in the black area.

The plates B to E were strong enough in the interlayer bonding between the vitreous selenium layer and the polyimide layer or between the polyimide layer and the aluminum plate, and were entirely satisfactory in the adhesive tape test described in copending U.S. Pat. application, Ser. No. 033,425, dated Oct. 24, 1968, now abandoned. However, the plate A demonstrated poor interlayer bonding between the vitreous selenium layer and the aluminum plate, and exhibited separation of these layers by the same adhesive tape test.

Example 2

A solution of polypyromellitimide (commercially available under the trade name "Pire ML Lacquer," Du Pont de Nemours & Co., U.S.A.) was applied to six cleaned aluminum plates of 1mm in thickness by a knife-blade in order to from a polypyromellitimide layer. Upon the polyimide layer which had been completely dried and cured, the vitreous selenium layers and the OPC layers were applied by the similar manners to those of Example 1. The materials used were also similar to those of Example 1.

The thicknesses of the polyimide layer, the selenium layer and the opc layer are listed in Table 2.

TABLE 2

Plate	Polyimide Layer	Se Layer	OPC Layer
F	5μ	0.3μ	9μ
G	"	0.5μ	"
H	"	0.7μ	"
I	"	1.2μ	"
J	"	1.5μ	"
K	"	2.1μ	"

The electrophotographic characteristics of the six plates F to K in Table 2 are shown as corresponding curves F to K in FIG. 3.

The TESI imaging test used in Example 1 was applied to those plates in which the optimum exposure was about 20 luxsecond in each plate. The resultant TESI images were all clear and showed no fog in the white background.

The plates F to K were of sufficient strength in the interlayer adhesiveness between the vitreous selenium layer and the polyimide layer or between the polyimide layer and the aluminum plate, and were entirely satisfactory in the adhesive tape test employed in Example 1.

Example 3

A solution of polymellitimide (commercially available under the trade name "Pire ML Lacquer," Du Pont de Nemours & Co., U.S.A.) was applied to four cleaned aluminum plates of 1mm in thickness by a knife-blade in order to form a polypyromellitimide layer. Upon the polyimide layer which had been completely dried and cured, the vitreous selenium layers and OPC layers were applied in a manner similar to those of Example 1. The materials used were also similar to those of Example 1.

The thicknesses of the polyimide layer, the selenium layer and the OPC layer are listed in Table 3.

TABLE 3

Plate	Polyimide Layer	Se Layer	OPC Layer
L	5μ	0.2–0.4μ	5μ
M	"	"	10μ
N	"	"	15μ
O	"	"	20μ

The electrophotographic characteristics of the four plates L to O in Table 3 are shown as corresponding curves F to K in FIG. 3.

The TESI imaging test used in Example 1 was applied to those plates. The optimum exposure was about 20 lux·second in each plate. The resultant TESI images were all clear and showed no fog in the white background.

Example 4

A cleaned copper plate was coated with a polypyromellitimide solution (commercially available under the trade name "Pire ML Lacquer," Du Pont de Nemours & Co., U.S.A.) and was then dried and cured. The cured thickness of the polyimide layer was 5μ, and a vitreous selenium layer of 0.2 to 0.4μ thickness was vacuum-deposited thereon.

Finally the OPC solution used in Example 1 was further applied onto the selenium surface. The dried thickness of the OPC top layer was 8μ.

The electrophotographic characteristics of the four-layered plate identified as plate P is shown in curve P in FIG. 5. For comparison, a three-layered photosensitive plate comprising from the bottom up, a cleaned copper backing of 1mm in thickness, a vitreous selenium layer of 0.2 to 0.4μ in thickness, and an OPC layer of 8μ in thickness, was made by using the same raw materials and manufacturing process as the four-layered plate.

While the four-layered plate P still exhibited the same electrophotographic characteristics even at 1 year after preparation date, the three-layered plate showed no photosensitivity after 2 days.

Example 5

Aluminum layer of 0.06μ in thickness was vacuum-deposited on a 75μ thick polyethylene terepIthalate film (commercially available as a trade name "Metalumy," Toray Co., Japan), and the surface thereof was coated with a polypyromellitimide solution (commercially available under the trade name "Pire ML Lacquer," Du Pont de Nemours & Co., U.S.A.) and was then dried and cured. The cured thickness of the polyimide layer was 5μ.

A vitreous selenium layer having a varied thickness ranging from 0.15 to 0.3μ was vacuum-deposited onto the polyimide layer in a similar manner to those of Examples 1 and 2.

The OPC solution used in Examples 1 and 2 was further applied to the selenium layer and then completely dried. The dried thickness of the OPC layer was 10μ.

The electrophotographic photosensitive film, identified as film Q, comprising five layers was completed as described above, and then was subjected to a measurement of electrophotographic characteristics. It was as shown in FIG. 6.

The curves Q–I and Q–II correspond to the electrophotographic characteristics of the virgin film Q and after being subjected to 3,500 cycles of the electrophotographic process, respectively.

The TESI imaging test used in Example 1 was also carried out with the reused film Q. The high contrast and no fog image were obtained by exposing an imaging light of 20 lux·second in white area.

The film Q was flexible and even when folded to an angle of 170° C, the film showed no separation within its interlayers.

Example 6

An aluminum foil of 7μ in thickness was laminated onto a thick paper with an emulsion type adhesive, and the surface thereof was coated with a polypyromellitimide solution (commercially available as "Pire ML Lacquer," Du Pont de Nemours & Co., U.S.A.), and was then dried and cured.

A vitreous selenium layer having a varied thickness ranging from 0.15 to 0.3μ was vacuum-deposited onto the polyimide layer in a similar manner to those of Examples 1 and 2.

The OPC solution used in Examples 1 and 2 was further applied to the selenium layer and then was completely dried. The dried thickness of the OPC layer was 8μ. The completed photosensitive plate is identified as plate R.

The electrophotographic characteristic of the plate R is shown as a curve-R in FIG. 7.

It is understood from the curve-R that the plate is suitable for Carlson's electrophotographic process.

Example 7

A polypyromellitimide solution (commercially available under the trade name "Traynese No. 2000,", Toray Co., Japan) was applied to a cleaned aluminum plate of 1mm thickness, and was then dried and cured. The cured thickness of the polyimide layer was 6μ.

A vitreous selenium layer having a varied thickness ranging from 0.2 to 0.4μ was vacuum-deposited onto the polyimide layer by the similar manner to those of Examples 1 and 2.

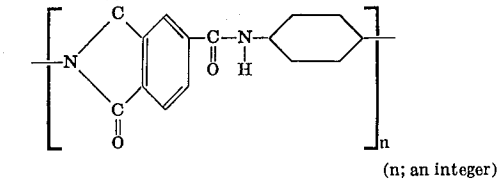
The OPC solution used in Examples 1 and 2 was further applied to the selenium layer and was then completely dried. The dried thickness of the OPC layer was 10μ.

The completed electrophotographic photosensitive plate which was identified as plate S was then subjected to a measurement of electrophotographic characteristics. It was as shown in FIG. 8.

The interlayer bonding between the selenium layer and the polyimide layer or between the polyimide layer and the aluminum plate was strong enough for the adhesive tape test used in Example 1. Even when the plate S was folded rectangularly, the plate showed no separation within its interlayers.

Example 8

A solution including mainly a polyimide (commercially available under the trade name "Trayness No. 1000" Toray Co., Japan) which is defined by the following chemical formula



(n; an integer)

was applied to a cleaned aluminum plate of 1 mm in thickness and was then dried and cured. The cured thickness of the polyimide layer was 5μ.

A vitreous selenium layer having a varied thickness ranging from 0.2 to 0.4μ was vacuum-deposited onto the polyimide layer in a manner similar to those of Examples 1 and 2.

The OPC solution used in Examples 1 and 2 was further applied to the selenium layer and ten completely dried. The dried thickness of the OPC layer was 10μ.

The completed electrophotographic plate, identified as plate T, was subjected to the measurement of the electrophotographic characteristics. The result is shown as curve T in FIG. 9.

The interlayer bonding between the selenium layer and the polyimide layer or between the polyimide layer and the aluminum plate was sufficiently strong in the adhesive tape test used in Example 1.

Example 9

An electrophotographic photosensitive plate having four layers, identified as plate V, was prepared in the same manner as those of Examples 1 and 2.

The raw materials used for the backing plate, the polyimide layer and the selenium layer were quite similar to those used in Example 1. The organic photoconductive insulating layer for the plate V was made by coating a solution which consisted of 10g of 1,3-diphenyl-5-(p-methoxy)-pyrazoline, 10g of polystyrene (commercially available under the trade name "Diarex HF-55-106," Mitsubishi-Monsanto Chemical Co., Japan) and 90g of monochlorobenzene.

For comparison, a three-layered photosensitive plate, named plate U, having no polyimide layer was also prepared. The thicknesses of the layers of the completed plate U and V were listed in Table 4.

TABLE 4

Plate	Polyimide Layer	Se Layer	OPC Layer
U	none	0.2–0.4μ	10μ
V	5μ	"	"

The electrophotographic characteristics of the plate U and V are shown as corresponding curves U and V in FIG. 10, respectively.

The interlayer bonding in plate U and V were tested by the adhesive tape test used in Example 1.

The coating of the plate U having the two layers, e.g., selenium layer and the OPC layer, were easily peeled in a combined form from its aluminum backing plate. On the contrary, the coating of the plate V having the polyimide layer, the selenium layer and the OPC layer were sufficiently strong in the same adhesive tape test.

Example 10

A solution of polypyromellitimide (commercially available under the trade name "Pire ML Lacquer," Du Pont de Nemours & Co., U.S.A.) was coated on a cleaned aluminum plate of 1mmm in thickness and then dried and cured. The cured polyimide layer was 5μ in thickness.

A vitreous selenium layer of 0.2 to 0.4μ in thickness was vacuum-deposited by the same manner as the Example 1.

An OPC solution which comprised 100g of brominated polyvinyl carbazole (cf. U.S. Pat. No. 3,421,891), 40g of polycarbonate (commercially available under the trade name "Iupiron", Mitsubishi Edogawa Chemical Co., Japan), 20g of an epoxy resin (commercially available as trade name "Epikote 828," Shell Chemical Co., Japan) and 1,200g of monochlorobenzene, was applied to the vitreous selenium surface, and dried completely. The completed electrophotographic photosensitive plate, identified plate W, was subjected to measurement of its electrophotographic characteristics. The result is shown as curve W in FIG. 11.

Further, the plate W was examined with the adhesive tape test used in Example 1, and the interlayer bonding was sufficiently strong in the test.

Example 11

A solution of polypyromellitimide (commercially available as a trade name "Pire ML Lacquer," Du Pont de Nemours & Co., U.S.A.) was coated on a cleaned aluminum plate of 1mm in thickness, and then dried and cured. The cured polyimide layer was 5μ in thickness.

A vitreous alloy layer of 95 molar percent selenium and 5 molar percent of tellurium was vacuum-deposited under 1×10^{-4} mmHg. The used selenium and tellurium were both 99.99 percent in purity. The evaporation method used was the alloy method, in which the evaporation source material was previously alloyed. Used here was a single tungsten basket type coil. The evaporated layer of the alloy was 0.5μ in thickness.

The OPC solution used in Example 1 was applied to the surface of the alloy layer and dried. The dried thickness of the OPC layer was 8μ.

The resultant electrophotographic photosensitive plate, identified as plate X, was subjected to measurement of its electrophotographic characteristics. The result is shown as curve-X in FIG. 12.

Further, the plate X was examined with the adhesive tape test used in Example 1, and the interlayer bonding was sufficiently strong enough in the test.

Example 12

A solution of polypyromellitimide (commercially available under the trade name "Pire ML Lacquer," Du Pont de Nemours & Co., U.S.A.) was applied to a cleaned aluminum plate of 1mm in thickness, and was then dried and cured. The cured polyimide layer was 5μ in thickness.

A vitreous selenium layer including sulfur was vacuum deposited onto the polyimide surface in a similar manner to those of Examples 1 and 2.

The source material was a mixture of 1g of selenium powder and 0.2g of sulfur, which was heated and evaporated in a tungsten basket coil under 1×10^{-4} mmHg.

The resultant selenium layer, including sulfur, was 0.5μ in thickness.

The OPC solution used in Example 1 was applied to the surface of the selenium layer including sulfur, and the dried OPC layer was 8μ in thickness.

The completed electrophotographic photosensitive plate, identified as plate Y, was subjected to measurement of its electrophotographic characteristics. The result is shown as curve Y in FIG. 13. From the curve Y, it is understood that the plate Y is useful in Carlson's electrophotographic process.

Example 13

A solution of 100g of Pire ML lacquer (used in Example 1) including 5g of sucrose acetate-isobutyrate was applied to a cleaned aluminum plate of 1mm in thickness, and was then dried and cured.

A vitreous selenium layer of 0.2 to 0.4μ in thickness was vacuum-deposited in a similar manner to those of Examples 1 and 2.

An OPC solution used in Example 1 was further applied to the surface of the selenium layer and dried. The dried OPC layer was 8μ thick.

The completed electrophotographic photosensitive plate, identified as plate Z, was subjected to a measurement of its electrophotographic characteristics. The result is shown as curve Z in FIG. 14. From the curve Z, it is understood that the plate Z is useful in Carlson's electrophotographic process.

Example 14

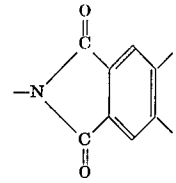
A surface of a polyimide film of 12μ in thickness (commercially available under the trade name "Kapton," Du Pont de Nemours & Co., U.S.A.) was coated with a copper metal layer by vacuum-evaporation under 1×10^{-4} mmHg. The white light transmittance of the copper layer was 50 percent. The copper layer was then iodized by contacting the copper layer with an iodine vapor in order to make a copper-iodide transparent electrode layer. Such an iodization process as used herein is described in U.S. Pat. No. 2,756,165.

Upon the opposite untreated surface of the polyimide film, a vitreous selenium layer of 0.2 to 0.4μ in thickness was vacuum-deposited in a manner similar to those of Examples 1 and 2. Further, upon the selenium layer, the OPC solution used in Example 1 was applied. The dried OPC layer was 10μ in thickness.

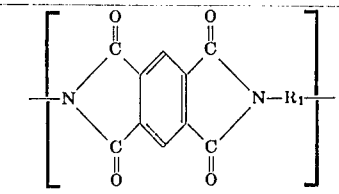
The completed four-layered electrophotographic photosensitive film, identified as film AB, was a flexible and reddish colored transparency. The electrophotographic characteristic of the film AB is shown as curve AB in FIG. 15. From the curve AB, it is understood that the film AB is useful in Carlson's electrophotographic process.

We claim:

1. An electrophotographic photosensitive plate comprising, from the top, a first layer comprising transparent organic photoconductive insulating material, a second layer comprising vitreous selenium, a third layer comprising a polyimide material, of which third layer has a thickness less than 15 microns, and a fourth layer of an electrically conductive backing, wherein said polyimide material in said third layer has the recurring unit



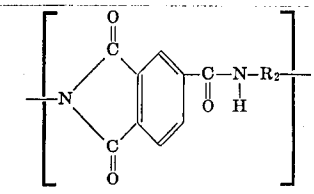
2. A multilayered electrophotographic photosensitive plate, defined by claim 1, wherein said polyimide material has the following chemical formula:



wherein:

R_1 is selected from hydrogen atom, aromatic group, alkyl group, alkyl-aryl group and diphenyl ether group, and n is an integer.

3. A multilayered electrophotographic photosensitive plate defined by claim 1, wherein said polyimide material has the following chemical formula:



wherein:

R_2 is selected from hydrogen atom, aromatic group, alkyl group, alkyl-aryl group and diphenyl ether group, and n is an integer.

4. A multilayered electrophotographic photosensitive plate defined by claim 1, wherein said third layer, including polyimide material, has a thickness range of 2 to 8 microns inclusive.

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