(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 0 743 561 A2

(12) EUROPEAN PATENT APPLICATION

(43) Date of publication:
20.11.1996 Bulletin 1996/47

(51) Int. Cl.[6]: **G03G 5/06**

(21) Application number: **96107814.4**

(22) Date of filing: **15.05.1996**

(84) Designated Contracting States:
DE FR GB IT

(30) Priority: **17.05.1995 JP 141420/95**

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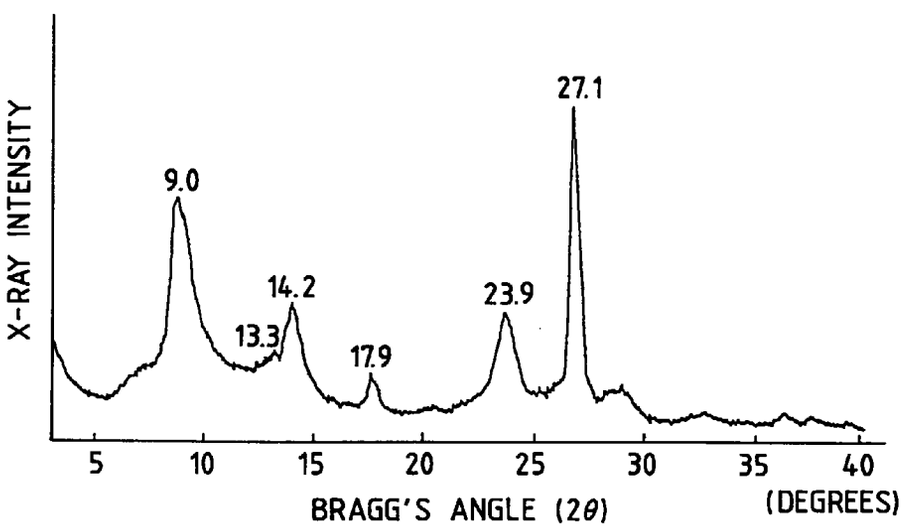
(54) Electrophotographic photosensitive member, process cartridge, and electrophotographic apparatus

(57) An electrophotographic photosensitive member is disclosed which has a photosensitive layer on a conductive support. The photosensitive layer contains an oxytitanium phthalocyanine and an azo pigment represented by the following Formula (1):

$$Ar\text{-}(N{=}N\text{-}Cp)_n.$$

Also, disclosed are a process cartridge and an electrophotographic apparatus using the electrophotographic photosensitive member.

FIG. 1



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Description

BACKGROUND OF THE INVENTION

Field of the Invention

This invention relates to an electrophotographic photosensitive member having a photosensitive layer containing a specific charge generating material, and also relates to a process cartridge, and an electrophotographic apparatus, having such an electrophotographic photosensitive member.

Related Background Art

Electrophotographic photosensitive members making use of organic photoconductive materials have been greatly improved in their sensitivity and durability (or running performance) as a result of the advancement of function-separated photosensitive members comprising and superposingly formed of a charge generation layer containing a charge-generating material and a charge transport layer containing a charge-transporting material, and have widely been put into practical use. In particular, in recent years, in place of conventional impact type printers, non-impact type printers to which electrophotography is applied are in wide use as terminal unit printers. These are chiefly laser beam printers having lasers as light sources. As the light sources, semiconductor lasers are used in view of the cost, the size of apparatus and so forth. Semiconductor lasers prevalingly used at present have an oscillation wavelength of as long as 790 plus-minus 20 nm, and hence electrophotographic photosensitive members having sufficient sensitivities in such a long-wavelength region have been developed.

As materials having sensitivities in such a long-wavelength region, metal-free phthalocyanines and metal phthalocyanines are known in the art, and have been put into practical use as charge-generating materials. In particular, among such phthalocyanine compounds, oxytitanium phthalocyanine has a very high sensitivity.

However, the electrophotographic photosensitive members making use of oxytitanium phthalocyanine are sought to have much better potential stability and much better photomemory (potential attenuation at portions irradiated by fluorescent lighting). Accordingly, it is proposed, e.g., to make the the charge generation layer thiner or to decrease the proportion of the oxytitanium phthalocyanine in the charge generation layer. Such measures, however, may cause a lowering of sensitivity.

For the purpose of obtaining a panchromatic sensitivity, it is proposed in Japanese Patent Application Laid-open No. 3-37656 to use oxytitanium phthalocyanine and a bisazo pigment in combination. Such a bisazo pigment, however, is little effective for the improvement in the above properties.

SUMMARY OF THE INVENTION

An object of the present invention is to provide an electrophotographic photosensitive member having a high sensitivity, a superior potential stability and superior photomemory characteristics when repeatedly used.

Another object of the present invention is to provide a process cartridge, and an electrophotographic apparatus, having such an electrophotographic photosensitive member.

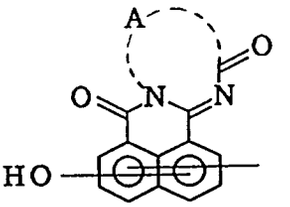
The present invention provides an electrophotographic photosensitive member comprising a conductive support and a photosensitive layer provided on the conductive support;

said photosensitive layer containing an oxytitanium phthalocyanine and an azo pigment represented by the following Formula (1).

Formula (1): $Ar\text{-}(N{=}N\text{-}Cp)_n$

wherein Ar represents a substituted or unsubstituted aromatic hydrocarbon group which may be bonded through a linking group, or a substituted or unsubstituted heterocyclic group which may be bonded through a linking group; Cp represents a coupler residual group having a phenolic hydroxyl group, at least one of said Cp representing a coupler residual group represented by the following Formula (2); and n represents an integer of 2 to 4.

Formula (2):



wherein A represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon ring or a divalent group of a substituted or unsubstituted heterocyclic ring containing a nitrogen atom in the ring.

The present invention also provides a process cartridge, and an electrophotographic apparatus, having the electrophotographic photosensitive member described above.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 shows an X-ray diffraction pattern of CuKα characteristics of I-type oxytitanium phthalocyanine.

Fig. 2 shows an X-ray diffraction pattern of CuKα characteristics of A-type oxytitanium phthalocyanine.

Fig. 3 shows an X-ray diffraction pattern of CuKα characteristics of B-type oxytitanium phthalocyanine.

Fig. 4 shows an X-ray diffraction pattern of CuKα characteristics of Y-type oxytitanium phthalocyanine.

Fig. 5 schematically illustrates the construction of an electrophotographic apparatus having a process cartridge having the electrophotographic photosensitive member of the present invention.

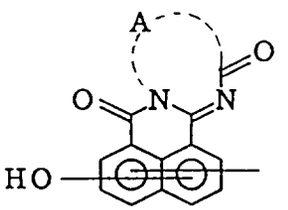
DESCRIPTION OF THE PREFERRED EMBODIMENTS

The electrophotographic photosensitive member of the present invention has a photosensitive layer on a conductive support, and the photosensitive layer contains an oxytitanium phthalocyanine and an azo pigment represented by the following Formula (1).

Formula (1): $Ar\{N=N-Cp)_n$

wherein Ar represents a substituted or unsubstituted aromatic hydrocarbon group which may be bonded through a linking group, or a substituted or unsubstituted heterocyclic group which may be bonded through a linking group; Cp represents a coupler residual group having a phenolic hydroxyl group, at least one of said Cp representing a coupler residual group represented by the following Formula (2); and n represents an integer of 2 to 4.

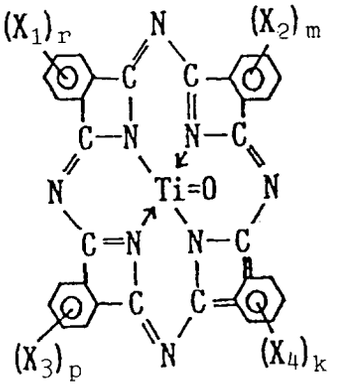
Formula (2):



wherein A represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon ring or a divalent group of a substituted or unsubstituted heterocyclic ring containing a nitrogen atom in the ring.

The present invention can be effective not only in that a superior potential stability and superior photomemory characteristics can be achieved but also in that, even though this specific azo pigment has no sensitivity around 800 nm at all, the sensitivity around 800 nm possessed by the oxytitanium phthalocyanine is enhanced by the chemical sensitizing action of the azo pigment.

The oxytitanium phthalocyanine used in the present invention has the following structure.



wherein X_1, X_2, X_3 and X_4 represents Cl or Br; and k, m, p and r are each an integer of 0 to 4.

The oxytitanium phthalocyanine may have any crystal form. It may preferably include the A type as disclosed in U.S. Patent No. 4,664,997, etc., the B type as disclosed in U.S. Patent No. 4,728,592, etc., the I type as disclosed in U.S. Patent No. 5,132,197, etc, and the Y type as disclosed in Japanese Patent Application Laid-open No. 64-17066. In particular, I type crystals are preferred. X-ray diffraction patterns of CuKα characteristics of oxytitanium phthalocyanines of the respective crystal forms are shown in Figs. 2, 3, 1 and 4, respectively. The oxytitanium phthalocyanine can be synthesized and can be made to have the above crystal forms by known methods disclosed in the above patent publications or by other methods.

As for the azo pigment represented by Formula (1), the group represented by Ar in the formula includes, for example, aromatic hydrocarbon rings such as benzene, naphthalene, fluorene, phenanthlene, anthracene and pyrene; heterocyclic rings such as furan, thiophene, pyridine, indole, benzothiazole, carbazole, acridone, dibenzothiophene, benzoxazole, oxadiazole and thiazole; and the above aromatic hydrocarbon rings or heterocyclic rings bonded directly or through an aromatic group or non-aromatic group, as exemplified by groups such as biphenyl, binaphthyl, diphenylamine, triphenylamine, N-methyldiphenylamine, fluorenone, phenanthrenequinone, anthraquinone, benzanthrone, terphenyl, diphenyloxadiazole, stilbene, distyrylbenzene, azobenzene, azoxybenzene, phenylbenzoxazole, diphenylmethane, diphenylsulfone, diphenyl ether, benzophenone, tetraphenyl-p-phenylenediamine, tetraphenylbenzidine, N-phenyl-2-pyridylamine and N,N-diphenyl-2-pyridylamine.

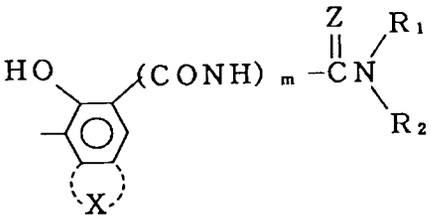
The substituent the Ar may have, includes alkyl groups such as methyl, ethyl, propyl and butyl; alkoxy groups such as methoxy, ethoxy and propoxy; halogen atoms such as fluorine, chlorine, iodine and bromine; alkylamino groups such as dimethylamino and diethylamino; a hydroxyl group; a nitro group; a cyano group; and halomethyl groups.

In Formula (2), the group represented by A includes, for example, groups such as o-phenylene, 2,3-naphthylene, 2,3-pyrazin-di-yl, 3,4-pyrazol-di-yl, 2,3-pyridin-di-yl, 4,5-pyridin-di-yl and 4,5-imidazol-di-yl.

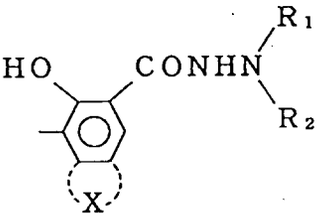
The substituent the A may have, includes alkyl groups such as methyl, ethyl, propyl and butyl; alkoxy groups such as methoxy, ethoxy and propoxy; halogen atoms such as fluorine, chlorine, iodine and bromine; a nitro group; a cyano group; and halomethyl groups. In particular, A may preferably be an unsubstituted o-phenylene.

In the group represented by Cp in Formula (1), the coupler residual group that may also be present other than the coupler residual group represented by Formula (2) includes, for example, groups represented by the following Formulas (3) to (8).

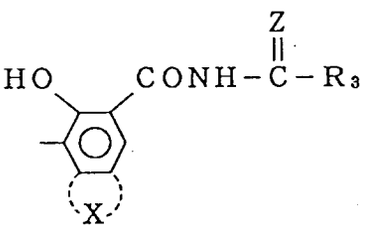
Formula (3):



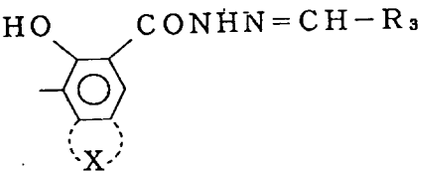
Formula (4):



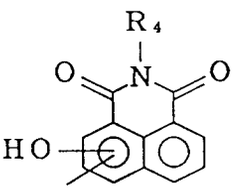
Formula (5):



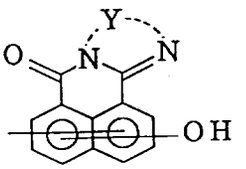
Formula (6):



Formula (7):



Formula (8):



X in Formulas (3), (4), (5) and (6) represents a group of atoms necessary to combine with the benzene ring to form a substituted or unsubstituted aromatic ring or a substituted or unsubstituted heterocyclic ring, where the aromatic ring and heterocyclic ring include, for example, a naphthalene ring, an anthracene ring, a carbazole ring, a benzcarbazole ring and a dibenzofuran ring. Y in Formula (8) represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon or a divalent group of a substituted or unsubstituted heterocyclic ring containing a nitrogen atom in the ring, specifically including groups such as o-phenylene, o-naphthylene, perinapthylene, 1,2-anthrylene, 3,4-pyrazol-di-yl, 2,3-pyridin-di-yl, 4,5-pyridin-di-yl, 6,7-indazol-di-yl and 6,7-quinolin-di-yl. R_1 and R2 in Formulas (3) and (4) each represent a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted aralkyl group or a substituted or unsubstituted heterocyclic group. R_1 and R_2 may also combine each other to form a substituted or unsubstituted cyclic amino group through the nitrogen atom. R_3 in Formulas (5) and (6) represents a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted aralkyl group or a substituted or unsubstituted heterocyclic group. R_4 in Formula (7) represents a hydrogen atom, a substituted or unsubstituted alkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted aralkyl group or a substituted or unsubstituted heterocyclic group. The above alkyl group includes groups such as methyl, ethyl and propyl; the aryl group, groups such as phenyl, naphthyl and anthryl; the aralkyl group, groups such as benzyl and phenethyl; the heterocyclic group, groups such as pyridyl, thienyl, carbazolyl, benzimidazolyl and benzothiazolyl; and the cyclic amino group containing a nitrogen atom in the ring, groups such as pyrrole, pyrroline, pyrrolidine, pyrrolidone, indole, indoline, carbazole, imidazole, pyrazole, pyrazoline, oxazine and phenoxazine. The substituent includes halogen atoms such as fluorine, chlorine, iodine and bromine; alkyl groups such as methyl, ethyl and propyl; alkoxyl groups such as methoxy and ethoxy; alkylamino groups such as dimethylamino and diethylamino; a phenylcarbamoyl group; a nitro group; a cyano group; and halomethyl groups such as trifluoromethyl. Z in Formulas (3) and (5) represents an oxygen atom or a sulfur atom, and m represents 0 or 1.

In Formula (2), n represents an integer of 2 to 4, and may preferably be 2 or 3.

Preferable examples of the azo pigment represented by Formula (1) of the present invention are shown below. The azo pigment used in the present invention is by no means limited to these. In the following, the specific structures of the exemplary pigments are each set forth by first giving a basic formula and subsequently its variations only.

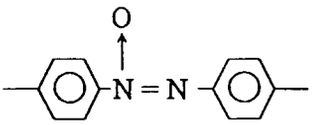
The coupler having the coupler residual group represented by Formula (2) is, when synthesized as described later, usually obtained in the form of a mixture of isomers. Accordingly, its examples are also given in the form of mixtures. For example, in pigment 1, what is meant by

"Basic formula 1:

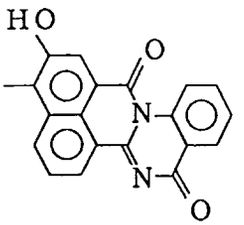
$$Cp_1\text{-}N{=}N\text{-}Ar\text{-}N{=}N\text{-}Cp_2$$

Pigment 1

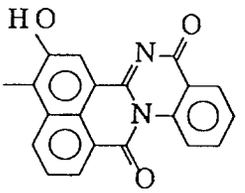
Ar:



Cp_1 and Cp_2:

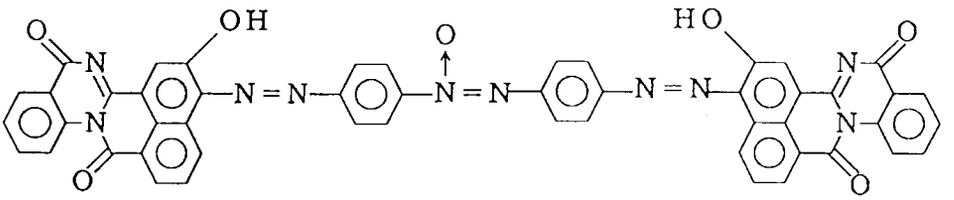


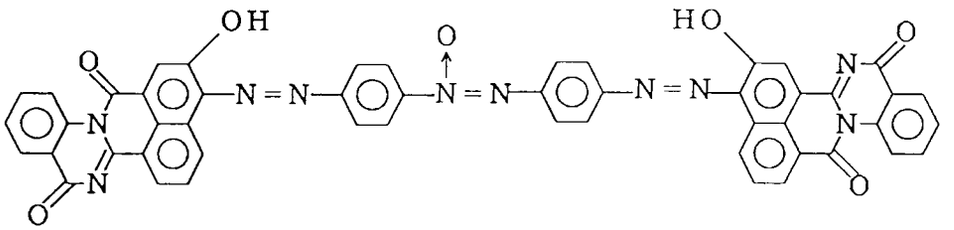
and

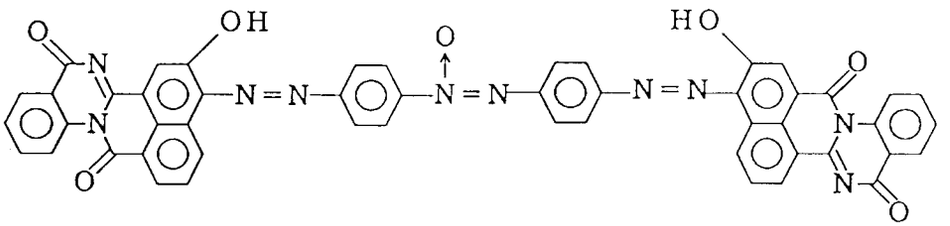


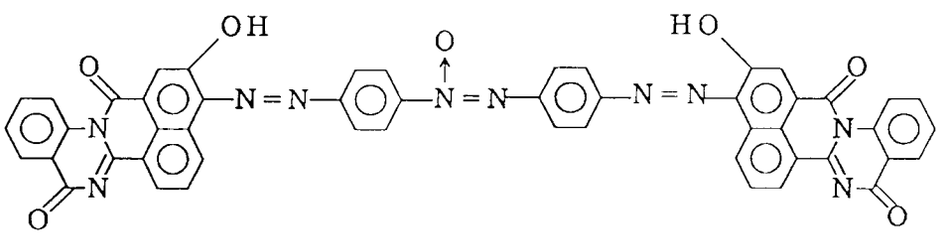
"

is that pigment 1 is a mixture of the compounds shown below.







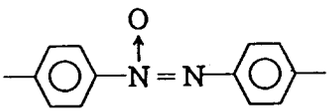


Basic Formula 1:

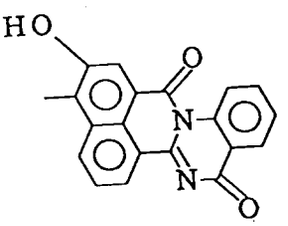
$$Cp_1\text{-}N{=}N\text{-}Ar\text{-}N{=}N\text{-}Cp_2$$

Exemplary Pigment 1

Ar:



Cp_1 and Cp_2:



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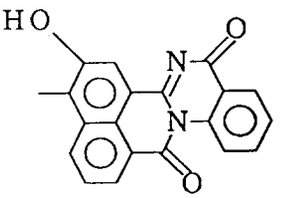
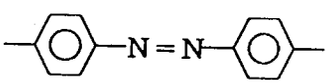
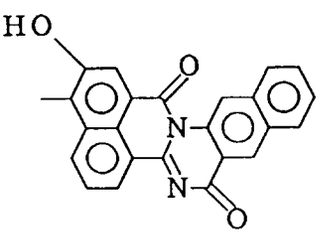


Exemplary Pigment 2

Ar:



Cp_1 and Cp_2:



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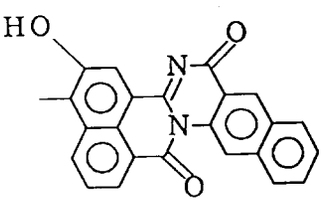
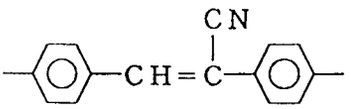
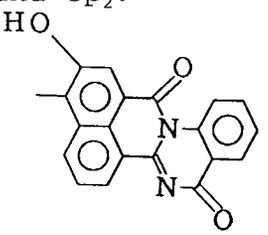


Exemplary Pigment 3

Ar:



Cp_1 and Cp_2:



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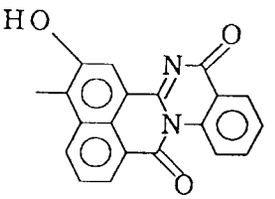
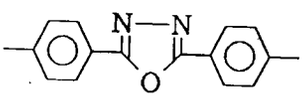
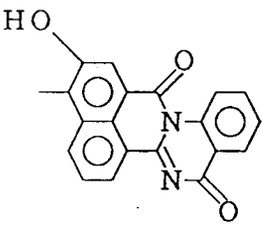


Exemplary Pigment 4

Ar:



Cp_1 and Cp_2:



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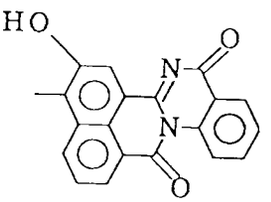
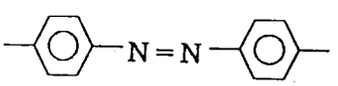
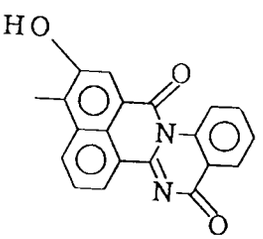


Exemplary Pigment 5

Ar:



Cp_1 and Cp_2:



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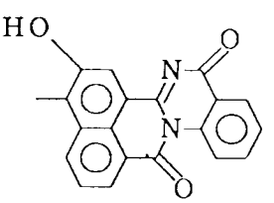
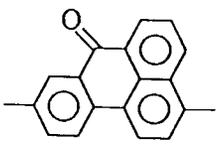
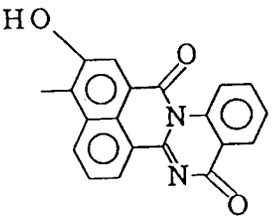


Exemplary Pigment 6

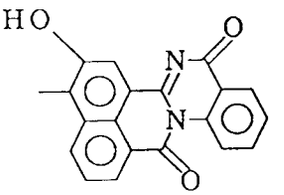
Ar:



Cp_1 and Cp_2:

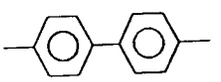


and

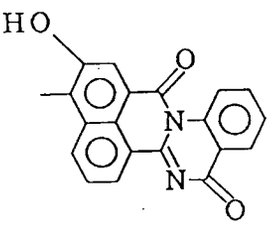


Exemplary Pigment 7

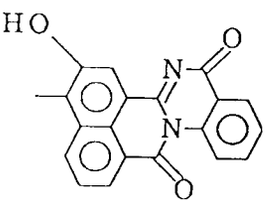
Ar:



Cp_1 and Cp_2:

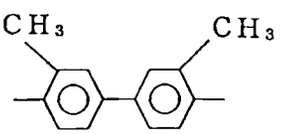


and

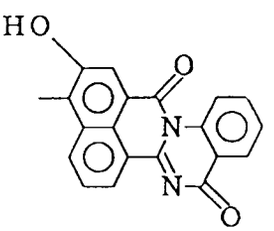


Exemplary Pigment 8

Ar:



Cp_1 and Cp_2:



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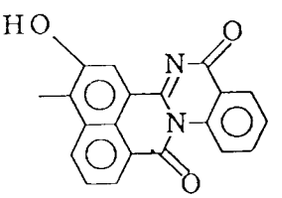
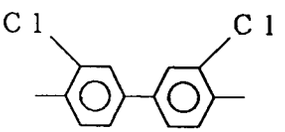
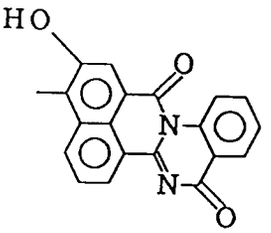


Exemplary Pigment 9

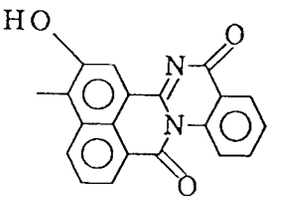
Ar:



Cp_1 and Cp_2:

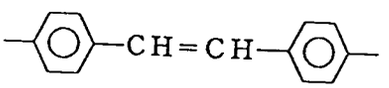


and

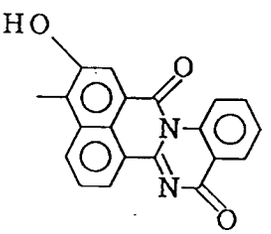


Exemplary Pigment 10

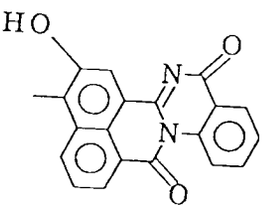
Ar:



Cp_1 and Cp_2:

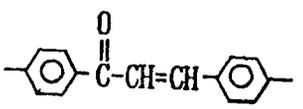


and

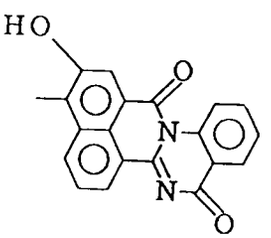


Exemplary Pigment 11

Ar:



Cp_1 and Cp_2:



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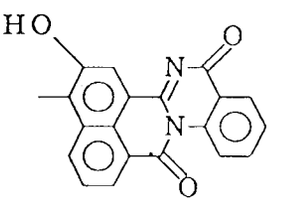
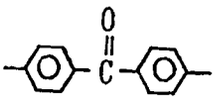
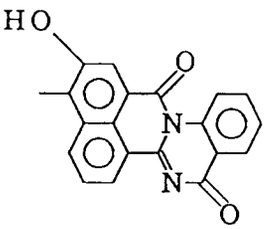


Exemplary Pigment 12

Ar:



Cp_1 and Cp_2:



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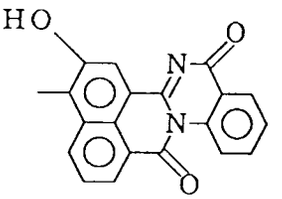
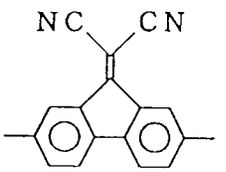
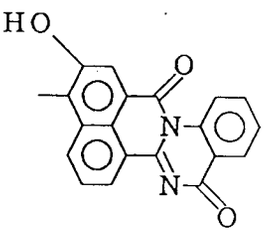


Exemplary Pigment 13

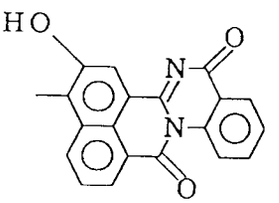
Ar:



Cp_1 and Cp_2:



and



Exemplary Pigment 14

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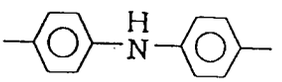
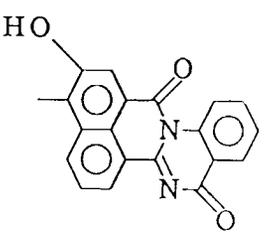


Cp_1 and Cp_2:



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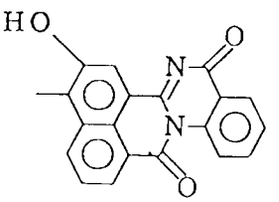
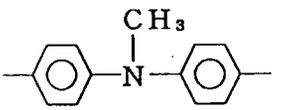
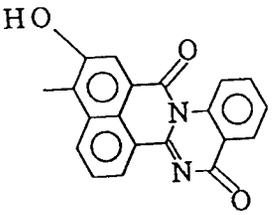


Exemplary Pigment 15

Ar:



Cp_1 and Cp_2:



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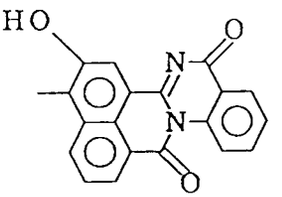
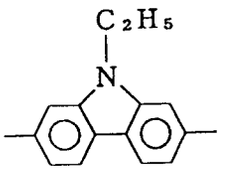
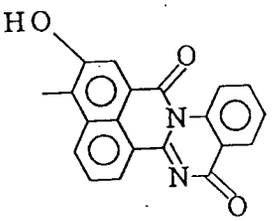


Exemplary Pigment 16

Ar:



Cp_1 and Cp_2:



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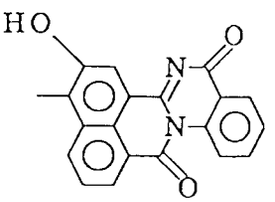
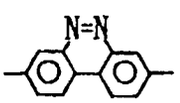
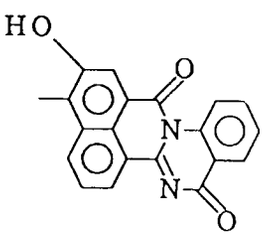


Exemplary Pigment 17

Ar:



Cp_1 and Cp_2:



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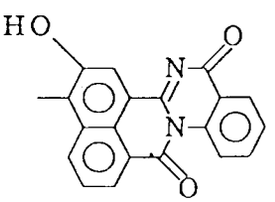
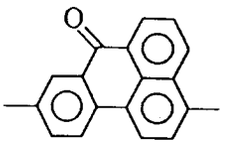
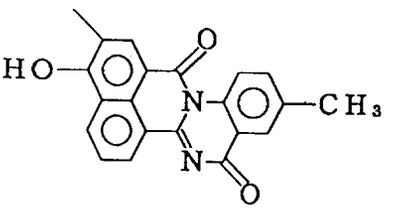


Exemplary Pigment 18

Ar:



Cp_1 and Cp_2:



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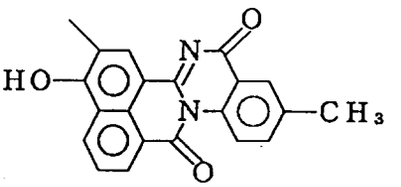
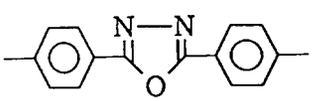
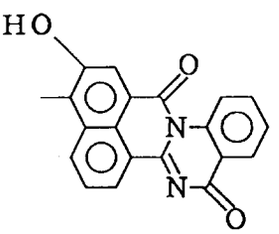


Exemplary Pigment 19

Ar:



Cp_1:



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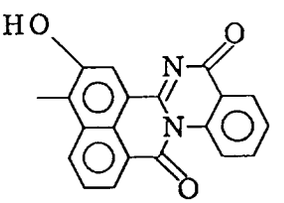
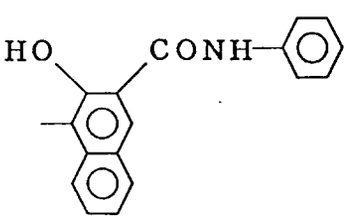
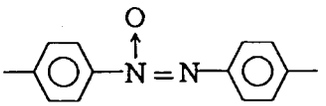


Cp_2:

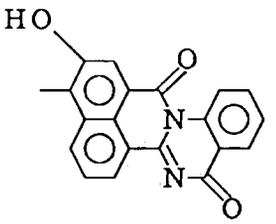


Exemplary Pigment 20

Ar:



Cp_1:



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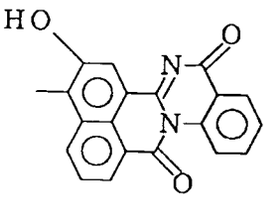
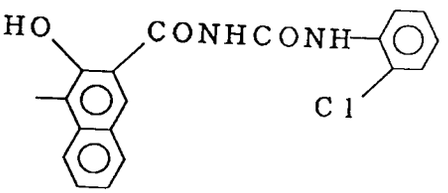
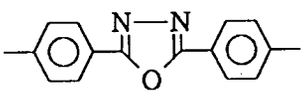


Cp_2:

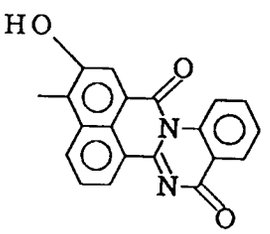


Exemplary Pigment 21

Ar:



Cp_1:



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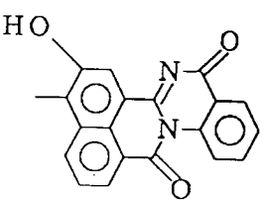
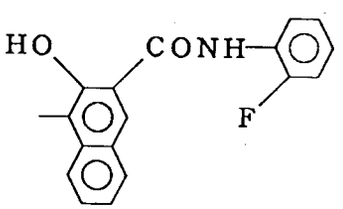
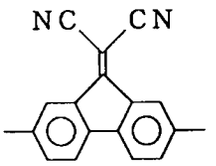


Cp_2:

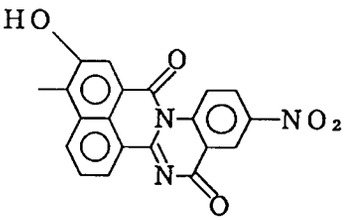


Exemplary Pigment 22

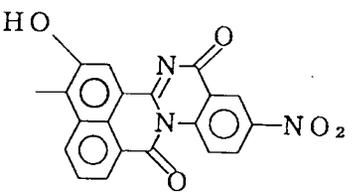
Ar:



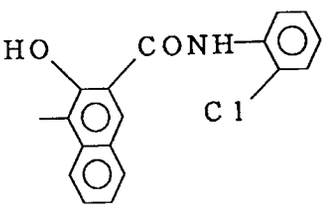
Cp_1:



and

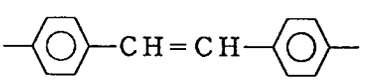


Cp_2:

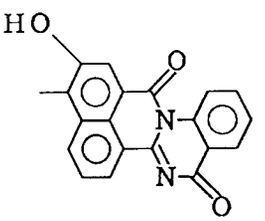


Exemplary Pigment 23

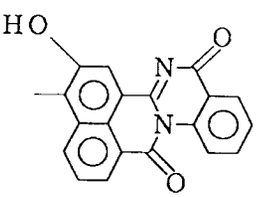
Ar:



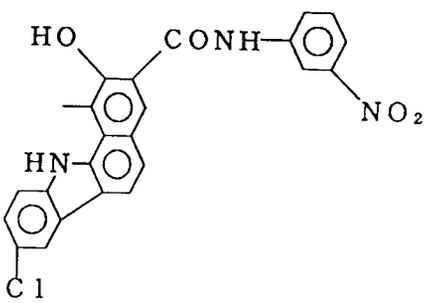
Cp_1:



and

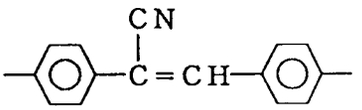


Cp_2:

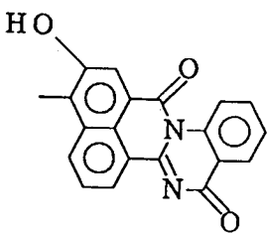


Exemplary Pigment 24

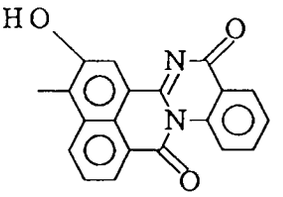
Ar:



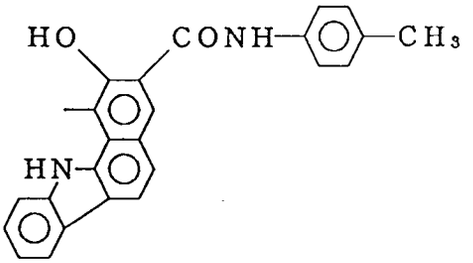
Cp_1:



and

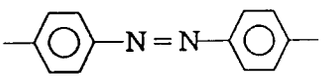


Cp_2:

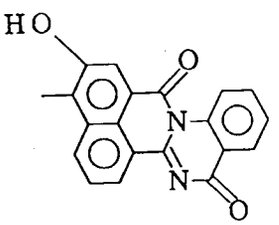


Exemplary Pigment 25

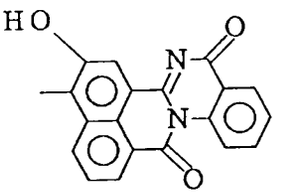
Ar:



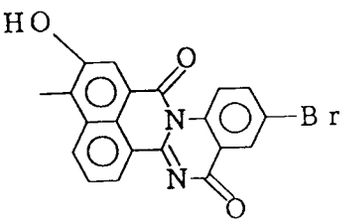
Cp_1:



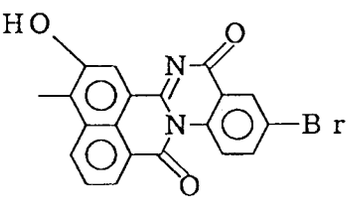
and



Cp_2:

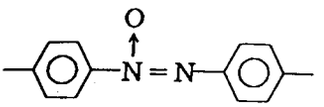


and

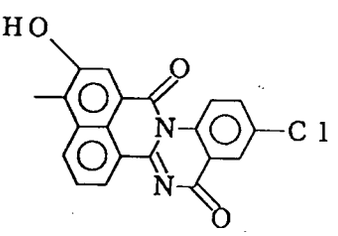


Exemplary Pigment 26

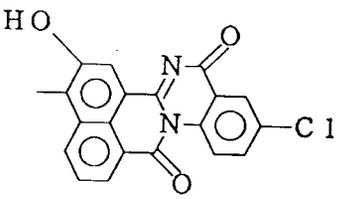
Ar:



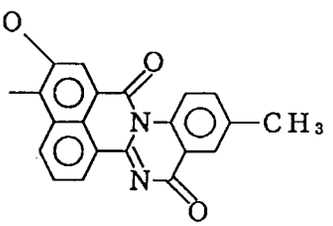
Cp_1:



and



Cp_2:



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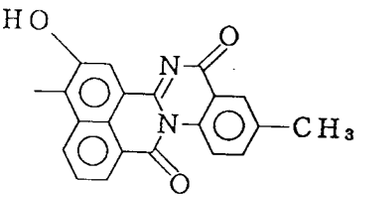
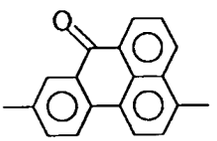
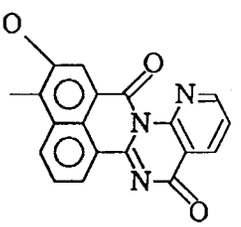


Exemplary Pigment 27

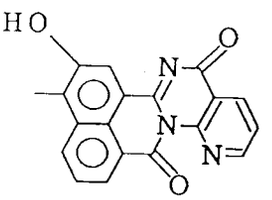
Ar:



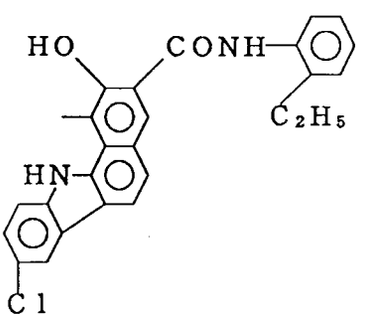
Cp_1:



and

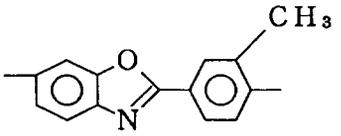


Cp_2:

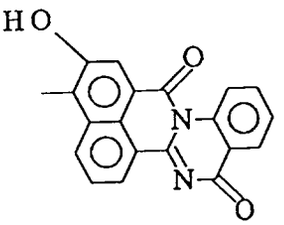


Exemplary Pigment 28

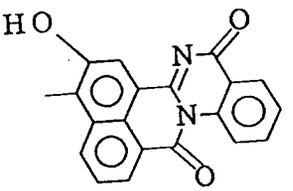
Ar:



Cp_1 and Cp_2:

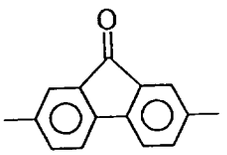


and

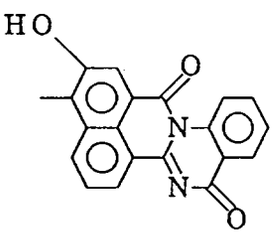


Exemplary Pigment 29

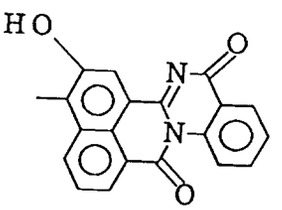
Ar:



Cp_1 and Cp_2:

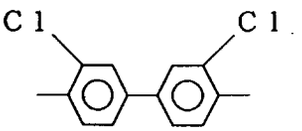


and

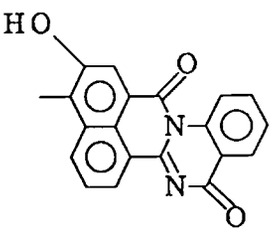


Exemplary Pigment 30

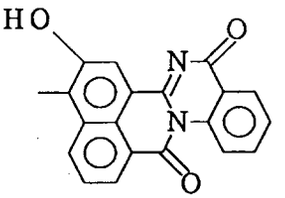
Ar:



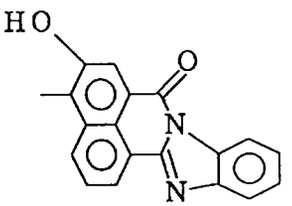
Cp_1:



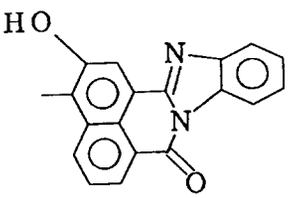
and



Cp_2:

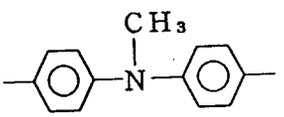


and

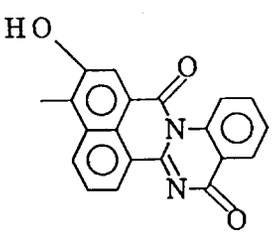


Exemplary Pigment 31

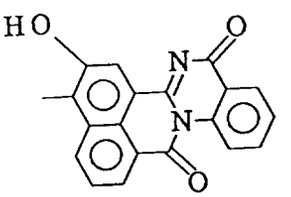
Ar:



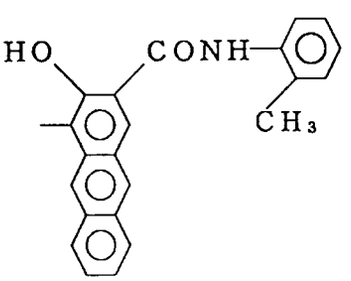
Cp_1:



and

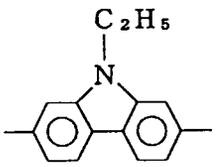


Cp_2:

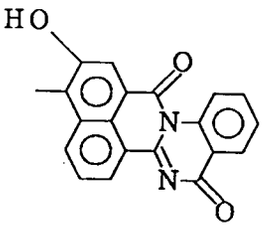


Exemplary Pigment 32

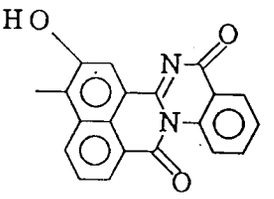
Ar:



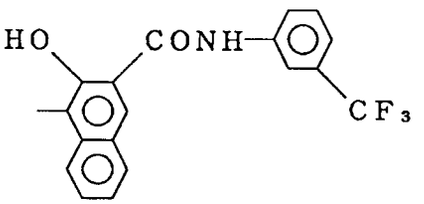
Cp_1:



and

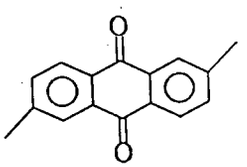


Cp_2:

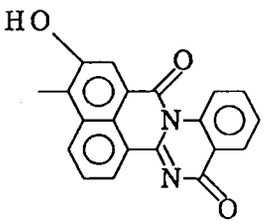


Exemplary Pigment 33

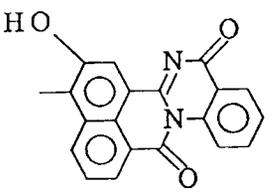
Ar:



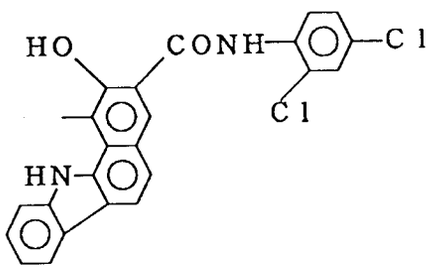
Cp_1:



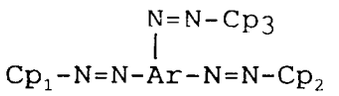
and



Cp_2:

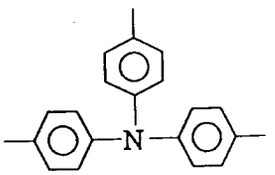


Basic Formula 2:

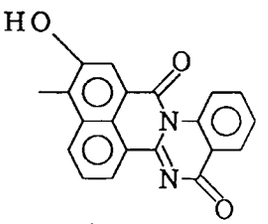


Exemplary Pigment 34

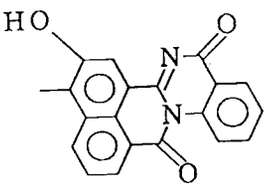
Ar:



Cp_1, Cp_2 and Cp_3:

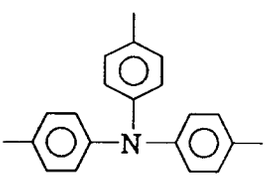


and

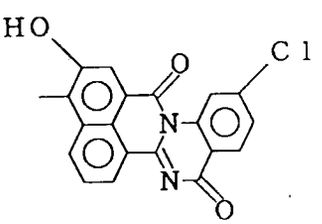


Exemplary Pigment 35

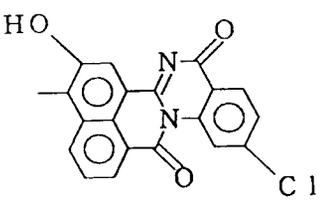
Ar:



Cp_1, Cp_2 and Cp_3:

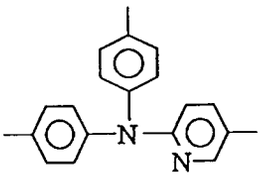


and

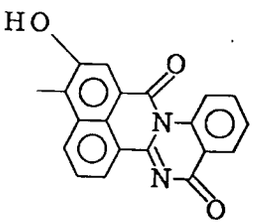


Exemplary Pigment 36

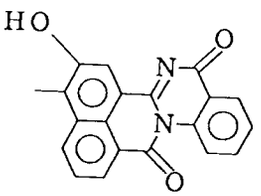
Ar:



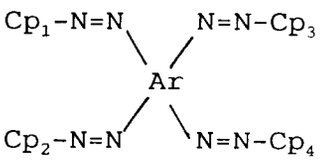
Cp_1, Cp_2 and Cp_3:



and

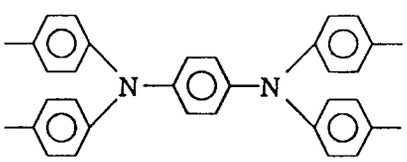


Basic Formula 3:

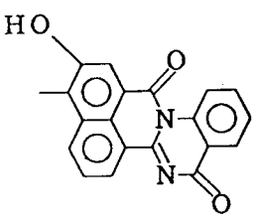


Exemplary Pigment 37

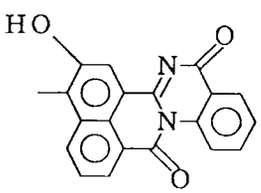
Ar:



Cp_1, Cp_2, Cp_3 and Cp_4:

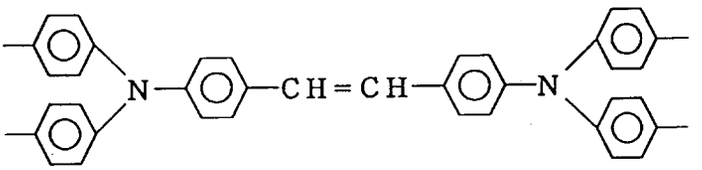


and

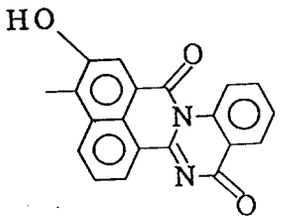


Exemplary Pigment 38

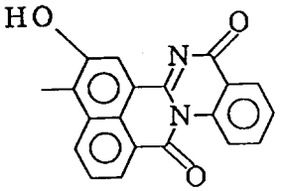
Ar:



Cp_1, Cp_2, Cp_3 and Cp_4:



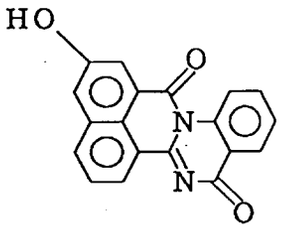
and



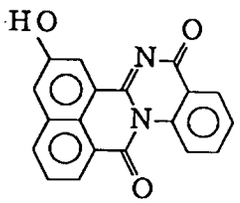
In the present invention, among these examples, the compounds in which all Cp's are coupler residual groups represented by Formula (2) are particularly preferred. Exemplary Pigments 1, 3, 4, 5, 6, 28 and 29 are more preferred.

The coupler having the coupler residual group represented by Formula (2) can be synthesized by subjecting hydroxy-1,8-naphthalic acid anhydride and the corresponding aminoarylcarboxylic acid amide to dehydration condensation in a suitable solvent. The solvent used includes nitrobenzene, dichlorobenzene, trichlorobenzene and xylene. Phosphorus oxychloride or polyphosphoric acid may be added as a condensation agent. The coupler having the coupler residual group represented by Formula (2), thus synthesized, is obtained in the form of a mixture of isomers represented by the following Formulas (2-a) and (2-b). Either isomer can be used in the present invention.

Formula (2-a)



Formula (2-b)



Synthesis Example 1 (Synthesis of Coupler)

To 130 ml of nitrobenzene, 10.0 g of 3-hydroxy-1,8-naphthalic acid anhydride and 7.0 g of o-aminobenzamide were added to carry out reflux with heating for 12 hours. After the reaction mixture was left to cool, the precipitate was filtered, and dispersed and washed with toluene and then with methanol, followed by reprecipitation repeated twice using an N,N-dimethylformamide/methanol solution. Thereafter, the precipitate was again dispersed and washed with methanol, followed by drying to give 3.5 g of a mixture of the couplers represented by Formulas (2-a) and (2-b). m.p.: 310-315°C; mass spectrum: m/z = 314.

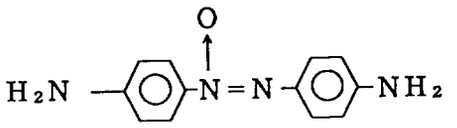
The azo pigment represented by Formula (1) can be readily obtained by;

(a) converting the corresponding amine into a diazo form, followed by coupling with the coupler having the coupler residual group represented by Formula (2), in the presence of an alkali in an aqueous system; or
(b) converting a diazonium salt into a borofluoride or a zinc chloride complex, followed by coupling with the coupler in the presence of a base such as sodium acetate, triethylamine or N-methylmorpholine in an organic solvent such as N,N-dimethylformamide or dimethylsulfoxide. When disazo pigments in which a coupler residual group other than the coupler residual group represented by Formula (2) is present together in the molecule are synthesized, they can be synthesized by;

(a) subjecting 1 mol of a tetrazolium salt firstly to coupling with 1 mol of the coupler having the coupler residual group represented by Formula (2) and subsequently to coupling with 1 mol of other coupler; or
(b) converting a diamine one amino group of which has been protected with an acetyl group or the like, into a diazo form, followed by coupling with the coupler having the coupler residual group represented by Formula (2), thereafter hydrolyzing the protective group using hydrochloric acid or the like, and again converting the compound into a diazo form, followed by coupling with other coupler. Trisazo pigments and tetrakisazo pigments in which a coupler residual group other than the coupler residual group represented by Formula (2) is present together in the molecule can also be synthesized in the same manner.

Synthesis Example 2 (Synthesis of Exemplary Pigment 1)

In a 300 ml beaker, 150 ml of water, 20 ml (0.23 mol) of concentrated hydrochloric acid and 7.3 g (0.032 mol) of the following diamine compound



were put, and the mixture was cooled to 0°C, to which a solution prepared by dissolving 4.6 g (0.067 mol) of sodium nitrite in 10 ml of water was dropwise added in 10 minutes while its liquid temperature was kept at 2°C or below. After stirring for 15 minutes, the reaction mixture was filtered with carbon, and then a solution prepared by dissolving 10.5 g (0.096 mol) of sodium borofluoride in 90 ml of water was dropwise added in the reaction mixture with stirring. The borofluoride precipitated was collected by filtration, and then washed with cold water, followed by washing with acetonitrile and then drying at room temperature under reduced pressure. Yield: 12.8 g, 94%.

Next, in a 5 lit. beaker, 3 lit. of DMF (N,N-dimethylformamide) was put, and then 13.2 g (0.042 mol) of a mixture of the couplers represented by Formulas (2-a) and (2-b) was dissolved therein. The resulting solution was cooled to have a liquid temperature of 5°C. Thereafter, 8.5 g (0.020 mol) of the borofluoride previously obtained was dissolved therein and subsequently 5.1 g (0.050 mol) of triethylamine was dropwise added in 5 minutes. After stirring for 2 hours, the pigment precipitated was collected by filtration, and then washed with DMF four times and with water three times, followed by freeze-drying. Yield: 14.2 g, 81%.

The electrophotographic photosensitive member of the present invention comprises a conductive support having thereon a photosensitive layer containing the oxytitanium phthalocyanine and azo pigment represented by Formula (1). The photosensitive layer may have any known form. Particularly preferred is a photosensitive layer of a function-separated type, comprised of a charge generation layer containing the oxytitanium phthalocyanine and azo pigment represented by Formula (1) and a charge transport layer superposingly formed thereon containing a charge-transporting material.

The charge generation layer of the present invention may be either a layer containing both the oxytitanium phthalocyanine and the azo pigment represented by Formula (1) or a layer formed by superposing layers respectively containing the both.

When the oxytitanium phthalocyanine and the azo pigment are mixed, they may preferably be in a weight ratio of oxytitanium phthalocyanine/azo pigment of from 95/5 to 70/30. If mixed in a weight ratio greater than 95/5, the intended improvement may be less effective, and on the other hand in a weight ratio smaller than 70/30, the sensitivity may lower on the side of long wavelength.

In the case when a charge generation layer containing the oxytitanium phthalocyanine or a charge generation layer containing the azo pigment are superposed, they may take either a form wherein the layer containing the azo pigment is in contact with the charge transport layer or a form wherein the layer containing the oxytitanium phthalocyanine is in contact with the charge transport layer. The latter form can be more effective in the present invention, and is preferred. The layer containing the oxytitanium phthalocyanine may preferably be formed in a layer thickness of from 0.1 to 0.5 μm. The layer containing the azo pigment need not be formed in so a large thickness, and can be well effective when formed in a thickness of about 0.05 μm to about 0.2 μm.

The charge generation layer can be formed by depositing the oxytitanium phthalocyanine and the azo pigment of the present invention on a conductive support, or by coating on a conductive support by a known method a coating solution prepared by dispersing these in a suitable solvent together with a binder resin. The layer may preferably be formed in a thin layer having a thickness not larger than 5 μm, and particularly preferably from 0.1 to 1 μm. The binder resin used in the formation of this layer by coating can be selected from a vast range of insulating resins or organic photoconductive polymers. It may preferably include polyvinyl butyral, polyvinyl benzal, polyarylates, polycarbonates, polyesters, phenoxy resins, cellulose resins, acrylic resins and polyurethanes. It may be used in an amount of not more than 80% by weight, and particularly preferably not more than 40% by weight, based on the total weight of the charge generation layer. The solvent used may preferably be selected from those which dissolve the above resin and do not dissolve the charge transport layer and subbing layer described later. It may specifically include ethers such as tetrahydrofuran and 1,4-dioxane; ketones such as cyclohexanone and methyl ethyl ketone; amides such as N,N-dimethylformamide; esters such as methyl acetate and ethyl acetate; aromatics such as toluene, xylene and chlorobenzene; alcohols such as methanol, ethanol and 2-propanol; and aliphatic halogenated hydrocarbons such as chloroform, methylene chloride, dichloroethylene, carbon tetrachloride and trichloroethylene.

The charge transport layer is superposed on or beneath the charge generation layer, and can be formed by coating a coating solution prepared by dissolving a charge-transporting material in a solvent optionally together with a suitable binder resin. It may preferably have a layer thickness of from 5 to 40 μm, and particularly preferably from 15 to 30 μm.

The charge-transporting material can be roughly grouped into an electron transporting material and a hole transporting material. The electron transporting material includes electron attractive materials as exemplified by 2,4,7-trinitrofluorenone, 2,4,5,7-tetranitrofluorenone, chloroanil and tetracyanoquinodimethane, or those obtained by forming

these electron attractive materials into polymers. The hole transporting material includes polycyclic aromatic compounds such as pyrene and anthracene; heterocyclic compounds such as compounds of a carbazole type, an indole type, an imidazole type, an oxazole type, a thiazole type, an oxadiazole type, a pyrazole type, a pyrazoline type, a thiadiazole type or a triazole type; hydrazone compounds such as p-diethylaminobenzaldehyde-N,N-diphenylhydrazone and N,N-diphenylhydrazino-3-methylidene-9-ethylcarbazole; triarylamine compounds such as tri-p-tolylamine, 4-(di-p-tolylamino)-biphenyl, 2-(di-p-tolyl)-amino-9,9'-dimethylfluorenone and 1-di-p-tolyl-aminopyrene; styryl compounds such as α-phenyl-4'-N,N-diphenylaminostilbene and 5-[4-(di-p-tolylamino)benzilidene]-5H-dibenzo[a,d]cyclo heptene; benzidine compounds; triarylmethane compounds; triphenylamine; or polymers having a group comprising any of these compounds as a backbone chain or side chain as exemplified by poly-N-vinylcarbazole and polyvinylanthracene. In addition to these organic charge-transporting materials, it is also possible to use inorganic materials such as selenium, selenium-tellurium, amorphous silicon and cadmium sulfide. These charge-transporting materials can be used alone or in combination of two or more kinds.

A binder resin may be used when the charge-transporting material has no film forming properties. It may specifically include insulating resins such as acrylic resins, polyarylates, polyesters, polycarbontes, polystyrene, an acrylonitrile/styrene copolymer, polyacrylamides, polyamides and chlorinated rubbers; and organic photoconductive polymers such as poly-N-vinylcarbazole and polyvinylanthracene.

The conductive support used in the present invention are exemplified by those made of aluminum, an aluminum alloy, copper, zinc, stainless steel, vanadium, molybdenum, chromium, titanium, nickel, indium, gold and platinum. Besides, it is possible to use supports comprised of plastics (as exemplified by polyethylene, polypropylene, polyvinyl chloride, polyethylene terephthalate and acrylic resins) having a film formed by vacuum deposition of any of these metals or alloys, supports comprising any of the above plastics, metals or alloys covered thereon with conductive particles (as exemplified by carbon black and silver particles) together with a suitable binder, and supports comprising plastics or paper impregnated with the conductive particles. The support may be in the form of a drum, a sheet or a belt, and should preferably be made to have a form most suited for electrophotographic apparatus to which it is applied.

In the present invention, a subbing layer having a barrier function and an adhesion function may be provided between the conductive support and the photosensitive layer. The subbing layer can be formed using casein, polyvinyl alcohol, nitrocellulose, polyamides such as nylon 6, nylon 66, nylon 610, copolymer nylon and alkoxymethylated nylon, polyurethanes, aluminum oxide, etc. The subbing layer may preferably have a layer thickness of not more than 5 μm, preferably from 0.1 μm to 3 μm.

As another embodiment of the present invention, the electrophotographic photosensitive member may include those having a photosensitive layer in which the oxytitanium phthalocyanine, the azo pigment of the present invention and the charge-transporting material as described above are incorporated in the same layer. In this instance, a charge-transfer complex comprised of poly-N-vinylcarbazole and trinitrofluorenone may be used as the charge-transporting material. The electrophotographic photosensitive member of this embodiment can be formed by coating a solution prepared by dissolving in a suitable resin solution the oxytitanium phthalocyanine, the azo pigment of the present invention and the charge-transporting material or charge-transfer complex as described above, followed by drying.

In both the electrophotographic photosensitive members, the azo pigment represented by Formula (1) may be either amorphous or crystalline in its crystal form. If necessary, the azo pigment represented by Formula (1) may also be used in combination of two or more kinds, or in combination of a known charge-generating material.

In the present invention, it is possible to further provide on the photosensitive layer a protective layer mainly composed of a resin.

The electrophotographic photosensitive member of the present invention can be not only utilized in electrophotographic copying machines, but also widely used in the field in which the electrophotography is applied as exemplified by laser beam printers, CRT printers, LED printers, liquid crystal printers, laser beam engravers and facsimile machines.

Fig. 5 schematically illustrates the construction of an electrophotographic apparatus having a process cartridge having the charging member of the present invention.

In Fig. 5, reference numeral 1 denotes an electrophotographic photosensitive member, which is rotatingly driven around an axis 2 in the direction of an arrow at a given peripheral speed. The photosensitive member 1 is uniformly charged on its periphery to a positive or negative, given potential through a primary charging means 3. The photosensitive member thus charged is then photoimagewise exposed to light 4 emitted from an imagewise exposure means (not shown) for slit exposure or laser beam scanning exposure. In this way, electrostatic latent images are successively formed on the periphery of the photosensitive member 1.

The electrostatic latent images thus formed are subsequently developed by toner by the operation of a developing means 5. The resulting toner-developed images are then successively transferred by the operation of a transfer means 6, to the surface of a transfer medium 7 fed from a paper feed section (not shown) to the part between the photosensitive member 1 and the transfer means 6 in the manner synchronized with the rotation of the photosensitive member 1.

The transfer medium 7 on which the images have been transferred is separated from the surface of the photosensitive member, is led through an image fixing means 8, where the images are fixed, and is then printed out of the apparatus as a copied material (a copy).

The surface of the photosensitive member 1 after the transfer of images is brought to removal of the toner remaining after the transfer, through a cleaning means 9. Thus the photosensitive member is cleaned on its surface, further subjected to charge elimination by pre-exposure light 10 emitted from a pre-exposure means (not shown), and then repeatedly used for the formation of images. When the primary charging means is a contact charging means making use of a charging roller or the like, the pre-exposure is not necessarily required.

In the present invention, the apparatus may be constituted of a combination of plural components integrally joined as a process cartridge from among the constituents such as the above electrophotographic photosensitive member 1, primary charging means 3, developing means 5 and cleaning means 9 so that the process cartridge is detachable from the body of the electrophotographic apparatus such as a copying machine or a laser beam printer. For example, at least one of the primary charging means 3, the developing means 5 and the cleaning means 9 may be integrally supported in a cartridge together with the electrophotographic photosensitive member 1 to form a process cartridge 11 that is detachable from the body of the apparatus through a guide means such as a rail 12 provided in the body of the apparatus.

In the case when the electrophotographic apparatus is used as a copying machine or a printer, the light 4 of the imagewise exposure is light reflected from, or transmitted through, an original, or light irradiated by the scanning of a laser beam, the driving of an LED array or the driving of a liquid crystal shutter array according to signals obtained by reading an original and converting the information into signals.

The present invention will be described below in greater detail by giving Examples.

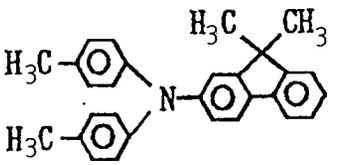
Example 1

50 parts (parts by weight; the same applies hereinafter) of titanium oxide powder coated with tin oxide, containing 10% of antimony oxide, 25 parts of resol type phenol resin, 20 parts of methyl cellosolve, 5 parts of methanol and 0.002 parts of silicone oil (a polydimethylsiloxane-polyoxyalkylene copolymer; weight average molecular weight: 3,000) were dispersed for 2 hours by means of a sand mill making use of glass beads of 1 mm diameter. The dispersion thus obtained was dip-coated on an aluminum cylinder (80 mm diameter and 360 mm long), followed by drying at 140°C for 30 minutes to form a conductive layer with a layer thickness of 20 μm.

On this conductive layer, a solution prepared by dissolving 5 parts of a 6-66-610-12 polyamide quaterpolymer in a mixed solvent of 70 parts of methanol and 25 parts of butanol was dip-coated, followed by drying to form a subbing layer with a layer thickness of 1 μm.

To a solution prepared by dissolving 10 parts of polyvinyl butyral (trade name: S-LEC BX-1; available from Sekisui Chemical Co., Ltd.) in 400 parts of cyclohexanone, 7 parts of I-type oxytitanium phthalocyanine having distinctive peaks at Bragg's angles (2θ ±0.2°) of 9.0°, 14.2°, 23.9° and 27.1° in an X-ray diffraction pattern of CuKα characteristics (its diffraction pattern is shown in Fig. 1) and 3 parts of an azo pigment Exemplary Pigment 1 were added. The resulting mixture was dispersed for 2 hours by means of a sand mill making use of glass beads of 1 mm diameter. To the dispersion thus obtained, 400 parts of ethyl acetate was added to dilute it. The dispersion thus obtained was dip-coated on the above subbing layer, followed by drying at 80°C for 10 minutes to form a charge generation layer with a layer thickness of 0.25 μm.

Next, 10 parts of a charge-transporting material having the following structural formula:



and 10 parts of bisphenol-Z polycarbonate (weight average molecular weight: 49,000) were dissolved in 60 parts of chlorobenzene. The resulting solution was dip-coated on the charge generation layer, followed by drying at 110°C for one hour to form a charge transport layer with a layer thickness of 20 μm. Thus, an electrophotographic photosensitive member was produced.

Comparative Example 1

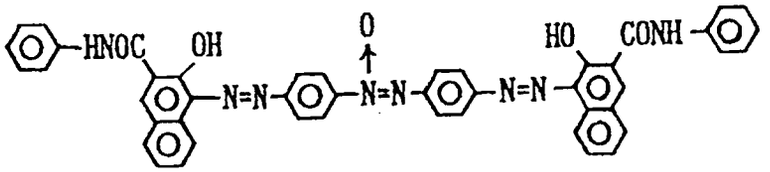
An electrophotographic photosensitive member was produced in the same manner as in Example 1 except that the I-type oxytitanium phthalocyanine was used in an amount of 10 parts and the azo pigment was not used.

Comparative Example 2

An electrophotographic photosensitive member was produced in the same manner as in Example 1 except that the azo pigment was not used.

Comparative Example 3

An electrophotographic photosensitive member was produced in the same manner as in Example 1 except that the azo pigment was replaced with an azo pigment represented by the following structural formula.



Performance Evaluation

Electrophotographic photosensitive members produced in Example 1 and Comparative Examples 1 to 3 were each set on a laser beam printer (trade name: LBP-SX; manufactured by CANON INC.) to make evaluation of their performances. First, each photosensitive member was made to have a dark-portion potential of -700 V, and then exposed to light emitted from a 802 nm laser to measure laser light intensity necessary for providing a light-portion potential of -150 V, i.e., to measure sensitivity. Also, the amount of change in dark-portion potential (ΔV_D) and amount of change in light-portion potential (ΔV_L) in the course of continuous 5,000 A4 size sheet printing were also measured. An instance where the value of the amount of change is negative indicates that the absolute value of the potential became smaller after the printing, and an instance where it is positive, that the absolute value became larger.

Photomemory characteristics were also evaluated. Stated specifically, first, using a fluorescent lamp (trade name: FV_L18 White Light Lamp; manufactured by Matsushita Electric Industrial Co., Ltd.), the electrophotographic photosensitive members produced in Example 1 and Comparative Examples 1 to 3 were each partially exposed to light at an intensity of illumination of 1,500 luxes for 5 minutes, and, after the exposure, left to stand at room temperature for 5 minutes (non-exposed portions were formed by screening the light). Thereafter, the photosensitive members thus treated were each set on the above laser beam printer to measure potential difference between the light-portion potential at non-exposed portions and that at exposed portions (photomemory = |potential at non-exposed portion - potential at exposed portion|).

Results thus obtained are shown in Table 1.

Table 1

	TiOPc*/azo pigment ratio (wt/wt)	Pigment/binder resin ratio (wt/wt)	Sensitivity	Amount of change in potential		Photomemory
				ΔV_D	ΔV_L	
			($\mu J/cm^2$)	(V)	(V)	(V)
Example:						
1	7/3	10/10	0.21	-10	+5	10
Comparative Example:						
1	10/0	10/10	0.20	-60	-30	60
2	7/0	7/10	0.27	-40	-15	45
3	7/3	10/10	0.29	-25	+40	40

* Oxytitanium phthalocyanine

As is seen from these results, in Example 1, a sensitivity equal to that in Comparative Example 1 is obtained and repetition performance and photomemory characteristics are improved, even though the oxytitanium phthalocyanine is in a smaller content than that in the latter. On the other hand, in Comparative Example 2, the sensitivity is fairly low and the repetition performance and photomemory characteristics are less improved, because the oxytitanium phthalocyanine is used in a small content and the azo pigment is not used. Comparative Example 3, in which a conventional bisazo pigment is used, the effect as in the present invention is not obtained.

Example 2

Dispersions for the charge generation layer were prepared in the same manner as in Example 1 except that a dispersion of the oxytitanium phthalocyanine and a dispersion of the azo pigment were separately prepared. Here, the pigment and binder resin in each dispersion was in a ratio of 10/10.

A conductive support and a subbing layer were formed in the same manner as in Example 1, and the azo pigment dispersion was coated on this subbing layer so as to be in a layer thickness of 0.1 μm, followed by drying, and then the oxytitanium phthalocyanine dispersion was further coated thereon so as to be in a layer thickness of 0.25 μm, followed by drying, to form a double-layer charge generation layer. The charge transport layer was formed thereon in the same manner as in Example 1. Thus, an electrophotographic photosensitive member was produced.

Example 3

An electrophotographic photosensitive member was produced in the same manner as in Example 2 except that the azo pigment charge generation layer and the oxytitanium phthalocyanine charge generation layer were formed in reverse order.

Evaluation was made on the electrophotographic photosensitive members produced in Examples 2 and 3, in the same manner as in Example 1. Results obtained are shown in Table 2. For comparison, the results of the electrophotographic photosensitive member produced in Comparative Example 1, which is an example in which no azo pigment is contained in the charge generation layer, are shown together.

Table 2

Electrophotographic photosensitive member	Sensitivity	Amount of change in potential		Photomemory
		ΔV_D	ΔV_L	
	($\mu J/cm^2$)	(V)	(V)	(V)
Example 2:	0.20	-10	0	15
Example 3:	0.24	-5	+20	5
Comparative Example 1:	0.20	-60	-30	60

As is seen from these results, in the present invention, the repetition performance and photomemory characteristics are improved, where, in view of sensitivity, the charge generation layer containing the azo pigment more preferably be provided on the side of the conductive support.

Examples 4 to 6

Electrophotographic photosensitive members corresponding to Examples 4, 5 and 6 were produced in the same manner as in Example 1 except that the I-type oxytitanium phthalocyanine was replaced with the A-type oxytitanium phthalocyanine, B-type oxytitanium phthalocyanine, Y-type oxytitanium phthalocyanine exhibiting the X-ray diffraction patterns as shown in Figs. 2, 3 and 4, respectively.

Comparative Examples 4 to 6

Electrophotographic photosensitive members corresponding to Comparative Examples 4, 5 and 6 were produced in the same manner as in Comparative Example 1 except that the I-type oxytitanium phthalocyanine was replaced with the A-type oxytitanium phthalocyanine, B-type oxytitanium phthalocyanine, Y-type oxytitanium phthalocyanine, respectively.

Evaluation was made on the electrophotographic photosensitive members produced in Examples 4 to 6 and Comparative Examples 4 to 6, in the same manner as in Example 1. Results obtained are shown in Table 3.

Table 3

TiOPc* crystal form	TiOPc*/azo pigment ratio (wt/wt)	Pigment/binder resin ratio (wt/wt)	Sensitivity	Amount of change in potential		Photomemory
				ΔV_D	ΔV_L	
			($\mu J/cm^2$)	(V)	(V)	(V)
Example:						
4 A type	7/3	10/10	0.37	-25	0	10
5 B type	7/3	10/10	0.36	-30	-15	15
6 Y type	7/3	10/10	0.25	-20	-10	20
Comparative Example:						
4 A type	10/0	10/10	0.37	-60	-20	45
5 B type	10/0	10/10	0.35	-70	-30	50
6 Y type	10/0	10/10	0.23	-50	-25	55

* Oxytitanium phthalocyanine

As is seen from these results, the present invention is effective without regard to the crystal form of the oxytitanium phthalocyanine.

Examples 7 to 16

Electrophotographic photosensitive members corresponding to Examples 7 to 16 were produced in the same manner as in Example 1 except that the azo pigment Exemplary Pigment 1 was replaced with azo pigments Exemplary Pigments 4, 29, 3, 28, 5, 20, 24, 26, 34 and 37, respectively, and evaluation was made similarly. Results obtained are shown in Table 2. For comparison, the results of Example 1 and Comparative Example 1 are shown together.

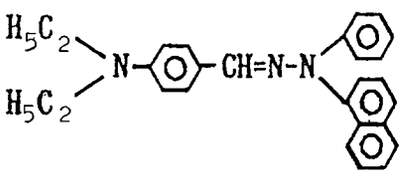
Table 4

	Exemplary Pigment	Sensitivity	Amount of change in potential		Photomemory
			ΔV_D	ΔV_L	
		($\mu J/cm^2$)	(V)	(V)	(V)
Example:					
7	4	0.22	-10	+5	15
8	29	0.20	-5	+10	5
9	3	0.23	-25	0	20
10	28	0.23	-20	-10	10
11	5	0.21	-10	+10	25
12	20	0.22	-20	-10	15
13	24	0.23	-15	-10	25
14	26	0.23	-5	+10	20
15	34	0.21	-10	0	10
16	37	0.23	-25	-10	25
Example:					
1	1	0.21	-10	+5	10
Comparative Example:					
1	-	0.20	-60	-30	60

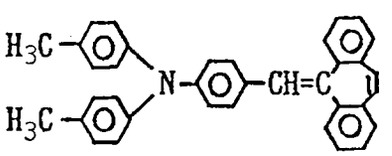
Examples 17 to 20

Electrophotographic photosensitive members corresponding to Examples 17 to 20 were produced in the same manner as in Example 1 except that the charge-transporting materials were respectively replaced with the compounds represented by the following structural formulas.

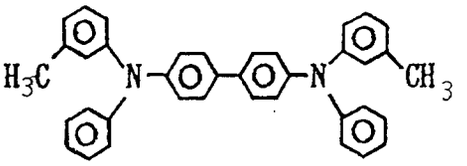
Charge-transporting material used in Example 17:



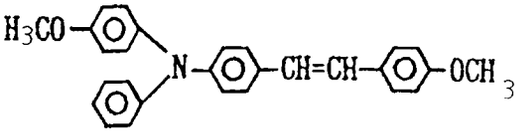
Charge-transporting material used in Example 18:



Charge-transporting material used in Example 19:



Charge-transporting material used in Example 20:



Comparative Examples 7 to 10

Electrophotographic photosensitive members corresponding to Comparative Examples 7 to 10 were produced in the same manner as in Comparative Example 1 except that the charge-transporting materials were respectively replaced with those used in Examples 17 to 20.

Evaluation was made on the electrophotographic photosensitive members produced in Examples 17 to 20 and Comparative Examples 7 to 10, in the same manner as in Example 1. Results obtained are shown in Table 5.

Table 5

Electrophotographic photosensitive member	Sensitivity	Amount of change in potential		Photomemory
		ΔV_D	ΔV_L	
	($\mu J/cm^2$)	(V)	(V)	(V)
Example 17:	0.24	-5	+10	0
Comparative Example 7:	0.22	-40	-15	40
Example 18:	0.21	-15	-5	5
Comparative Example 8:	0.20	-60	-25	55
Example 19:	0.21	-20	-10	15
Comparative Example 9:	0.21	-50	-30	60
Example 20:	0.25	-25	+10	25
Comparative Example 10:	0.23	-70	-45	65

As is seen from these results, the present invention is effective without regard to the type of the charge-transporting material.

An electrophotographic photosensitive member is disclosed which has a photosensitive layer on a conductive support. The photosensitive layer contains an oxytitanium phthalocyanine and an azo pigment represented by the following Formula (1):

$$Ar\text{-}(N{=}N\text{-}Cp)_n.$$

Also, disclosed are a process cartridge and an electrophotographic apparatus using the electrophotographic photosensitive member.

Claims

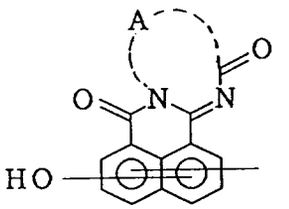
1. An electrophotographic photosensitive member comprising a conductive support and a photosensitive layer provided on the conductive support;

said photosensitive layer containing an oxytitanium phthalocyanine and an azo pigment represented by the following Formula (1).

Formula (1): $Ar\{N=N-Cp\}_n$

wherein Ar represents a substituted or unsubstituted aromatic hydrocarbon group which may be bonded through a linking group, or a substituted or unsubstituted heterocyclic group which may be bonded through a linking group; Cp represents a coupler residual group having a phenolic hydroxyl group, at least one of said Cp representing a coupler residual group represented by the following Formula (2); and n represents an integer of 2 to 4.

Formula (2):



wherein A represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon ring or a divalent group of a substituted or unsubstituted heterocyclic ring containing a nitrogen atom in the ring.

2. An electrophotographic photosensitive member according to claim 1, wherein all Cp's in Formula (1) are groups represented by Formula (2).

3. An electrophotographic photosensitive member according to claim 2, wherein the A in Formula (2) is an unsubstituted o-phenylene.

4. An electrophotographic photosensitive member according to claim 3, wherein the n in Formula (1) is 2 or 3.

5. An electrophotographic photosensitive member according to claim 1, wherein said oxytitanium phthalocyanine has distinctive peaks at Bragg's angles ($2\theta \pm 0.2°$) of 9.0°, 14.2°, 23.9° and 27.1° in an X-ray diffraction pattern of CuKα characteristics.

6. An electrophotographic photosensitive member according to claim 4, wherein said oxytitanium phthalocyanine has distinctive peaks at Bragg's angles ($2\theta \pm 0.2°$) of 9.0°, 14.2°, 23.9° and 27.1° in an X-ray diffraction pattern of CuKα characteristics.

7. A process cartridge comprising an electrophotographic photosensitive member and at least one means selected from the group consisting of a charging means, a developing means and a cleaning means;

said electrophotographic photosensitive member comprising a conductive support and a photosensitive layer provided on the conductive support;

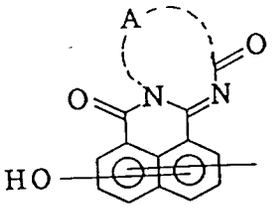
said photosensitive layer containing an oxytitanium phthalocyanine and an azo pigment represented by the following Formula (1).

Formula (1): $Ar\{N=N-Cp\}_n$

wherein Ar represents a substituted or unsubstituted aromatic hydrocarbon group which may be bonded through a linking group, or a substituted or unsubstituted heterocyclic group which may be bonded through a linking group; Cp represents a coupler residual group having a phenolic hydroxyl group, at least one of said Cp representing a

coupler residual group represented by the following Formula (2); and n represents an integer of 2 to 4.

Formula (2):



wherein A represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon ring or a divalent group of a substituted or unsubstituted heterocyclic ring containing a nitrogen atom in the ring; and

said means selected from the group consisting of a charging means, a developing means and a cleaning means being integrally supported in and detachable from the body of an electrophotographic apparatus.

8. A process cartridge according to claim 7, wherein all Cp's in Formula (1) are groups represented by Formula (2).

9. A process cartridge according to claim 8, wherein the A in Formula (2) is an unsubstituted o-phenylene.

10. A process cartridge according to claim 9, wherein the n in Formula (1) is 2 or 3.

11. A process cartridge according to claim 7, wherein said oxytitanium phthalocyanine has distinctive peaks at Bragg's angles (2θ ±0.2°) of 9.0°, 14.2°, 23.9° and 27.1° in an X-ray diffraction pattern of CuKα characteristics.

12. A process cartridge according to claim 10, wherein said oxytitanium phthalocyanine has distinctive peaks at Bragg's angles (2θ ±0.2°) of 9.0°, 14.2°, 23.9° and 27.1° in an X-ray diffraction pattern of CuKα characteristics.

13. An electrophotographic apparatus comprising an electrophotographic photosensitive member, a charging means, an exposure means, a developing means and a transfer means;

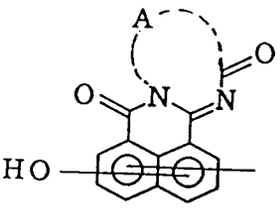
said electrophotographic photosensitive member comprising a conductive support and a photosensitive layer provided on the conductive support;

said photosensitive layer containing an oxytitanium phthalocyanine and an azo pigment represented by the following Formula (1).

Formula (1): $Ar\{N=N-Cp)_n$

wherein Ar represents a substituted or unsubstituted aromatic hydrocarbon group which may be bonded through a linking group, or a substituted or unsubstituted heterocyclic group which may be bonded through a linking group; Cp represents a coupler residual group having a phenolic hydroxyl group, at least one of said Cp representing a coupler residual group represented by the following Formula (2); and n represents an integer of 2 to 4.

Formula (2):



wherein A represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon ring or a divalent group of a substituted or unsubstituted heterocyclic ring containing a nitrogen atom in the ring.

14. An electrophotographic apparatus according to claim 13, wherein all Cp's in Formula (1) are groups represented by Formula (2).

15. An electrophotographic apparatus according to claim 14, wherein the A in Formula (2) is an unsubstituted o-phenylene.

16. An electrophotographic apparatus according to claim 15, wherein the n in Formula (1) is 2 or 3.

17. An electrophotographic apparatus according to claim 13, wherein said oxytitanium phthalocyanine has distinctive peaks at Bragg's angles ($2\theta \pm 0.2°$) of 9.0°, 14.2°, 23.9° and 27.1° in an X-ray diffraction pattern of CuKα characteristics.

18. An electrophotographic apparatus according to claim 16, wherein said oxytitanium phthalocyanine has distinctive peaks at Bragg's angles ($2\theta \pm 0.2°$) of 9.0°, 14.2°, 23.9° and 27.1° in an X-ray diffraction pattern of CuKα characteristics.

FIG. 1

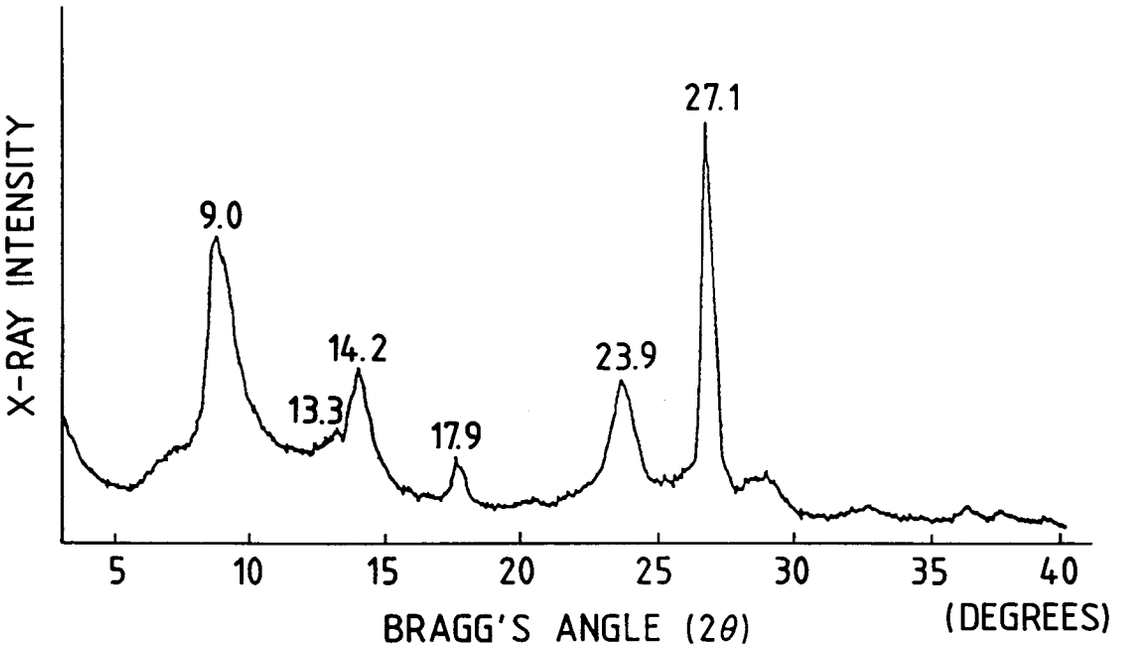

X-RAY INTENSITY
9.0
14.2
13.3
17.9
23.9
27.1
5
10
15
20
25
30
35
40
BRAGG'S ANGLE (2θ)
(DEGREES)

FIG. 2

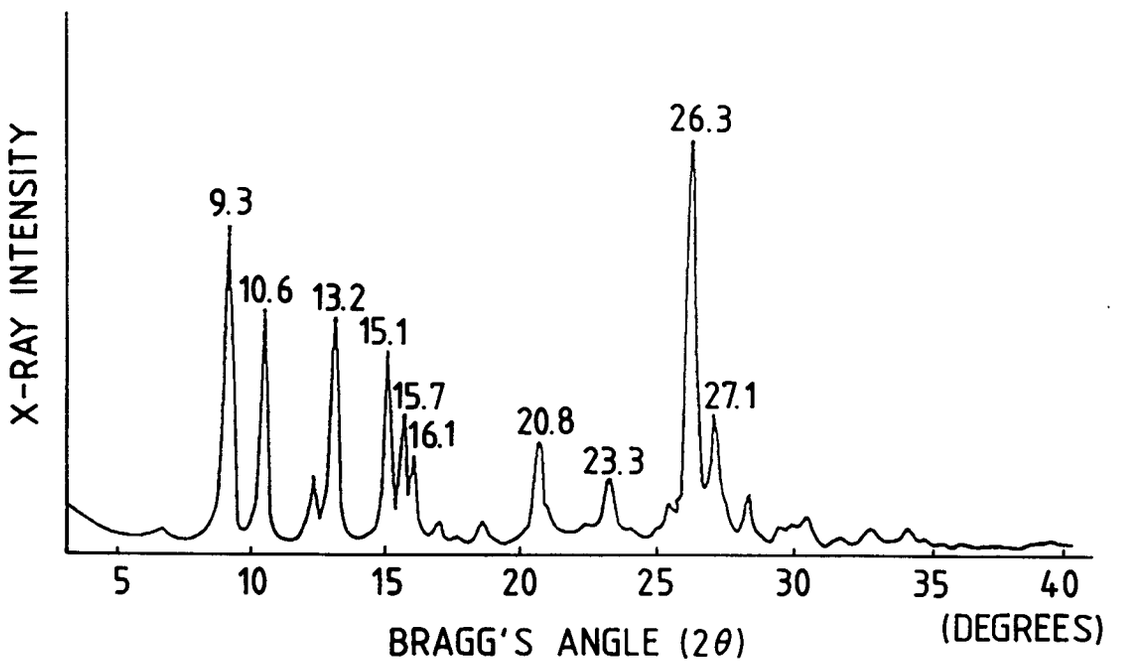

X-RAY INTENSITY
9.3
10.6
13.2
15.1
15.7
16.1
20.8
23.3
26.3
27.1
5
10
15
20
25
30
35
40
BRAGG'S ANGLE (2θ)
(DEGREES)

FIG. 3

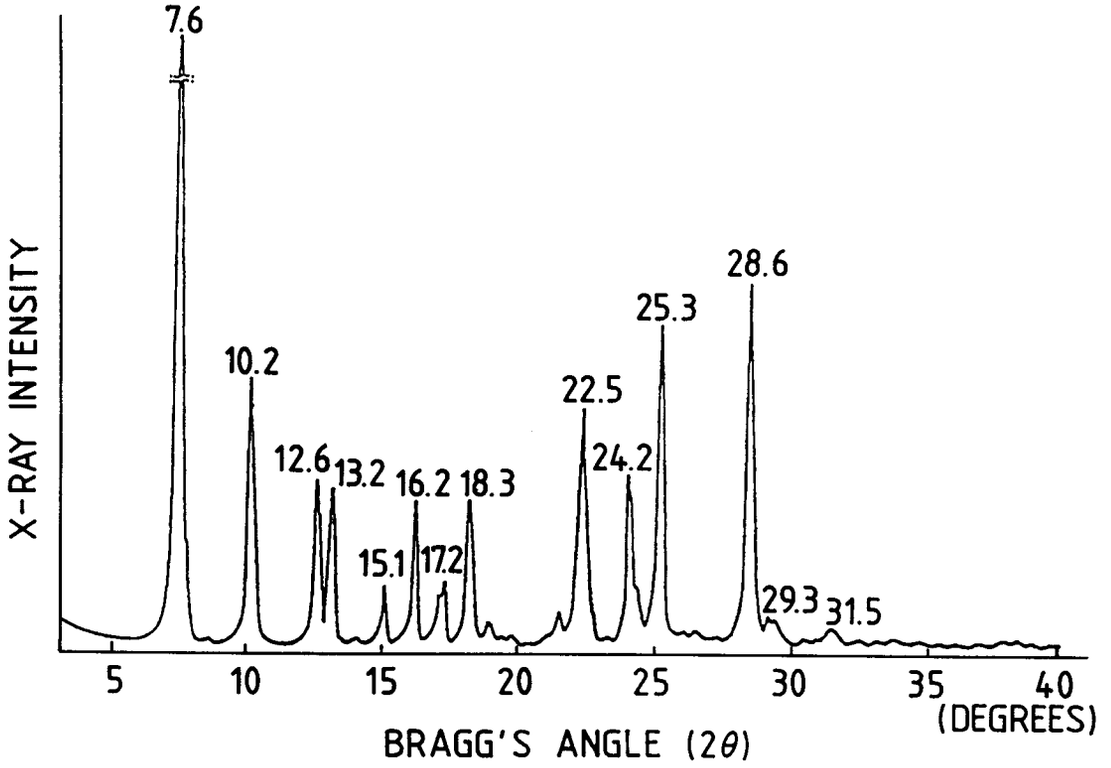

7.6
10.2
12.6
13.2
16.2
18.3
15.1
17.2
22.5
24.2
25.3
28.6
29.3
31.5
X-RAY INTENSITY
5
10
15
20
25
30
35
40
(DEGREES)
BRAGG'S ANGLE (2θ)

FIG. 4

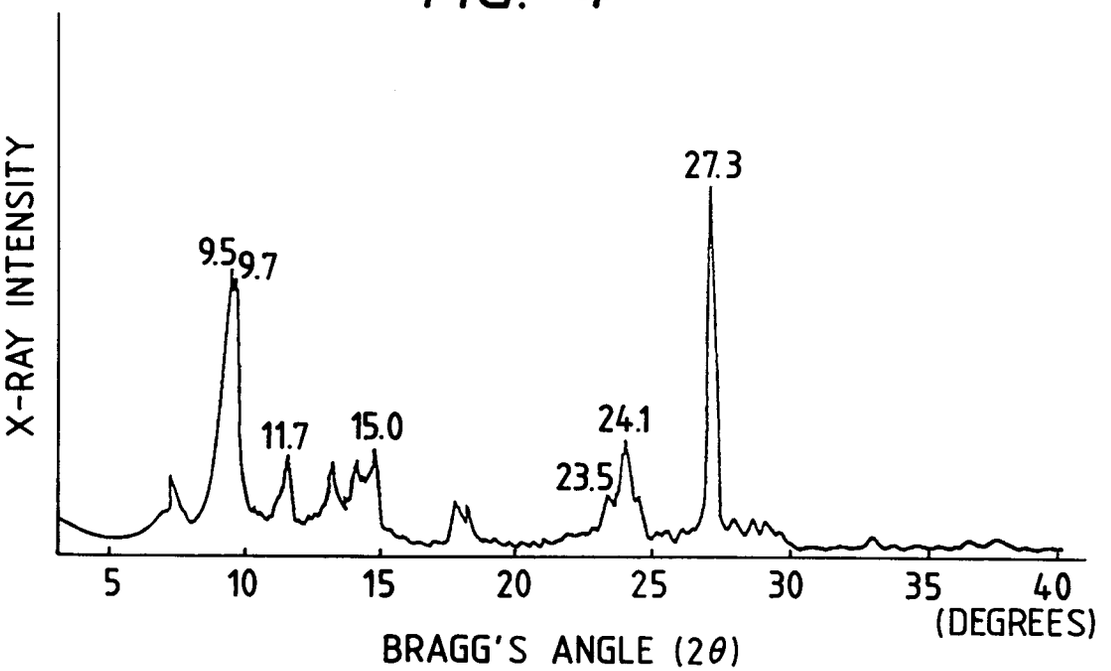

9.5
9.7
11.7
15.0
23.5
24.1
27.3
X-RAY INTENSITY
5
10
15
20
25
30
35
40
(DEGREES)
BRAGG'S ANGLE (2θ)

FIG. 5