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ELECTROPHOTOGRAPHIC MEMBER

Wilhelm Neugebauer, Martha Tomanek, and Hans Behmenburg, all of Wiesbaden-Biebrich, Germany, assignors, by mesne assignments, to Azoplate Corporation, Murray Hill, N.J., a corporation of New Jersey
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This invention relates to photographic reproduction and more particularly to electrophotographic processes, namely processes in which an electrostatic latent image is produced by utilizing the property of photoconduction (i.e. a variable conductivity dependent on the intensity of illumination). The electrostatic latent image may be produced in a conventional exposure operation, for example by means of a lens-projected image or by contact-printing techniques, whereby a non-visible electrostatic charge pattern (the so-called electrostatic latent image) is created on a surface. In this pattern the charge density at any point is related to the intensity of illumination obtaining at that point during the exposure. The latent image may be developed—i.e. rendered visible—by means of an electroscopic powder, such as a colored synthetic resin powder, and the resulting visible image may be fixed by rendering the powder permanently adherent to a support on which the image is desired, for example in suitable cases by heating to soften or melt the powder particles and/or the surface of the image support, or by application of an electric field, or with volatile solvents.

In electrophotographic processes the electrostatic latent image is commonly formed on the surface of a photoconductive insulating layer carried on a support. For example, material comprising such support and photoconductive layer may be sensitized by applying a uniform surface charge to the free surface of the photoconductive layer, for example, by means of a corona discharge, which charge is retained owing to the substantial insulating character, i.e. the low conductivity, of the layer in the dark. On exposure, as described above, the photoconductive property of the layer causes an increase in conductivity in the illuminated areas to an extent which is directly proportional to the intensity of illumination. As a result, the surface charge in the illuminated areas leaks away while the charge in the unilluminated areas remains. This result constitutes the aforementioned charge pattern or electrostatic latent image.

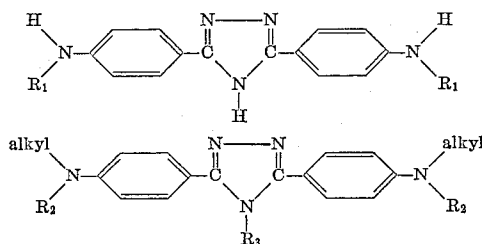
Electrophotographic processes have become of increasing importance in recent years, especially, for example, in connection with office duplicating, where offering as they do a wholly dry copying process, they are greatly preferred. Much interest has been aroused and much work has been done in this field, particularly in respect to obtaining suitable materials for forming a support as well as for forming a photoconductive insulating layer on the support. Although many efforts have been made to obtain materials capable of fulfilling modern requirements in respect to range of use, reliability, simplicity in handling, potential light sensitivity and keeping qualities, none has proved adequate. Among photoconductive substances used hitherto, selenium has been the most notable; however, it too does not meet the required standards.

The present invention has solved the above problems

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and provides a material for use in electrophotographic processes which is capable of being rendered light sensitive by applying an electrostatic charge thereto. It comprises a support carrying a photoconductive insulating layer, which layer includes at least one photoconductive substance consisting of a 2,5-bis-(p-amino-phenyl)-1,3,4-triazole compound.

According to preferred embodiments of the invention, the photoconductive substances are of general formulae



wherein

R₁ represents a hydrogen atom or an alkyl, cycloalkyl or acetyl radical,

R₂ represents an alkyl or acetyl radical, and

R₃ represents a hydrogen atom or an alkyl radical.

These general formulae include, for example, 2,5-bis[4'-amino-phenyl-(1')]-1,3,4-triazole, 2,5-bis-[4'-monoalkyl-amino-phenyl-(1')]-1,3,4-triazoles, 2,5-bis-[4'-monoacetyl-amino-phenyl-(1')]-1,3,4-triazoles, 2,5-bis-[4'-monoacetyl-amino-phenyl-(1')]-1,3,4-triazoles, and also 2,5-bis[4'-amino-phenyl-(1')]-1,3,4-triazoles in which the hydrogen atom in the 1-position is substituted by an alkyl radical.

The 2,5-bis-(p-aminophenyl)-1,3,4-triazole compounds have very good photoconductivity and are particularly suitable for the preparation of homogeneous layers having very long shelf-life. The compounds are colorless and fluoresce in daylight or in ultraviolet light.

The photoconductive substances are preferably applied to the support in the form of solutions in organic solvents, such as for example benzene, acetone, methylene chloride, or glycol monomethyl ether. Mixtures of two or more of the 1,3,4-triazole substances of this series in such solution may be used. Mixtures of solvents may also be used. Among the organic solvents are, for example, benzene, acetone, methylene chloride and glycol monomethyl-ether. Mixtures of two or more photoconductive substances and mixtures of solvents may also be used. It is also possible to employ the photoconductive substances in association with other organic photoconductive substances.

It has further been found advantageous in preparing the photoconductive insulating layers to use the photoconductive substance or substances in association with a resin, synthetic polymer or other organic colloid, for example:

(a) Natural and synthetic resins, e.g. balsam resins, phenol resins and other resins modified with colophony, coumarone resins and indene resins and the substances covered by the collective term "synthetic lacquer resins," which includes processed natural substances such as cellulose ether, see the *Kunststoffaschenbuch*

(Plastics Pocket Book), published by Saechtling-Zebrowski (11th edition, 1955, page 212 onwards);

(b) Polymers (including co-polymers) such as the polyvinyl chlorides, polyvinyl acetate, polyvinyl acetals, polyvinyl alcohols, polyvinyl ethers, polyacrylic and polymethacrylic esters, and polystyrene and isobutylene polymers;

(c) Polycondensates, e.g. polyesters, such as phthalate resins, alkyd resins, maleic acid resins, colophony esters of mixed higher alcohols, phenol-formaldehyde resins, particularly colophony-modified phenol-formaldehyde condensates, urea-formaldehyde resins, melamine-formaldehyde condensates, aldehyde resins, ketone resins of which particular mention is to be made of AW-2 resins of the firm Badische Anilin- und Sodafabrik, zylene-formaldehyde resins and polyamides; and

(d) Polyadducts, such as polyurethanes.

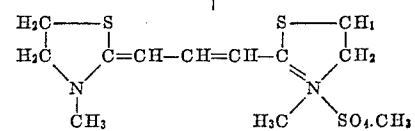
When the photoconductive substances are employed in association with organic colloids, the proportion of resin to photoconductive substance can be varied very greatly. The use of mixtures of approximately equal parts by weight of resin or other colloid and photoconductive substance has been found generally advantageous. When solutions of such mixtures of approximately equal parts by weight of resin and photoconductive substance are used, in most cases the said solutions give homogeneous trans-

solutions of polyamides in aqueous alcohols. Aqueous dispersions of other substances suitable for the pretreatment of the paper surface may also be used.

Solutions of the photoconductive substances in organic solvents as described above, with or without the added organic colloids aforementioned, can be applied to the supports in known manner (for example, by spraying, direct application, or whirl coating), and the supports thus coated, dried, so that an even photoconductive layer is formed thereon.

While the layers are, in themselves, non-light-sensitive, by applying a positive or negative electrostatic charge thereto, by means, for example, of a corona discharge, the layers are rendered light-sensitive and can be used with long-wave U.V. light of 3,600 to 4,000 A.U. in producing electrostatic latent images as described above. Very good images may be obtained by a short exposure under a master to a high-pressure lamp.

Although the layers when charged are but slightly sensitive to light in the visible spectrum, it has further been found that their spectral sensitivity can be extended into the visible part of the spectrum by the addition to the layers of sensitizers, preferably in the proportion one to three percent weight for weight of photoconductive substance. The most suitable sensitizers are dyestuff compounds, a number of which are given by way of example in the following table:

Dyestuff Group	Dyestuff Compound	Reference (Schultz' Farbstofftabellen, 7th ed., Vol. 1 (1931).)
Triarylmethane dyes.....	Brilliant Green..... Victoria Blue B..... Methyl Violet..... Crystal Violet..... Acid Violet 6B.....	No. 760 (p. 314). No. 822 (p. 347). No. 783 (p. 327). No. 785 (p. 329). No. 831 (p. 351).
Xanthene dyes: Rhodamines.....	Rhodamine B..... Rhodamine 6G..... Rhodamine G extra..... Sulphorhodamine B..... True acid Eosin G.....	No. 864 (p. 365). No. 866 (p. 366). No. 865 (p. 366). No. 863 (p. 364). No. 870 (p. 368).
Phthaleins.....	Eosin S..... Eosin A..... Erythrosin..... Phloxin..... Rose Bengal..... Fluorescein.....	No. 883 (p. 375). No. 881 (p. 374). No. 886 (p. 376). No. 890 (p. 378). No. 889 (p. 378). No. 880 (p. 373).
Thiazine dyes.....	Methylene Blue.....	No. 1038 (p. 440).
Acridine dyes.....	Acridine yellow..... Acridine orange..... Trypaflavine.....	No. 901 (p. 383). No. 908 (p. 387). No. 906 (p. 386).
Quinoline dyes.....	Pinacyanol..... Cryptocyanine.....	No. 924 (p. 396). No. 927 (p. 397).
Quinone dyestuffs: Anthraquinones.....	Alizarin..... Alizarin red S..... Quinizarine.....	No. 1141 (p. 499). No. 1145 (p. 502). No. 1148 (p. 504).
Cyanine dyes.....		

parent colourless layers on drying that can be considered as solid solutions.

The support may be of any material suitable for use in electrophotographic processes, for example, aluminium or other metal plates or foils, glass plates, paper sheets or webs, or plastic foil, especially foils made of electrically conductive resins. If paper is to be used as a support for the photoconductive layer, it is preferable that it shall have been pretreated against penetration by the coating solution, for example, with methyl-cellulose in aqueous solution; polyvinyl alcohol in aqueous solution; a solution in acetone and methyl-ethyl-ketone of a mixed polymer of acrylic acid methyl ester and acrylonitrile; or with

The layer-carrying supports may be utilized for the production of images by electrophotographic means, for example, as follows: When the photoconductive layer has been charged, by means of, for example, a corona discharge with a charging apparatus maintained at 6000 volts, the thus sensitized layer is exposed to light under a master or by episcopic or diascopic projection and is then dusted over in known manner with a suitable developing agent such as a resin powder colored with carbon black. We prefer a resin developer obtained by fusing 30 parts by weight of polystyrene (K-Wert 55), 30 parts by weight of a maleic acid resin modified with rosin and sold under the registered trademark "Beckacite K-105," and 30 parts

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by weight or carbon black, and subsequently finely grinding the fused mass. The image that now becomes visible can easily be wiped off, and therefore needs to be fixed; it can, for example, be heated briefly to approximately 120° C. by means of an infrared radiator. The temperature need not be as high as this if the heat treatment is carried out in the presence of vapors of solvents such as trichloroethylene, carbon tetrachloride or ethyl alcohol. The powdered image can also be fixed by means of steam. From positive masters, positive images of good contrast are produced.

After being fixed, these electrophotographic images can be converted into printing plates; the support, e.g. the paper or plastic foil, is wiped over with a solvent for the photoconductive layer, e.g. alcohol, or acetic acid and then rinsed with water and rubbed in with greasy ink in known manner. In this way positive printing plates are obtained which can be set up in an offset machine and used for printing. They give very long runs.

If transparent supports are used, the electrophotographic images can also be used as masters for the production of further copies of any sort of light-sensitive sheets. In this respect, the photoconductive compounds to be used, as provided by the invention, are superior to substances used hitherto, such as selenium or zinc oxide, inasmuch as the latter give cloudy layers because solid solutions cannot be produced with such materials and only suspensions are possible.

Where translucent supports are used for photoconductive layers such as are provided by the invention, reflex images can also be produced. The possibility of a reflex copy is also an advantage over the known art.

Moreover, the photoconductive layers prepared as provided in the invention have a further important advantage in that they can be charged positively as well as negatively. With positive charging the images are particularly good while there is negligible evolution of ozone, which is injurious to health and is very considerable with negative charging.

The 2,5-bis-[4'-aminophenyl-(1')]-1,3,4-triazole compounds corresponding to the aforementioned general formulae may generally be prepared as follows: The corresponding 2,5-bis-[4'-amino-phenyl-(1')]-1,3,4-oxadiazole compound is submitted to prolonged heating with formamide in the proportion of about 100 g. of formamide to 10 g. of the oxadiazole compound to temperatures of 180 to 190° C.; the reaction mixture is then poured into water, whereupon a precipitate is formed, which is separated by filtration from the mother liquor. The yield of the triazole compound is almost quantitative.

The 1,3,4-oxadiazole compounds used as starting material as described above in preparing the 1,3,4-triazole compounds may themselves be prepared in manner known per se from 2 mol. of a corresponding p-amino-benzene-carboxylic acid and 1 mol. of a hydrazine salt, by means of prolonged heating in the presence of a dehydrating agent. The preparation of the respective oxadiazole compounds has been set forth in French Patent No. 1,080,107.

If the oxadiazoles which are used as starting materials contain non-substituted hydrogen atoms in the two amino groups, formylation takes place at these points. If the N-formylamino-phenyl-1,3,4-triazoles so obtained are boiled with 10% hydrochloric acid, hydrolysis of the formyl groups occurs and the required 2,5-bis(amino-phenyl)-1,3,4-triazoles with primary or secondary amino groups can be obtained by neutralizing the hydrochloride solution with sodium hydroxide.

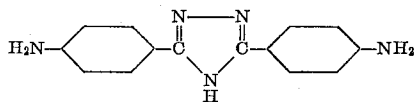
To substitute the hydrogen atom in the 1-position of the 1,3,4-triazole compounds, by an alkyl radical, the starting triazole may be dissolved in an aqueous alcoholic solution of caustic alkali and submitted to the action of the corresponding dialkyl sulfate.

Examples of 1,3,4-triazole compounds which may be

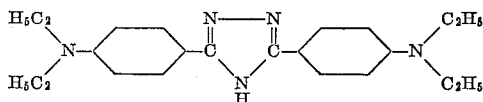
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used according to the invention as photoconductive substances are as follows:

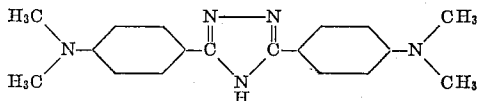
Formula 1



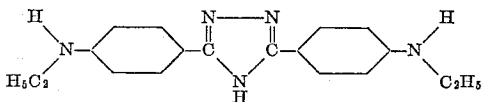
Formula 2



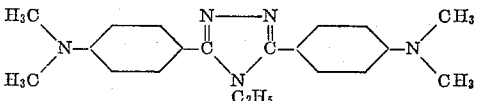
Formula 3



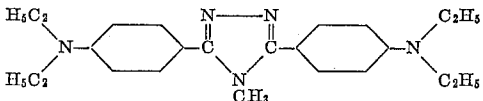
Formula 4



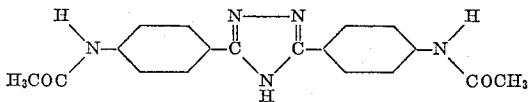
Formula 5



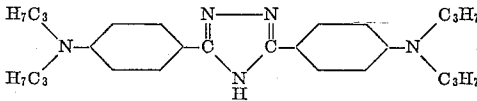
Formula 6



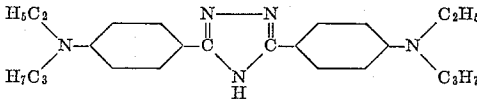
Formula 7



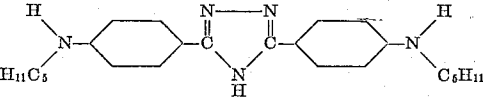
Formula 8



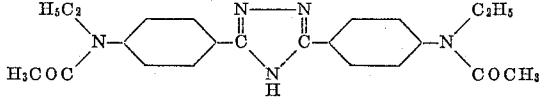
Formula 9



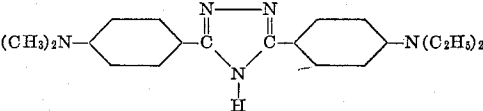
Formula 10



Formula 11



Formula 12



2,5-bis-[4'-amino-phenyl-(1')]-1,3,4-triazole corresponding to Formula 1 (melting point 252-254° C.) may be obtained by heating 10 g. of 2,5-bis-[4'-amino-phenyl-(1')]-1,3,4-oxadiazole with 100 g. of formamide for 8 hours at 180-190° C. The reaction product is poured into water and the resulting precipitate, a formylamino

compound, is separated by suction from the mother liquor, and then boiled for one hour with 10% hydrochloric acid. The formyl groups are thereby hydrolyzed. The acid solution is neutralized with sodium hydroxide to produce the amino compound. The amino compound is purified by recrystallization from 80% ethyl alcohol.

2,5 - bis - [4' - N,N - diethylamino - phenyl-(1')]-1,3,4-triazole corresponding to Formula 2 (melting point 198-200° C.) may be prepared from 2,5-bis-[4'-diethylamino-phenyl-(1')]-1,3,4-oxadiazole, by heating for 12 hours at 180-190° C. with ten times its weight of formamide. The reaction product is poured into water to form a precipitate which is dried and purified by recrystallization from a benzene/petroleum ether mixture.

2,5 - bis - [4' - N,N - dimethylamino - phenyl - (1')]-1,3,4-triazole corresponding to Formula 3 (melting point 255-256° C.) can be prepared in analogous manner to the compound corresponding to Formula 2, by boiling 2,5-bis - [4' - N,N - dimethylamino - phenyl - (1')]-1,3,4-oxadiazole, for 10 hours with ten times its weight of formamide. The compound is purified by recrystallization from 96% ethyl alcohol.

2,5 - bis - [4' - N - ethylamino - phenyl - (1')]-1,3,4-triazole corresponding to Formula 4 (melting point 168-170° C.) may be prepared from 2,5-bis-[4'-N-ethylamino-phenyl-(1')]-1,3,4-oxadiazole, by heating at 180-190° C. with ten times its amount by weight of formamide. The compound thus produced is 2,5-bis-[4'-(N-formyl-ethylamino)-phenyl-(1')]-1,3,4-triazole. Two hours' boiling of this reaction product boiled for two hours with 10% hydrochloric acid to hydrolyze the formyl groups, and the compound corresponding to Formula 4 is precipitated from the hydrochloride solution by means of sodium hydroxide.

1 - ethyl - 2,5 - bis - [4' - N,N - dimethylamino - phenyl-(1')]-1,3,4-triazole corresponding to Formula 5 (melting point 131° C.) can be prepared by dissolving 30 g. of the compound corresponding to Formula 3 in aqueous alcoholic potassium hydroxide and reacting the resulting solution with 20 g. of diethyl sulfate for two hours at 60° C. When the reaction is complete, the reaction mixture is diluted with water. The precipitate formed is separated by suction from the mother liquor, washed in water and recrystallized from 96% ethyl alcohol.

1 - methyl - 2,5 - bis - [4' - N,N - diethylamino - phenyl-(1')]-1,3,4-triazole corresponding to Formula 6 (melting point 132-133° C.) can be prepared in analogous manner to the compound corresponding to Formula 5, by methylating 36 g. of the triazole compound corresponding to Formula 2, at 60° C. with 15 g. of dimethyl sulfate in aqueous alcoholic potassium hydroxide.

2,5 - bis - [4' - (N - acetyl - amino) - phenyl - (1')]-1,3,4-triazole corresponding to Formula 7; this compound does not melt below 300° C. and can be prepared by boiling 25 g. of the compound corresponding to Formula 1 for three hours with 250 g. of acetic anhydride. The reaction mixture is poured into water, the liquid is filtered off, and the residue 2,5-bis-[4'-(N-acetyl-amino)-phenyl-(1')]-1,3,4-triazole is washed with water and recrystallized from 96% ethyl alcohol.

2,5 - bis - [4' - N,N - di - n - propyl - amino - phenyl-(1')]-1,3,4-triazole corresponding to Formula 8 (melting point 196-199° C.) can be prepared by boiling 2,5-bis-[4'-N,N-di-n-propyl-amino-phenyl-(1')]-1,3,4-oxadiazole, for six hours with ten times its weight of formamide. The reaction product is recrystallized from 96% ethyl alcohol.

2,5 - bis - [4' - N - ethyl - N - n - propyl - amino - phenyl-(1')]-1,3,4-triazole corresponding to Formula 9 (melting point 119-121° C.) can be prepared by boiling 2,5-bis-[4' - N - ethyl - N - n - propyl - amino - phenyl - (1')]-1,3,4-oxadiazole for ten hours with eight times its weight of formamide. The reaction product obtained is recrystallized from 70% ethyl alcohol.

2,5 - bis - [4' - (N - iso - amyl - amino) - phenyl - (1')]-1,3,4-triazole corresponding to Formula 10 (melting point 130-131° C.) can be prepared by heating 2,5-bis-[4'-(N-

iso-amyl-amino)-phenyl-(1')]-1,3,4-oxadiazole, for ten hours with ten times its weight of formamide. The reaction product is boiled for one hour under reflux with 10% hydrochloric acid and, after cooling, the reaction mixture containing hydrochloric acid is neutralized with sodium hydroxide. The precipitate that thereupon settles out is separated by suction from the mother liquor and recrystallized from 80% ethyl alcohol.

2,5 - bis - [4' - (N - ethyl - N - acetyl - amino) - phenyl-(1')]-1,3,4-triazole corresponding to Formula 11 (melting point 128-130° C.) can be prepared by boiling 30 g. of the compound corresponding to Formula 4, for three hours with 250 g. of acetic anhydride. The reaction mixture is poured into water, the liquid is neutralized with ammonia and the resulting colorless precipitate is recrystallized from 96% ethyl alcohol.

2,5 - bis - [4' - N - cyclohexyl - amino - phenyl - (1')]-1,3,4-triazole can be prepared by boiling 2,5-bis-[4'-N-cyclohexyl-amino-phenyl-(1')]-1,3,4-oxadiazole, for ten hours with ten times its weight of formamide. When the reaction is complete, the reaction mixture is poured into water, and the precipitate is filtered and dried. The formyl compound resulting from boiling with formamide is hydrolyzed by boiling for two hours with 10% hydrochloric acid. After cooling, the hydrochloride solution is neutralized with ammonia and the N-cyclohexylamino compound precipitates out. It is recrystallized from 96% ethyl alcohol.

Embodiments of the invention by way of example and methods of producing the same will now be set forth in the following specific examples:

Example 1

1 g. of the compound corresponding to Formula 6 and 1 g. of colophony formaldehyde resin (e.g. the product marketed under the registered trademark "Corepal" 140 by Farbenfabriken Bayer A.G., Leverkusen) are dissolved in 30 g. of benzene; the solution is applied to paper, the surface of which has been pretreated against the penetration of organic solvents, and the paper then dried.

With the paper thus coated a direct positive image can be produced by the electrophotographic process by providing the dry coating on the paper with a positive electric charge by means of a corona discharge exposed under a positive master to the light of a high-pressure mercury lamp, and then dusting over with resin powder colored with carbon black in manner known per se. A positive image is formed, which can be fixed by slight heating to produce a permanent image having excellent contrast. The background of the paper is brightened by the substance coated thereon.

If in the above-described process a transparent support, such as a cellulose acetate foil, is used instead of paper, an electrophotographic positive image produced thereon can be used as a master for further reproduction on light-sensitive sheets.

Example 2

1 g. of the compound corresponding to Formula 2 and 2 g. of coumarone resin (e.g. the coumarone resin 701/70 produced by the Gesellschaft für Teerverwertung, Duisburg-Meiderich) are dissolved in 30 g. of methylene chloride and the solution is applied to an aluminium foil. After evaporation of the solvent, a coating is left on the surface of the foil adhering firmly thereto.

With the thus-coated aluminium foil, images can be produced from masters by the electrophotographic process in known manner per se, and paper prints with good contrast can be obtained therefrom by the transfer process.

Example 3

0.5 g. of the compound corresponding to Formula 1, 1.5 g. of the compound corresponding to Formula 3 and 1 g. of the synthetic resin known as Kunstharz AW-2 (produced by Badische Anilin- und Soda-Fabrik A.G.,

Ludwigshafen am Rhein) are dissolved in 50 g. of benzene and the solution is applied to a paper prepared in accordance with U.S. Patents 2,534,650, 2,681,617 or 2,559,610. After evaporation of the solvent a coating is left which adheres firmly to the surface of the foil. In the electrophotographic process, direct positive images with good contrast are produced from masters with this coated paper. After being fixed by heating these images can be converted into a printing plate, if the paper is wiped over with 96% ethyl alcohol, rinsed well with water and rubbed with greasy ink in the presence of 1% phosphoric acid. Positive printing plates are obtained which can be set up in an offset machine and used for printing.

Example 4

0.5 g. of the compound corresponding to Formula 4, 1 g. of the compound corresponding to Formula 2 and 1 g. of resin-modified maleic acid resin (e.g. the product marketed under the trade name "Beckacite" K-125 by Reichhold Chemie A.G., Hamburg) are dissolved in 30 g. of benzene and applied to a superficially-roughened aluminium foil. After evaporation of the solvent, a coating is left which adheres firmly to the surface of the foil. Subsequent procedure is as described in Example 1; after the powder image has been fixed a positive image is obtained on the aluminium foil. This image can be converted into a printing plate by applying to the aluminium foil a 50% acetic acid solution, rinsing with water and then rubbing in 1% phosphoric acid and greasy ink. A positive printing plate is obtained, which can be set up in an offset machine and used for printing.

Example 5

1 g. of the compound corresponding to Formula 5, 1 g. of a phenol-formaldehyde resin modified with colophony (e.g. the resin produced by polycondensation and marketed under the registered trademark "Rhenophen," by Rheinpreussen G.m.b.H., Homberg (Ndrh.)) and 0.01 g. of Victoria blue B are dissolved in 30 g. of benzene. This solution is applied to a paper which is light-permeable, but not transparent, and the surface of which has been pretreated against the penetration of organic solvents, and the paper is then dried. The product "Rhenophen" is described on page 32 of the German publication "Lackrohstoff-Tabellen," by Erich Karsten (1955).

After the paper has been positively charged by means of a corona discharge, it is placed, coated side down, on a page of a book which may be printed on both sides, and exposed for two seconds to the light of a 100-watt incandescent lamp. Exposure takes place through the back of the light-permeable, but not transparent paper, a sheet of black paper being placed against the back of the page.

After exposure, the image formed on the light-permeable, but not transparent paper foil is developed by dusting over with a carbon black-resin powder, and a positive mirror-image with very good contrast is obtained. It can be transferred by pressure to a suitable paper or to a plastic foil to form a correct image. The correct image can also be produced by means of an electric field in manner known per se. If the paper or plastic foil is transparent, this image can be used as an intermediate original for further reproduction, for example, on photoprinting paper.

Example 6

The procedure as in Example 1 is followed except that the coating solution used consists of 1 g. of the compound corresponding to Formula 7, 0.03 g. of rhodamine 6G and 1 g. of ketone formaldehyde resin (e.g. that known as "Kunstharz EM," produced by polycondensation by Rheinpreussen G.m.b.H., Homberg (Ndrh.)), in 30 g. of glycol monomethyl ether. Positive images with good contrast are obtained in electrophotographic image production. By episcopic projection, images can be pro-

duced also from pages printed on both sides. Half and whole tones are reproduced with good contrast. The product "Kunstharz EM" is described on page 80 of "Lackrohstoff-Tabellen."

Example 7

The procedure as in Example 1 is followed except that the coating solution used consists of 0.5 g. of the compound corresponding to Formula 2 and 0.5 g. of the compound corresponding to Formula 6 in 30 g. of glycol monomethyl ether. Positive images with very good contrast are obtained in electrophotographic image production.

Example 8

2 g. of the compound corresponding to Formula 8 are dissolved in 30 cc. of glycol monomethyl ether and the solution applied to an aluminium foil. Evaporation of the solvent leaves a coating which adheres firmly to the surface of the foil. Subsequent procedure is as described in Example 1, whereby a positive image is obtained on the aluminium foil when the electrophotographically-produced image has been dusted over with resin and fixed by heat or treated with trichloro-ethylene vapor. The image-bearing aluminium foil can be converted into a printing plate, by wiping over the image side of the foil a 96% ethyl alcohol solution, rinsing with water and rubbing with greasy ink and 1% phosphoric acid. A positive printing plate is obtained which may be set up in an offset machine and used for printing.

Example 9

1 g. of the compound corresponding to Formula 9 and 1 g. of the compound corresponding to Formula 10 are dissolved in a mixture of 20 cc. of glycol monomethyl ether and 10 cc. of benzene and the solution is applied to transparent paper foil the surface of which has been pretreated against penetration of organic solvents, and the paper is then dried. By an electrophotographic process, images with good contrast are produced on this coated transparent paper, which images can be fixed by heat or by treatment with trichloroethylene vapor. The product can then be used as an intermediate original for further reproduction, e.g. for photoprinting on sensitized paper.

Example 10

The procedure as in Example 1 is followed except that the coating solution consists of 2 g. of the compound corresponding to Formula 2 in 30 cc. of glycol monomethyl ether. Positive images with very good contrast are obtained by an electrophotographic process as heretofore described.

Example 11

The procedure as in Example 1 is followed, except that the coating solution consists of 1 g. of the compound corresponding to Formula 11 and 1 g. of colophony in 30 g. of glycol monomethyl ether. Positive images are obtained by an electrophotographic process as heretofore described.

Example 12

1 g. of the compound corresponding to Formula 2, 1 g. of a resin modified with zinc (for example "Zinkresinat 357," a product prepared by Lehmann & Crebert, Mannheim-Rheinau), and 0.02 g. of acid violet 6BN are dissolved in 30 g. of benzene. The solution is applied to paper and dried. After being negatively charged by means of a corona discharge, the paper is exposed for ¼ second under a positive master to the light of a 100-watt incandescent lamp at a distance of about 15 cm. and dusted with a resin powder colored with carbon black as in Example 1. A positive image appears which is fixed by heat. The product "Zinkresinat 357" is described on page 25 of "Lackrohstoff-Tabellen."

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Example 13

The procedure as in Example 1 is followed, except that the coating solution is prepared with 30 g. of glycol monomethyl ether containing 1 g. of the compound corresponding to Formula 9 and 1 g. of partly polymerized natural resin (e.g. the natural resin about 40% polymerized marketed by the American firm Hercules Powder Company, Wilmington, under the trade name Hercules "Poly Pale" resin). Positive images are obtained. The product "Poly Pale" is described on page 24 of "Lackrohstoff-Tabellen."

Instead of Hercules "Poly Pale," a resin that is about 80% dimerized abietic acid (for example, that marketed by the same American firm under the trade name Hercules "Dymerex") can be used, or a hydrated resin, such as the resin produced as a mixture of dihydro and tetrahydro-abietic acid by the Hercules Powder Company and marketed under the trade name Hercules "Staybelite." The product "Dymerex" is described on page 24 of "Lackrohstoff-Tabellen."

Example 14

A paper base is coated with a solution in 100 g. of acetone of 10 g. of a product prepared by after-chlorination of polyvinyl chloride (such as the material marketed by Dynamit A. G., Rheinfelden, under the registered trademark "Rhenoflex"). The coated solution is dried. When dried, the coated paper is further coated with a solution of 1 g. of the compound corresponding to Formula 2 and 1 g. of ketone resin (such as that known as "Kunstharz AP," marketed by Chemische Werke Huls Aktiengesellschaft, Marl), in 30 g. of glycol monomethyl ether and again dried. The paper is then further processed as in Example 1 and gives an image with very good contrast even after one-second exposure under a master to the light of a 100-watt incandescent lamp at a distance of about 15 cm.

Example 15

1 g. of the compound corresponding to Formula 12 and 1 g. of a resin modified with zinc (e.g. that marketed by Robert Kraemer, Bremen, under the trade name "Erkazit Zinkharz 165") are dissolved in 30 g. of benzene. The solution is coated on to paper and dried. The coated paper is further processed as described in Example 1. A positive image is formed which is fixed by steam treatment. The product "Erkazit Zinkharz 165" is described on page 24 of Lackrohstoff-Tabellen."

Example 16

1 g. of the compound corresponding to Formula 2, 1 g. of ketone formaldehyde resin (e.g. the product marketed by the Chemische Werke Huls A.G., Marl, under the trade name "Kunstharz SK"), and 0.03 g. of rhodamine B are dissolved in 30 g. of glycol monomethyl ether. The solution is coated onto paper and dried. In the electrophotographic process this paper, which after coating is pink in color, can be used for the production of direct positive images. After the coated paper has been positively charged by means of a corona discharge, it is exposed under a film diapositive, e.g. to a 100 watt incandescent lamp at a distance of about 15 cm. for 1/2 second, and dusted with resin powder colored with carbon black in manner known per se. The positive image formed is fixed by slight heating. It is characterized by excellent contrast, the background having become practically colorless after the heating. If the rhodamine B is omitted from the coating solution, other conditions being the same, the positively charged paper has to be exposed under the master for 6 seconds. The product "Kunstharz SK" is described on page 80 of "Lackrohstoff-Tabellen."

Example 17

10 g. of after-chlorinated polyvinyl chloride, e.g. the product marketed by Dynamit Aktiengesellschaft vormals

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Alfred Nobel & Co., Troisdorf, Werk Rheinfelden, under the registered trademark "Rhenoflex," are dissolved in 100 g. of methyl-ethyl-ketone. To this solution are added first 10 g. of the compound corresponding to Formula 12 in 50 g. of toluene and then 0.011 g. of rhodamine B extra (Schultz, "Farbstofftabellen," 7th edition, 1st vol. (1931), No. 864) dissolved in 2 g. of methyl alcohol. With the solution thus obtained, which has a kinetic viscosity of about 20.8 centistokes (17.1 centipoises), paper is machine coated by means of a cast-coating device. The thickness of the coating obtained is about 6 microns. On this paper direct positive images are produced by the electrophotographic process in the manner described in Example 1. Light-sensitivity is very great. From double-sided printed sheets images very rich in contrast can be produced episcopically with an exposure of 1 second to a light of about 30 lux.

The compound corresponding to Formula 12 is produced when 20 g. of 2-[4'-diethyl-amino-phenyl-(1')]-5-[4''-dimethyl-amino-phenyl-(1'')] - 1,3,4 - oxadiazole are heated with 200 g. of formamide for 10 hours at 180-190° C. After the reaction mixture has been cooled, it is poured into water, and the reaction product is separated by suction and recrystallized from 80% ethyl alcohol.

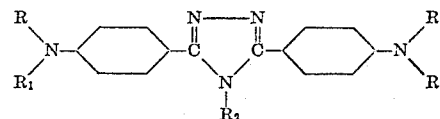
Example 18

10 g. of a product prepared by after-chlorination of polyvinyl chloride, e.g. the plastic marketed by Dynamit Aktiengesellschaft, Rheinfelden, under the registered trademark "Rhenoflex," are dissolved in 100 g. of methyl-ethyl-ketone. To this solution there are added first a solution of 10 g. of the compound corresponding to Formula 2 in 50 g. of toluene and then a solution of 0.004 g. of ethyl violet (Schultz, "Farbstofftabellen," 7th edition, 1st vol. (1931), No. 787) in 2 g. of methyl alcohol. With the solution thus obtained, which has a kinetic viscosity of approximately 20.8 centistokes (17.1 centipoises) a paper base is machine coated. The thickness of the coating should, for favorable results, be about 6 microns. After the coating has been dried the paper is negatively charged by means of a corona discharge and then a latent image is produced episcopically on this paper from a double-sided printed page. The paper is now treated on its coated side with a developer consisting of small glass balls and a finely divided mixture of resin and carbon black. The black colored resin adheres to the parts of the layer not struck by light during exposure and a positive image becomes visible; this is slightly heated and thereby made permanent (fixed). It shows good contrast.

Obviously many modifications and variations of the present invention are possible in the light of the above teachings. It is, therefore, to be understood that within the scope of the appended claims the invention may be practiced otherwise than as specifically described.

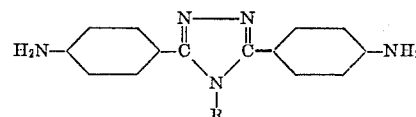
What is claimed is:

1. A 2,5-bis-(p-aminophenyl)-1,3,4-triazole.
2. A compound having the formula



in which R, R₁, R₃, and R₄ are selected from the group consisting of hydrogen, alkyl, cycloalkyl, and acyl radicals, and R₂ is selected from the group consisting of hydrogen and alkyl radicals.

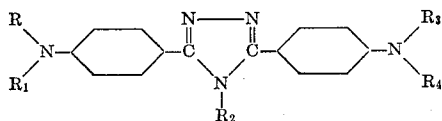
3. A compound having the formula



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in which R is selected from the group consisting of hydrogen and lower alkyl.

4. A compound having the formula



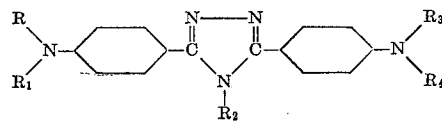
in which R, R₁, R₃, and R₄ are lower alkyl radicals, and R₂ is selected from the group consisting of hydrogen and lower alkyl radicals.

5. An electrophotographic material comprising a conductive support layer and a photoconductive insulating layer, the latter containing a 2,5-bis-(p-aminophenyl)-1,3,4-triazole.

6. An electrophotographic material according to claim 5 in which the photoconductive layer contains a dyestuff sensitizer.

7. An electrophotographic material according to claim 5 in which the photoconductive layer contains an organic colloid.

8. An electrophotographic material comprising a conductive support layer and a photoconductive insulating layer, the latter containing a compound having the formula

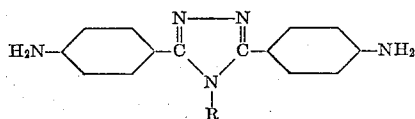


in which R, R₁, R₃, and R₄ are selected from the group consisting of hydrogen, alkyl, cycloalkyl, and acyl radicals, and R₂ is selected from the group consisting of hydrogen and alkyl radicals.

9. An electrophotographic material according to claim 8 in which the photoconductive layer contains a dyestuff sensitizer.

10. An electrophotographic material according to claim 4 in which the photoconductive layer contains an organic colloid.

11. An electrophotographic material comprising a conductive support layer and a photoconductive insulating layer, the latter containing a compound having the formula

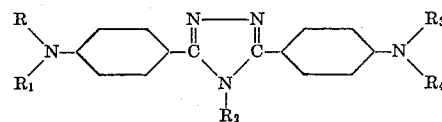


in which R is selected from the group consisting of hydrogen and lower alkyl.

12. An electrophotographic material according to claim 4 in which the photoconductive layer contains a dyestuff sensitizer.

13. An electrophotographic material according to claim 11 in which the photoconductive layer contains an organic colloid.

14. An electrophotographic material comprising a conductive support layer and a photoconductive insulating layer, the latter containing a compound having the formula



in which R, R₁, R₃, and R₄ are lower alkyl radicals, and R₂ is selected from the group consisting of hydrogen and lower alkyl radicals.

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15. An electrophotographic material according to claim 14 in which the photoconductive layer contains a dyestuff sensitizer.

16. An electrophotographic material according to claim 14 in which the photoconductive layer contains an organic colloid.

17. A process according to claim 37 in which the photoconductive layer contains a dyestuff sensitizer.

18. A process according to claim 37 in which the photoconductive layer contains an organic colloid.

19. A process according to claim 38 in which the photoconductive layer contains a dyestuff sensitizer.

20. A process according to claim 38 in which the photoconductive layer contains an organic colloid.

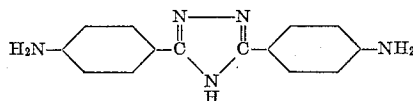
21. A process according to claim 39 in which the photoconductive layer contains a dyestuff sensitizer.

22. A process according to claim 39 in which the photoconductive layer contains an organic colloid.

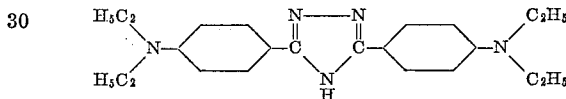
23. A process according to claim 40 in which the photoconductive layer contains a dyestuff sensitizer.

24. A process according to claim 40 in which the photoconductive layer contains an organic colloid.

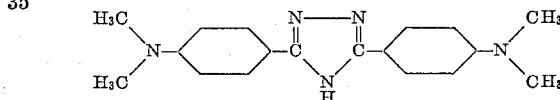
25. A compound having a formula



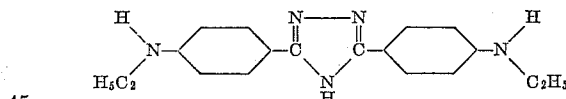
26. A compound having a formula



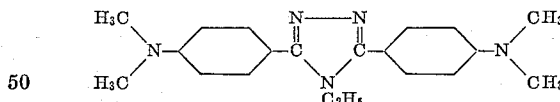
27. A compound having a formula



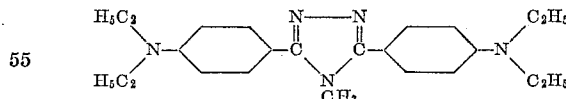
28. A compound having a formula



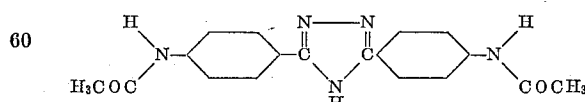
29. A compound having a formula



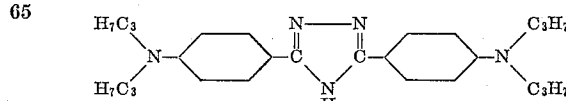
30. A compound having a formula



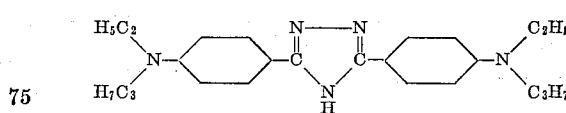
31. A compound having a formula



32. A compound having a formula

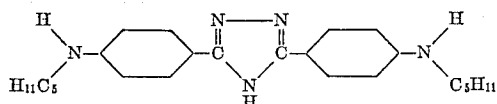


33. A compound having a formula:

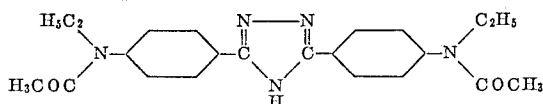


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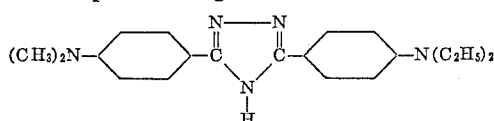
34. A compound having the formula:



35. A compound having the formula:

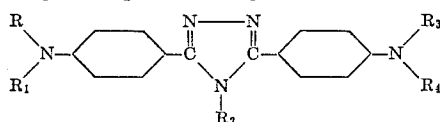


36. A compound having the formula:



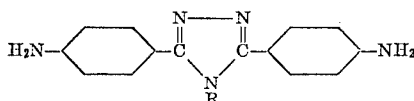
37. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a 2,5-bis-(p-aminophenyl)-1,3,4-triazole.

38. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula



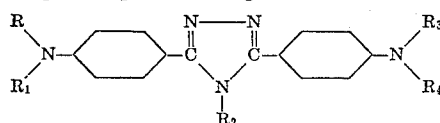
in which R, R₁, R₃ and R₄ are lower alkyl radicals, and R₂ is selected from the group consisting of hydrogen and lower alkyl radicals.

39. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula



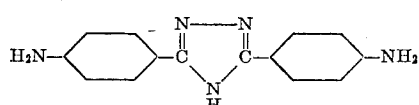
in which R is selected from the group consisting of hydrogen and lower alkyl radicals.

40. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula



in which R, R₁, R₃, and R₄ are lower alkyl radicals and R₂ is selected from the group consisting of hydrogen and lower alkyl radicals.

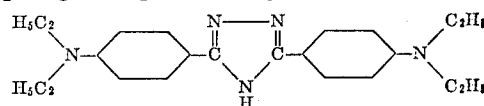
41. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula



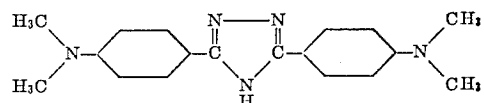
42. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and

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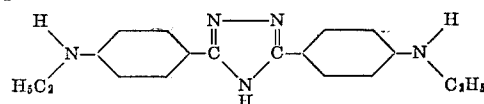
developing the resulting image, the photoconductive layer comprising a compound having the formula



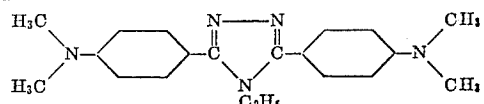
43. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula



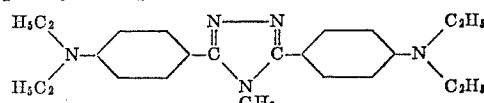
44. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula



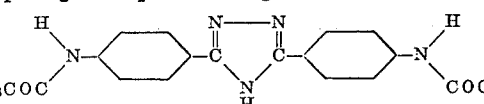
45. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula



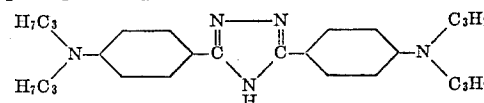
46. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula



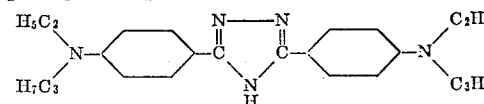
47. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula



48. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula

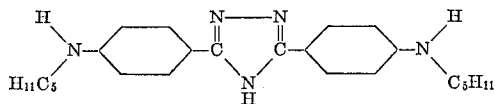


49. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula

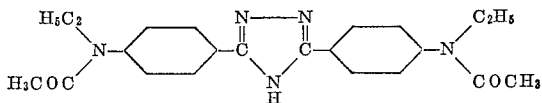


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50. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula



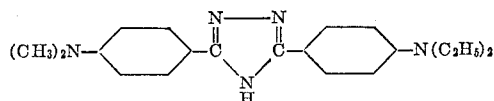
51. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and developing the resulting image, the photoconductive layer comprising a compound having the formula



52. A photographic reproduction process which comprises exposing an electrically charged, supported photoconductive insulating layer to light under a master and

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developing the resulting image, the photoconductive layer comprising a compound having the formula



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