

[54] **ELECTROPHOTOGRAPHIC RECORDING MATERIAL**
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[51] **Int. Cl.**..... **G03g 5/06**
[58] **Field of Search** **96/1 R, 1 PG, 1.3, 1.5,**
96/1.6; 252/501

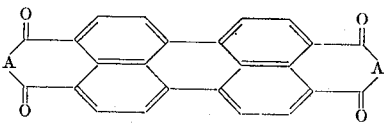
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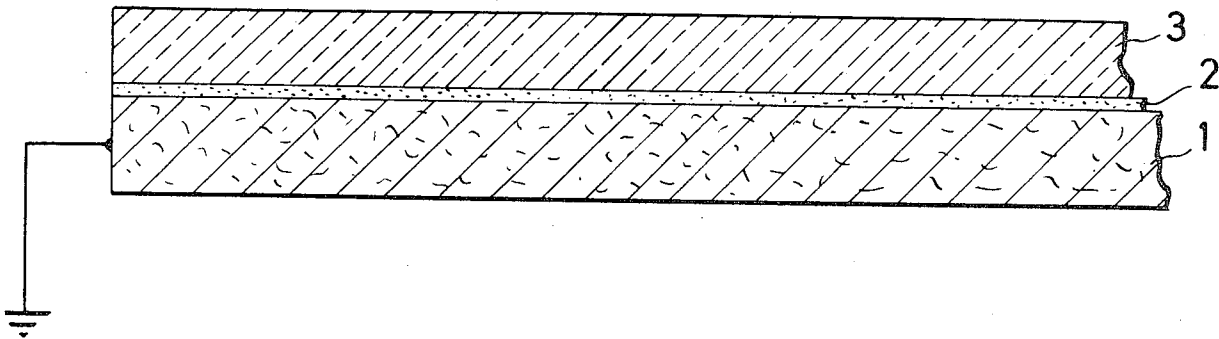
Primary Examiner—Roland E. Martin, Jr.
Attorney, Agent, or Firm—James E. Bryan

[57] **ABSTRACT**
This invention relates to an electrophotographic recording material consisting of an electroconductive support material and a photoconductive double layer of organic materials which consists of a homogeneous, opaque, charge carrier producing dyestuff layer of a compound corresponding to the general formula



wherein
A stands for O (oxygen) or =NR (imide nitrogen) in which
R stands for hydrogen, lower alkyls with one to four carbon atoms, aryl or substituted aryl, aralkyl, a heterocyclic group, or —NHR', with
R' being either phenyl, which may be substituted, or benzoyl,
and of a transparent top layer of insulating materials containing at least one charge transporting compound.

14 Claims, 3 Drawing Figures



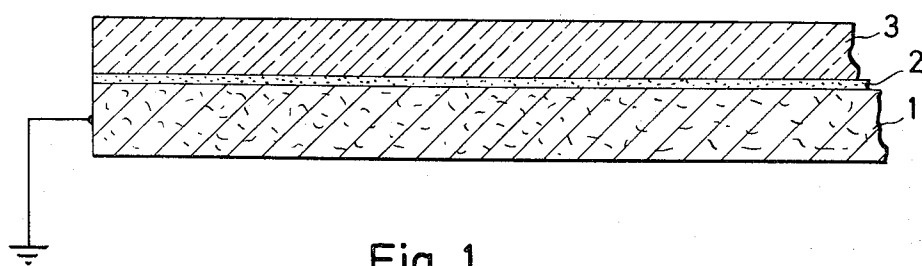


Fig. 1

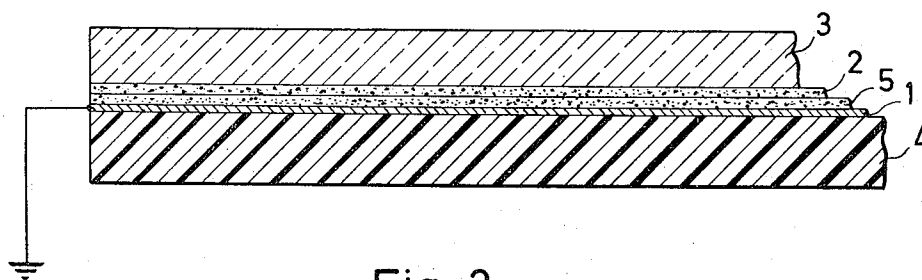
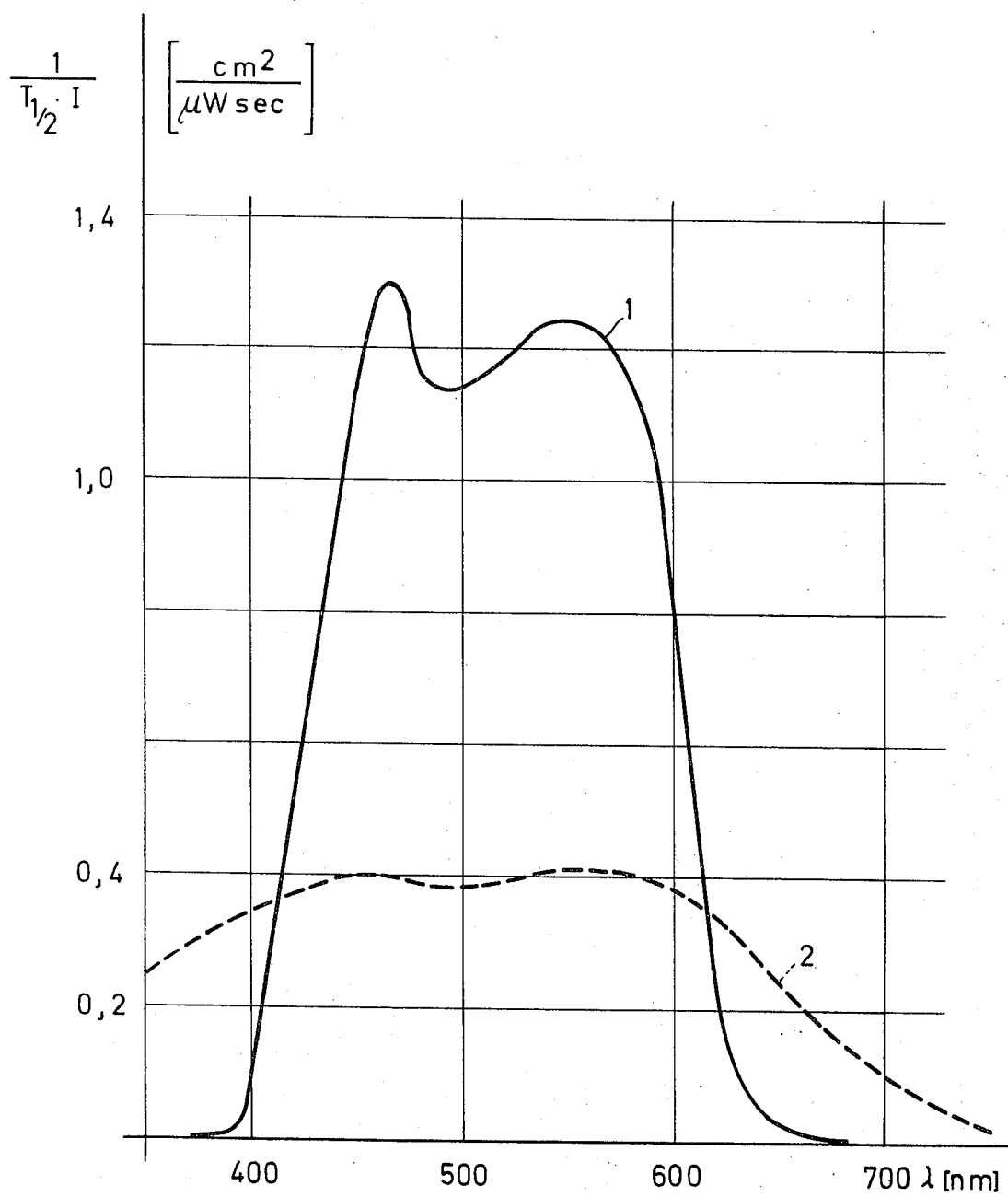
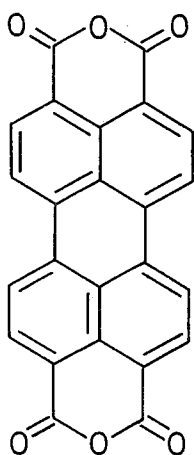


Fig. 2

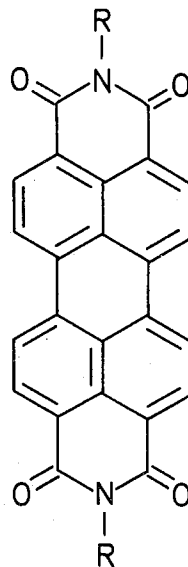
Fig. 3



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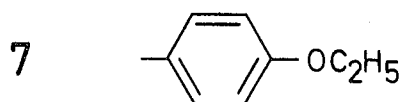
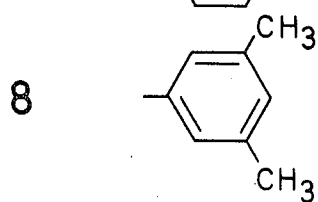
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3 R = CH₃

4 R = (CH₂)₃-CH₃

5 R = CH₂CH₂OH

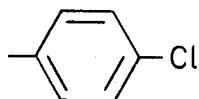
6 R = C₆H₅



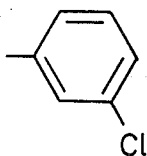
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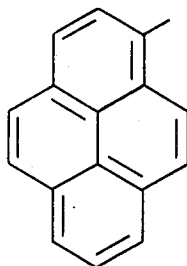
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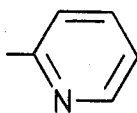
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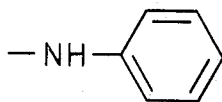
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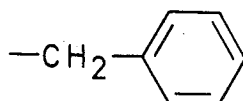
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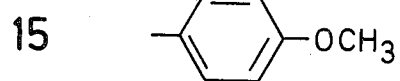
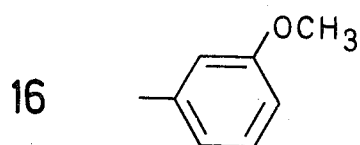
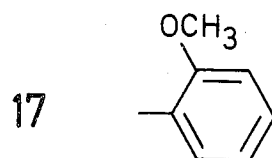
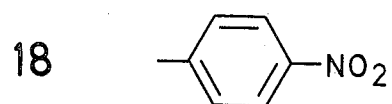
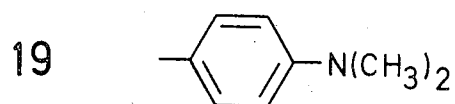


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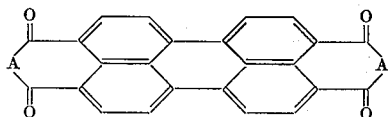
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ELECTROPHOTOGRAPHIC RECORDING MATERIAL

This invention relates to an electrophotographic recording material consisting of an electroconductive support material and a photoconductive double layer of organic materials which consists of a homogeneous, opaque, charge carrier producing dyestuff layer of a compound corresponding to the general formula



wherein

A stands for O (oxygen) or =NR (imide nitrogen) in which

R stands for hydrogen, lower alkyls with one to four carbon atoms, aryl or substituted aryl, aralkyl, a heterocyclic group, or —NHR', with

R' being either phenyl, which may be substituted, or benzoyl,

and of a transparent top layer of insulating materials containing at least one charge transporting compound.

It is known from German Offenlegungsschriften Nos. 1,597,877 and 1,797,342 for electrophotographic recording material to extend the spectral sensitivity of selenium layers to the red spectral range by a double layer arrangement, e.g. with phthalocyanine dispersion layers. Disadvantageous are the vacuum vapor depositions of selenium requiring high technical expenditure, the brittleness of comparatively thick selenium layers, the poor adhesion of adjacent heterogeneous constituents in these layers and the only difficulty realizable uniformly wetting coating with the corresponding dispersions. Furthermore, no optimum light-sensitivities can be achieved as a result of the absorption behavior and the different charge conducting mechanisms of selenium and phthalocyanine in the double layer arrangement.

From U.S. Pat. No. 3,573,906, for example, there are also known photoconductive double layers containing an organic, possibly photoconductive, insulating layer between the support material and the vapor-deposited selenium layer in order to impart adhesion. Such a layer construction, however, considerably hinders the necessary charge transport so that, in this case, too, no higher light-sensitivities are obtainable.

Furthermore, from German Auslegeschrift No. 1,964,817, it is known to provide vapor-deposited selenium layers with a layer of an organic, photoconductive insulating material which is substantially insensitive to light in the visible range of the spectrum. According to German Offenlegungsschrift No. 2,120,912, it has also been suggested to use those light-sensitive layer arrangements for electrophotographic recording materials which contain, as the charge carrier producing layer, an inorganic material, such as the sulfide, selenide, selenotelluride or telluride of cadmium or zinc, and, as the charge carrier transporting layer, an organic material with at least 20 per cent by weight of

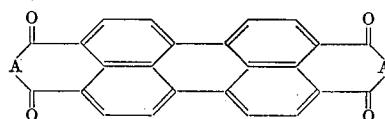
2,4,7-trinitro-9-fluorenone. A disadvantage of the production of these layers with inorganic photoconductors is the exact observation of the vapor deposition conditions of selenium or the exact adjustment of the mixtures in order to obtain a good photoconductive modification of the inorganic materials. Furthermore, the adhesion of selenium to conductive support material, such as to aluminum, is insufficient. Fatigue in repeated charge/exposure cycles does not allow the use in electrophotographic copying devices.

Japanese Pat. application No. 43-26710 already discloses photoconductive double layers of organic materials on a conductive support. According to that application, a lower, relatively thick layer of a considerably diluted homogeneous solution of a sensitizer in a binder is provided with an upper transparent light-sensitive layer. This layer construction, however, only offers a relatively low sensitivity increase only little meeting technical demands. Another known suggestion according to German Offenlegungsschrift No. 1,909,742 is to repeatedly pour a sensitizer solution over a photoconductive layer and to evaporate the solvent. A disadvantage thereof is the low mechanical resistance of the applied layer as a result of insufficient cohesion and adhesion of the applied sensitizer. Furthermore, repeated coating is cumbersome.

The construction of photoconductive double layers containing a dyestuff layer is also known, e. g. from Belgian Pat. Nos. 763,389 and 763,541, but for this layer construction, top layers are used which allow no sensitivities satisfying highest demands and, as regards adhesion between the dyestuff layer and the top layer, do not represent an optimization and are not sufficiently resistant to mechanical attack, e.g. in electrophotographic copying devices, particularly to that due to the cleaning of the photoconductive layer.

It is the object of the present invention to provide an organic photoconductor layer highly light-sensitive for the xerographic copying procedure which overcomes the described disadvantages and the adhesion of which between the various layers satisfies the highest technical demands, which exhibits no wear or fatigue and which, even after repeated use, may be used again rapidly.

The present invention provides an electrophotographic recording material consisting of an electroconductive support material with a photoconductive double layer of organic materials which consists of a homogeneous, opaque, charge carrier producing dyestuff layer of a compound corresponding to the general formula



wherein

A stands for O (oxygen) or =NR (imide nitrogen) in which

R stands for hydrogen, lower alkyls with one to four carbon atoms, aryl or substituted aryl, aralkyl, a heterocyclic group, or —NHR' , with

R' being either phenyl, which may be substituted, or benzoyl,

and of a transparent top layer of insulating materials with at least one charge transporting compound, which is characterized in that the transparent top layer consists of a mixture of a binder with a charge transporting, monomer, heterocyclic compound substituted by at least one dialkyl amino group or two alkoxy groups and having an extended π -electron system or with a condensation product of 3-bromo-pyrene and formaldehyde.

By means of the invention, it is possible to obtain highly light-sensitive, photoconductive double layers for the electrophotographic recording material of the invention which have a high mechanical resistance and may be arranged on a cylindrical drum, for example, or may circulate as an endless belt without exhibiting special signs of wear and thus are very suitable for the use in electrophotographic copying devices. The high light-sensitivity particularly results from the fact that the charge transporting compound present in the transparent top layer is sensitized by the charge carrier producing dyestuff layer in that the charge carriers, such as electrons or holes are taken by the top layer.

In a preferred embodiment, the organic dyestuff layer has a thickness in the range from about 0.005 to about 2 μm . High concentration of excited dyestuff molecules is achieved thereby in the dyestuff layer and at the boundary surface between the dyestuff layer and the top layer. Furthermore, the adhesion between the electroconductive support material and the top layer is not impaired.

In a preferred embodiment, the transparent top layer has a thickness in the range from about 5 to about 20 μm . This assures a sufficiently high charge.

The structure of the electrophotographic recording material according to the invention is shown in the attached FIGURES. FIG. 1 shows a material consisting of an electroconductive support 1, an organic dyestuff layer 2, and an organic transparent top layer 3. FIG. 2 shows a metallized plastic layer 1, 4 as the support, to which an intermediate layer 5 is applied which prevents the injection of charge carriers in the dark. This combination is coated with a photoconductive double layer consisting of the organic dyestuff layer 2 and an organic, transparent top layer 3.

Suitable electroconductive support materials are materials which hitherto have been used for this purpose, for example aluminum foils or transparent plastic supports to which aluminum, gold, copper, zinc, cadmium, indium, antimony, bismuth, tin, lead or nickel has been laminated or applied by vapor deposition. Generally, any support may be used which has been made sufficiently electroconductive.

An organic intermediate layer, or also a metal oxide layer produced by a thermal, anodic or chemical process, (such as, e.g., an aluminum oxide layer) may be applied to the electroconductive support. This layer serves the purpose to reduce the injection of charge carriers from the electroconductive support into the organic dyestuff layer. In addition thereto, it improves the adhesion of the dyestuff layer to the support. Besides the oxide layers already mentioned, such materials are used which are not noticeably dissolved when

the top layer is applied, e.g. polyamide resins or polyvinyl phosphonic acid.

An organic intermediate layer may have a thickness of approximately 1 μm , a metal oxide layer may range in thickness from about 10^2 to 10^4 Å.

The organic dyestuff layer of perylene-3,4,9,10-tetracarboxylic acid derivatives of the electrophotographic recording material of the invention substantially determines the spectral light-sensitivity of the photoconductive double layer of the invention. The organic dyestuff layer must be extremely uniform since only its uniformity guarantees a uniform injection of charge carriers into the top layer. To achieve this object, the dyestuff layers are applied according to special coating methods. Such methods are the application by mechanically rubbing the most finely powdered dyestuff material into the electroconductive support material, the application by chemical deposition of a leuco-base to be oxidized, for example, the application of electrolytical or electrochemical processes or the gun spray method. The application preferably is performed, however, by vapor depositing the dyestuff in the vacuum. A tightly packed homogeneous coating is achieved thereby.

The tightly packed coating makes it unnecessary to produce thick dyestuff layers for achieving a high absorption. The tightly packed dyestuff molecules and the extremely low layer thickness permit, in a particularly advantageous manner, the transport of charge carriers so that it is completely sufficient to produce the charge carriers at the boundary layer only.

The application of the dyestuff layer by vapor deposition in the vacuum requires dyestuffs with thermal resistivity in the temperature range to be applied for vapor deposition. The high extinction of the dyestuff allows high concentration of excited dyestuff molecules. Excitation (1) and charge separation (2) take place in the dyestuff layer according to the following reaction equations:



with

S — dyestuff molecule

S^x — excited dyestuff molecule, and

$\cdot S^+$, $\cdot S^-$ — dyestuff radical ions.

At the boundary surface between the organic dyestuff layer and the transparent top layer, reactions of the excited dyestuff molecules or the resulting charge carriers in the form of the dyestuff radical ions with the molecules of the charge transport effecting compound in the top layer are possible according to the following equations:



with

F_1 — donor molecule

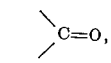
F_2 — acceptor molecule

$\cdot F_1^+$, $\cdot F_2^-$ — donor or acceptor radical ion.

At the boundary surface, sensitizing reactions take place between the transparent top layer and the organic dyestuff layer. The top layer thus is a sensitized organic photoconductor at least in the area of the boundary surface, which leads to the surprisingly high photoconductivity.

Reactions 3 and 5 proceed preferably when the π -electron system in the top layer is a compound which, as a donor compound, easily can release electrons. This is the case with 2,5-bis-(p-diethylamino-phenyl)-1,3,4-oxadiazole, for example. Reactions 4 and 6 are preferably possible with a substance in the top layer which, as an electron acceptor, easily accepts electrons, e.g. 2,4,7-trinitrofluorenone or N-t-butyl-3,6-dinitro-naphthalimide.

By means of the specific embodiment of the invention it is sufficient for the efficiency of the dyestuff when, besides its intense absorption, it only has either electron-attracting substituents, e.g.

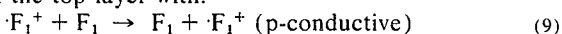
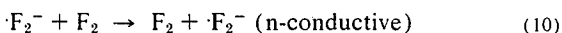


—NO₂, halogen, or electron-repelling substituents, e.g. —NH₂, —N-alkyl₂ or —O-alkyl, depending on whether it is preferably suitable for reactions 3,5 or 4,6. The invention permits charge carrier transport fostered by a particularly low expenditure of energy within the tightly packed dyestuff layer according to the following reactions:



In all conventional sensitizing processes, however, transport via the dyestuff molecules present in low concentration is impeded by their large distance from one another.

Analogous is the procedure of the charge transport in the top layer with:



The practical consequence of reactions 1 to 10 is that, in the use of electron donors in the top layer, the double layer arrangement is negatively charged so that reactions 3,5,8,9 can proceed. In the inverse case, layers with electron acceptance in the top layer are positively charged so that reactions 4,6,7, and 10 can proceed.

As far as their preparation is not mentioned in this application, the derivatives of perylene-3,4,9,10-tetracarboxylic acid, in particular the anhydride and the diimide derivatives, are known. They may be obtained according to condensation methods which are known in organic chemistry, e.g. by reacting perylene-3,4,9,10-tetracarboxylic acid or the corresponding anhydride at elevated temperatures with primary amines carrying different substituents. Thus, German Patent Specification No. 1,067,157 describes the preparation of N,N'-di-(3,5-dimethylphenyl)-peryene-3,4,9,10-tetracarboxylic acid-diimide (Formula 8) and in German Auslegeschrift No. 1,113,773 the synthesis of N,N'-di-(4-ethoxyphenyl)-peryene-3,4,9,10-tetracarboxylic acid diimide (Formula 7) is described. The pigments tested in connection with this application

were purified by the methods described in the respective patents.

N,N'-di-(4-chlorophenyl)-peryene-3,4,9,10-tetracarboxylic acid diimide (Formula 9) and N,N'-dimethyl-peryene-3,4,9,10-tetracarboxylic acid diimide (Formula 3) are described in "Colour Index" IInd Edition, Vol. 3, 1956, on page 3543, under Nos. 71.135 and 71.130.

Depending on their substitution at the imide nitrogen, the color of these pigment dyestuffs is either red (Formula 7), scarlet (Formula 8), chestnut brown (Formula 3), Bordeaux red (Formula 2) or violet-tinged red (Formula 12). The spectral light-sensitivity of the materials during photoconduction is mainly determined by the absorption behavior of these dyestuffs in the visible range of the spectrum.

It was surprising that in an arrangement such as that shown in the attached FIGS. 1 and 2, i.e., with an electroconductive support 1, an organic dyestuff layer 2, an organic top layer 3 facilitating the charge transport, or a metal laminated plastic foil 1,4 and an intermediate layer 5 prohibiting injection of the charge carriers, the perylene-3,4,9,10-tetracarboxylic acid derivatives together with the organic top layers of the invention display a very high degree of photosensitivity in the visible range of the spectrum. The multi-layer photoconductor layers of the invention are compared to a photoconductor layer containing a donor-acceptor complex, as described in Belgian Patent Specification No. 699,851.

The layer according to the Belgian Patent consists of polyvinyl carbazole ("Luvican M 170", a product of BASF, Ludwigshafen, Germany) and 2,4,7-trinitrofluorenone at a molar ratio of 1 : 1 (thickness of the layer approx. 12 μ m). In Example 4, a double layer having a layer thickness of 10 to 12 μ m is compared with this layer under identical conditions, determining the half time ($T_{1/2}$ msec) for the respective wave length range by negative charging and exposure under an XBO 150 xenon lamp, interposing monochromatic filters (line filters, halftime range 10–12 nm, marketed by Schott and Gen., Mainz, Germany).

By plotting the reciprocal values of the products of halftime ($T_{1/2}$ msec) and light-intensity I, expressed in μ W/cm² against the wave length in nm, a curve is produced for the spectral light-sensitivity which is shown in the attached FIG. 3. The reciprocal value of $T_{1/2}$. I is the light-energy, per unit area, which must be irradiated to discharge the layer to half its original charge U_0 . FIG. 3 shows clearly that the light-sensitivity of the double layer (curve 1) within the wave length range from 415 to 625 nm is substantially higher than that of the known polyvinyl carbazole/2,4,7-trinitrofluorenone layer (curve 2) with which it is compared.

The transparent top layer of organic insulating materials with at least one charge transporting compound is described as follows:

The transparent top layer has a high electric resistance and prevents in the dark the flowing off of the electrostatic charge. Upon exposure to light, it transports the charges produced in the organic dyestuff layer.

If it is to be negatively charged, the transparent top layer preferably consists of a mixture of an electron donor compound and a binder. But when the electrographic recording material is to be used for positive charge the transparent top layer consists of a mixture of an electron acceptor compound and a binder.

Consequently, in the transparent top layer there are used compounds for charge transport which are known as electron donors or electron acceptors. They are used together with binders or adhesives adapted to the compound for charge transport as regards charge transport, film property, adhesion, and surface characteristics. Furthermore, conventional sensitizers or substances forming charge transfer complexes may be present. But they can only be used in so far as the necessary transparency of the top layer is not impaired. Finally, other usual additives such as levelling agents, plasticizers, and adhesives may also be present.

Suitable compounds for charge transport are especially those organic compounds which have an extended π -electron system, e.g. monomer aromatic heterocyclic compounds.

Monomers employed in accordance with the invention are those which have at least one dialkyl amino group or two alkoxy groups. Particularly proved have heterocyclic compounds, such as the oxadiazole derivatives, mentioned in German Pat. No. 1,058,836. An example thereof is in particular the 2,5-bis-(p-diethylaminophenyl)-oxadiazole-1,3,4. Further suitable monomer electron donor compounds are, for example, triphenyl amine derivatives, benzo-condensed heterocycles, pyrazoline or imidazole derivatives, as well as triazole and oxazole derivatives, as disclosed in German Pat. Nos. 1,060,260 and 1,120,875.

Formaldehyde condensates of various aromatic compounds, e.g. the formaldehyde condensate of 3-bromopyrene, may also be used.

Besides these mentioned compounds having predominantly a p-conductive character, it is also possible to use n-conductive compounds. These so-called electron acceptors are known from German Pat. No. 1,127,218, for example. Compounds such as 2,4,7-trinitrofluorenone or N-t-butyl-3,6-dinitronaphthalimide have proved particularly suitable.

Suitable binders with regard to flexibility, film properties, and adhesion are natural and synthetic resins. Examples thereof are in particular polyester resins, e.g. those marketed under the names "Dynapol" (Dynamit Nobel), "Vitel" (Goodyear), which are copolyesters of iso- and terephthalic acid with glycol. Silicone resins are those known under the names SR of General Electric Comp. or Dow 804 of Dow Corning Corp., U.S.A., and which are three-dimensionally cross-linked phenylmethyl siloxanes or the so-called "reactive" resins, e.g. the so-called "DD" lacquers consisting of an equivalent mixture of polyesters or polyethers containing hydroxyl groups and polyfunctional isocyanates, e.g. of the "Desmophen" or "Desmodur" type marketed by Bayer AG, Leverkusen, Germany, have proved particularly suitable. Furthermore, copolymers of styrene and maleic acid anhydride, e.g. those known under the name "Lytron," Monsanto, and polycarbonate resins, e.g. the resins known by the name of "Lexan Grade" of General Electric, U.S.A. may be used.

The mixing ratio of charge transporting compound to binder may vary. Relatively certain limits are given, however, by the requirement for maximum photosensitivity, i.e., for the biggest possible portion of charge transporting compound, and for crystallization to be prevented, i.e., for the biggest possible portion of binder. A mixing ratio of about 1 : 1 parts by weight has proved preferable, but mixing ratios from about 3 : 1

to 1 : 4 or above, depending on the particular case, are also suitable.

The conventional sensitizers to be used additionally may advantageously foster charge transport. Moreover, they may produce charge carriers in the transparent top layers. Suitable sensitizers are, for example, Rhodamine B extra, "Schultz, Farbstofftabellen" (dyestuff tables), 1st volume, 7th edition, 1931, No. 864, page 365, Brilliant Green, No. 760, page 314, Crystal Violet, No. 785, page 329, and Cryptocyanine, No. 927, page 397. In the same sense as act the sensitizers may also act added compounds which form charge transfer complexes with the charge transporting compound. Thus, it is possible to achieve another increase of the photosensitivity of the described double layers. The quantity of added sensitizer or of the compound forming the charge transfer complex is so determined that the resulting donor acceptor complex with its charge transfer band still is sufficiently transparent for the organic dyestuff layer below. Optimum concentration is at a molar donor/acceptor ratio of about 10 : 1 to about 100 : 1 and vice versa.

The addition of adhesives as binders to the charge transporting compounds already yields a good photosensitivity. In this case, low-molecular polyester resin, such as Adhesive 49000, Du Pont, has proved particularly suitable.

In the described manner, the top layers have the property to render possible a high charge with a small dark discharge. Whereas in all conventional sensitizations an increase of the photosensitivity is connected with an increase of the dark current, the arrangement of the invention can prevent this parallelity. The layers are thus usable in electrophotographic copying devices with low copying speeds and very small lamp energies as well as in those with high copying speeds and correspondingly high lamp energies.

The invention will now be described more in detail by reference to the following examples:

EXAMPLE 1

For the preparation of electrophotographic conductor layers, N,N'-bis-(3,5-dimethylphenyl)-perylene-3,4,9,10-tetracarboxylic acid diimide (Formula 8) (prepared in accordance with German Patent Specification No. 1,067,157) is deposited at a reduced pressure of 10^{-3} to 10^{-4} mm of mercury (pump type A1 of Pfeiffer, Wetzlar, Germany) and at a temperature of 390°C within 5 minutes on a $75\text{ }\mu\text{m}$ thick aluminum foil mounted at a distance of 18 cm. The resulting pigment layer is homogeneous, glossy, and covers the supporting material completely.

Solutions of the following composition are applied, separately, to the dyestuff layers thus produced:

- 4.0 g of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 of a melting point of $149^{\circ}\text{--}150^{\circ}\text{C}$, 4.0 g of a polyester resin, e.g. "Dynapol" L 206 (a product of Dynamit Nobel AG, Troisdorf, Germany) dissolved in 40 ml of tetrahydrofuran,
- the same components as in (a), + 0.4 g of 3,5-dinitrobenzoic acid (melting point 206°C)
- the same components as in (a), + 0.004 g of Brilliant Green (Color Index No. 42040).

The centrifuge is so adjusted that the resulting top layers have a thickness of about 8 to $12\text{ }\mu\text{m}$. The layers are superficially dried with a fan and then thoroughly

dried by heating them for about 5 minutes to 110°–120° C. Homogeneous, smooth layers are thus produced.

In order to measure its photo-sensitivity, each of the coated plates is charged to a negative potential by passing it three times through a charging apparatus (Type AG 56, of Kalle Aktiengesellschaft, Wiesbaden-Biebrich, Germany) adjusted to 7.5 kV. Subsequently, the layers are exposed to the light of a xenon lamp (Osram, type XBO 150). The intensity of illumination in the plane of measurement is approximately 300 lux.

Altitude of charge and curve of photo-induced light decay of the photoconductive layer are measured by means of an electrometer (type 610 B of Keithley Instruments, U.S.A.) through a transparent probe, using the method described by Arneth and Lorenz in "Reprographie 3," 199 (1963). The results are compiled in the table below.

The layers are characterized by stating their altitude of charge (V) and their half time $T \frac{1}{2}$, i.e., the time in which the charge drops to half its initial value ($T \frac{1}{2}$ — in msec).

For comparison, the photo-sensitivity of the respective top layer without a dyestuff layer is also measured (zero layer). For this purpose, the corresponding solution is whirl-coated in such a manner upon a 75 μ m thick aluminum plate that the resulting layer has a thickness of 10–12 μ m and the plate is then subjected to the same drying and measuring steps.

Layer or Double layer, respectively	Dyestuff	U_0 (V) Negative charge	$T \frac{1}{2}$ (msec)
0, a	—	1200	525
0, b	—	1350	1710
0, c	—	1400	280
1, a	8	1000	49
1, b	8	880	56
1, c	8	810	47

EXAMPLE 2

Two photoconductor layers are prepared as follows:

Homogeneous dyestuff layers are produced by vacuum depositing perylene-3,4,9,10-tetracarboxylic acid anhydride (Formula 1) (BASF, Ludwigshafen, Germany) under the following conditions:

Vacuum: 10^{-3} – 10^{-4} mm of mercury

Temperature: 380° C

Duration of vacuum deposition: 2 to 3 minutes

Supporting material: 100 μ m thick aluminum foil

a. The resulting intensively red-colored dyestuff layer is whirl-coated with a solution comprising 4.0 g of 2,4,7-trinitrofluorenone (melting point 175° C) and 4.0 g of a polyester resin (e.g. "Dynapol L 206") in 40 ml of tetrahydrofuran in such a manner that a top layer of a thickness of about 10 μ m is produced.

b. A solution comprising 4.0 g of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4, 4.0 g of a polyester resin (e.g. "Dynapol L 206") and 0.4 g of 3,5-dinitrobenzoic acid is whirl-coated on top of the vacuum deposited dyestuff layer. The thickness of the layer is likewise 10 μ m.

Layers containing N,N'-dimethyl-perylene-3,4,9,10-tetracarboxylic acid diimide (C.I. 71 130) (Formula 3) as the dyestuffs are coated in the same manner.

The double layers thus produced are dried during 5 minutes in a recirculated-air drier at 100° to 120° C.

The top layers have a homogeneous, glossy surface and are measured as described in Example 1, once with a positive charge, once with a negative charge. The results are as follows:

Double Layer	Dyestuff (Formula)	U_0 (V)		$T \frac{1}{2}$ (msec.)	
		negative charge	positive charge	negative charge	positive charge
a	1	1050	520	2100	46
b	1	900	420	48	150
a	3	880	880	2320	62
b	3	930	775	24	262

EXAMPLE 3

In order to test the electrophotographic properties, top layers of about 10 μ m thickness are applied to the dyestuff layers described below, by whirl-coating them with a solution comprising 4.0 g of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 and 4.0 g of a polyester resin (e.g. "Dynapol L 206") in 40 ml of tetrahydrofuran, followed by 5 minutes drying at 110° to 120° C.

At a reduced pressure of 10^{-3} to 10^{-4} mm of mercury the following dyestuffs are vacuum-deposited under the conditions stated in the table:

		Time(min)	Temp.(° C)
30	a) perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 2) (C.I. 71 129)	2	400
35	b) N,N'-Di-n-butyl-perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 4) — Belgium Patent Specification No. 662 395	2	330
40	c) N,N'-Di- β -hydroxyethyl-perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 5) — German Patent Specification No. 1 916 196	3	400
45	d) N,N'-Di-(4-ethoxyphenyl)-perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 7) — C.I. 71 145	4	380
50	e) N,N'-Di-(4-chlorophenyl)-perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 9) — C.I. 71 135	5	420
55	f) N,N'-Di-(3-chlorophenyl)-perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 10) — German Patent Specification No. 1 067 548	4	440
	g) N,N'-Di-pyrenyl-perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 11)	9	420
	h) N,N'-Di-(2-pyridyl)-perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 12) — German Patent Specification (DAS) No. 1 132 272	3	380

Dyestuff No. 11 (N,N'-dipyrenyl-perylene-3,4,9,10-tetracarboxylic acid-diimide) is prepared by heating 39.2 g of perylene-3,4,9,10-tetracarboxylic acid and 46 g of 3-aminopyrene for several hours at 180°–200° C in 400 g of quinoline and 3 cm³ of concentrated sulfuric acid. After the reaction mixture has cooled, the dyestuff is drawn off by cold suction filtration, washed with quinoline and methanol, and finally dried. The resulting dyestuff has a melting point above 315° C.

The organic photoconductor double layers are measured as described in Example 1. The results are listed in the following table:

Double Layer	Dyestuff Formula No.	U_o (V) negative charge	$T \frac{1}{2}$ (msec)
a	2	1175	41
b	4	1275	24
c	5	1650	46
d	7	760	69
e	9	720	48
f	10	1025	93
g	11	1125	89
h	12	875	12
Zero Layer	—	1200	525

In Example 3 h, the photosensitivity of the material is determined by means of another measuring instrument. The photoconductor layer is placed on a slowly rotating disk and passed through a charging apparatus (corona adjustment 7.0 kV, grid 1.5 kV) to the exposure station, where it is arrested. The photoconductor layer is then exposed to the light of a xenon lamp (Osram, type XBO 150), with a heat absorbing glass (type KG3, Schott & Gen., Mainz, Germany) and a neutral filter of 15 percent transparency being interposed. The intensity of illumination in the plane of measurement is $270 \mu\text{W}/\text{cm}^2$. The altitude of charge and the curve of photo-induced light decay of the photoconductive layer are measured by an oscillographic device over an electrometer (type 610 CR, Keithley Instruments, U.S.A.) and a transparent probe. Evaluation is as described in Example 1.

EXAMPLE 4

The dependence of the photo-sensitivity on the thickness of the top layer is demonstrated by the following example: N,N'-dimethyl-perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 3) is vacuum deposited in the manner described in Example 1 upon an aluminum coated polyester film, the conditions of vacuum deposition being:

Reduced pressure of 10^{-3} to 10^{-4} mm of mercury

Temperature of 380°C

Duration: 2 to 3 minutes.

By varying the speed of rotation of a plate whirler, layers of varying thicknesses, viz. (a) of a thickness from 5 to $7 \mu\text{m}$, and (b) of a thickness from 10 to $12 \mu\text{m}$, are then applied from an 18 percent solution of equal parts by weight of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 and a polyester resin (e.g. "Dynapol L 206") in tetrahydrofuran, the thickness of the layers being determined by a measuring instrument marketed by Carl Mahr, Esslingen, Germany. The photosensitivity is determined as described in Example 1.

Double Layer	U_o (V) negative charge	$T \frac{1}{2}$ (msec)
a	760	13-14
b	990	22

The dark decay of the two layers is measured by means of a dyntest apparatus marketed by ECE, Giesen, Germany. The following values result:

Double Layer	U_o (V)	ΔU_b (1 sec)	ΔU_d (2 sec)
a	1000	110	175
b	1580	50	110

EXAMPLE 5

A solution comprising 4 g of 2-phenyl-4-(2-chlorophenyl)-5-(4-diethylaminophenyl)-oxazole (German Patent Specification No. 1,120,875) and 4 g of a polyester resin (e.g. "Dynapol L 206") in 40 ml of tetrahydrofuran is whirlcoated onto a $100 \mu\text{m}$ thick aluminum foil carrying a dyestuff layer of N,N'-dimethylperylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 3). After drying, homogeneous, glossy top layers of a thickness, respectively, of about 5 and about $10 \mu\text{m}$ are obtained.

The photosensitivity of the double layers is measured by the method described in Example 3 h and compared with a zero layer (top layer, but no dyestuff layer).

Layer	Thickness	Dyestuff Formula No.	U_o (V) negative charge	$T \frac{1}{2}$ (msec)
Double Layer	$5 \mu\text{m}$	3	380	9
Double Layer	$10 \mu\text{m}$	3	750	16
Zero-Layer	$10 \mu\text{m}$	—	800	750

EXAMPLE 6

A solution consisting of high molecular weight poly-N-vinyl carbazole (PVCa) (e.g. "Luvican M 170," a product of BASF, Ludwigshafen, Germany), an acceptor compound, e.g. 2,4,7-trinitrofluorenone (TNF) in varying molar ratios, and about 10 percent of an adhesive agent (e.g. a polyester resin, such as "Adhesive 49000," a product of DuPont) is whirlcoated as a top layer upon a $100 \mu\text{m}$ thick aluminum foil coated with a layer of N,N'-dimethyl-perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 3) by vacuum deposition. Since donor-acceptor complexes of a drak red-brown color are produced when polyvinyl carbazole and 2,4,7-trinitrofluorenone are combined, it is obvious to use only a relatively small quantity of activated poly-N-vinyl carbazole. A particularly favorable mol ratio between PVCa and TNF is in the range from about 1:0.10 to 1:0.001. For the following examples, the ratios given in the table were selected and the photosensitivities of single and double layers were compared. The top layers have a thickness of approximately $6 \mu\text{m}$.

The photosensitivity was determined by the method described in Example 1 (xenon lamp, 300 lux in the plane of measurement).

Layer	PVCa/TNF molar ratio	U_o (V) negative charge	$T \frac{1}{2}$ (msec)
single layer of PVCa/TNF	1:0.1	650	70

-Continued

Layer	PVCa/TNF molar ratio	U_0 (V) negative charge	$T_{1/2}$ (msec)
single layer of PVCa/TNF	1 : 0.05	680	100
single layer of PVCa/TNF	1 : 0.01	640	255
double layer of N,N'-dime- thylperylimide + PVCa/TNF	1 : 0.1	600	44
do.	1 : 0.05	570	38
do.	1 : 0.01	700	45

EXAMPLE 7

A solution containing 2.0 g of a bromo-pyrene resin, prepared by condensing 3-bromopyrene [see Org. Synth., Vol. 48 (1968) page 30] with formaldehyde in glacial acetic acid, and 1.0 g of a polyester resin (e.g. "Dynapol L 206") is applied to a 100 μ m thick aluminum foil coated with a layer of N,N'-dimethylperylene-3,4,9,10-tetracarboxylic acid diimide (Formula 3). The dry top coating has a thickness of about 6 μ m.

When measuring the photosensitivity as described in Example 1, a half-time ($T_{1/2}$) of 23 msec results from a negative charge of 820 V.

EXAMPLE 8

A top layer consisting of equal parts by weight of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 and one of the binders listed below is applied to an aluminum foil with an N,N'-dimethyl-perylene-3,4,9,10-tetracarboxylic acid diimide layer vacuum-deposited thereon under the conditions stated in Example 4. The top layer in each case is about 10 μ m thick. The photosensitivity is measured by the method described in Example 3 h (270 μ W/cm²).

- | | |
|----|---|
| a) | polyester resin ("Dynapol L 206") (50 %)
negative charge of 875 V
half-time $T_{1/2}$ = 9 msec |
| b) | styrene/maleic acid anhydride copolymer
(e.g. "Lytron 820", 50 %), a product of
Monsanto, U.S.A.)
negative charge of 850 V
$T_{1/2}$ = 21 msec |
| c) | "Lytron 820" (40 %)
chlorinated diphenyl resin (e.g. "Chlophenarz" W,
10 %, a product of Bayer AG, Leverkusen, Germany)
negative charge of 790 V
$T_{1/2}$ = 12 msec |
| d) | three-dimensionally cross-linked phenyl-methyl-
siloxane (e.g. silicone resin "SR 182" (60 % in
toluene) a product of General Electric, U.S.A.)
negative charge of 825 V
$T_{1/2}$ = 9 msec |
| e) | polycarbonate resin (e.g. "Lexan Grade 141", a
product of General Electric, U.S.A.)
negative charge of 1050 V
$T_{1/2}$ = 12 msec |

EXAMPLE 9

Polyurethane lacquers, e.g. the "DD" lacquers developed by Bater AG, Leverkusen, Germany, have been found to be particularly suitable for the preparation of the top layers. A component containing hydroxyl groups ("Desmophen"-type) and an isocyanate component ("Desmodur"-type) form the base materials, and according to the manufacturer's statement (see

"Bayer-Produkte für die Lackindustrie," Vol. 2, "Desmodur/Desmophen") equivalent quantities of both components are employed. An aluminum foil with an N,N'-dimethylperylene-3,4,9,10-tetracarboxylic acid-diimide coating is used, which is provided with a top layer consisting of 1 part by weight of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 and 1 part of the "DD"-lacquer component. After having dried for 5 to 15 minutes at 120° C, the coated layers have a thickness of about 10 μ m. The following "DD"-components are employed:

- | | |
|----|---|
| a) | 2.0 g of "Desmophen 1100"
3.0 g of "Desmodur HL"
negative charge of 500 to 600 V
$T_{1/2}$ = 11 msec |
| b) | 3.0 g of "Desmophen 650"
2.4 g of "Desmodur N"
negative charge of 485 V
$T_{1/2}$ = 13.5 msec |
| c) | 1.5 g of "Desmophen 550 U"
3.3 g of "Desmodur L"
negative charge of 785 V
$T_{1/2}$ = 8 msec |

EXAMPLE 10

For the preparation of an organic intermediate layer (FIG. 2), a 1 percent solution of a polyamide resin (e.g. "Elvamide 8061," a product of DuPont, U.S.A.) in a 1 : 1 mixture of n-propanol and methanol is applied to a polyester film of a thickness of 75 μ m laminated to aluminum and then dried. The intermediate layer has a thickness of less than 1 μ m and a weight of about 0.2 to 0.3 g/m².

The thus precoated supporting material is then provided, by vacuum-deposition, with a layer of

- | | |
|----|--|
| a) | perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 2) and a layer of |
| b) | N,N'-dimethyl-perylene-3,4,9,10-tetracarboxylic acid diimide, respectively, (Formula 3). |

These dyestuff layers are then coated with top layers of about 10 μ m thickness which consist of 1 part by weight of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 and 1 part by weight of a polyester resin, e.g. "Dynapol L 206." The photosensitivity is measured by the method described in Example 3 h (xenon lamp, light intensity in the plane of measurement 750 μ W/cm²). The following values result:

Layer	U_0 (V) negative charge	$T_{1/2}$ (msec)	Dark Decay ΔU_0 after 2 sec
a)	1050	10	120
b)	750	8	70

The dark decay was determined by means of a dyntest-90 device (ECE Giessen, Germany) 2 seconds after the layers had been charged to saturation.

EXAMPLE 11

Oxide layers of 400 and 800 Å. thickness, respectively, are produced on a 100 μ m thick aluminum foil by anodic oxidation. A dyestuff layer of N,N'-dimethylperylene-3,4,9,10-tetracarboxylic acid diimide (Formula 3) is applied to these oxide layers by vacuum deposition, and the dyestuff layers are then coated with a solution of 1 part by weight of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 and 1 part by

weight of a polyester resin, (e.g. "Dynapol L 206") so that the resulting top layer has a thickness of approximately 10 μm . Measurement of the photo-sensitivity by the method described in Example 3 h yields the following values (xenon lamp, light-intensity 615 $\mu\text{W}/\text{cm}^2$):

Thickness of Al_2O_3 layer	U_0 (V) negative charge	$T \frac{1}{2}$ (msec)	Dark Decay ΔU_D after 2 sec
about 400 Å	870	16.5	60
about 800 Å	900	14	80

EXAMPLE 12

Dyestuff layers of N,N'-dimethylperylene-3,4,9,10-tetracarboxylic acid diimide (Formula 3) are produced in accordance with Example 4 and then coated on a centrifuge with a solution containing 81.4 percent of polyvinyl carbazole and 18.6 percent of an adhesive polyester resin (e.g. "Adhesive 49000") in tetrahydrofuran so that the resulting top layers have a thickness, respectively, of 6 to 10 μm after drying. The photosensitivity is determined as described in Example 1 (xenon lamp, light-intensity in the plane of measurement 300 lux):

Layer	Dyestuff	Thickness of Top Layer	U_0 (V) negative charge	$T \frac{1}{2}$ (msec)
Double Layer	3	6 μm	780	64
Double layer	3	10 μm	1150	150
Zero layer	—	10 μm	950	4200

EXAMPLE 13

75 μm thick polyester films are provided, by vacuum deposition, with aluminum, gold, copper, zinc, cadmium, indium, antimony, nickel and tin layers, respectively, and these layers are then coated, according to the conditions stated in Example 4, with a dyestuff layer consisting of N,N'-dimethylperylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 3).

The dyestuff layers are then coated with a top layer of about 10 μm thickness consisting of 1 part by weight of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 and 1 part by weight of a polyester resin, e.g. "Dynapol L 206," in tetrahydrofuran. The photosensitivity is measured as described in Example 3 h (xenon lamp, light intensity of 270 $\mu\text{W}/\text{cm}^2$):

Metal	U_0 (V) negative charge	$T \frac{1}{2}$ (msec)	Dark Decay ΔU_D after 2 sec
Al	860	9	200
Au	750	10	115
Cu	875	8	160
Zn	925	9	160
Cd	775	24	160
In	900	7	135
Sb	250	19	80
Ni	800	11	140
Sn	925	8	260

EXAMPLE 14

N,N'-dimethylperylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 3) is vacuum deposited as a dyestuff layer on an aluminum foil of 70 μm thickness in a high-vacuum deposition apparatus (type CVE-20 S, marketed by Bendix Vacuum division, Friedberg, Germany) at a pressure of approximately 5×10^{-5} mm of mercury. The distance between the heated vessel containing the dyestuff and the aluminum support is 15 cm; the temperature during the vacuum deposition process, measured on the surface of the dyestuff by means of a nickel/chromium thermoelement is between 250° and 290° C. The support is disposed on a drum which rotates at a speed of 20 revolutions per minute during the vacuum deposition process. By varying the duration of the vacuum deposition process, while maintaining the other coating conditions, dyestuff layers of different thicknesses are produced. The weight of the dyestuff layers is determined by weighing the material once with the layer and once after removal of the layer.

The dyestuff layers produced in this manner are then coated with a solution containing 1 part by weight of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 and 1 part by weight of a polyester resin (e.g. "Dynapol L 206") in tetrahydrofuran as the solvent.

The thickness of the top layers is about 10 μm thickness in a high-vacuum deposition apparatus (type CVE-20 S, marketed by Bendix Vacuum division, Friedberg, Germany) at a pressure of approximately 5×10^{-5} mm of mercury. The distance between the heated vessel containing the dyestuff and the aluminum support is 15 cm; the temperature during the vacuum deposition process, measured on the surface of the dyestuff by means of a nickel/chromium thermoelement is between 250° and 290° C. The support is disposed on a drum which rotates at a speed of 20 revolutions per minute during the vacuum deposition process. By varying the duration of the vacuum deposition process, while maintaining the other coating conditions, dyestuff layers of different thicknesses are produced. The weight of the dyestuff layers is determined by weighing the material once with the layer and once after removal of the layer.

The dyestuff layers produced in this manner are then coated with a solution containing 1 part by weight of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 and 1 part by weight of a polyester resin (e.g. "Dynapol L 206") in tetrahydrofuran as the solvent.

The thickness of the top layers is about 10 μm after drying (5 minutes at 120° C).

The photosensitivity is determined by means of a xenon lamp (light-intensity approximately 750 $\mu\text{W}/\text{cm}^2$).

The bond strength of the laminate consisting of aluminum support, dyestuff layer and top coating, is tested by crumpling the material.

Layer	Weight of dyestuff layer (g/m ²) (approx.)	Bond Strength	U_0 (V) negative charge	$T \frac{1}{2}$ (msec)	Dark Decay ΔU_D after 2 sec
1	0.01	good	850	12	80
2	0.02	good	925	10	90
3	0.03	good	975	8	95
4	0.05	good	1000	8	95

-Continued

Layer	Weight of dyestuff layer (g/m ²) (approx.)	Bond Strength	U _o (V) negative charge	T ½ (msec)	Dark Decay ΔU _D after 2 sec
5	0.08	good	900	8	90
6	0.10	good	950	7	100
7	0.13	good	950	7	130

The dark decay is measured by a dyntest-90 instrument of ECE, Giessen, Germany.

EXAMPLE 15

The photosensitivity of a double layer material may be determined, inter alia, by the position taken by the substituent in the phenyl group of N,N'-diphenyl-perylene-3,4,9,10-tetracarboxylic acid-diimide (Formula 6 of U.S. Pat. No. 3,340,264). Therefore, derivatives in which a methoxy group is in the p-, m- or o-position (compounds for Formulae 15, 16 and 17) are applied for 2 minutes by vacuum deposition to 100 μm thick aluminum foils at a temperature between 410° and 430° and a reduced pressure of 10⁻³ to 10⁻⁴ mm of mercury. The phenyl derivative itself can be coated by vacuum deposition during 2 minutes at 380° C.

A solution containing 1 part by weight of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 and 1 part by weight of a polyester resin, e.g. "Dynapol L 206," is applied to these dyestuff layers.

After drying, the coated layers had a thickness of about 10 μm.

The photosensitivity is determined as described in Example 3 (xenon lamp, light-intensity of 750 μW/cm²).

Substituent	Formula No.	U _o (V) negative charge	T ½ (msec)	Dark Decay ΔU _D after 2 sec
-phenyl	6	950	24	175
-p-methoxy-phenyl (C.I. 71 140)	15	475	56	260
-m-methoxy-phenyl	16	500	14	240
-o-methoxy-phenyl	17	525	700	170

EXAMPLE 16

Condensation products prepared as described in French Patent Specification No. 1,580,624 from perylene-3,4,9,10-tetracarboxylic acid anhydride and phenylhydrazine (Formula 13), or benzylamine (Formula 14), or p-nitroaniline (Formula 18), or p-dimethylaminoaniline (Formula 19) display a high degree of photosensitivity when arranged in a double layer material according to the invention.

The layers are vacuum deposited as described in Example 1.

Dyestuff layer	Duration of Vacuum Deposition (minutes)	Temperature during Vacuum Deposition
1) Formula 13	3	380° C

-Continued

Dyestuff layer	Duration of Vacuum Deposition (minutes)	Temperature during Vacuum Deposition
2) Formula 14	1.5	380° C
3) Formula 18	7	420-470° C
4) Formula 19	5	420-430° C

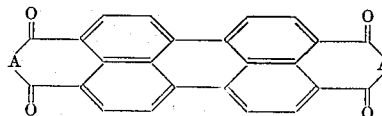
The dyestuff layers thus produced are coated with a solution of 1 part by weight of 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4 and 1 part by weight of a polyester resin, e.g. "Dynapol L 206." After drying, the layer has a thickness of approximately 10 μm. The photosensitivity is determined as described in Example 3 (xenon lamp, light-intensity of 750 μW/cm²).

No.	Dyestuff of the Double Layer	U _o (V) negative charge	T ½ (msec)	Dark Decay ΔU _D after 2 sec
1	Formula 13	820	31	140
2	Formula 14	1050	9	175
3	Formula 18	550	32	230
4	Formula 19	975	26	190

It will be obvious to those skilled in the art that many modifications may be made within the scope of the present invention without departing from the spirit thereof, and the invention includes all such modifications.

What is claimed is:

1. Electrophotographic recording material comprising an electroconductive support and a photoconductive double layer of organic materials thereon, said double layer being composed of a tightly packed, homogeneous, uniform, opaque, charge carrier producing dyestuff layer of a compound corresponding to the general formula



wherein

A is oxygen or =NR (imide nitrogen) in which

R is hydrogen, lower alkyl with one to four carbon atoms, an aryl group, an N-substituted heterocyclic group, or —NHR', with

R' being either a phenyl group or benzoyl, and a transparent top layer of insulating materials containing at least one charge transporting compound, in which the transparent top layer is composed of a mixture of a binder with a charge-transporting, monomeric heterocyclic compound having an extended π-electron system which is substituted by at least one dialkylamino group or by two alkoxy groups, or with a condensation product of 3-bromopyrene and formaldehyde.

2. Electrophotographic material according to claim 1, in which the heterocyclic compound is selected from

the group consisting of oxazoles, oxadiazoles, triazoles, imidazoles and pyrazoles.

3. Electrophotographic material according to claim 1, in which the heterocyclic compound is an oxadiazole.

4. Electrophotographic material according to claim 1, in which the heterocyclic compound is 2,5-bis-(4-diethylaminophenyl)-oxadiazole-1,3,4.

5. Electrophotographic material according to claim 1, in which the heterocyclic compound is an oxazole.

6. Electrophotographic material according to claim 1, in which the heterocyclic compound is 2-phenyl-4-(2-chlorophenyl)-5-(4-diethylaminophenyl)-oxazole.

7. Electrophotographic material according to claim 1, in which the dyestuff layer has a thickness from about 0.005 to about 2 μm and the transparent top layer has a thickness from about 5 to about 20 μm .

8. Electrophotographic material according to claim 1, in which the transparent top layer consists of a 1 : 1 mixture of the charge-transporting compound and the binder.

9. Electrophotographic material according to claim 1, in which the binder is selected from the group con-

sisting of polyesters or copolyesters, silicone resins, reactive resins of polyethers or polyesters and polyfunctional isocyanates, styrene/maleic anhydride copolymers, and polycarbonate resins.

10. Electrophotographic material according to claim 1, in which the binder is a copolymer of isophthalic acid or terephthalic acid and glycol.

11. Electrophotographic material according to claim 1, in which the binder is a styrene/maleic anhydride copolymer.

12. Electrophotographic material according to claim 1, in which the binder is a low molecular weight polyester resin.

13. Electrophotographic material according to claim 1, in which the transparent top layer additionally contains sensitizers and/or substances forming charge-transfer complexes.

14. Electrophotographic material according to claim 1, in which an insulating intermediate layer is disposed between the support and the photoconductive double layer.

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UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,871,882

Dated March 18, 1975

Inventor(s) Wolfgang Wiedemann

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

Column 12, line 55, "an" should read - - - and - - -.

Column 13, line 62, "Bater" should read - - - Bayer - - -.

Column 16, lines 27 through 49 should be deleted.

Signed and sealed this 27th day of May 1975.

(SEAL)

Attest:

RUTH C. MASON
Attesting Officer

C. MARSHALL DANN
Commissioner of Patents
and Trademarks