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

(11) Publication number: **0 456 257 A1**

(12) EUROPEAN PATENT APPLICATION

(21) Application number: **91107617.2**

(22) Date of filing: **10.05.91**

(51) Int. Cl.[5]: **G03C 7/32**, G03C 7/333, G03C 7/305

(30) Priority: **10.05.90 JP 120820/90**
07.02.91 JP 36568/91

(43) Date of publication of application:
13.11.91 Bulletin 91/46

(84) Designated Contracting States:
DE FR GB IT NL

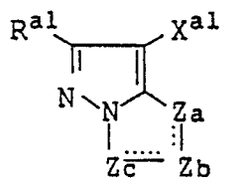
(71) Applicant: **FUJI PHOTO FILM CO., LTD.**
210 Nakanuma Minami Ashigara-shi
Kanagawa(JP)

(72) Inventor: **Mihayashi, Keiji**
c/o Fuji Photo Film Co., Ltd., No. 210, Nakanuma
Minami Ashigara-shi, Kanagawa(JP)
Inventor: **Ichijima, Seiji**
c/o Fuji Photo Film Co., Ltd., No. 210, Nakanuma
Minami Ashigara-shi, Kanagawa(JP)

(74) Representative: **Hansen, Bernd, Dr.rer.nat. et al**
Hoffmann, Eitle & Partner Patentanwälte
Arabellastrasse 4 Postfach 81 04 20
W-8000 München 81(DE)

(54) Silver halide color photographic material.

(57) A silver halide color photographic material is provided comprising a support having thereon (i) at least a silver halide emulsion layer containing (ii) a yellow-colored cyan coupler capable of undergoing reaction with an oxidation product of an aromatic primary amine developing agent to release a group containing a water-soluble 6-hydroxy-2-pyridon-5-ylazo group, a water soluble 2-acylaminophenylazo group, a water soluble 2-sulfonamidophenylazo group, a water soluble 5-aminopyrazol-4-ylazo on a water soluble pyrazolon-4-ylazo group and (iii) a coupler represented by the general formula (A):



(A)

wherein R^{a1} represents a hydrogen atom or substituent; X^{a1} represents a hydrogen atom or a group capable of being separated from the compound of formula (A) upon a coupling reaction with an oxidation product of an aromatic primary amine developing agent; Za, Zb and Zc each represents a methine group, substituted methine group, =N- group or -NH- group; one of Za-Zb bond and Zb-Zc bond is a double bond and the other is a single bond; if Zb-Zc bond is a carbon-carbon double bond, it may be a part of an aromatic ring; R^{a1} or X^{a1} may form a dimer or higher polymer; and if Za, Zb or Zc is a substituted methine, it may form a dimer or higher polymer. A preferred embodiment further comprises a compound represented by the general formula (II):

$$A - \{(L1)_a - (B)_m\}_p - (L2)_n - DI \qquad (II)$$

wherein A represents a group which is capable of undergoing a reaction with an oxidation product of an aromatic

EP 0 456 257 A1

primary amine developing agent to cause cleavage of A from $\{(L1)_a - (B)_m\}_p - (L2)_n - DI$; L1 represents a group which undergoes cleavage of the bond between L1 and the group to its right after cleavage of the bond between L1 and A as viewed in the general formula (I); B represents a group which undergoes reaction with an oxidation product of a developing agent to cause cleavage of the bond between B and the group to its right side as viewed in the general formula (I); L2 represents a group which causes cleavage of the bond between L2 and DI after cleavage of the bond of L2 to the group to its left as viewed in the general formula (I); DI represents a development inhibitor; and p represents an integer from 0 to 2, with the proviso that if p is 2, the two $\{(L1)_a - (B)_m\}$ groups are the same or different.

FIELD OF THE INVENTION

The present invention relates to a silver halide color photographic material. More particularly, the present invention relates to a silver halide color photographic material comprising yellow colored cyan couplers and pyrazoloazole couplers.

BACKGROUND OF THE INVENTION

One of the properties required for silver halide color photographic materials (hereinafter simply referred to as "light-sensitive material" as necessary) is color reproductivity.

One approach for improving color reproducibility is to use colored couplers in, e.g., color negative light-sensitive materials for picture taking, to correct for undesired absorption of colored dye images. These colored couplers are disclosed in many publications and patents, for instance Research Disclosure No. 17643, VII-G.

Colored couplers used for the correction of undesired absorption of cyan images include those which exhibit a maximum absorption wavelength between about 500 nm and 600 nm in the visible light range and undergo a coupling reaction with an oxidation product of an aromatic primary amine developing agent to form a cyan dye image which exhibits a maximum absorption wavelength between about 630 nm and 750 nm.

However, a cyan dye image also has an absorption in the visible light range of 400 to 500 nm. If these undesired absorptions are also corrected for by the so-called yellow-colored cyan couplers, an effect which photographically approximates the interimage effect developed from a cyan colored image layer and a yellow colored image layer can be obtained, probably providing an advantage in color reproduction. This possibility is described in JP-A-61-221748 and JP-A-1-319774 (the term "JP-A" as used herein means an "unexamined published Japanese patent application").

However, this appoach does not necessarily provide satisfactory properties, due to its low coupling reactivity and small molecular extinction coefficient. This approach is also disadvantageous in that photographic properties such as the preservability of light-sensitive material and the stability of latent images are subject to great fluctuation. Thus, this approach is not yet suitable for practical use.

Magenta couplers for color negative light-sensitive materials for picture taking include, primarily, 5-pyrazolone couplers.

However, colored images obtained from 5-pyrazolone magenta couplers exhibit a secondary absorption in the vicinity of about 430 nm in the short wavelength side. This secondary absoption drastically impairs color reproducibility. Therefore, color negative light-sensitive materials for picture taking usually comprise yellow colored magenta couplers to correct for the undesired absorbption.

In recent years, pyrazoloazole couplers have been rapidly developed. Examples of such pyrazoloazole couplers include pyrazolotriazole couplers described in U.S. Patents 3,725,067, 4,562,146, 4,607,002, 4,675,280, 4,840,886, 4,621,046, and 4,659,652, and JP-A-61-65243, JP-A-61-65245, JP-A-61-65246, and JP-A-61-65247, pyrazolotetrazole couplers as described in JP-A-60-33552, and pyrazolopyrazole couplers as described in JP-A-60-43659.

These couplers provide colored images having colors ranging from magenta to cyan depending on the substituents incorporated therein. These colored images exhibit no secondary absorption peak in the short wavelength range as observed in the absorption spectrum of colored images obtained from the above mentioned 5-pyrazolone couplers. These colored images have the further great advantage that they exhibit a small absorbance in the short wavelength side in the blue light range. Thus, these couplers can be said to exhibit excellent color reproductivity.

However, these couplers are disadvantageous in that they do not provide a high coupling reactivity and are subject to great fluctuation in photographic properties after storage at elevated temperature and humidity after preparation of light-sensitive material or after storage at elevated temperature and humidity for a prolonged period of time between the time of picture taking and development. This makes it difficult to put this approach into practical use.

There is therefore a need for a light-sensitive material which provides a high coupling reactivity with yellow colored cyan couplers, a high molecular extinction coefficient and improvements in preservability of light-sensitive material and in stability of latent images, even with pyrazoloazole couplers which exhibit a small absorbance by colored images in the short wavelength area.

SUMMARY OF THE INVENTION

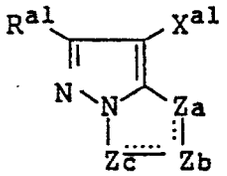
It is therefore one object of the present invention to provide a silver halide color photographic material which exhibits improved color reproducibility.

It is another object of the present invention to provide a silver halide color photographic material which provides improvements in color reproducibility as well as in inhibition of fluctuation of photographic properties and latent images during preservation.

It is a further object of the present invention to provide a silver halide color photographic material which provides improved fastness of color images obtained and a good balance in fastness of three color images, i.e., cyan, magenta and yellow.

These and other objects of the present invention will become more apparent from the following detailed description and examples.

The above objects of the present invention are accomplished with a silver halide color photographic material comprising a support having thereon (i) at least a silver halide emulsion layer (ii) a yellow-colored cyan coupler capable of undergoing a reaction with an oxidation product of an aromatic primary amine developing agent to release a group containing a water-soluble 6-hydroxy-2-pyridon-5-ylazo group, a water soluble 2-acylaminophenylazo group, a water soluble 2-sulfonamidophenylazo group, a water soluble 5-aminopyrazol-4-ylazo group or a water soluble pyrazolon-4-ylazo group and (iii) a coupler represented by the general formula (A):



(A)

wherein R^{a1} represents a hydrogen atom or substituent; X^{a1} represents a hydrogen atom or a group capable of being separated therefrom upon a coupling reaction with an oxidation product of an aromatic primary amine developing agent; Za, Zb and Zc each represents a methine group, substituted methine group, =N- group or -NH- group; one of the Za-Zb and Zb-Zc bonds is a double bond and the other is a single bond; if the Zb-Zc bond is a carbon-carbon double bond, it may be a part of an aromatic ring; R^{a1} or X^{a1} may form a dimer or higher polymer; and if Za, Zb or Zc is a substituted methine, it may form a dimer or higher polymer.

DETAILED DESCRIPTION OF THE INVENTION

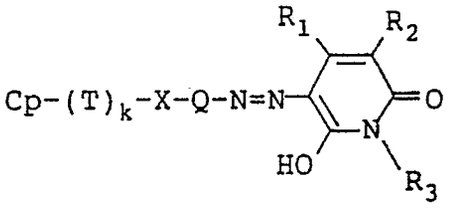
The yellow-colored cyan coupler to be used in the present invention will be further described hereinafter.

The yellow colored cyan coupler of the present invention is a cyan coupler which exhibits a maximum absorption wavelength between 400 nm and 500 nm in the visible absorption range and undergoes coupling with an oxidation product of an aromatic primary amine developing agent to form a cyan dye having a maximum absorption wavelength between 630 nm and 750 nm in the visible absorption range.

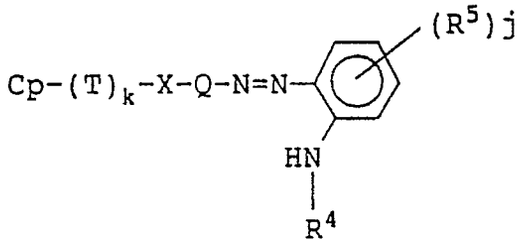
Examples of such yellow-colored cyan couplers include couplers disclosed in JP-A-61-221748 and JP-A-1-319744.

In addition to these yellow-colored cyan couplers, a cyan coupler capable of undergoing reaction with an oxidation product of an aromatic primary amine developing agent to release a group containing a water-soluble 6-hydroxy-2-pyridon-5-ylazo group, a water-soluble pyrazolon-4-ylazo group, a water-soluble 5-aminopyrazol-4-ylazo group, a water-soluble 2-acylaminophenylazo group, or a water-soluble 2-sulfonamidophenylazo group may be preferably used in view of color reproducibility.

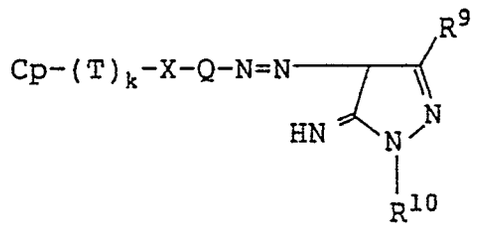
The yellow-colored cyan couplers of the present invention may be preferably represented by one of general formulae (CI) to (CIV):



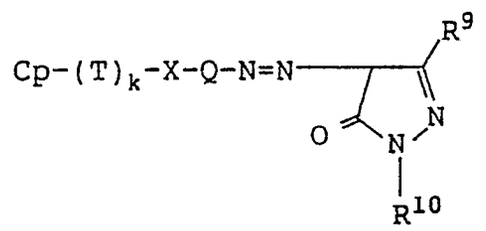
(CI)



(CII)



(CIII)

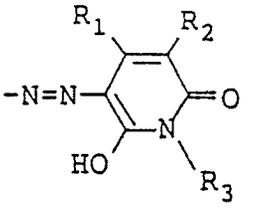


(CIV)

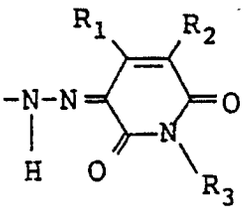
In the general formulae (CI) to (CIV), Cp represents a cyan coupler group (T is connected to the coupling position thereof), T represents a timing group, k represents an integer 0 or 1, X represents a divalent connecting group containing N, O or S by which $(T)_k$ and Q are connected to each other, and Q represents an arylene group or a divalent heterocyclic group.

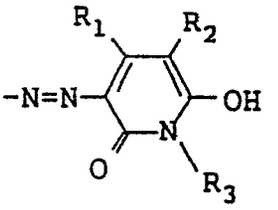
In the general formula (CI), R_1 and R_2 each independently represents a hydrogen atom, a carboxyl group, a sulfo group, a cyano group, an alkyl group, a cycloalkyl group, an aryl group, a heterocyclic group, a carbamoyl group, a sulfamoyl group, a carbonamido group, a sulfonamido group or an alkylsulfonyl group, and R_3 represents a hydrogen atom, an alkyl group, a cycloalkyl group, an aryl group or a heterocyclic group, with the proviso that at least one of T, X, Q, R_1, R_2 and R_3 of formula (CI) contains a water-soluble group (e.g., hydroxyl, carboxyl, sulfo, amino, ammoniumyl, phosphono, phosphino, hydroxysulfonyloxy).

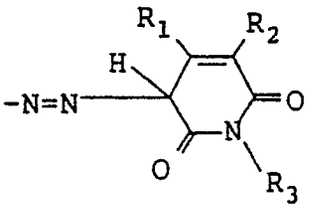
It will be recognized by one shilled in the art that

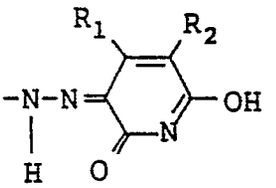


in the general formula (CI) can take any of the following tautomeric structures:

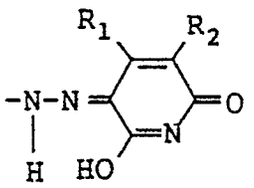




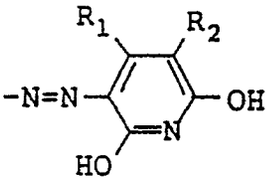




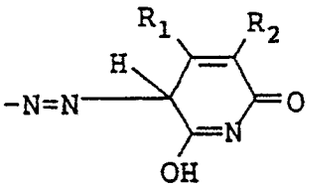
(when R_3 is a hydrogen atom)



(when R_3 is a hydrogen atom)



(when R_3 is a hydrogen atom)



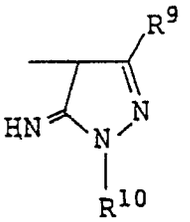
(when R_3 is a hydrogen atom)

These tautomeric structures are within the scope of general formula (CI) of the present invention.

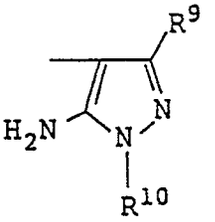
In the general formula (CII), R^4 represents an acyl group or sulfonyl group, R^5 represents a substitutable group, and j represents an integer from 0 to 4. When j is an integer from 2 to 4, the plurality of R^5 groups may be the same or different, with the proviso that at least one of T, X, Q, R_4, and R_5 contains a water-soluble group (e.g., hydroxyl, carboxyl, sulfo, phosphono, phosphino, hydroxysulfonyloxy, amino, ammoniumyl).

In the general formulae (CIII) and (CIV), R^9 represents a hydrogen atom, a carboxyl group, a sulfo group, a cyano group, an alkyl group, a cycloalkyl group, an aryl group, an alkoxy group, a cycloalkyloxy group, an aryloxy group, a heterocyclic group, a carbamoyl group, a sulfamoyl group, a carbonamide group, a sulfonamide group or an alkylsulfonyl group, and R^{10} represents a hydrogen atom, an alkyl group, a cycloalkyl group, an aryl group or a heterocyclic group, with the proviso that at least one of T, X, Q, R^9, and R^{10} contains a water-soluble group (e.g., hydroxyl, carboxyl, sulfo, phosphono, phosphino, hydroxysulfonyloxy, amino, ammoniumyl).

The groups



and

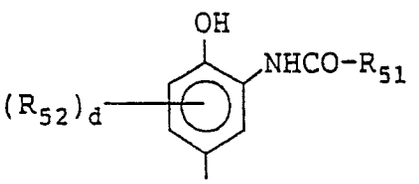


are in tautomeric relationship with each other and thus are the same compound.

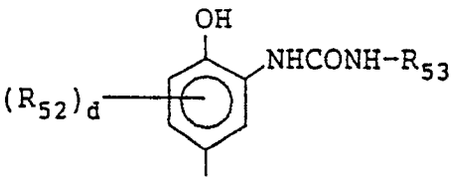
The compounds represented by the general formulae (CI) to (CIV) will be further described hereinafter.

Examples of the coupler group represented by Cp include known cyan coupler groups (e.g., phenolic, naphtholic, diphenylimidazolic, hydroxypyridinic, long wavelength-absorbing pyrazolotriazolic cyan coupler groups).

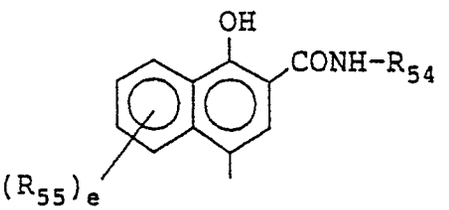
Preferred examples of Cp include coupler groups represented by the following general formulae (Cp-6), (Cp-7) and (Cp-8):



(Cp-6)



(Cp-7)



(Cp-8)

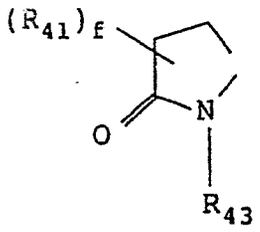
In these general formulae, the free bond extending from the coupling position indicates the position at which a coupling-separable moiety is connected to Cp.

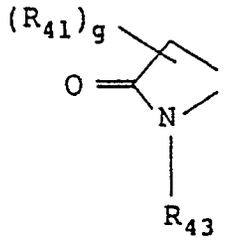
In these general formulae, if R_{51}, R_{52}, R_{53}, R_{54} or R_{55} contains a nondiffusing group, then R_{51}, R_{52}, R_{53}, R_{54} or R_{55} is selected so that the total number of carbon atoms contained in R_{51}, R_{52}, R_{53}, R_{54}, or R_{55} is from 8 to 40, preferably 10 to 30. When R_{51}, R_{52}, R_{53}, R_{54}, and R_{55} do not contain a non-diffusing group, the total number of carbon atoms contained therein is preferably 15 or less. In the case of bis type, telomer type or polymer type couplers, any of the above mentioned substituents represents a divalent group which connects repeating units. In this case, the total number of carbon atoms contained in these substituents may exceed the above specified ranges.

Hereinafter, R_{41} represents an aliphatic, aromatic or heterocyclic group, R_{42} represents an aromatic or heterocyclic group, and R_{43}, R_{44} and R_{45} each represents a hydrogen atom, an aliphatic group, aromatic group or a heterocyclic group.

R_{51}, R_{52}, R_{53}, R_{54}, R_{55}, d and e will be further described hereinafter.

R_{51} has the same meaning as R_{42}. R_{52} has the same meaning as R_{41} or represents $R_{41}CON(R_{43})$-, $R_{41}OCON(R_{43})$-, $R_{41}SO_2N(R_{43})$-, $(R_{43})(R_{44})$-NCON(R_{45})-, $R_{41}O$-, $R_{41}S$-, a halogen atom or $(R_{41})(R_{43})N$-. The suffix d represents an integer from 0 to 3. The suffix e represents an integer from 0 to 4. When d is plural, the plurality of R_{52} groups represent the same substituent or different substituents. R_{52} may be several divalent groups which are connected to each other to form a cyclic structure. Typical examples of divalent groups for the formation of a cyclic structure include:





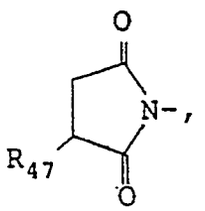
wherein f represents an integer from 0 to 4; and g represents an integer from 0 to 2; when e is plural, the plurality of R_{55} groups represent the same substituent or different substituents; R_{53} has the same meaning as R_{41}; R_{54} has the same meaning as R_{41}. R_{55} has the same meaning as R_{41} or represents $R_{41}OCONH$-, $R_{41}SO_2NH$-, $(R_{43})(R_{44})NCON(R_{45})$-, $(R_{43})(R_{44})NSO_2N(R_{45})$-, $R_{43}O$-, $R_{41}S$-, a halogen atom or $(R_{41})(R_{43})N$- group. When there is a plurality of R_{55} groups, they may be the same or different.

In the foregoing description, the aliphatic group is a C_{1-32}, preferably C_{1-22} saturated or unsaturated, acyclie or cyclic, straight-chain or branched, substituted or unsubstituted aliphatic hydrocarbon group. Typical examples of such an aliphatic group include methyl, ethyl, propyl, isopropyl, butyl, (t)-butyl, (i)butyl, (t)amino, hexyl, cyclohexyl, 2-ethylhexyl, octyl, 1,1,3,3-tetramethylbutyl, decyl, dodecyl, hexadecyl, and octadecyl groups.

The aromatic group is a C_{6-20} aromatic group, and preferably is a substituted or unsubstituted phenyl group or a substituted or unsubstituted naphthyl group.

The heterocyclic group is a C_{1-20}, preferably C_{1-7}, preferably 3- to 8-membered substituted or unsubstituted heterocyclic group containing a hetero atom selected from nitrogen, oxygen and sulfur atoms. Typical examples of such a heterocyclic group include 2-pyridyl, 2-thienyl, 2-furyl, 1,3,4-thiadiazol-2-yl, 2,4-dioxo-1,3-imidazolidin-5-yl, 1,2,4-triazol-2-yl, and 1-pyrazolyl.

If the above mentioned aliphatic hydrocarbon group, aromatic group and heterocyclic group contain substituents, typical examples of such substituents include a halogen atom, an $R_{47}O$- group, an $R_{46}S$- group, an $R_{47}CON(R_{48})$- group, an $(R_{47})(R_{48})NCO$- group, an $R_{46}OCON(R_{48})$- group, an $R_{46}SO_2N(R_{47})$- group, an $(R_{47})(R_{48})NSO_2$- group, an $R_{46}SO_2$- group, an $R_{47}OCO$- group, an $(R_{47})(R_{48})NCON(R_{49})$- group, groups having the same meaning as R_{46},



an $R_{46}COO$- group, an $R_{47}OSO_2$- group, a cyano group, and a nitro group, wherein R_{46} represents an aliphatic group, aromatic group or heterocyclic group, and R_{47}, R_{48} and R_{49} each represents an aliphatic group, aromatic group, heterocyclic group or hydrogen atom. The aliphatic group, aromatic group and heterocyclic group are as defined above in connection with R_{41}, R_{42}, R_{43} and R_{44}.

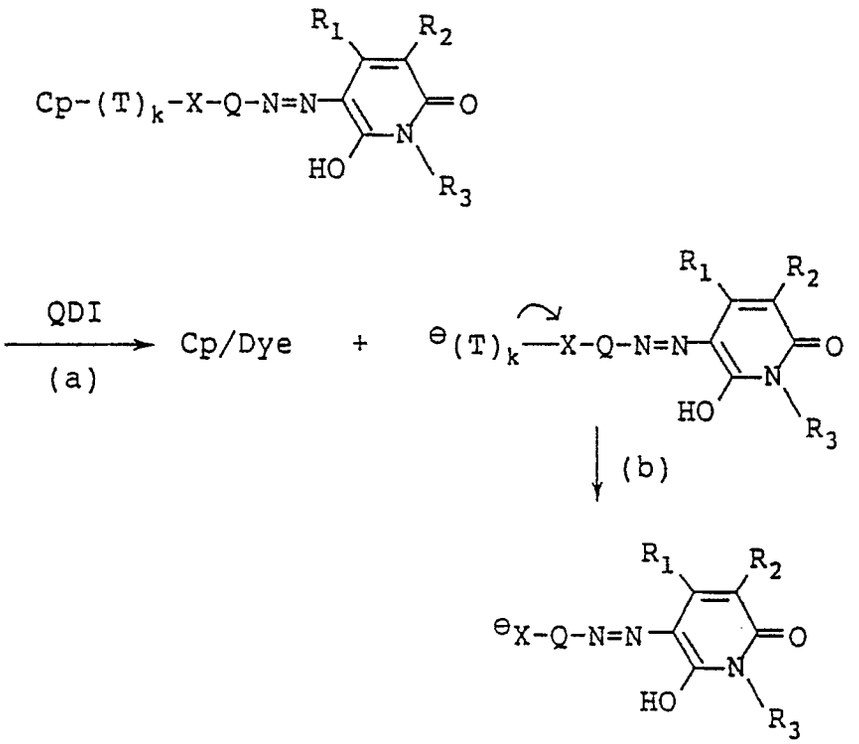
In the general formula (Cp-6), R_{51} is preferably an aliphatic group or aromatic group. R_{52} is preferably a chlorine atom, aliphatic group or $R_{41}CONH$- group. The suffix d is preferably 1 or 2. R_{53} is preferably an aromatic group.

In the general formula (Cp-7), R_{52} is preferably an $R_{41}CONH$- group. The suffix d is preferably 1. R_{54} is preferably an aliphatic group or aromatic group.

In the general formula (Cp-8), e is preferably 0 or 1. R_{55} is preferably an $R_{41}OCONH$- group, an $R_{41}CONH$-group or an $R_{41}SO_2NH$- group. These substituents may be preferably connected to the 5-position of the naphthol ring.

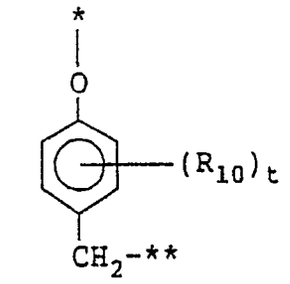
The timing group represented by T is a group which causes cleavage of its bond to X after cleavage of its bond to Cp by a coupling reaction of a coupler with an oxidation product of an aromatic primary amine developing agent. The timing group T is used for various purposes such as adjusting coupling reactivity, stabilizing couplers and adjusting the timing of release of the X containing moiety.

A reaction scheme of cleavage at a development processing, for example, in a case of (Cl), is shown below.

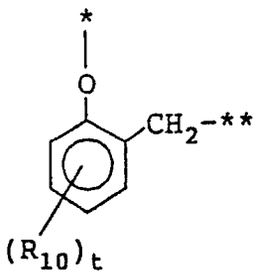


A reaction step (a) is based on a coupling reaction of quinonediimine (QDI) and a coupler, which is a well known reaction in the art. In a cleavage reaction of timing group comprising a step (b) (when q represents 1), the timing group and the cleavage reaction thereof are known in the art. For instance, timing groups (T-1) and (T-2) are disclosed in U.S. Patent 4,409,323, (T-3) is disclosed in U.S. Patent 4,421,845, (T-4) is disclosed in U.S. Patent 4,248,962, (T-5) is disclosed in U.S. Patent 4,652,516, (T-6) is disclosed in U.S. Patent 4,146,396 and (T-7) is disclosed in GB Patent 1,531,927.

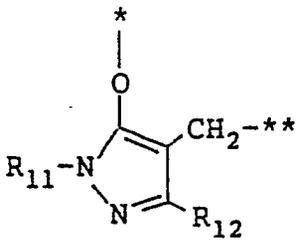
Examples of the timing group T include the following known groups (the marks * and ** indicate the position at which the timing group is connected to Cp and X, respectively):



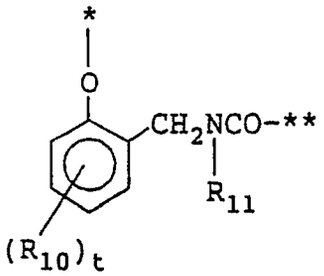
(T-1)



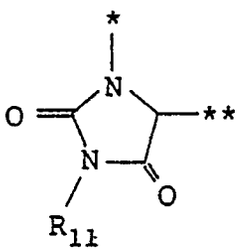
(T-2)



(T-3)



(T-4)

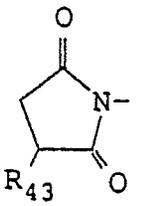


(T-5)

(T-6)

(T-7)

In these general formulae, R_{10} represents a group capable of substituting to the benzene ring, R_{11} has the same meaning as R_{41}, and R_{12} represents a hydrogen atom or substituent. The suffix t represents an integer 0 to 4. Examples of substituents represented by R_{10} and R_{12} include R_{41}, a halogen atom, $R_{43}O$-, $R_{43}S$-, $R_{43}(R_{44})NCO$-, $R_{43}OOC$-, $R_{43}SO_2$-, $R_{43}(R_{44})NSO_2$-, $R_{43}CON(R_{43})$-, $R_{41}SO_2N$-(R_{43})-, $R_{43}CO$-, $R_{41}COO$-, $R_{41}SO$-, nitro, $R_{43}(R_{44})NCON$-(R_{45})-, cyano, $R_{41}OCON(R_{43})$-, $R_{43}OSO_2$-, $R_{43}(R_{44})N$-, $R_{43}(R_{44})$-$NSO_2N(R_{45})$, and



groups. The terms R_{41}, R_{42}, R_{43}, R_{44}, and R_{45} are as defined above.

The suffix k represents an integer 0 or 1. In general, k is preferably 0, that is, Cp and X are preferably directly connected to each other.

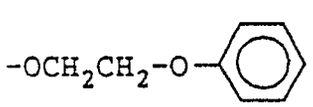
X is a divalent connecting group which is connected to the $(T)_k$ containing moiety via a N, O or S atom of X. Preferred examples of such a divalent connecting group include -O-, -S-, -OCO-, -OCOO-, -OCOS-, -OCONH-, $-OSO_2-$, $-OSO_2NH-$ or a nitrogen containing heterocyclic group which is connected to the $(T)_k$ containing moiety via its nitrogen atom (e.g., groups derived from pyrrolidine, piperidine, morpholine, piperadine, pyrrole, pyrazole, imidazole, 1,2,4-triazole, benzothiazole, succinimido, phthalimido, oxazolidin-2,4-dione, imidazolidin-2,4-dione, 1,2,4-triazolidin-3,5-dione), and connecting groups obtained by combining these groups with an alkylene group (e.g., 1,4-cyclohexylene), an arylene group (e.g., o-phenylene, p-phenylene), a divalent heterocyclic group (e.g., groups derived from pyridine, thiphene), -CO-, $-SO_2-$, -COO-, -CONH-, $-SO_2NH-$, $-SO_2O-$, -NHCO-, $-NHSO_2-$, -NHCONH-, $-NHSO_2NH-$, -NHCOO- group, etc. X is more preferably represented by the general formula (II):

$$*-X_1-(L-X_2)_m-** \quad (II)$$

wherein * indicates the position at which it is connected to the $(T)_k$ containing moiety; ** indicates the position at which it is connected to the Q containing moiety; X_1 represents -O- or -S-; L represents an alkylene group; X_2 represents a single bond, -O-, -S-, -CO-, $-SO_2-$, -OCO-, -COO-, -NHCO-, -CONH-, $-SO_2NH-$, $-NHSO_2-$, $-SO_2O-$, $-OSO_2-$, -OCOO-, -OCONH-, -NHCOO-, -NHCONH-, $-NHSO_2NH-$, -OCOS-, -SCOO-, $-OSO_2NH-$ or $-NHSO_2O-$; and m represents an integer from 0 to 3. The total number of carbon atoms contained in X is preferably from 0 to 12, more preferably, 0 to 8. X is most preferably $-OCH_2CH_2O-$.

Q represents an arylene group or divalent heterocyclic group. If Q is an arylene group, it may be a condensed ring or it may contain substituents (e.g., halogen atom, hydroxyl, carboxyl, sulfo, nitro, cyano, amino, ammonium, phosphono, phosphino, alkyl, cycloalkyl, aryl, carbonamido, sulfonamido, alkoxy, aryloxy, acyl, sulfonyl, carboxyl, carbamoyl, sulfamoyl). The total number of carbon atoms contained in Q is preferably in the range of 6 to 15, more preferably, 6 to 10. If Q is a divalent heterocyclic group, the heterocyclic group is a 3- to 8-membered, preferably 5-to 7-membered, single or condensed heterocyclic group containing at least one hetero atom selected from N, O, S, P, Se and Te atoms (e.g., groups derived from pyridine, thiophene, furan, pyrrole, pyrazole, imidazole, thiazole, oxazole, benzothiazole, benzoxazole, benzofuran, benzothiophene, 1,3,4-thiadiazole, indole, quinoline). The heterocyclic group may contain substituents (the same as those contained in the arylene group represented by Q). The number of carbon atoms contained in Q is preferably in the range of 2 to 15, and more preferably, 2 to 10. Q is most preferably a 1,4-phenylene group.

Accordingly, $-(T)_k-X-Q-$ is most preferably represented by the following formula:



If R_1, R_2 or R_3 in general formula (CI) is an alkyl group, the alkyl group may be either straight-chain or branched or may contain unsaturated bonds or substituents (e.g., a halogen atom, a hydroxyl, carboxyl, sulfo, phosphono, phosphino, cyano, alkoxy, aryl, alkoxycarbonyl, amino, ammoniumyl, acyl, carbonamide, sulfonamide, carbamoyl, sulfamoyl, or sulfonyl group).

If R_1, R_2 or R_3 is a cycloalkyl group, it is a 3- to 8-membered cycloalkyl group which may contain crosslinking groups, unsaturated bonds or substituents (the same substituents as those contained in the alkyl group represented by R_1, R_2 or R_3).

If R_1, R_2 or R_3 is an aryl group, it may be a condensed ring or may contain substituents such as those

contained in the alkyl group represented by R_1, R_2 or R_3, an alkyl and a cycloalkyl group.

If R_1, R_2 or R_3 is a heterocyclic group, it is a 3- to 8-membered, preferably 5- to 7-membered, single or condensed heterocyclic group containing at least one hetero atom selected from N, S, O, P, Se and Te atoms (e.g., imidazolyl, thienyl, pyrazolyl, thiazolyl, pyridyl, quinolinyl). The heterocyclic group may contain substituents (the same as those contained in the aryl group represented by R_1, R_2 or R_3).

In general formula (CI), the carboxyl group may be a carboxylate group, the sulfo group may be a sulfonate group, the phosphino group may be a phosphinate group, and the phosphono group may be a phosphonate group. Examples of paired (counter) ions contained in these groups include Li^+, Na^+, K^+ and ammonium.

R_1 is preferably a hydrogen atom, a carboxyl group, a C_{1-10} alkyl group (e.g., methyl, t-butyl, carbomethyl, 2-sulfomethyl, carboxymethyl, 2-carboxymethyl, 2-hydroxymethyl, benzyl, ethyl, isopropyl) or a C_{6-12} aryl group (e.g., phenyl, 4-methoxyphenyl, 4-sulfophenyl). Particularly preferred among these groups are a hydrogen atom, a methyl group, and a carboxyl group.

R_2 is preferably a cyano group, a carboxyl group, a C_{1-10} carbamoyl group, a C_{0-10} sulfamoyl group, a sulfo group, a C_{1-10} alkyl group (e.g., methyl, sulfomethyl), a C_{1-10} sulfonyl group (e.g., methylsulfonyl, phenylsulfonyl), a C_{1-10} carbonamide group (e.g., acetamide, benzamide) or a C_{1-10} sulfonamide group (e.g., methanesulfonamide, toluenesulfonamide). Particularly preferred among these groups are a cyano group, a carbamoyl group, and a carboxyl group.

R_3 is preferably a hydrogen atom, a C_{1-12} alkyl group (e.g., methyl, sulfomethyl, carboxymethyl, 2-sulfomethyl, 2-carboxymethyl, ethyl, n-butyl, benzyl, 4-sulfobenzyl) or a C_{6-15} aryl group (e.g., phenyl, 4-carboxyphenyl, 3-carboxyphenyl, 4-methoxyphenyl, 2,4-dicarboxyphenyl, 2-sulfophenyl, 3-sulfophenyl, 4-sulfophenyl, 2,4-disulfophenyl, 2,5-disulfophenyl). R_3 is more preferably a C_{1-7} alkyl group or a C_{6-10} aryl group.

R_4 in general formula (CII) is an acyl group represented by the general formula (III) or a sulfonyl group represented by the general formula (IV):

$$R_{11}CO- \quad (III)$$

$$R_{11}SO_2- \quad (IV)$$

In these formulae, R_{11} is an alkyl, cycloalkyl, aryl or heterocyclic groups.

The alkyl group represented by R_{11} may be either straight-chain or branched, or may contain unsaturated bonds or substituents (e.g., halogen atom, hydroxyl, carboxyl, sulfo, phosphono, phosphino, cyano, alkoxy, aryl, alkoxycarbonyl, amino, ammoniumyl, acyl, carbonamide, sulfonamide, carbamoyl, sulfamoyl, sulfonyl).

The cycloalkyl group represented by R_{11} may be a 3- to 8-membered cycloalkyl group or may contain crosslinking groups, unsaturated bonds or substituents (the same as those which can be contained in the alkyl group represented by R_{11}).

The aryl group represented by R_{11} may be a condensed ring or may contain substituents (e.g., the same substituents as those which can be contained in the alkyl group represented by R_{11}, and, in addition, an alkyl group, and a cycloalkyl group).

The heterocyclic group represented by R_{11} is a 3- to 8-membered, preferably 5- to 7-membered single or condensed heterocyclic group containing at least one hetero atom selected from N, S, O, P, Se and Te atoms (e.g., imidazolyl, thienyl, pyrazolyl, thiazolyl, pyridyl, quinolinyl). The heterocyclic group may contain substituents (the same as those which can be contained in the aryl group represented by R_{11}).

The carboxyl substituent may be a carboxylate group, the sulfo substituent may be a sulfonate group, the phosphino substituent may be a phosphinate group, and the phosphono substituent may be a phosphonate group. Examples of paired (counter) ions contained in these groups include Li^+, Na^+, K^+ and ammonium.

R_{11} is preferably a C_{1-10} alkyl group (e.g., methyl, carboxymethyl, sulfoethyl, cyanoethyl), a C_{5-8} cycloalkyl group (e.g., cyclohexyl, 2-carboxycyclohexyl) or a C_{6-10} aryl group (e.g., phenyl, 1-naphthyl, 4-sulfophenyl). Particularly preferred among these groups are a C_{1-3} alkyl group, and a C_6 aryl group.

R_5 is a substituent group, preferably an electron-donating group, particularly $-NR_{12}R_{13}$ or $-OR_{14}$. The position at which R_5 is connected to the benzene ring is preferably the 4-position. R_{12}, R_{13} and R_{14} each represents a hydrogen atom, an alkyl group, a cycloalkyl group, an aryl group or a heterocyclic group. R_{12} and R_{13} may together form a ring. The nitrogen-containing heterocyclic group is preferably aliphatic.

The suffix j represents an integer from 0 to 4, preferably 1 or 2, most preferably 1.

The alkyl group represented by R_9 or R_{10} may be either straight-chain or branched or may contain unsaturated bonds or substituents (e.g., halogen atom, hydroxyl, carboxyl, sulfo, phosphono, phosphino, cyano, alkoxy, aryl, alkoxycarbonyl, amino, ammoniumyl, acyl, carbonamide, sulfonamide, carbamoyl, sulfamoyl, sulfonyl).

The cycloalkyl group represented by R_9 or R_{10} may be a 3- to 8-membered cycloalkyl group or may contain crosslinking groups, unsaturated bonds or substituents (examples of the substituents are the same as those described above as substituents for the alkyl group represented by R_9 or R_{10}).

The aryl group represented by R_9 or R_{10} may be a condensed ring or may contain substituents (e.g., the same as those which can be contained in the alkyl group represented by R_9 or R_{10}, and in addition alkyl, or cycloalkyl).

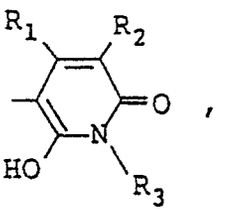
The heterocyclic group represented by R_9 or R_{10} is a 3- to 8-membered, preferably a 5- to 7-membered heterocyclic group, containing at least one hetero atom selected from N, S, O, P, Se and Te atoms (imidazolyl, thienyl, pyrazolyl, thiazolyl, pyridyl, quinolinyl). The heterocyclic group may contain substituents (the same as those contained in the aryl group represented by R_9 or R_{10}).

The carboxyl substituent may be a carboxylate group, the sulfo substituent may be a sulfonate group, the phosphino substituent may be a phosphinate group, and the phosphono substituent may be a phosphonate group. Examples of paired (counter) ions in these groups include Li^+, Na^+, K^+ and ammonium.

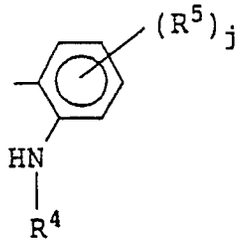
R_9 is preferably a cyano group, a carboxyl group, a C_{1-10} carbamoyl group, a C_{2-10} alkoxycarbonyl group, a C_{7-11} aryloxycarbonyl group, a C_{0-10} sulfamoyl group, sulfo group, a C_{1-10} alkyl group (e.g., methyl, carboxymethyl, sulfomethyl), a C_{1-10} sulfonyl group (e.g., methylsulfonyl, phenylsulfonyl), a C_{1-10} carbonamido group (e.g., acetamido, benzamido), a sulfonamido group (e.g., methanesulfonamido, toluenesulfonamido), an alkyloxy group (e.g., methoxy, ethoxy) or an aryloxy group (e.g., phenoxy). Particularly preferred among these groups are a cyano group, a carbamoyl group, an alkoxycarbonyl group, and a carboxyl group.

R_{10} is preferably a hydrogen atom, a C_{1-12} alkyl group (e.g., methyl, sulfomethyl, carboxymethyl, ethyl, 2-sulfoethyl, 2-carboxyethyl, 3-sulfopropyl, 3-carboxypropyl, 5-sulfopentyl, 5-carboxypentyl, 4-sulfobenzyl) or a C_{6-15} aryl group (e.g., phenyl, 4-carboxyphenyl, 3-carboxyphenyl, 2,4-dicarboxyphenyl, 4-sulfophenyl, 3-sulfophenyl, 2,5-disulfophenyl, 2,4-disulfophenyl). R_{10} is more preferably a C_{1-7} alkyl group or C_{6-10} aryl group.

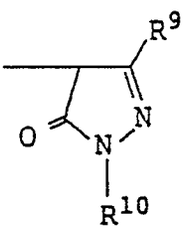
Specific examples of Cp, X, Q,



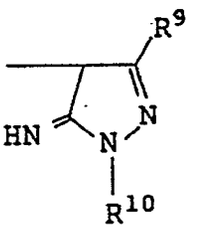
,



,

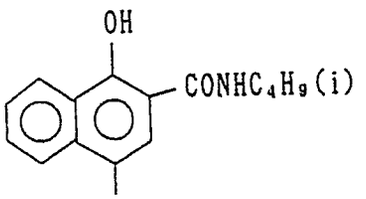


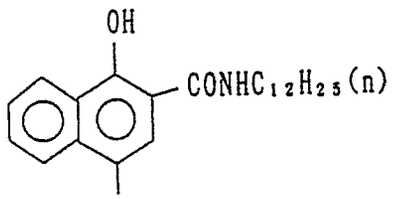
, and

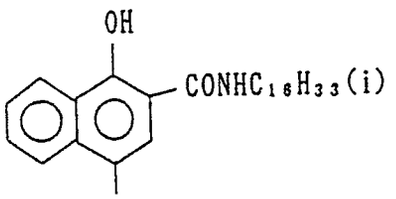


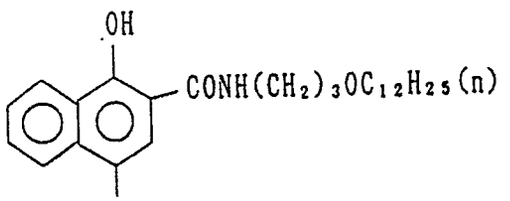
are be set forth below.

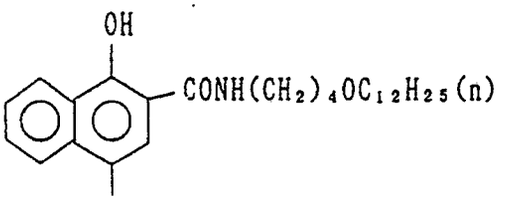
Examples of Cp:

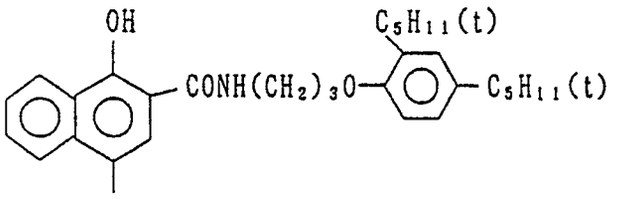


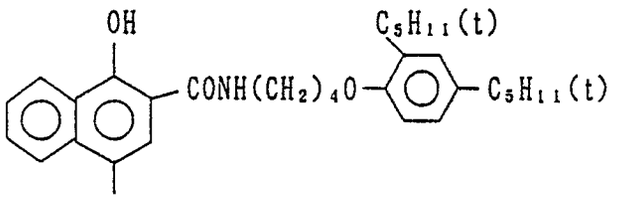


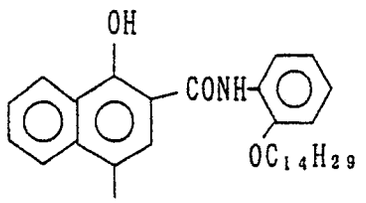


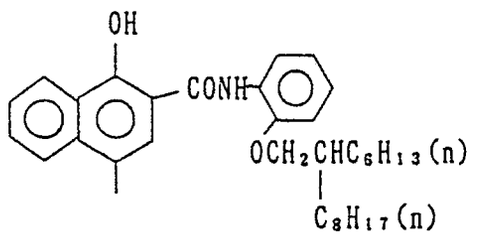


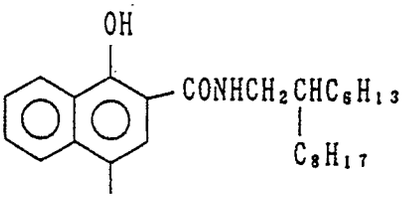

OH
CONH(CH2)4OC12H25(n)

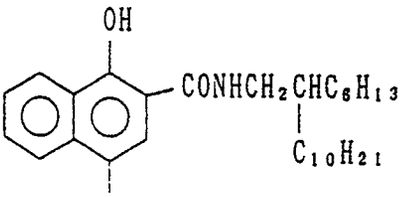

OH
C5H11(t)
CONH(CH2)3O
C5H11(t)

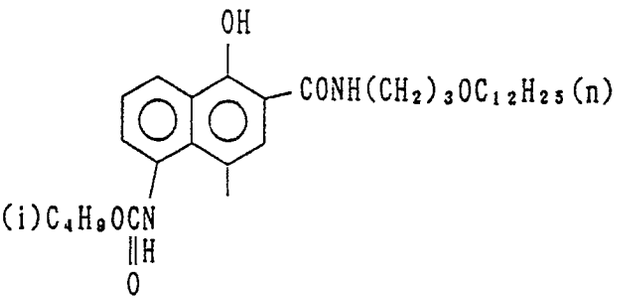

OH
C5H11(t)
CONH(CH2)4O
C5H11(t)

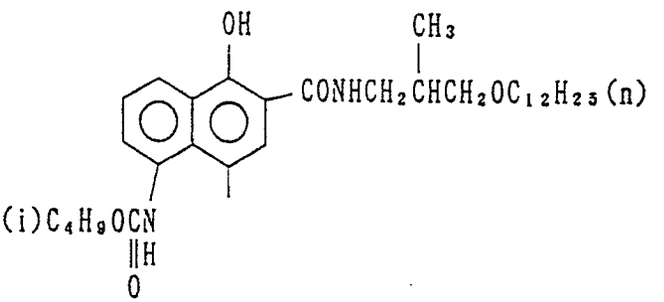

OH
CONH
OC14H29

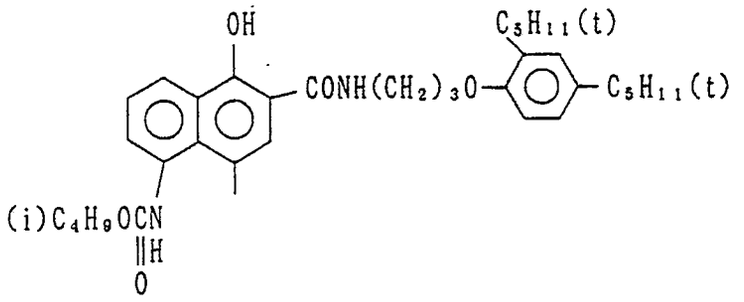

OH
CONH
OCH2CHC6H13(n)
C8H17(n)

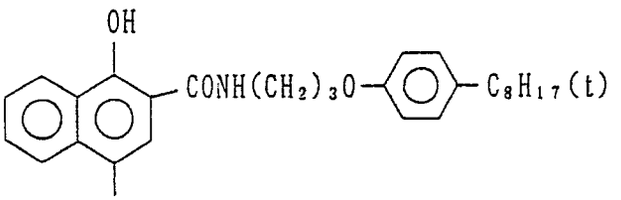


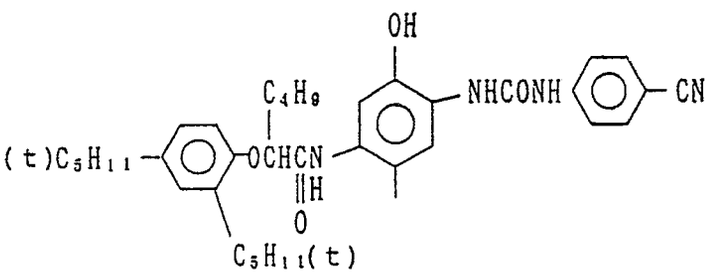


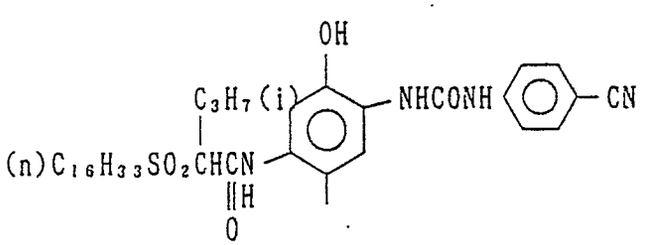


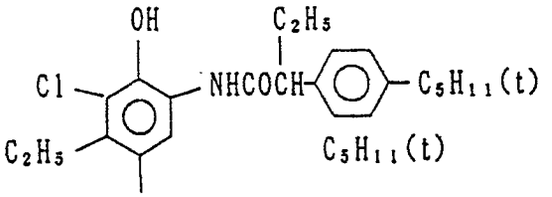


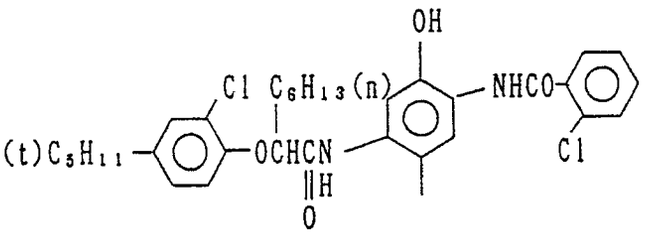


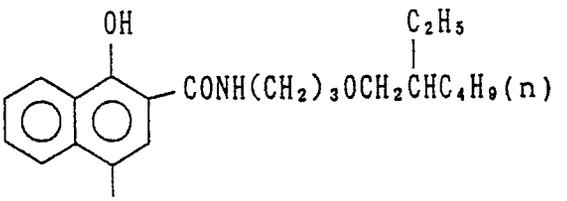


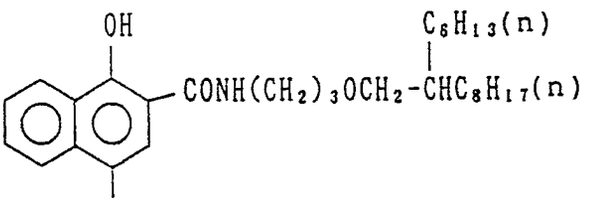


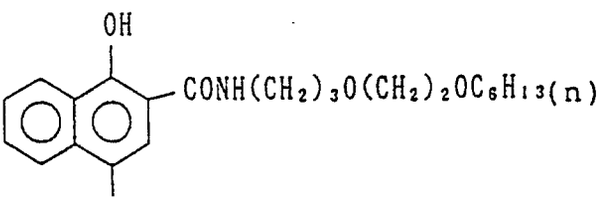


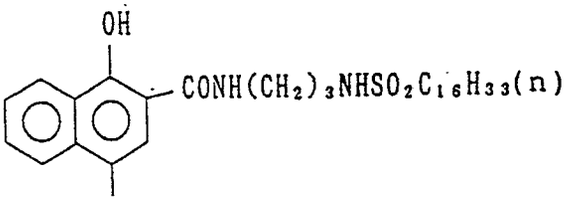


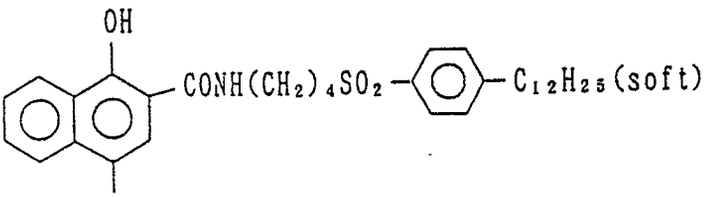


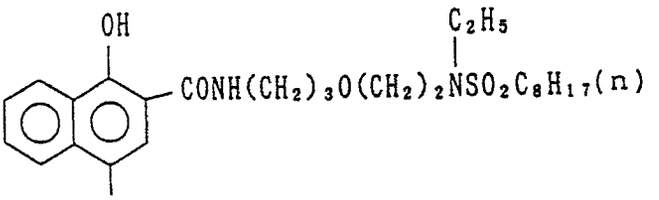


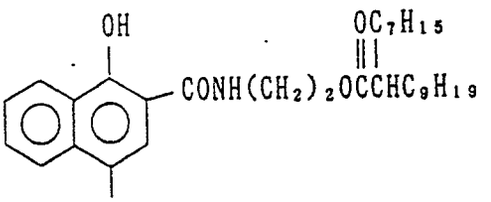


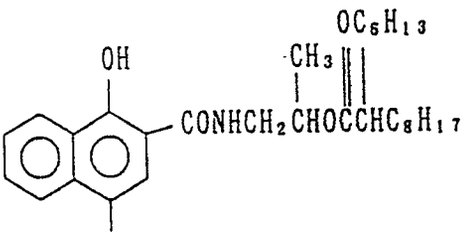








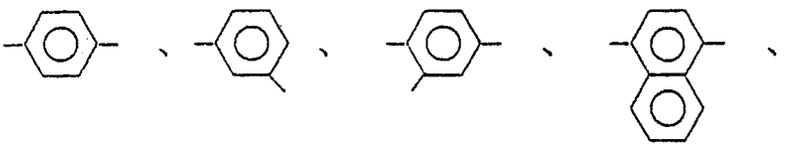


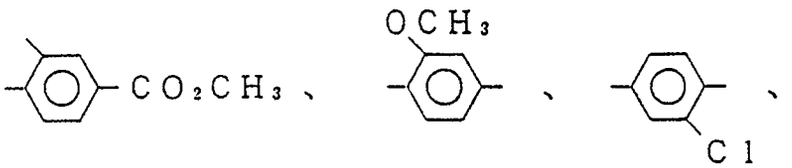


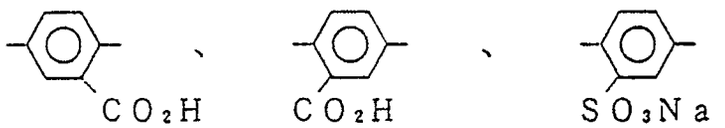
Examples of X:

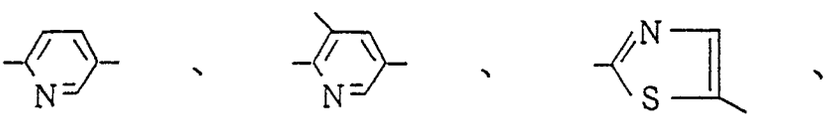
-O-, -S-, -OCH_2-, -OCH_2CH_2-, -OCH_2CH_2O-, -$OCH_2CH_2CH_2O$-, -$O(CH_2CH_2O)_2$-, -OCH_2CH_2S-, -OCH_2CH_2NHCO-, -$OCH_2CH_2NHSO_2$-, -$OCH_2CH_2SO_2$-, -OCH_2CH_2OCO-, -OCH_2CH_2CO-, -SCH_2CONH-, -SCH_2COO-, -$OCH(CH_3)CONH$-, -$OCH_2CH_2OSO_2$-, -OCO-, -$OCH_2CH(COOH)$-, -$OCH_2CH(COOH)CH_2$-, -$OCH_2CH(COOH)O$-, -$OCH(COOH)CH_2O$-, -$OCH_2CH(COOH)S$-, -$OCH_2CH(SO_3Na)O$-

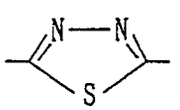
Examples of Q:



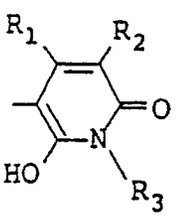




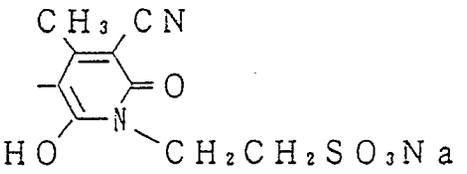


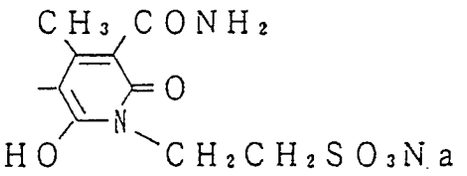


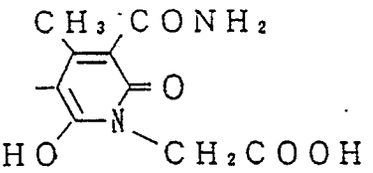
Examples of the group

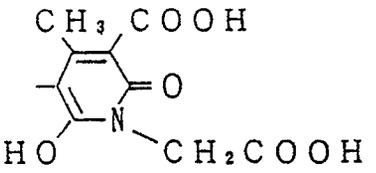


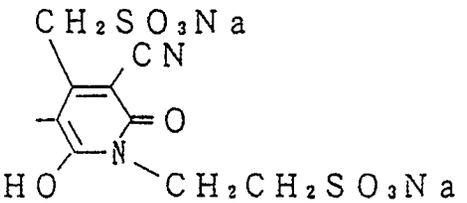
in formula (CI) are shown below.

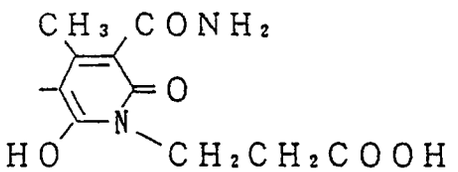


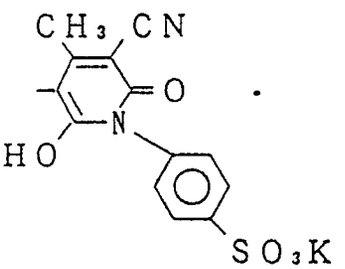


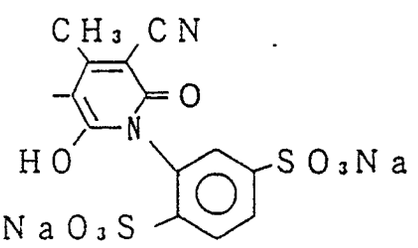


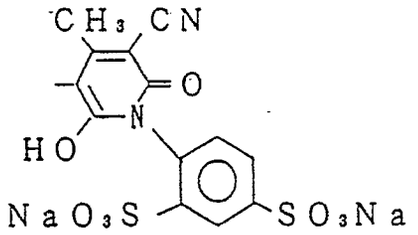


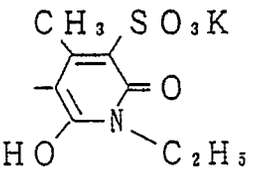


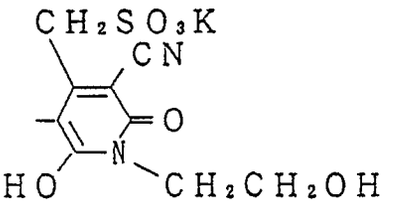


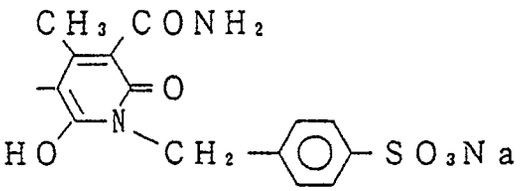


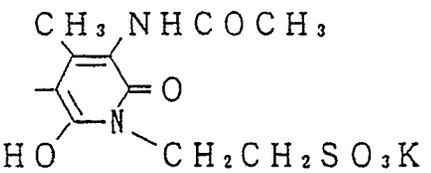


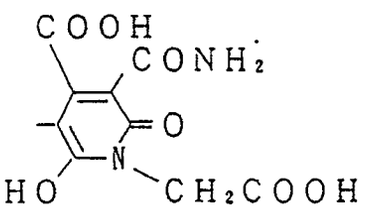


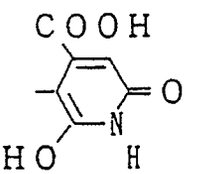


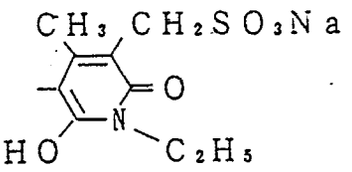


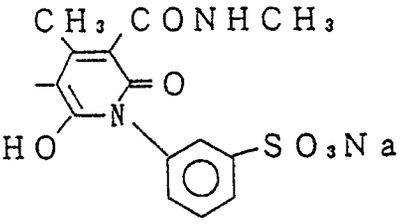


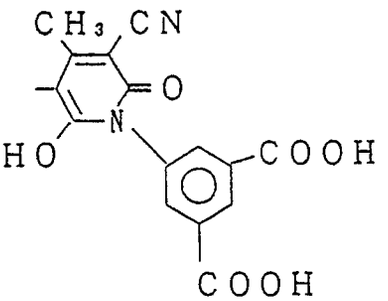


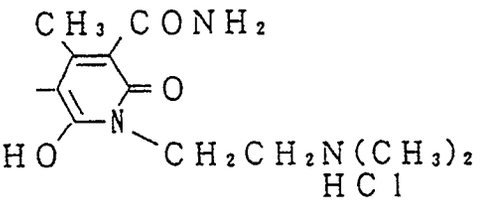


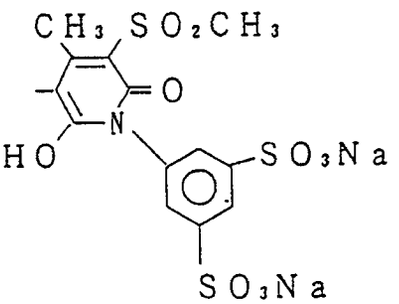


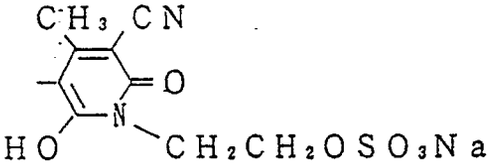


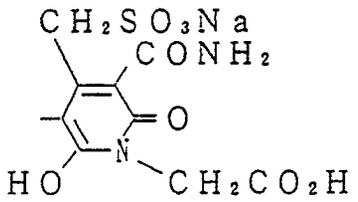


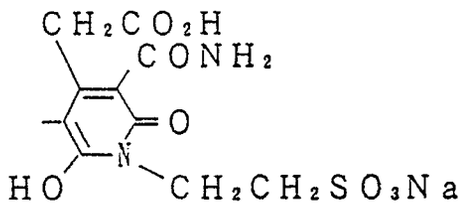


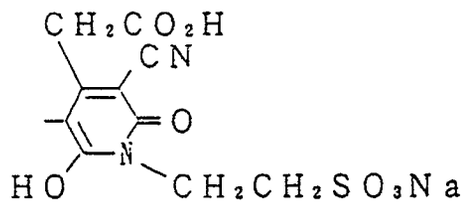


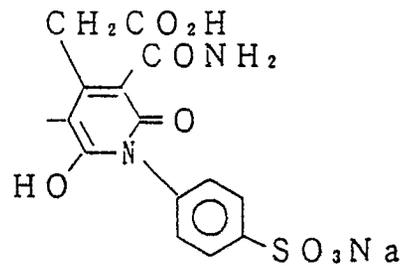


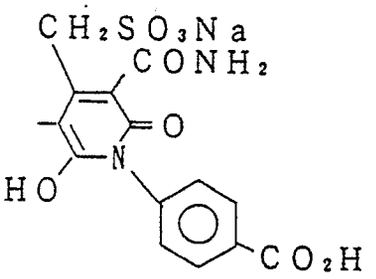


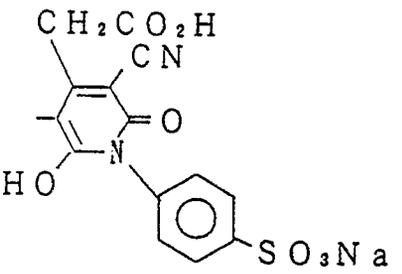


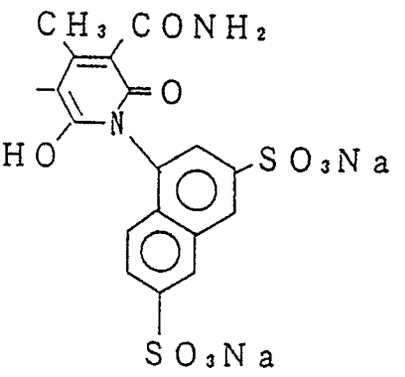


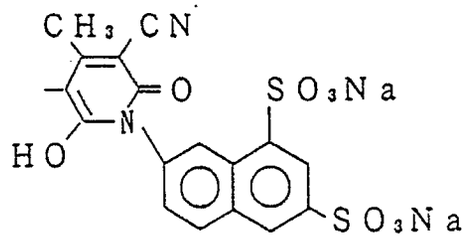


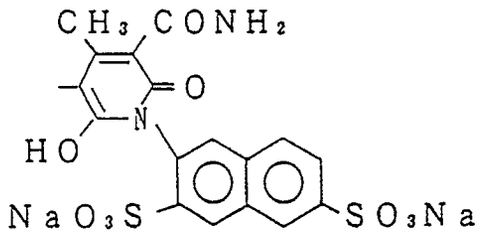


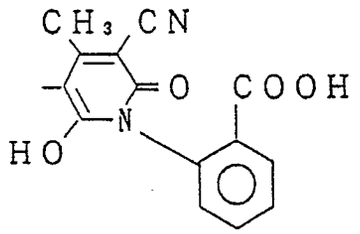


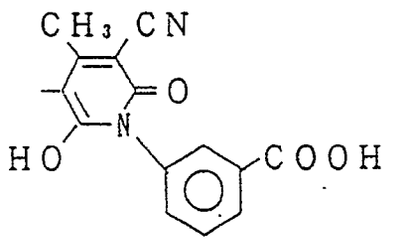


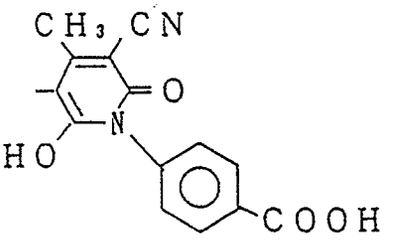


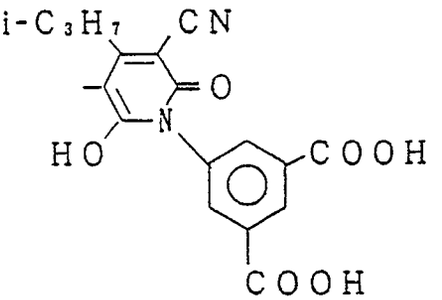


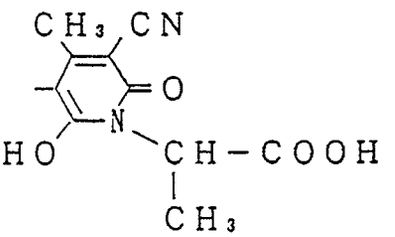


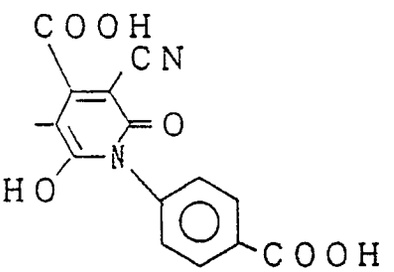




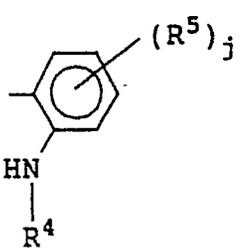




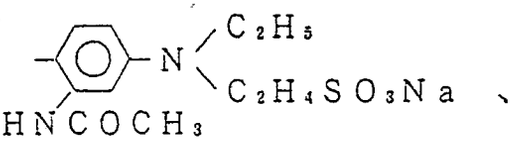


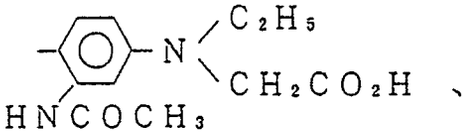


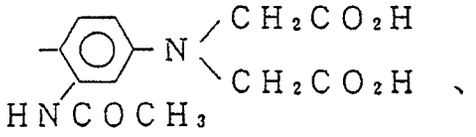
Examples of

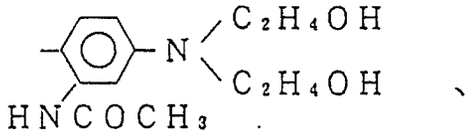


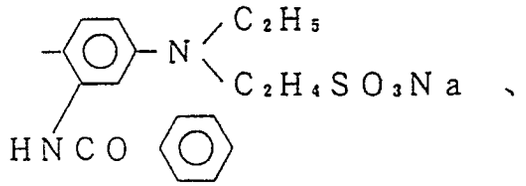
in formula (CII) include:

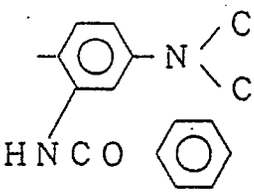




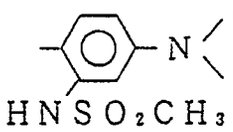




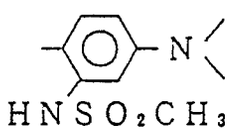




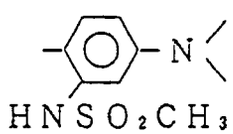
、



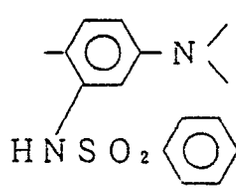
、



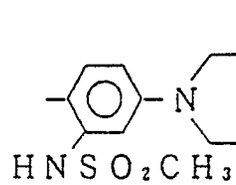
、



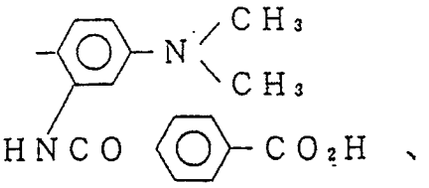
、

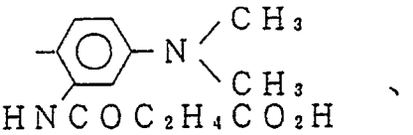


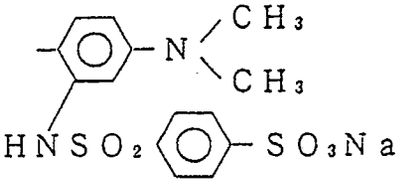
、



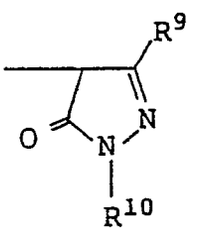
、



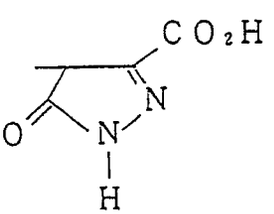


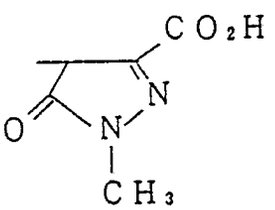


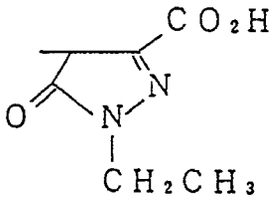
Examples of the group



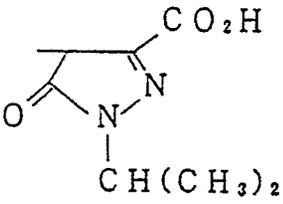
in formula (CIV) include:



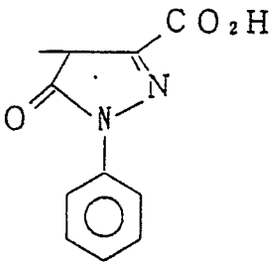




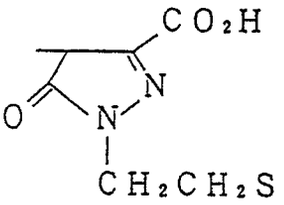
、



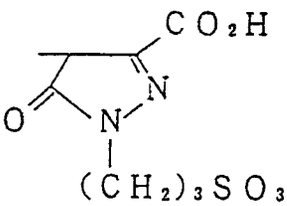
、



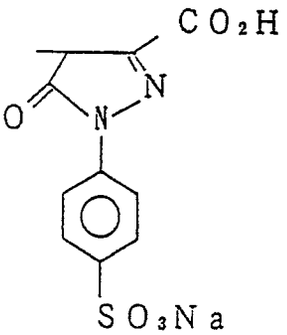
、



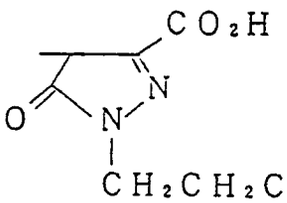
、



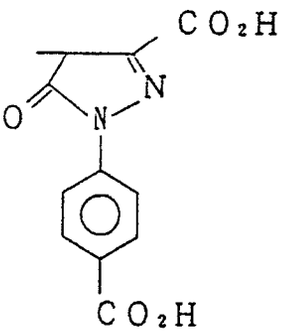
、



、

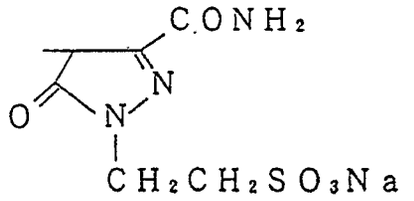


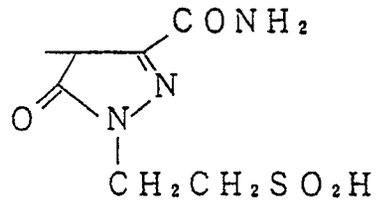
、

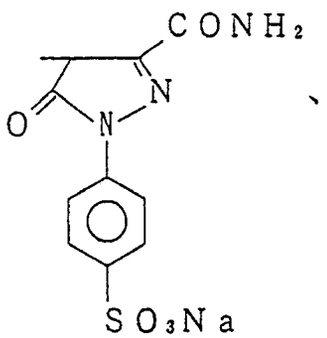


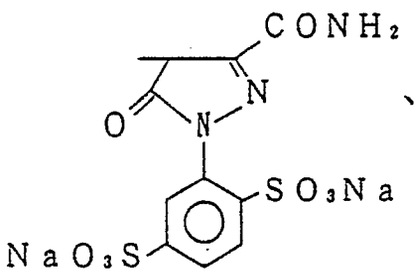
、

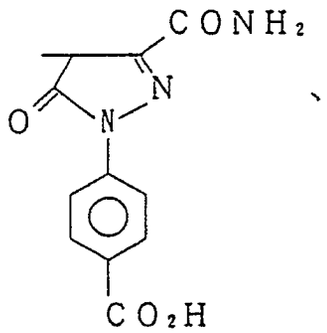
34

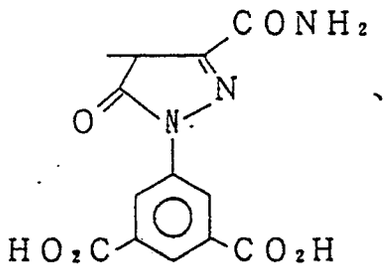


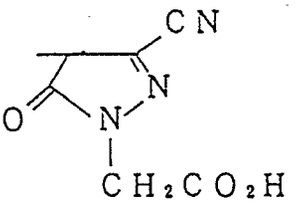




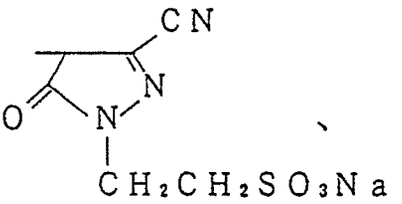




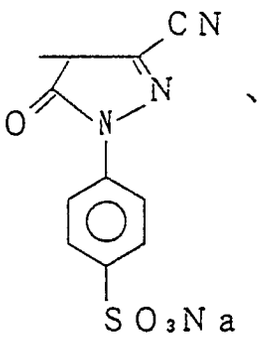




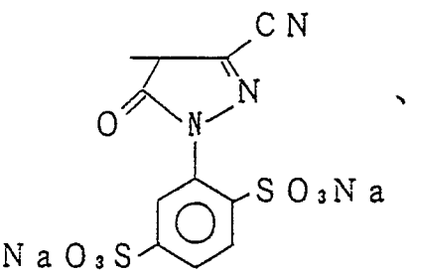
、



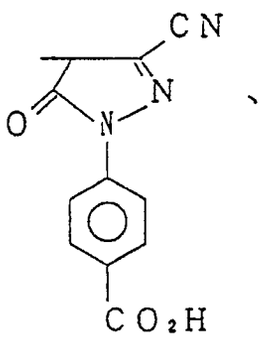
、



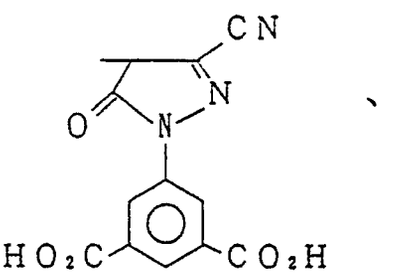
、



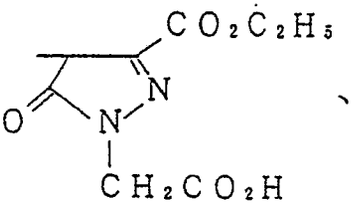
、

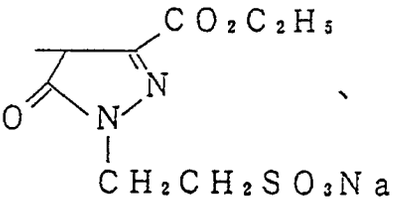


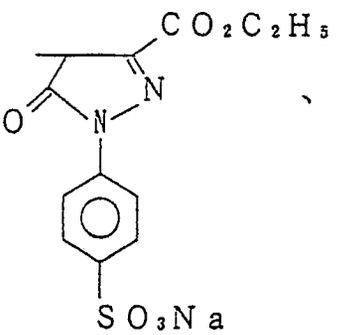
、

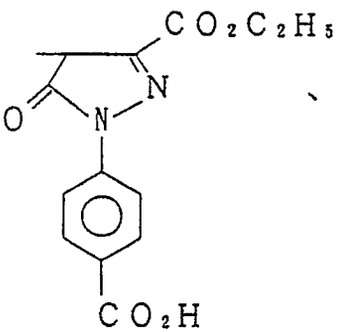


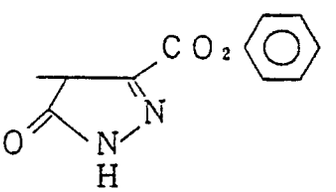
、

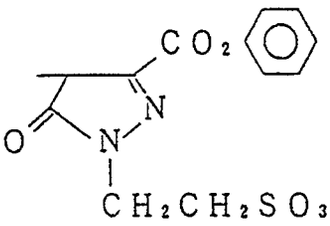


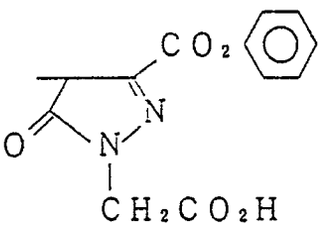


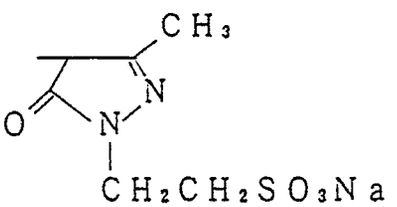


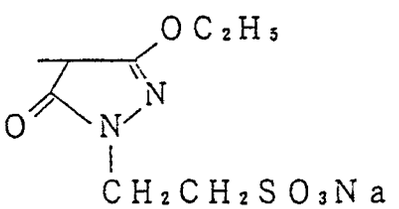


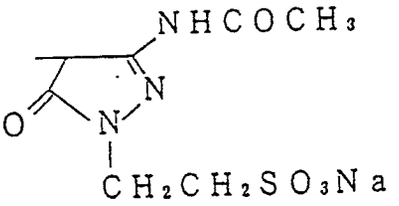


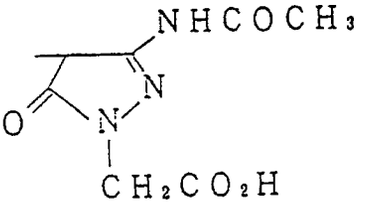


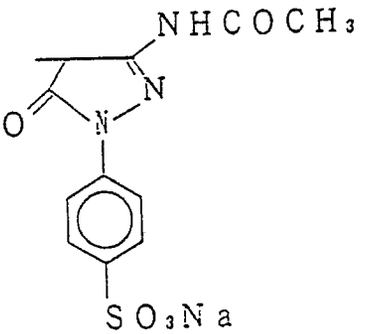


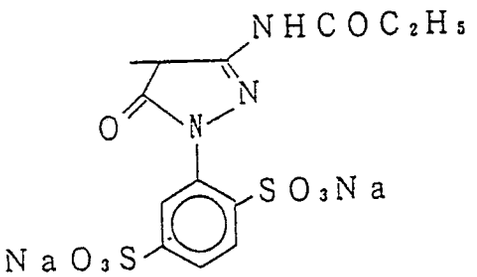


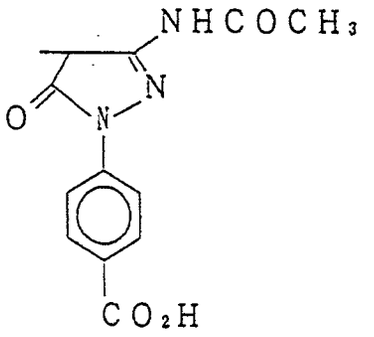


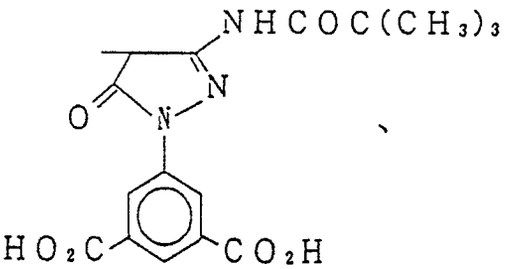


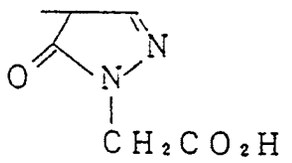


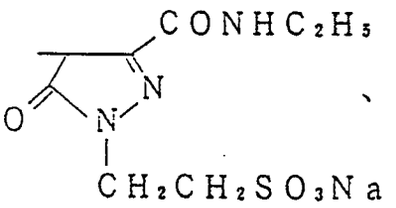




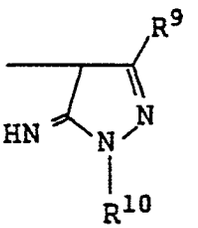




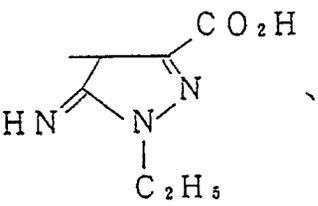


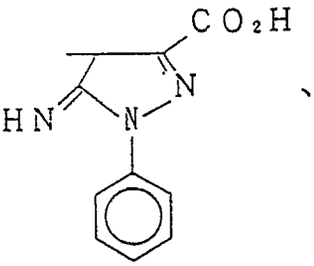


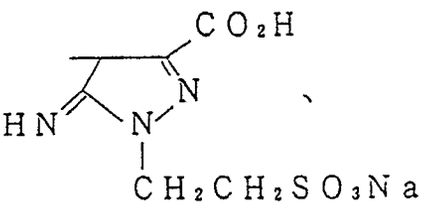
Examples of the group

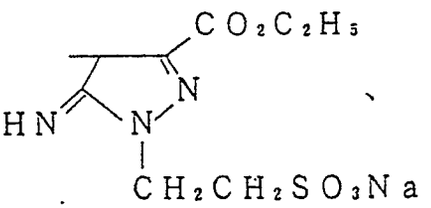


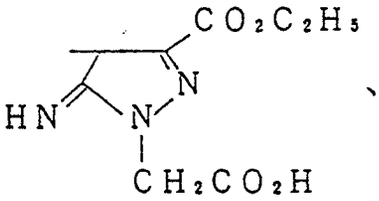
in formula (CIII) include:

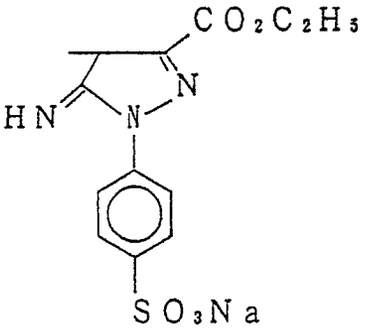


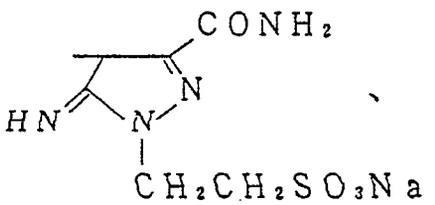


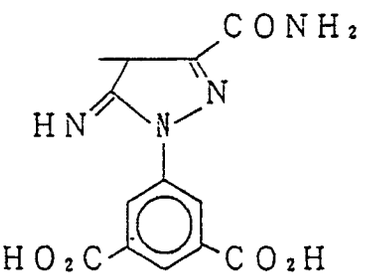


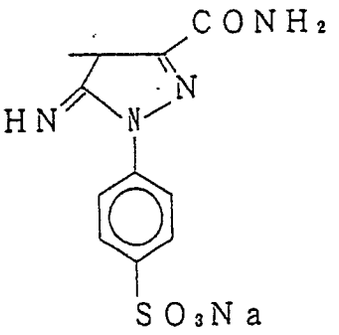


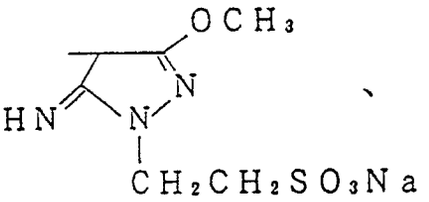


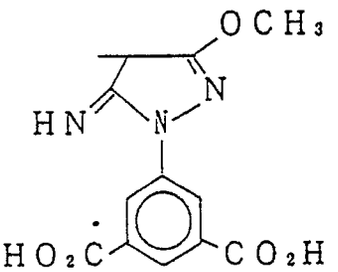


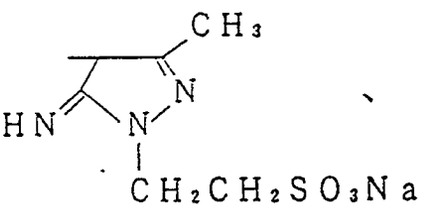


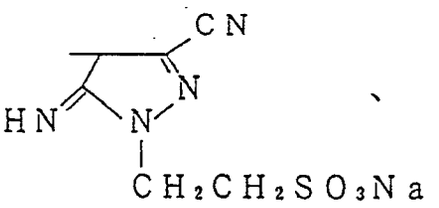


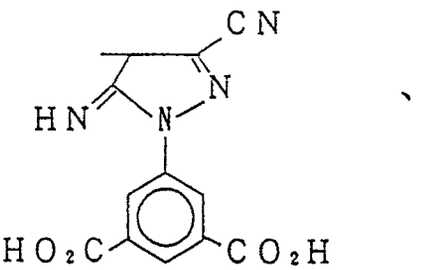


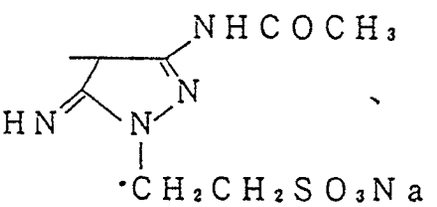






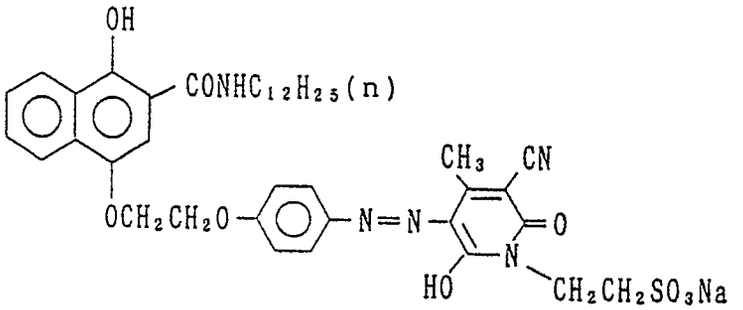




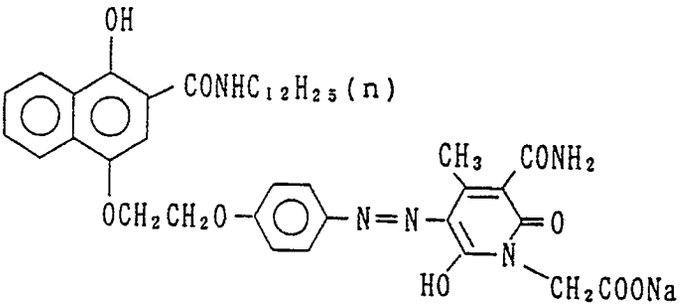


Examples of the yellow-colored couplers of the present invention will be set forth below:

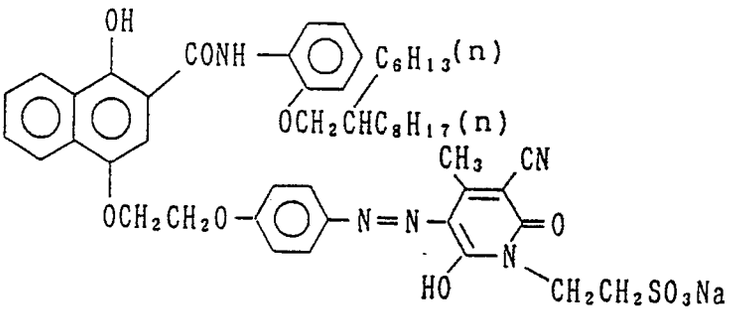
(YC－1)



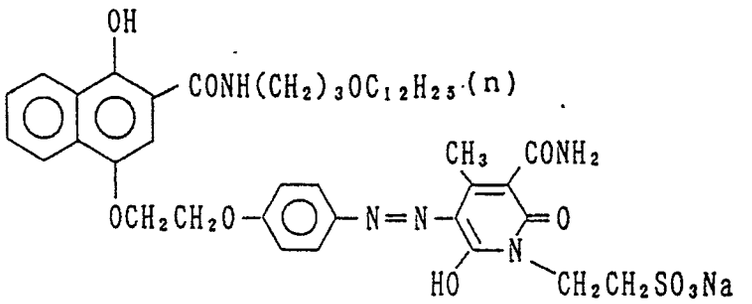
(YC－2)



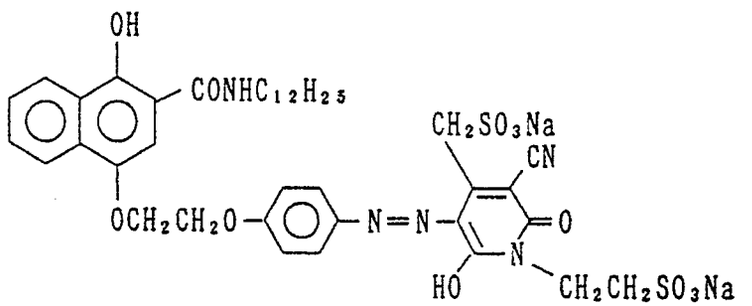
(YC－3)



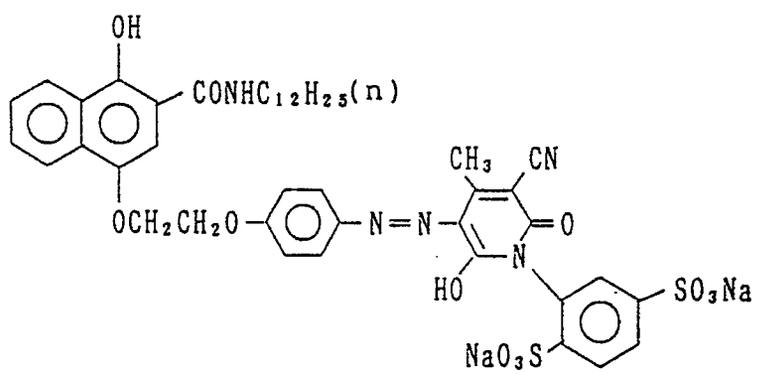
(YC－4)



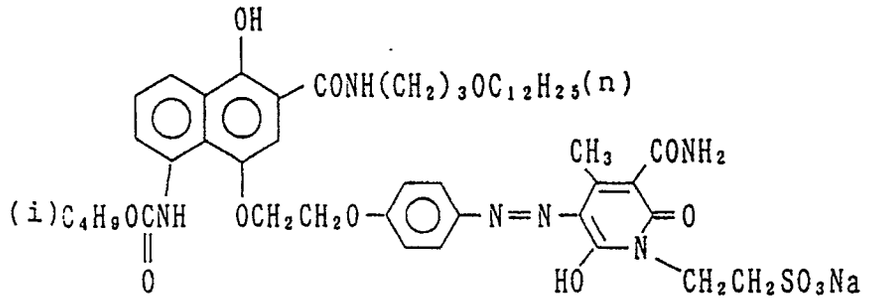
(YC－5)



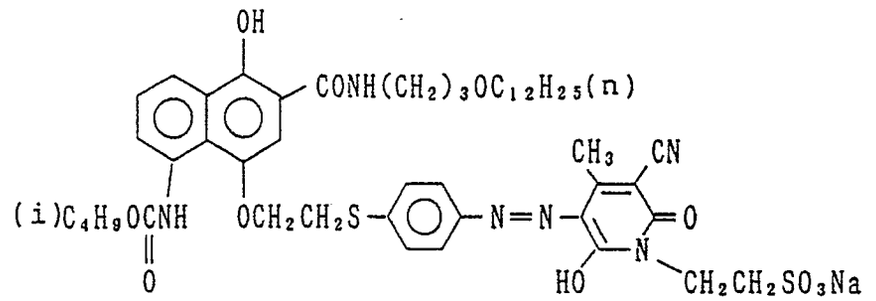
(YC－6)



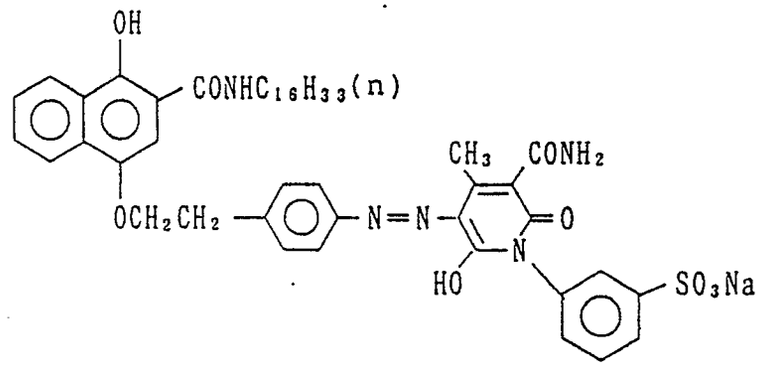
(YC－7)



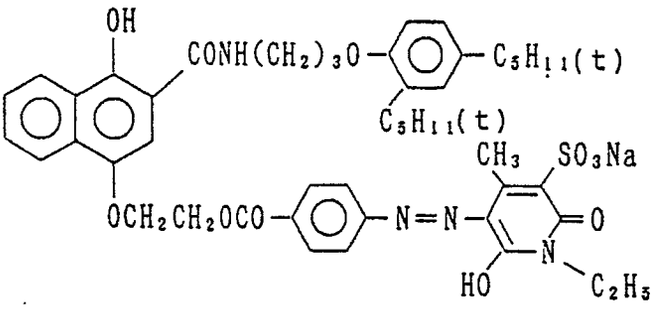
(YC－8)



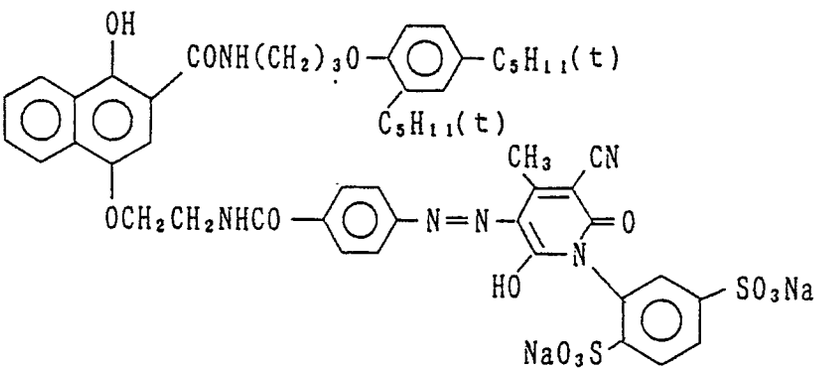
(YC－9)



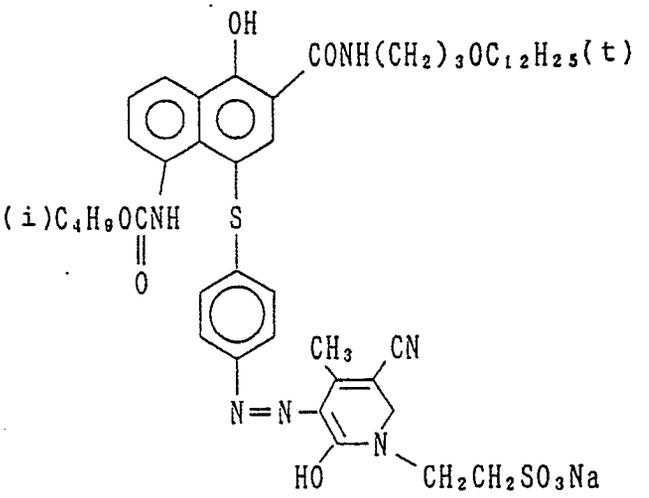
(YC－10)



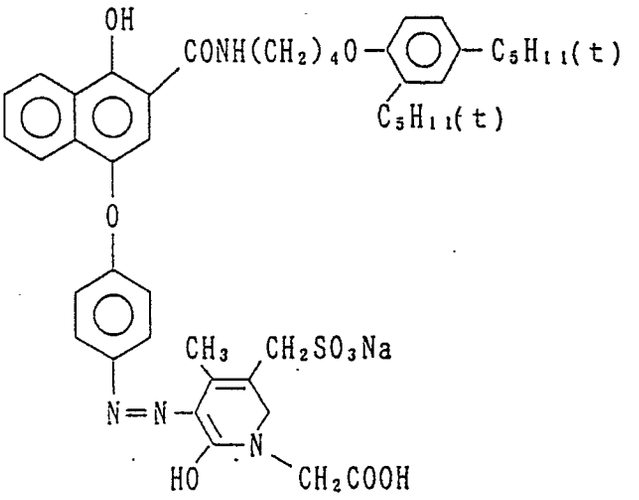
(YC－11)



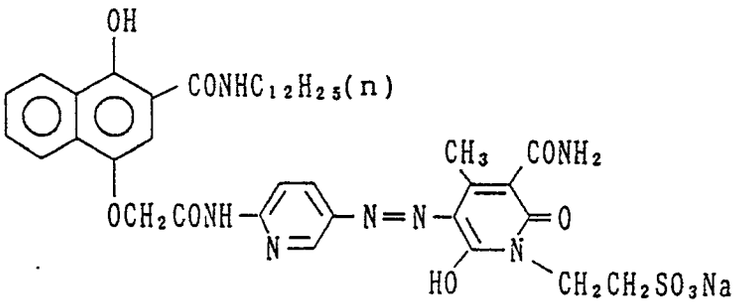
(YC－12)



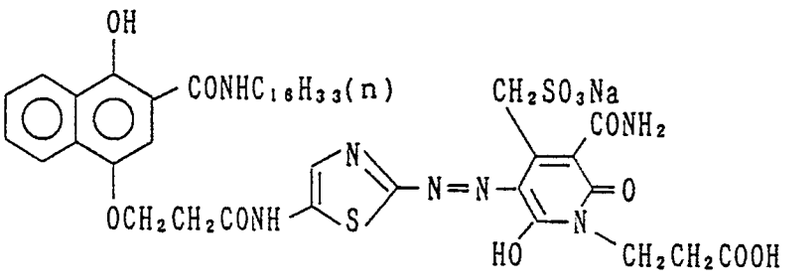
(YC－13)



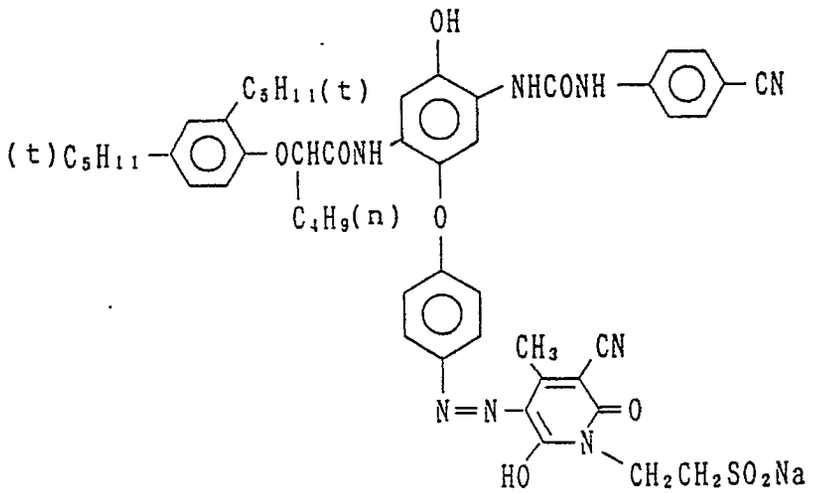
(YC－14)



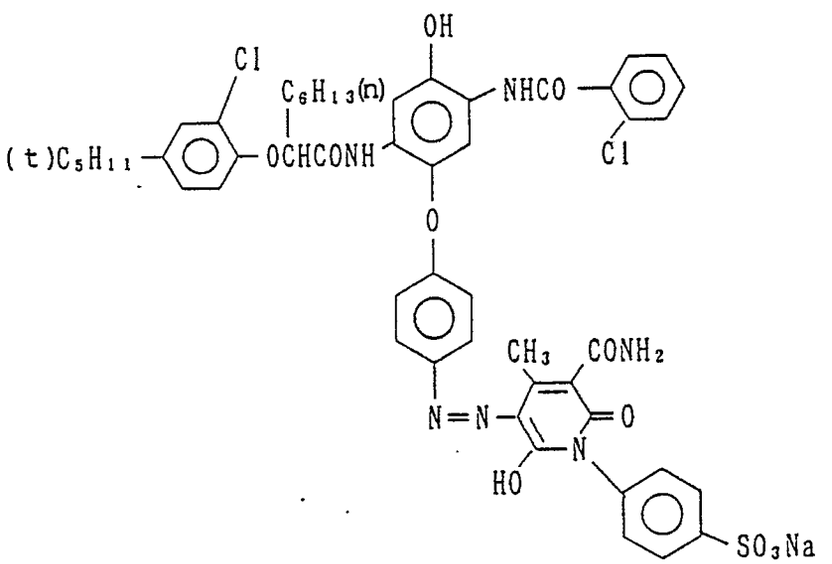
(YC－15)



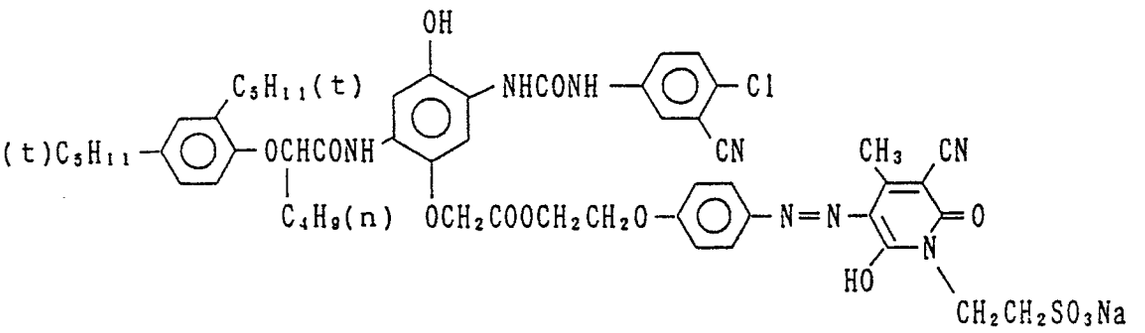
(YC－16)



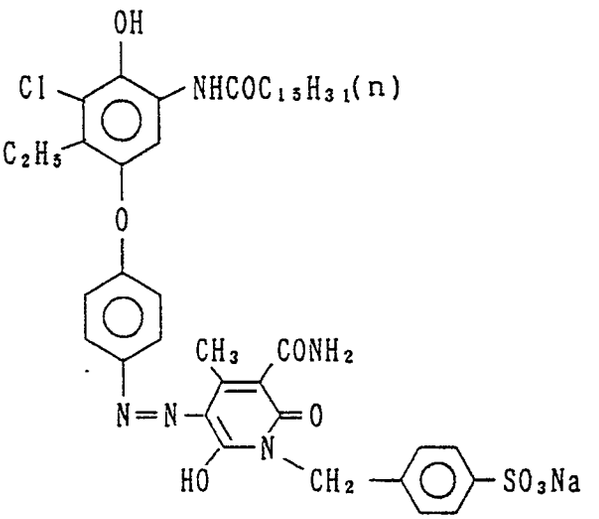
(YC－17)



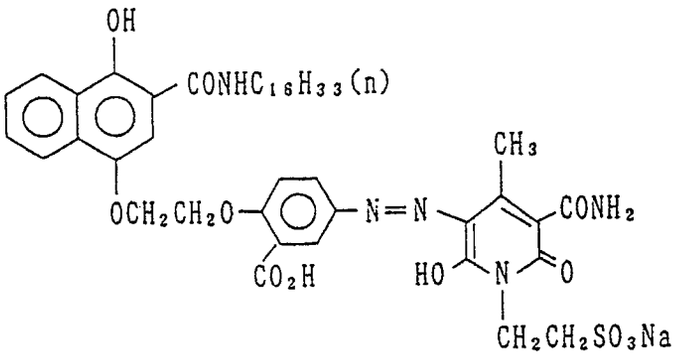
(YC－18)



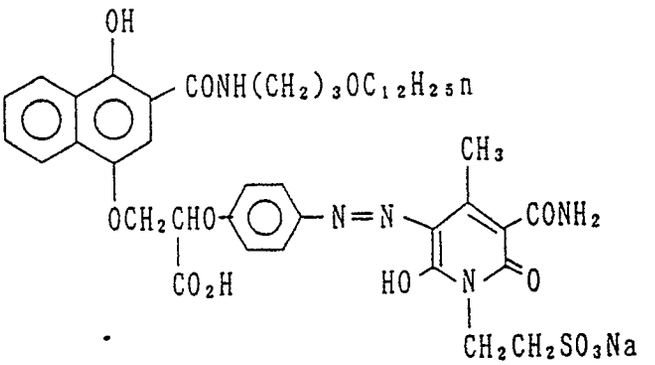
(YC－19)



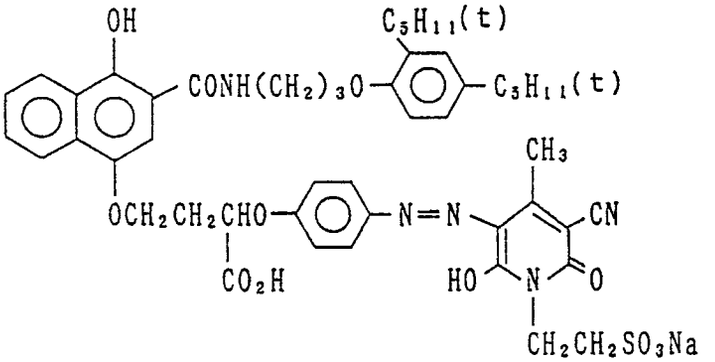
(YC－20)


OH
CONHC16H33(n)
CH3
OCH2CH2O
N=N
CONH2
CO2H
HO
N
O
CH2CH2SO3Na

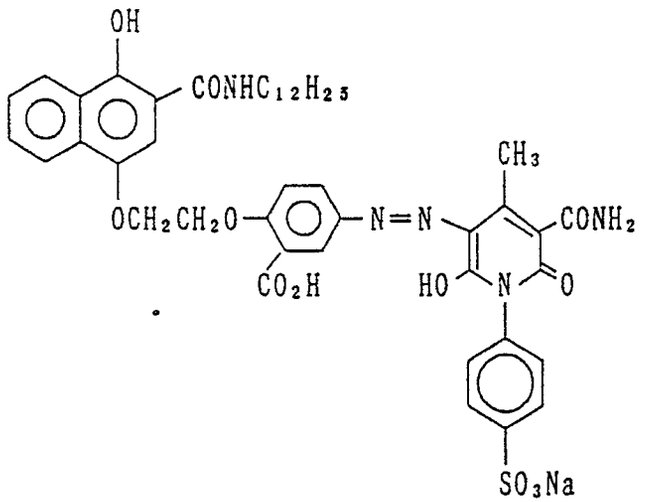
(YC－21)


OH
CONH(CH2)3OC12H25n
CH3
OCH2CHO
N=N
CONH2
CO2H
HO
N
O
CH2CH2SO3Na

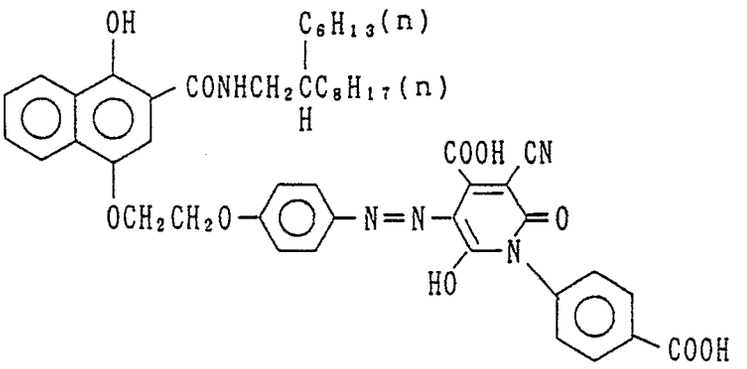
(YC－22)



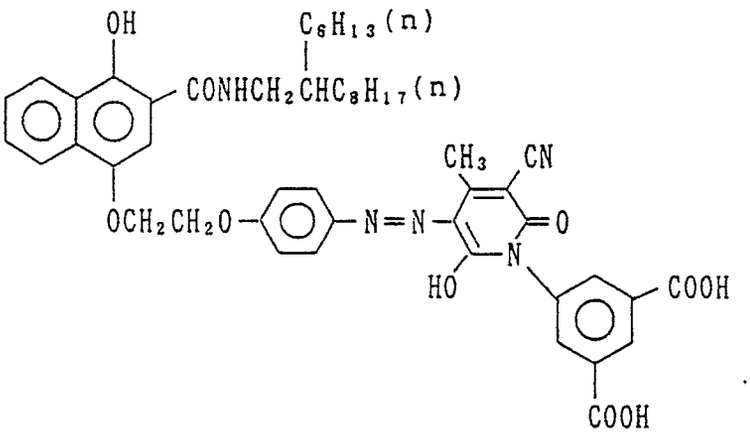
(YC－23)



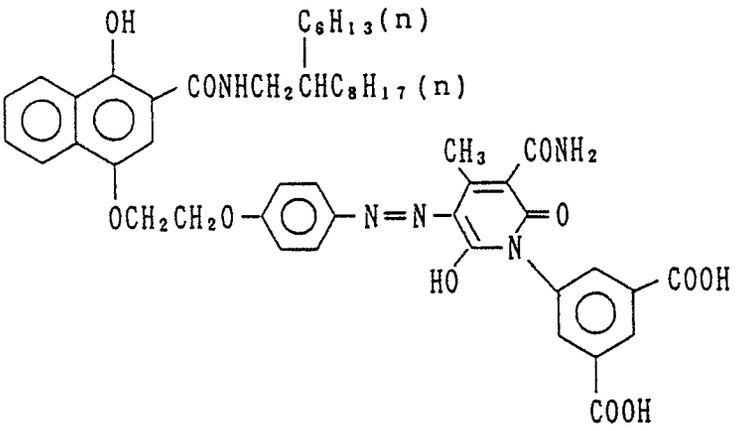
(YC－24)



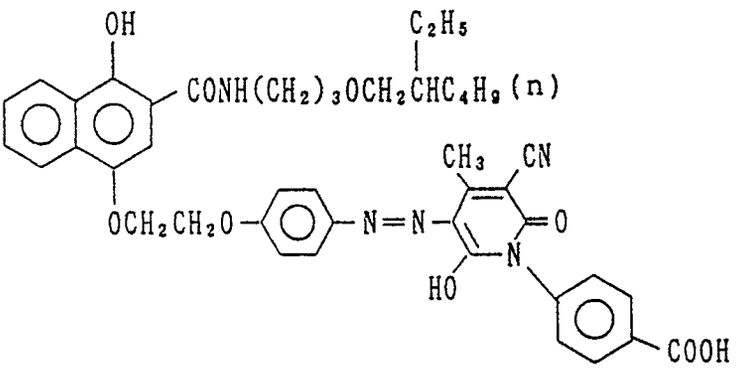
(YC－25)



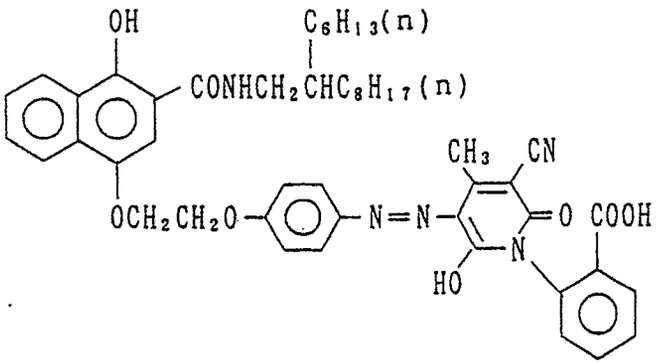
(YC－26)



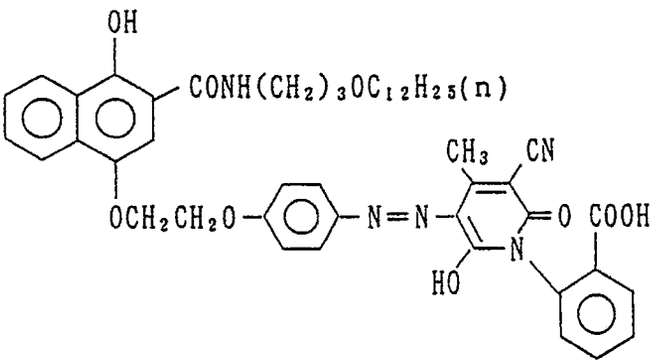
(YC－27)



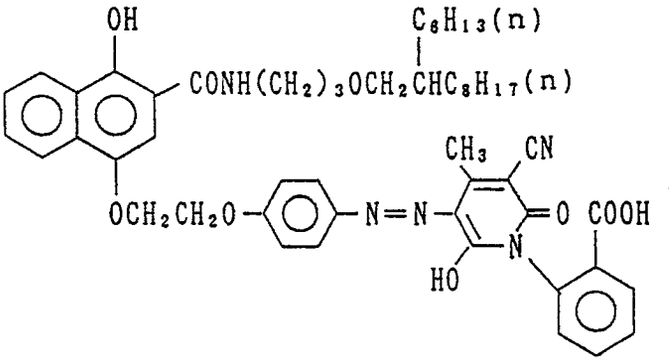
(YC－28)



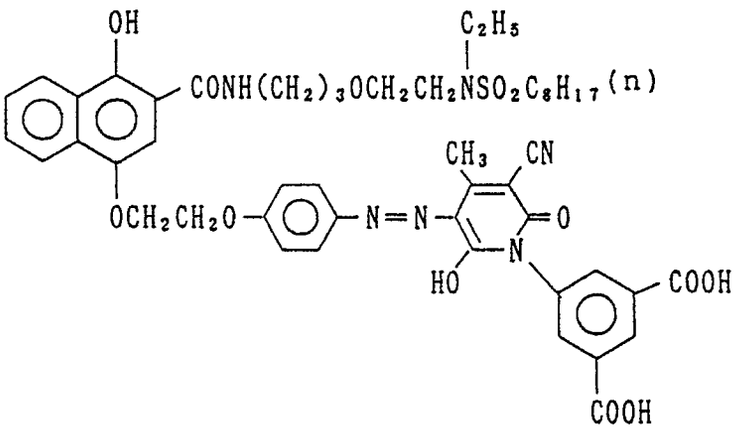
(YC－29)



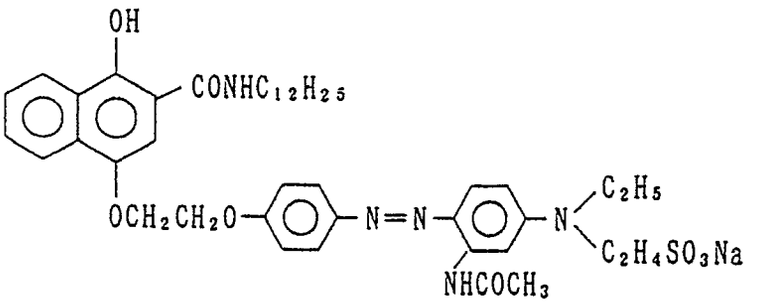
(YC－30)



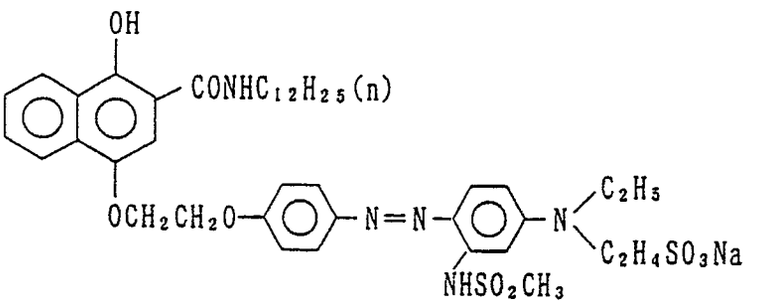
(YC－31)



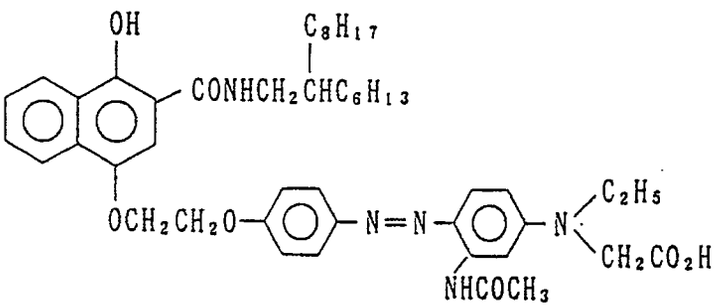
(YC－32)



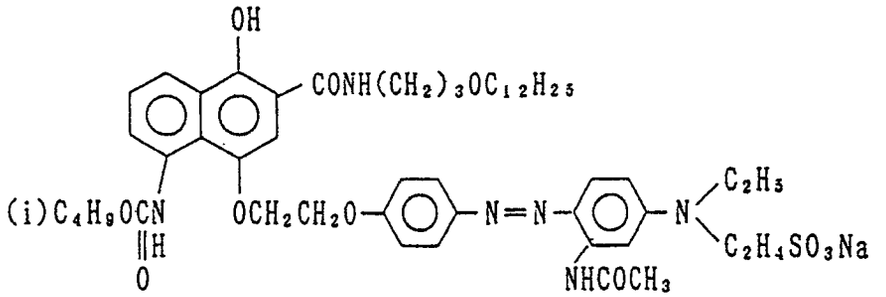
(YC－33)



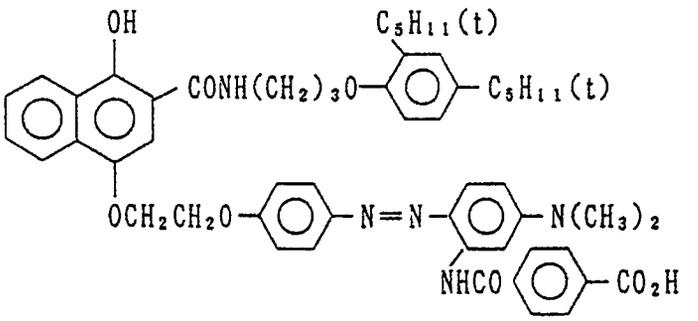
(YC－34)



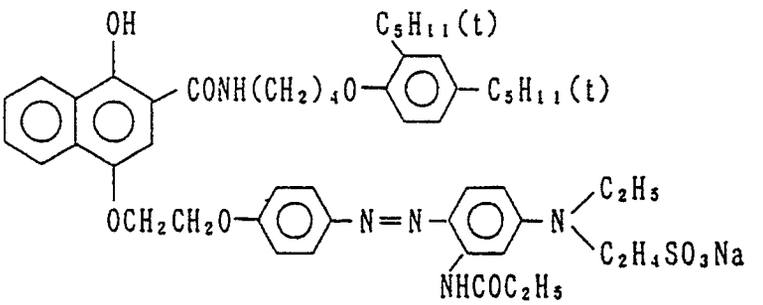
(YC－35)



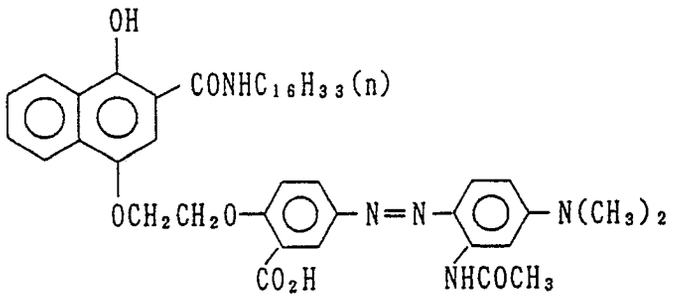
(YC－36)



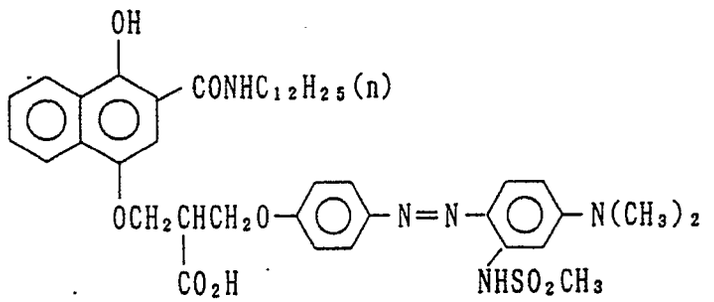
(YC－37)



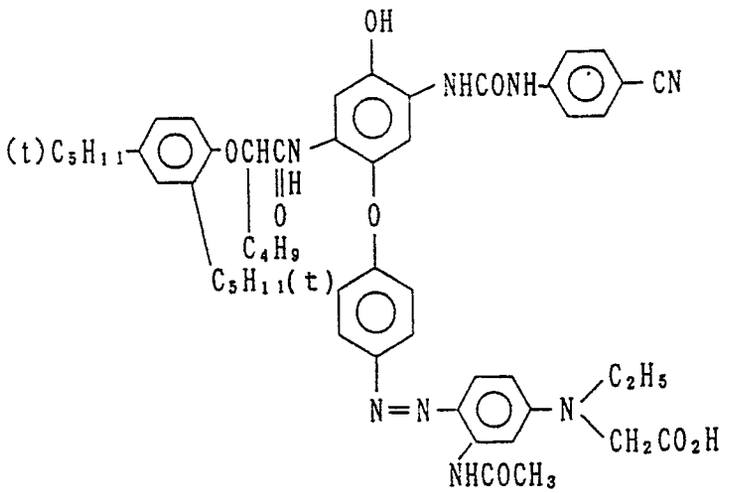
(YC－38)



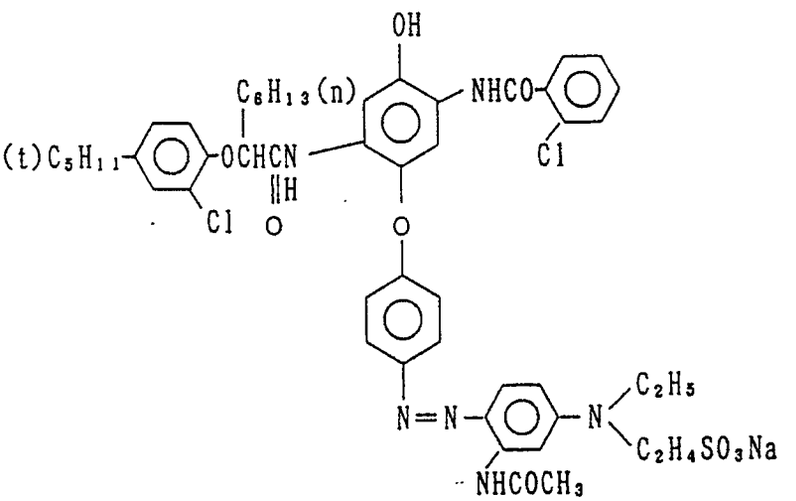
(YC－39)



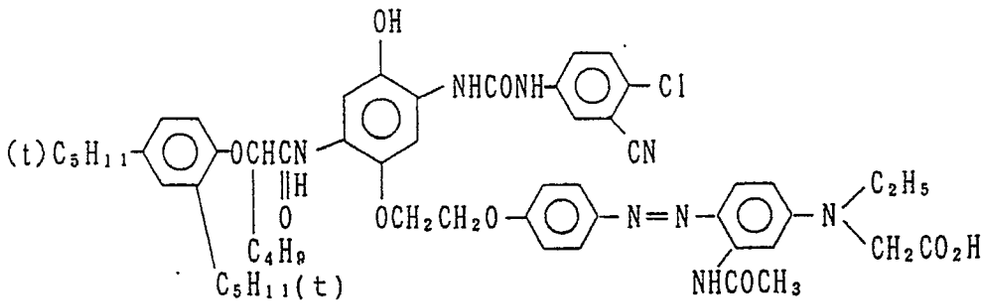
(YC－40)



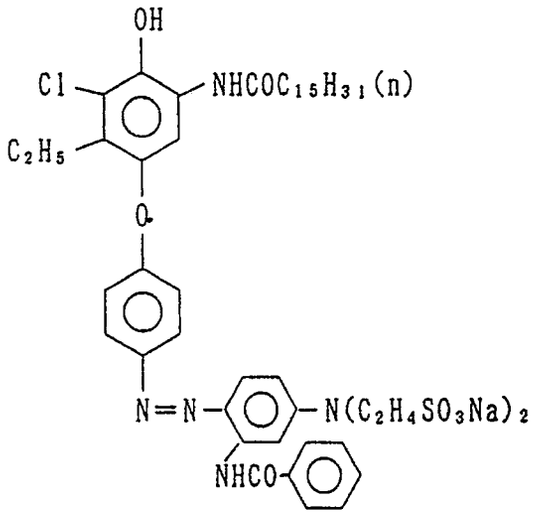
(YC－41)



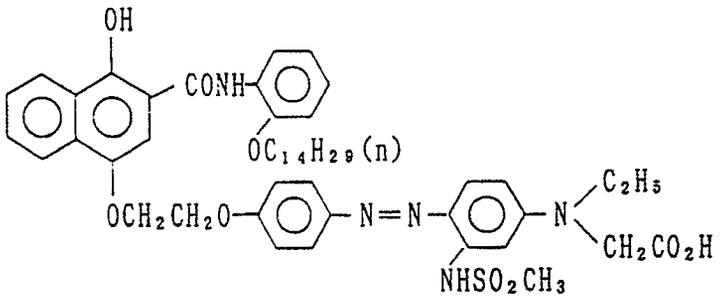
(YC－42)



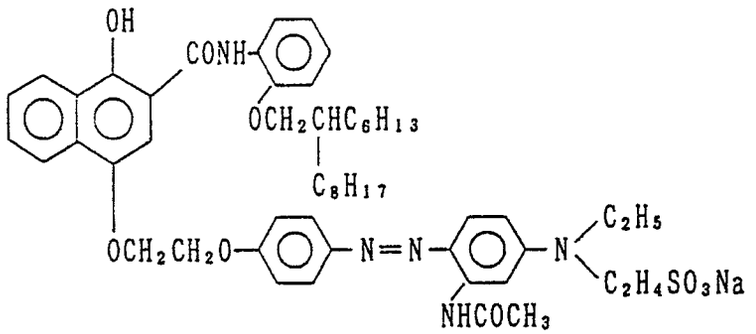
(YC－43)



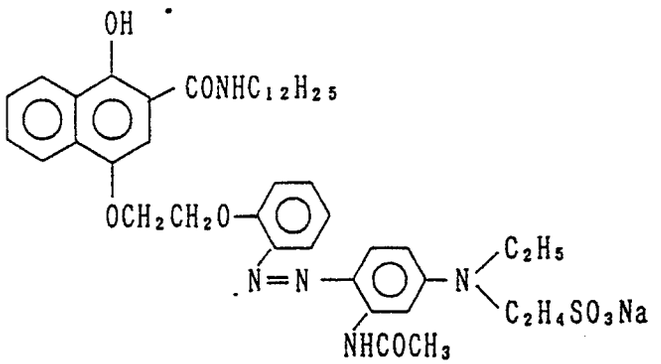
(YC－44)



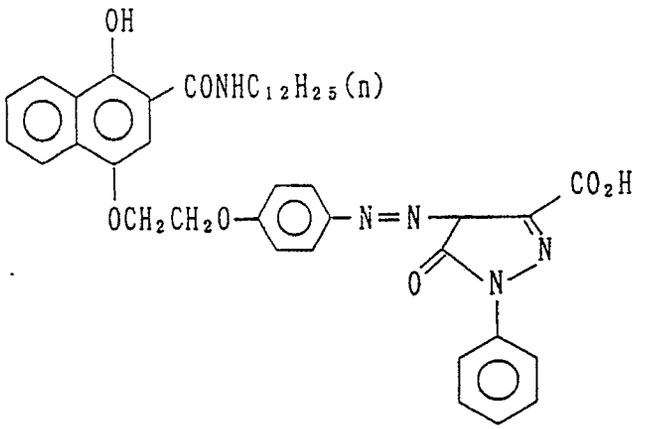
(YC－45)



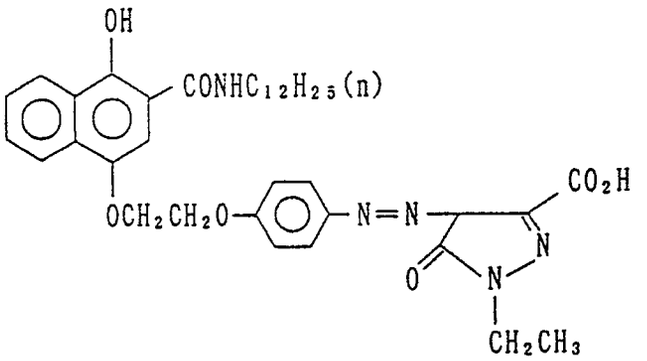
(YC－46)



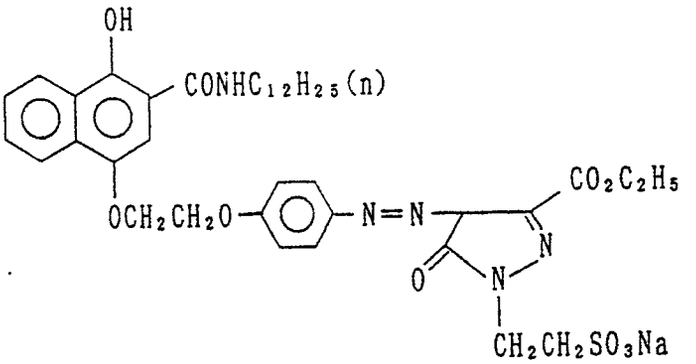
(YC－47)



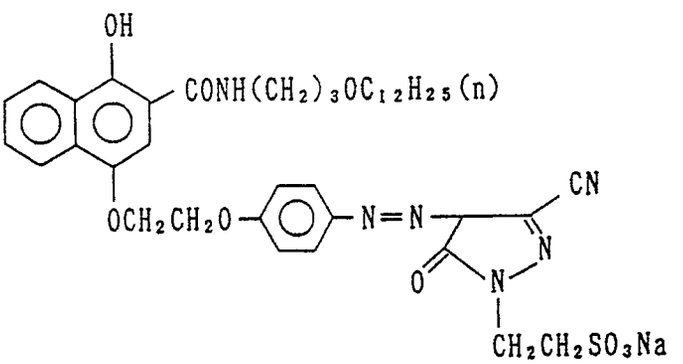
(YC－48)



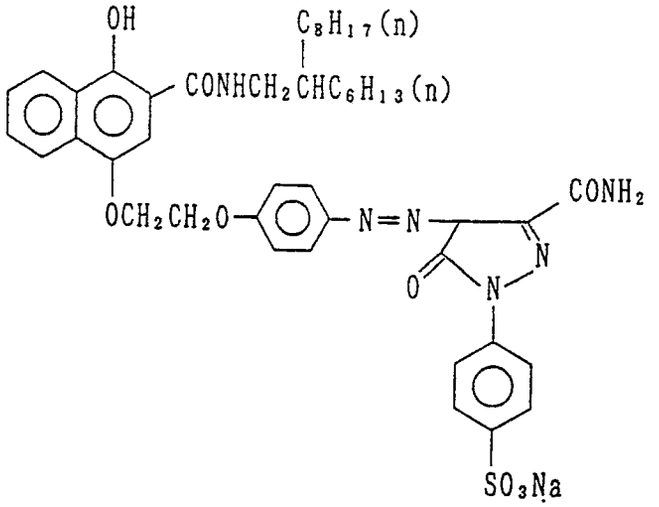
(YC－49)



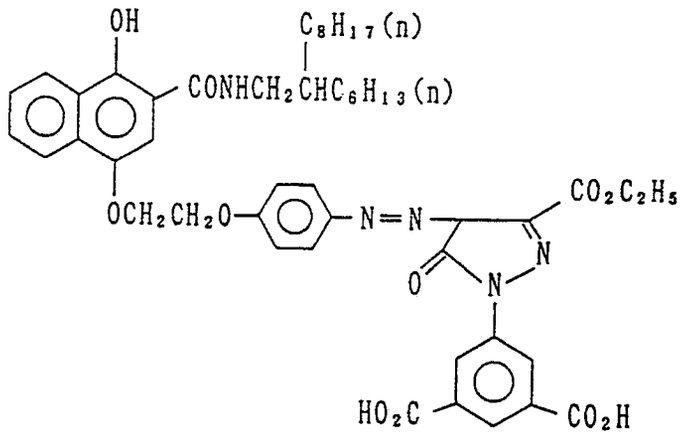
(YC－50)



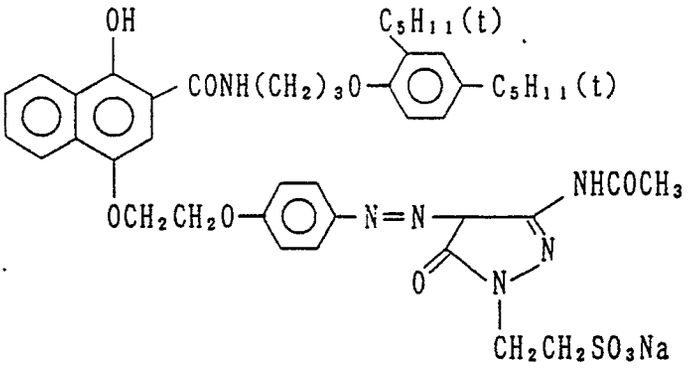
(YC－51)



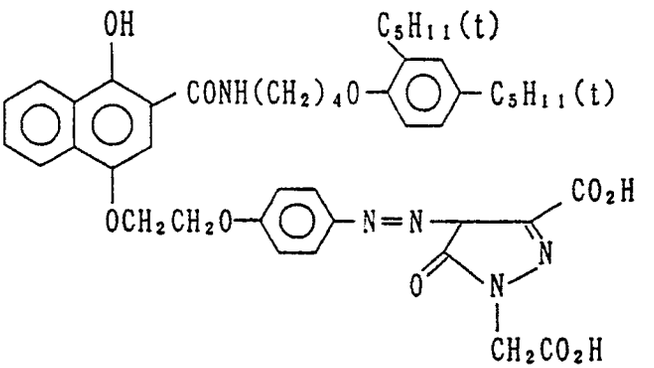
(YC－52)



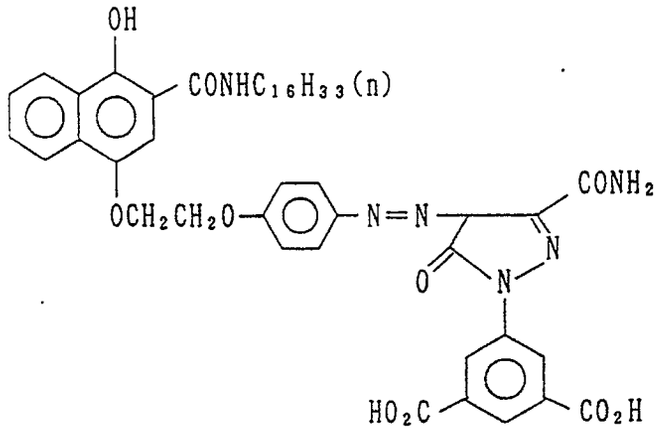
(YC－53)



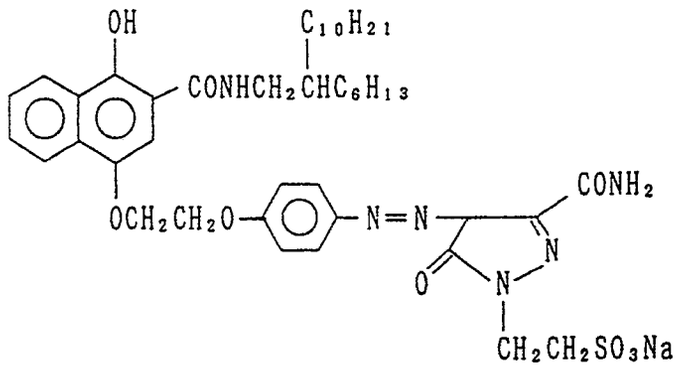
(YC－54)



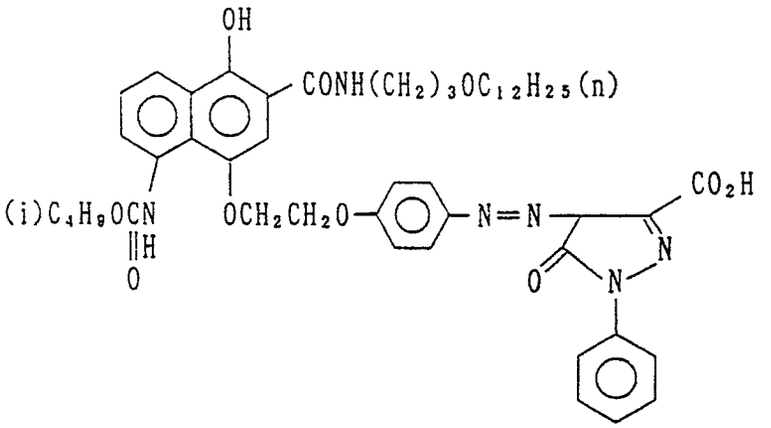
(YC－55)



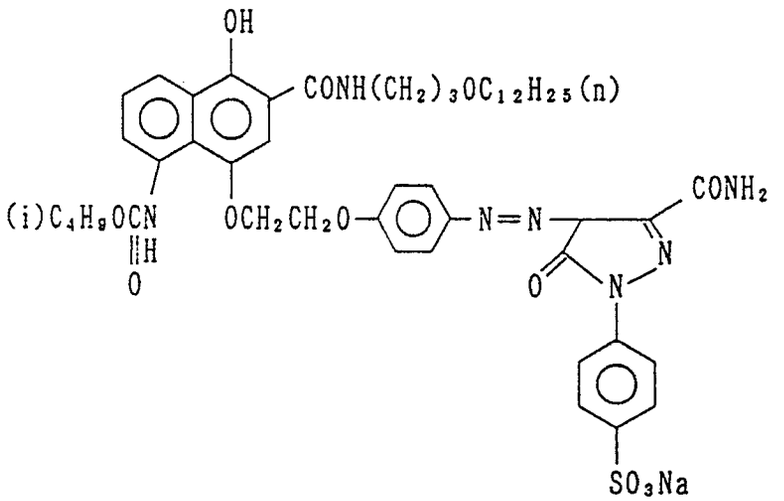
(YC－56)



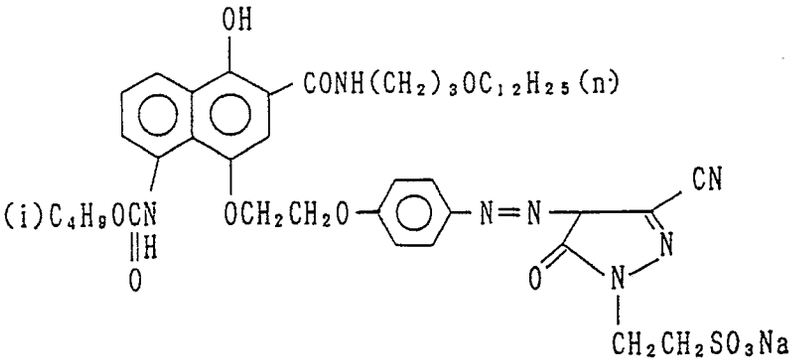
(YC－57)



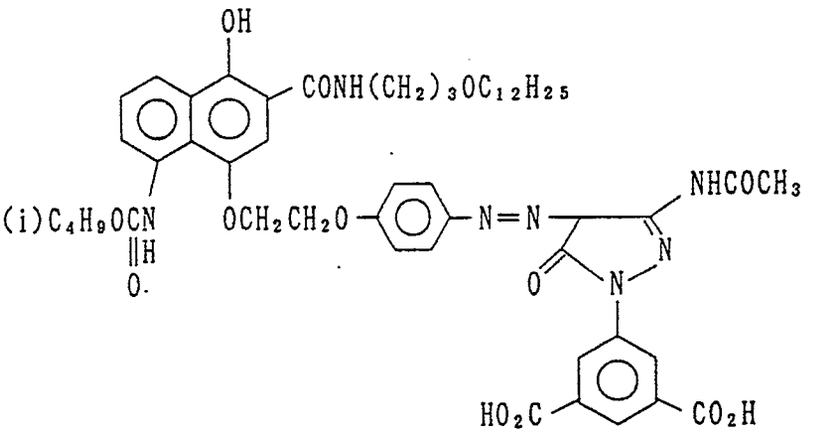
(YC－58)



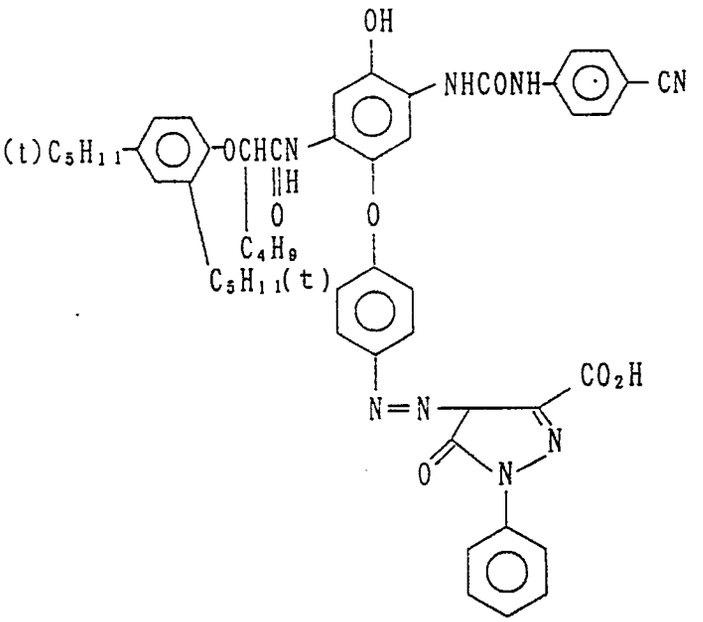
(YC－59)



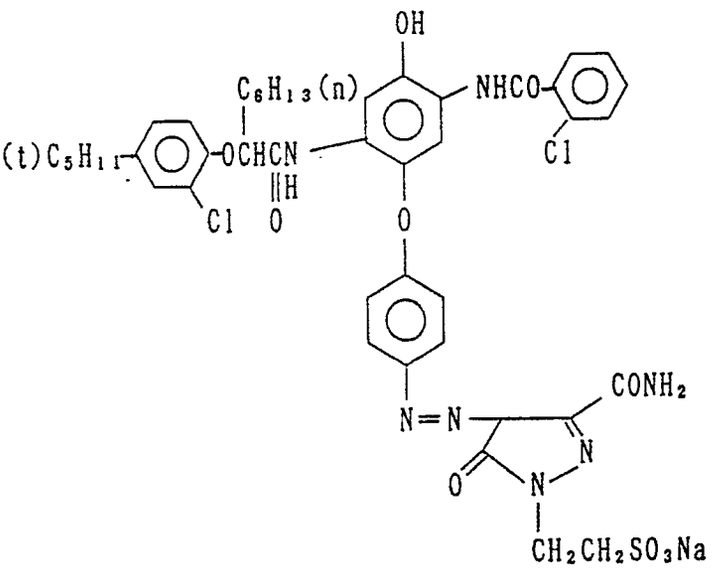
(YC－60)



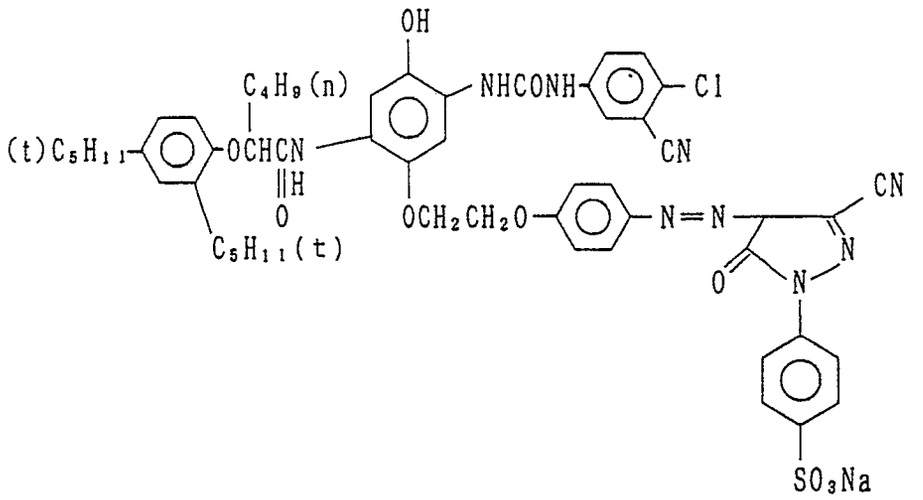
(YC－61)



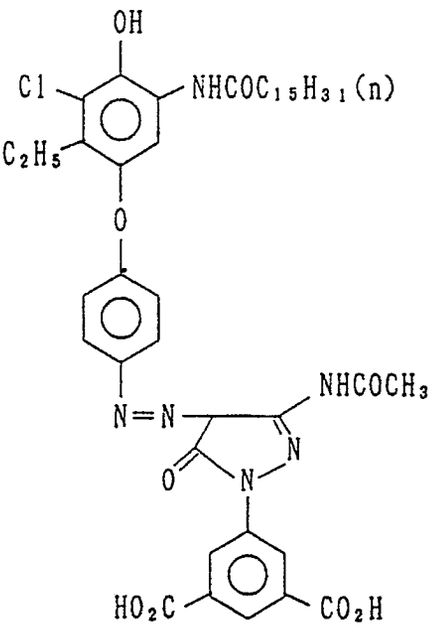
(YC－62)



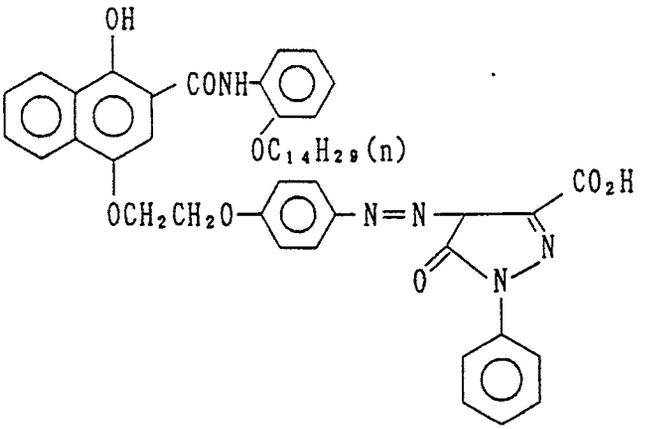
(YC-63)



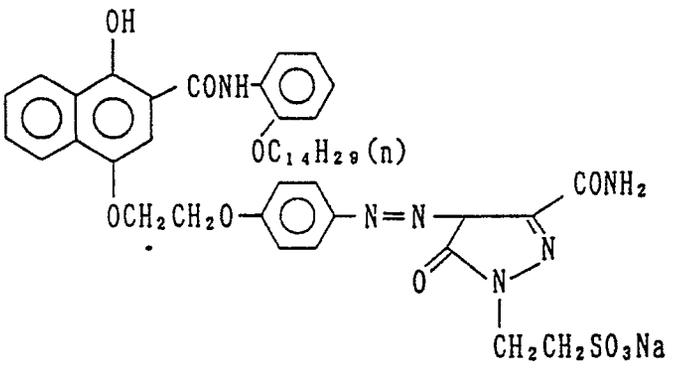
(YC-64)



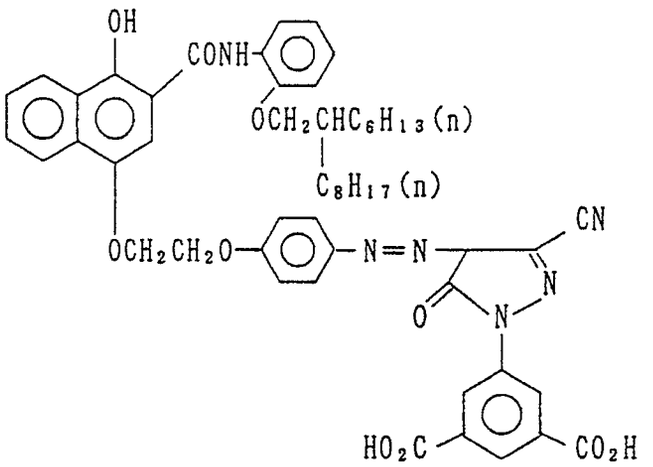
(YC－65)



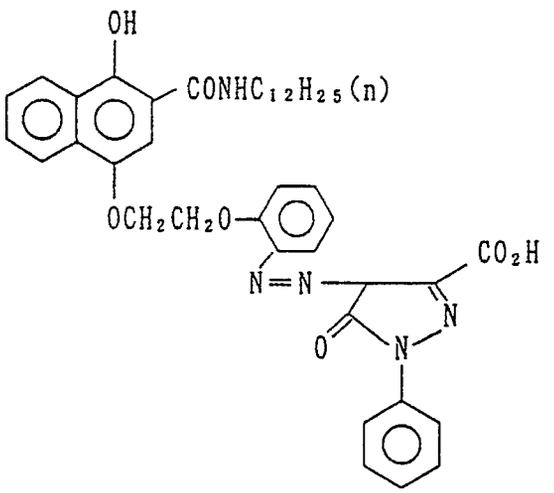
(YC－66)



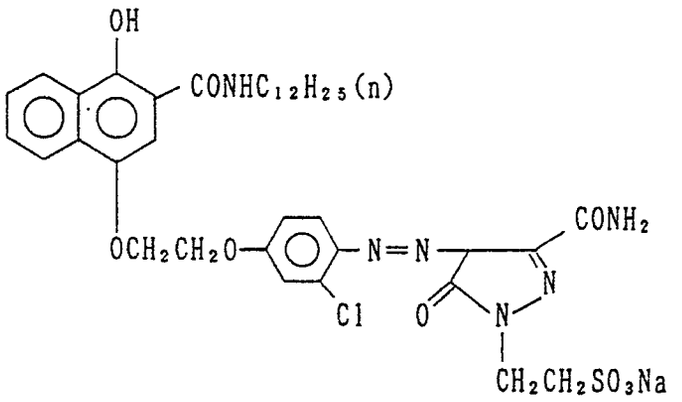
(YC－67)



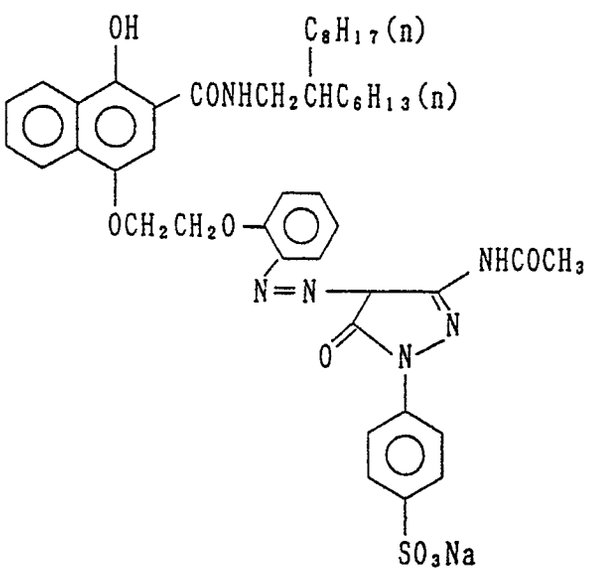
(YC－68)



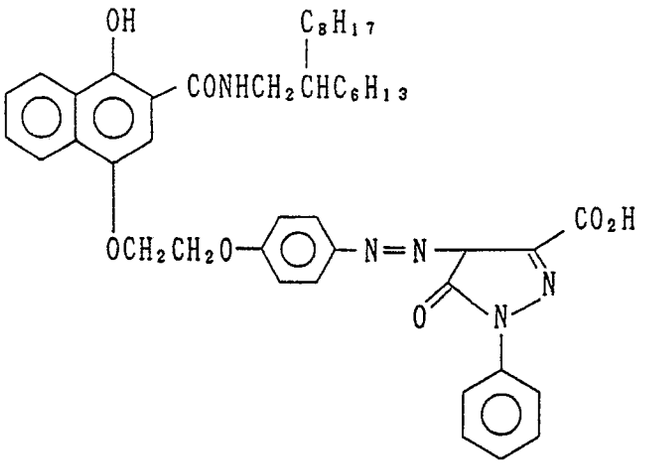
(YC-69)



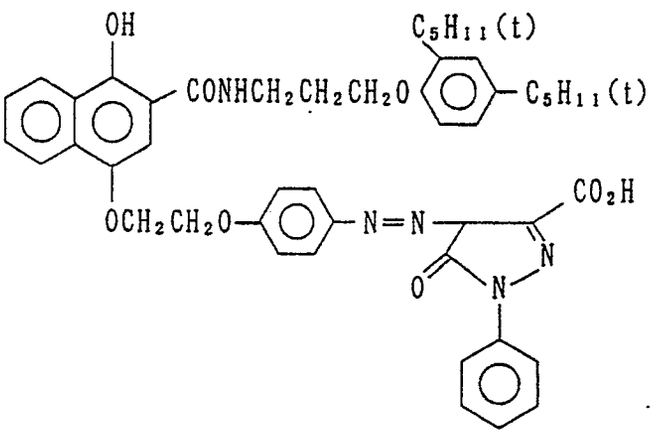
(YC-70)



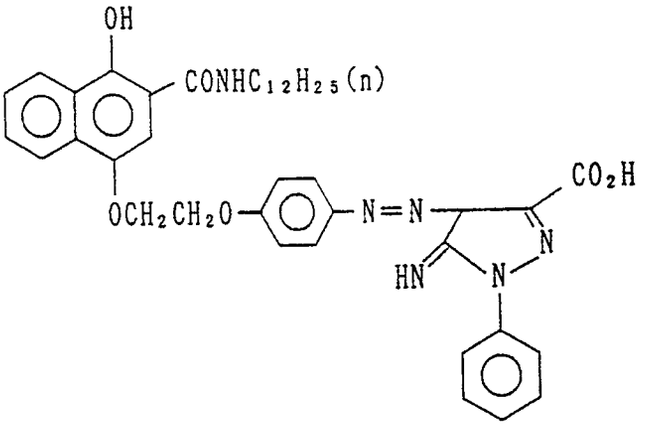
(YC-71)



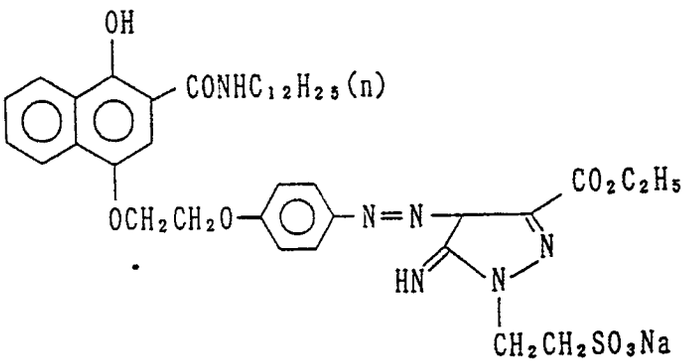
(YC-72)



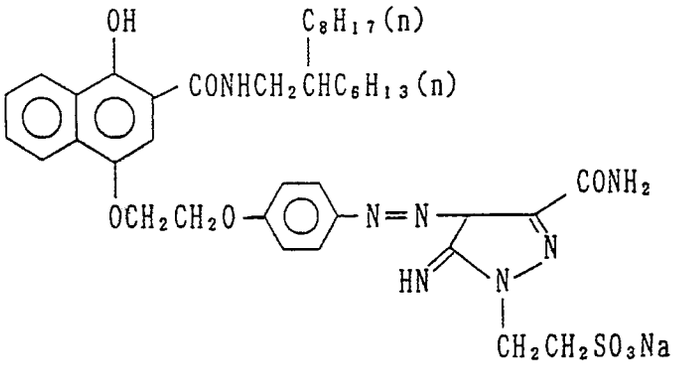
(YC－73)



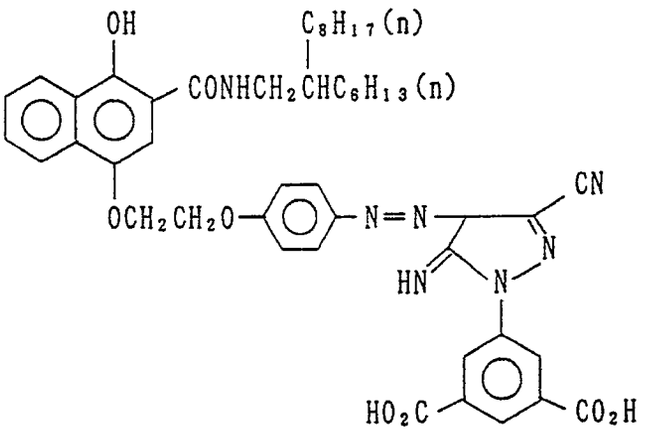
(YC－74)



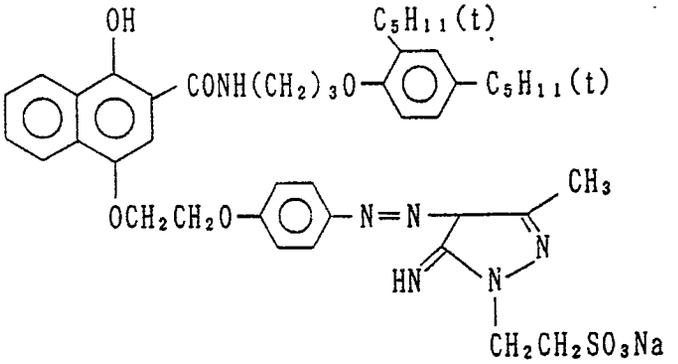
(YC－75)



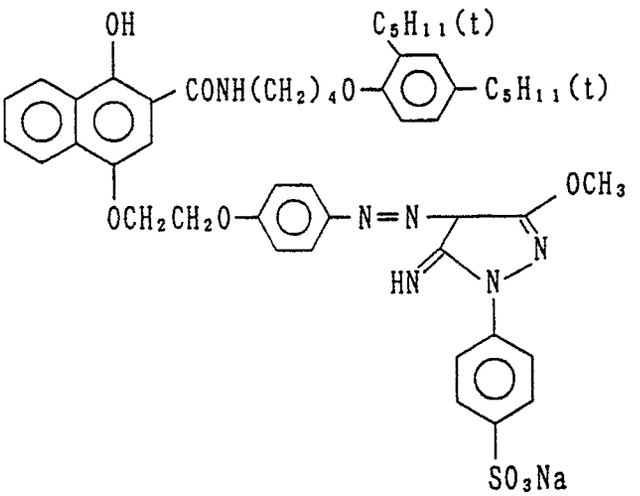
(YC－76)



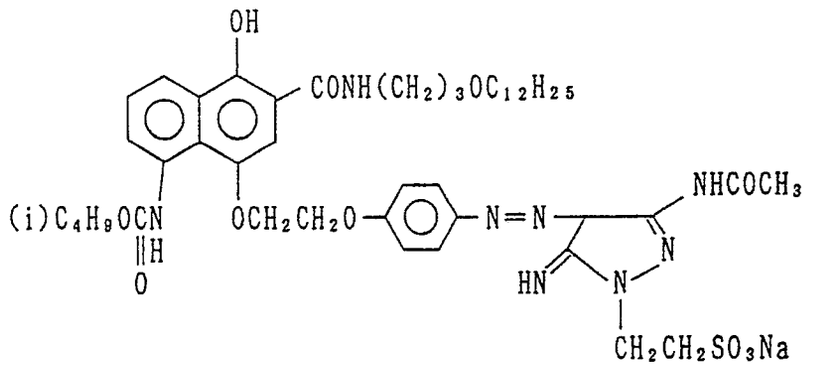
(YC－77)



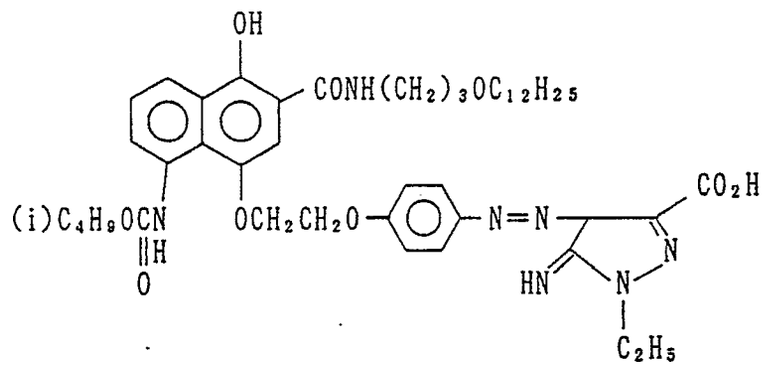
(YC－78)



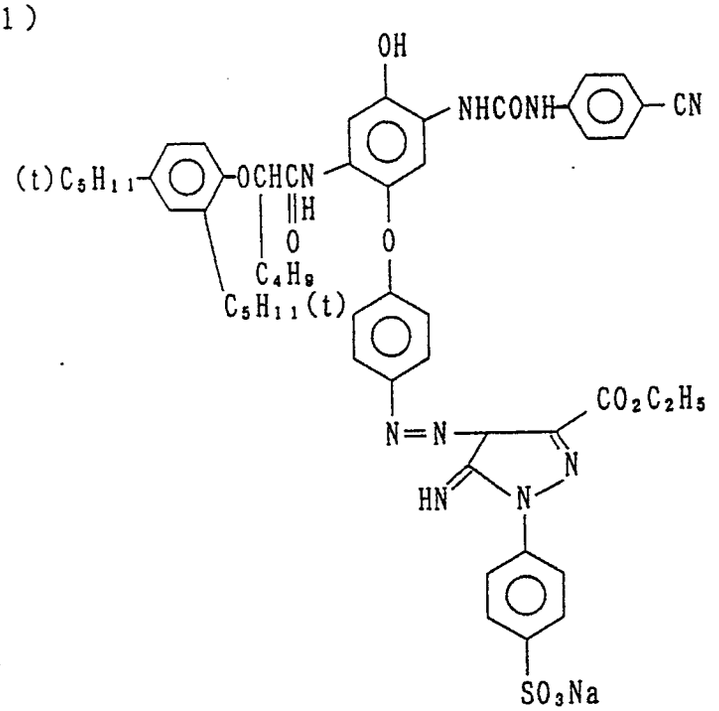
(YC－79)



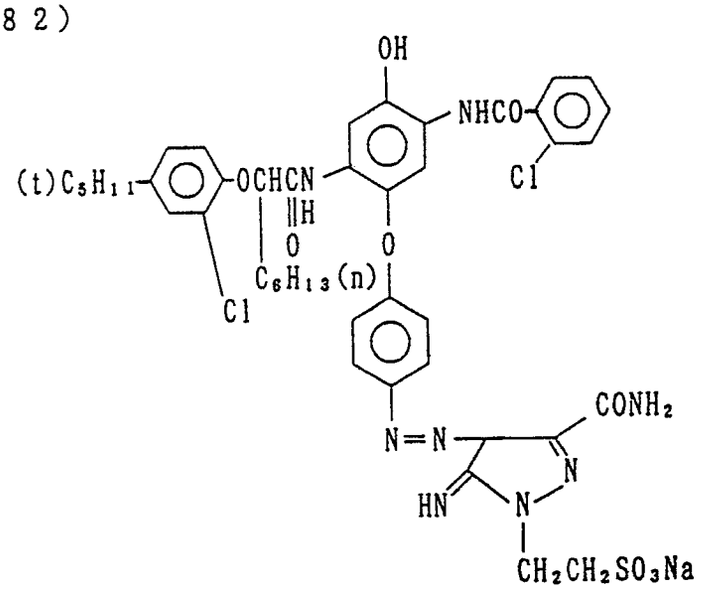
(YC－80)



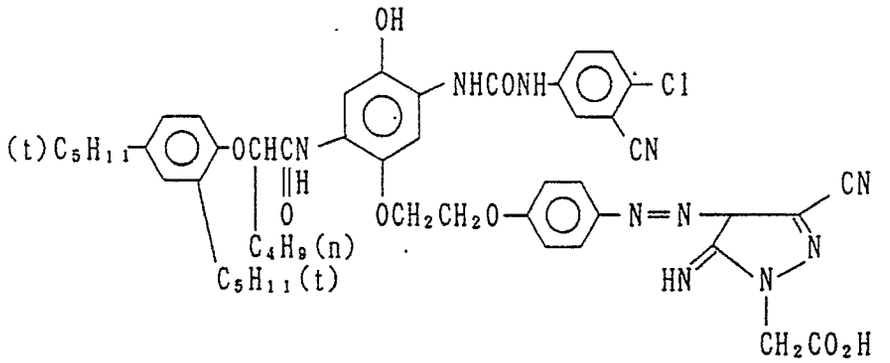
(YC－81)



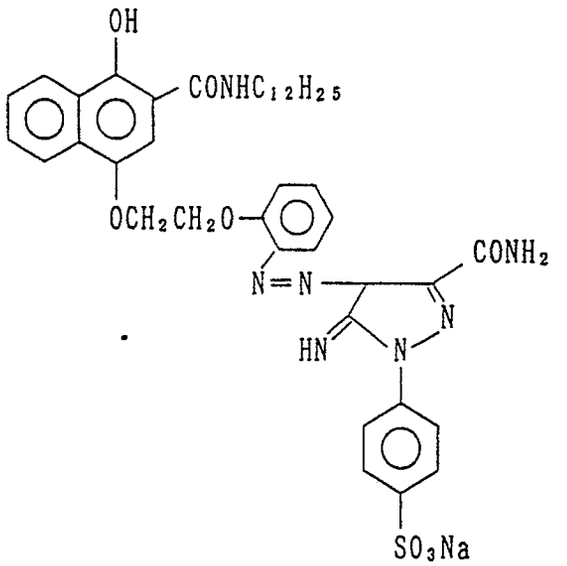
(YC－82)



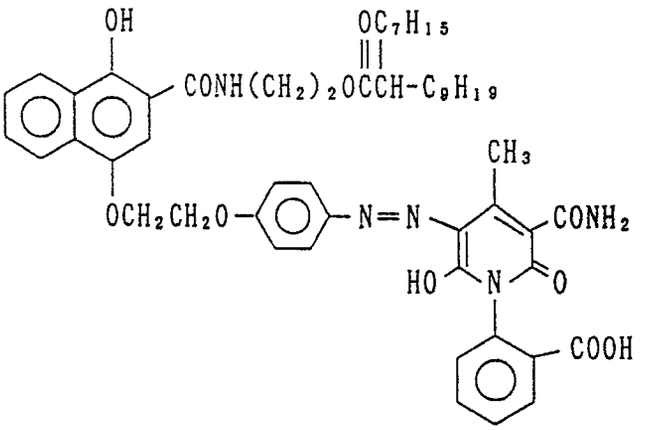
(YC-83)



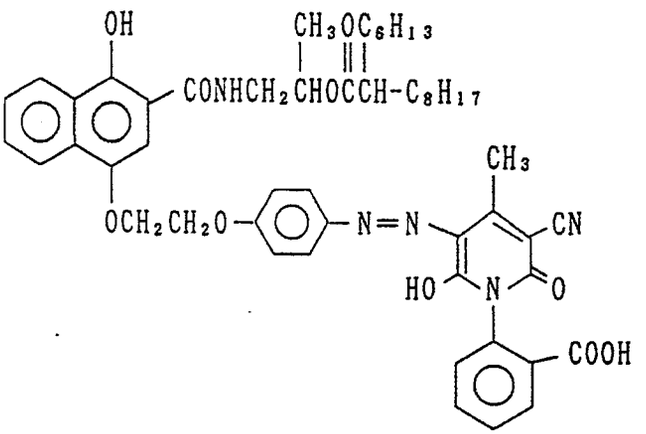
(YC-84)



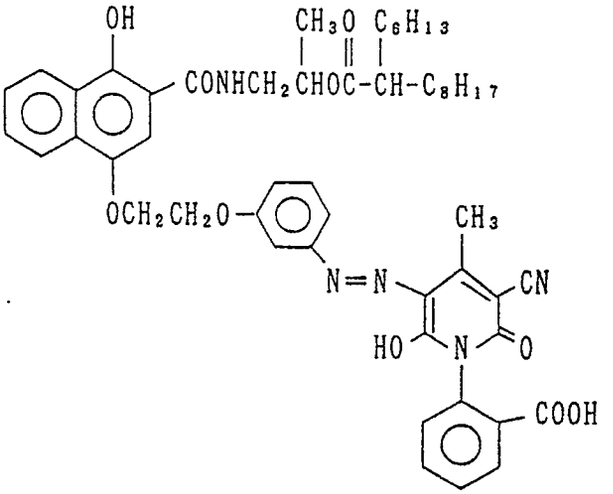
(YC－85)



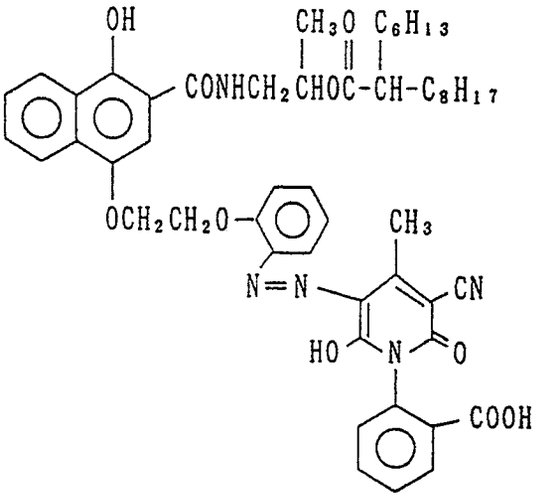
(YC－86)



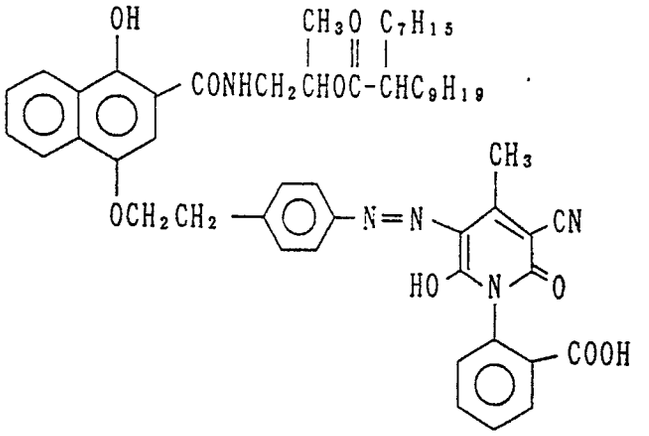
(YC－87)



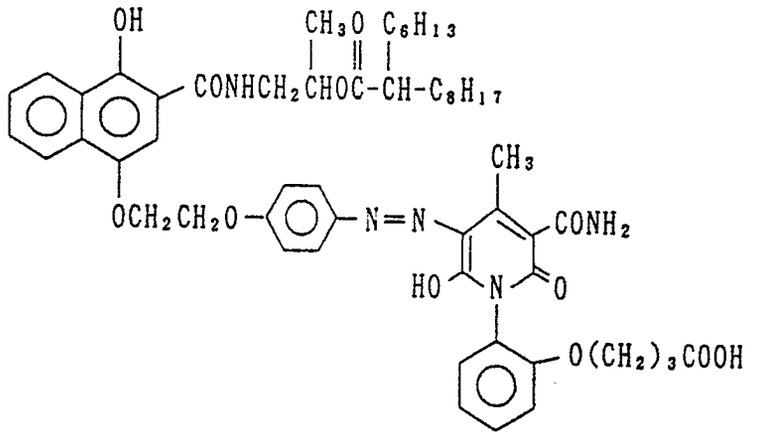
(YC－88)



(YC-89)



(YC-90)



The synthesis of the yellow-colored coupler of the present invention represented by the general formula (CI) can be normally accomplished by a diazo coupling reaction of a 6-hydroxy-2-pyridone with an aromatic diazonium salt or a heterocyclic diazonium salt having a coupler structure.

The synthesis of the former reaction component, i.e., the 6-hydroxy-2-pyridine, can be accomplished by any suitable method as disclosed in Klinsberg, (ed.) Heterocyclic Compounds--Pyridone and Its Derivatives--Part III, Interscience, (1962); Journal of the American Chemical Society, 1943, Vol. 65, page 449; Journal of the Chemical Technology & Biotechnology, 1986, Vol. 36, page 410; Tetrahedron, 1966, Vol. 22, page 445; JP-B-61-52827 (the term "JP-B" as used herein means an "examined Japanese patent publication"); West German Patents 2,162,612, 2,349,709, and 2,902,486; and U.S. Patent 3,763,170.

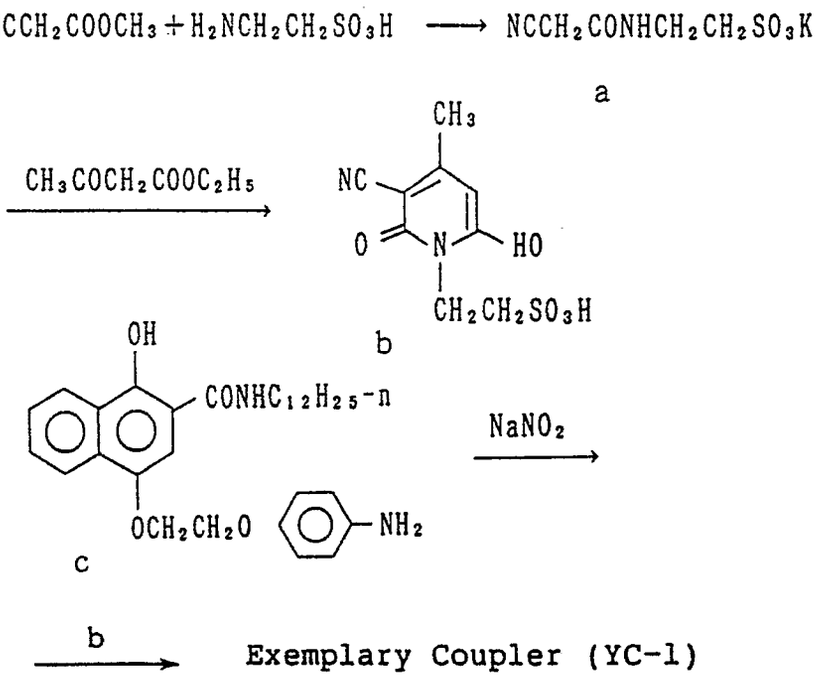
The synthesis of the latter reaction component, i.e., the diazonium salt, can be accomplished by any suitable method as disclosed in U.S. Patents 4,004,929, and 4,138,258, and JP-A-61-72244 and JP-A-61-273543.

The diazo coupling reaction of the 6-hydroxy-2-pyridone with the diazonium salt can be effected in a solvent such as methanol, ethanol, methyl cellosolve, acetic acid, N,N-dimethylformamide, N,N-dimethylacetamide, tetrahydrofuran, dioxane, water, or a mixture thereof. The base to be used in the reaction may be sodium acetate, potassium acetate, sodium carbonate, potassium carbonate, sodium hydrogencaronate, sodium hydroxide, potassium hydroxide, pyridine, triethylamine, tetramethylurea, tetramethyl guanidine or the like. The reaction temperature is normally in the range of -78°C to 60°C, preferably -20°C to 30°C.

Examples of the synthesis of yellow-colored couplers of the present invention will be set forth below.

SYNTHESIS EXAMPLE 1

Synthesis of Exemplary Coupler (YC-1)



Synthesis of Compound a

500 ml of methanol was added to 125.2 g of taurine and 66 g of potassium hydroxide. The mixture was heated with stirring. 110 g of methyl cyanoacetate was added dropwise to the material over a period of about 1 hour. The material was heated under reflux for 5 hours, and then allowed to stand overnight. The resulting crystal was filtered off, washed with ethanol, and then dried to obtain 202.6 g of Compound a in a crystal form.

Synthesis of Compound b

11.5 ml of water was added to 11.5 g of Compound a and 3.5 g of potassium carbonate. 7.8 g of ethyl acetate was then added dropwise to the mixture under heating with stirring over a steam bath. The system was further stirred for 7 hours. After the system was allowed to cool, 9.2 ml of concentrated hydrochloric acid was added thereto with stirring to effect crystallization. The resulting crystal was filtered off, washed with methanol, and then dried to obtain 10.4 g of Compound b in a crystal form.

Synthesis of Exemplary Compound (YC-1)

10.1 g of Compound c prepared by the synthesis method described in U.S. Patent 4,138,258 was dissolved in 60 ml of N,N-dimethylformamide and 60 ml of methyl cellosolve. 4.3 ml of concentrated hydrochloric acid was added to the solution under cooling with ice. A solution of 1.84 g of sodium nitrite in 5 ml of water was added dropwise to the system to prepare a solution of diazonium. 60 ml of methyl cellosolve and 20 ml of water were added to 7.8 g of Compound b and 8.2 g of sodium acetate. The diazonium solution was then added dropwise to the mixture with stirring under cooling with ice.

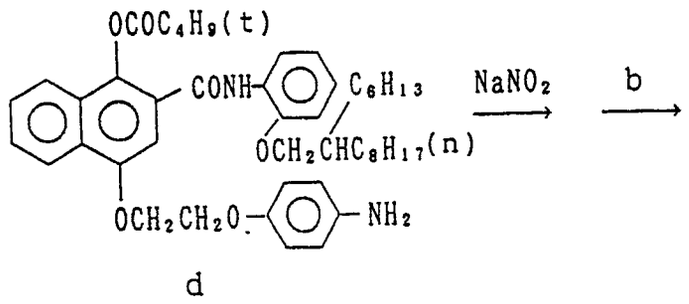
After the dropwise addition, the mixture was stirred further for 1 hour under ice-cooling, and then at room temperature for 2 hours. The resulting crystal was filtered off, washed with water, and then dried. The crystal was then dispersed in 500 ml of methanol. The dispersion was heated under reflux for 1 hour, and then allowed to cool. The crystal was filtered off, washed with methanol, and then dried to obtain 13.6 g of Exemplary Compound (YC-1) in the form of reddish crystal.

The melting point of the compound was 269 to 272 °C (decomposition). The structure of the compound

was confirmed by ^{1}H-NMR spectrum, mass spectrum and elementary analysis. The compound exhibited a maximum absorption wavelength of 457.7 nm and a molecular extinction coefficient of 41,300 in methanol. Thus, the compound exhibited excellent spectral absorption characteristics as a yellow-colored coupler.

SYNTHESIS EXAMPLE 2

Synthesis of Exemplary Coupler (YC-3)



d

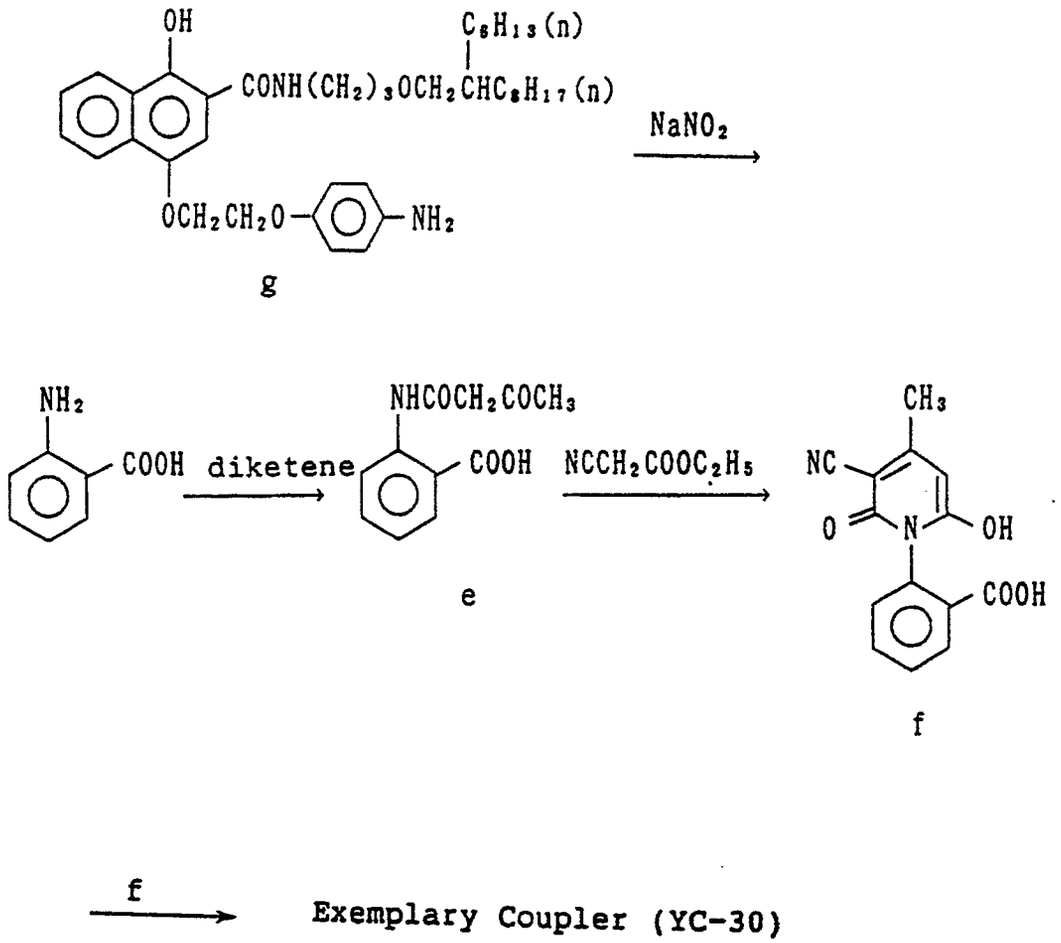
NaOH → Exemplary Coupler (YC-3)

19.2 g of Compound d prepared by the synthesis method as described in JP-A-62-85242 was dissolved in 75 ml of N,N-dimethylformamide and 75 ml of methyl cellosolve. 5.6 ml of concentrated hydrochloric acid was added to the solution with stirring under cooling with ice. A solution of 2.5 g of sodium nitrite in 5 ml of water was added dropwise to the system. After the completion of the dropwise addition, the system was further stirred for 1 hour under ice-cooling and then for 1 hour at room temperature to prepare a diazonium solution.

75 ml of methyl cellosolve and 26 ml of water were added to 10.1 g of Compound b (prepared as in Synthesis Example 1) and 10.7 g of sodium acetate. The diazonium solution was then added to the system with stirring under cooling with ice. After the dropwise addition was completed, the system was further stirred for 1 hour under ice-cooling and then for 2 hours at room temperature. The resulting crystal was filtered off. The crystal was then dispersed in 200 ml of methanol. A solution of 2.2 g of sodium hydroxide in 10 ml of water was added dropwise to the dispersion. The material was stirred for 3 hours. The system was neutralized with concentrated hydrochloric acid. The resulting crystal was washed with water and then with methanol, and then dried. The resulting crude crystal was purified with heat methanol in the same manner as in Synthesis Example 1 to obtain 14.8 g of Exemplary Coupler (YC-3). The melting point of the compound was 246 to 251 °C (decomposition). The structure of the compound was confirmed by ^{1}H-NMR spectrum, mass spectrum and elementary analysis. The compound exhibited a maximum absorption wavelength of 457.6 nm and a molecular extinction coefficient of 42,700 in methanol. Thus, the compound exhibited excellent spectral absorption characteristics as a yellow-colored coupler.

SYNTHESIS EXAMPLE 3

Synthesis of Exemplary Coupler (YC-30)



Synthesis of Compound e

137.1 g of anthranilic acid was added to 600 ml of acetonitrile. The mixture was heated with stirring. 92.5 g of diketene was added dropwise to the material over about 1 hour. The material was heated under reflux for 1 hour, and then cooled to room temperature. The resulting crystal was filtered off, washed with acetonitrile, and then dried to obtain 200.5 g of Compound e in a crystal form.

Synthesis of Compound f

199.1 g of Compound e, 89.2 g of ethyl cyanoacetate, and 344 g of 28% sodium methoxide were added to 0.9 ℓ of methanol. The reaction mixture was allowed to undergo reaction in an autoclave at a temperature of 120 °C for 8 hours. After being allowed to stand overnight, the reaction mixture was then concentrated under reduced pressure. 700 ml of water was added to the system. 230 ml of concentrated hydrochloric acid was added to the system so that the system was acidified. The resulting crystal was withdrawn by filtration. The resulting crude crystal was washed with a mixture of ethyl acetate and acetonitrile at an elevated temperature to obtain 152 g of Compound f.

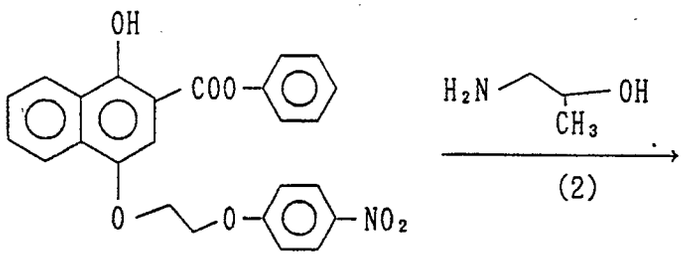
Synthesis of Exemplary Coupler (YC-30)

13.0 g of Compound g prepared by the synthesis method as described in U.S. Patent 4,138,258 was dissolved in 40 ml of N,N-dimethylformamide. 4.5 ml of concentrated hydrochloric acid was added to the solution under cooling with ice. A solution of 1.48 g of sodium nitrite in 5 ml of water was added dropwise to the system to prepare a solution of diazonium. 20 ml of N,N-dimethylformamide and 15 ml of water were added to 6.0 g of Compound f and 8 g of sodium acetate. The diazonium solution was added dropwise to the system with stirring under cooling with ice. After the completion of the dropwise addition, the system

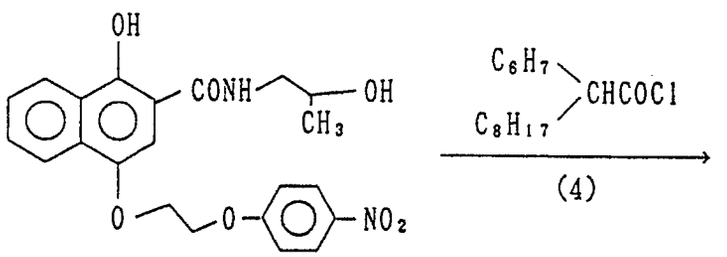
was further stirred at room temperature for 30 minutes. The system was acidified with hydrochloric acid. The system was extracted with ethyl acetate, washed with water, and then concentrated under reduced pressure. The concentrate was recrystallized from a mixture of ethyl acetate and methanol to obtain 13 g of Exemplary Coupler (YC-30) in the form of a yellow crystal. The melting point of the coupler was 154 to 156° C. The structure of the compound was confirmed by ^{1}H-NMR spectrum, mass spectrum and elementary analysis. The compound exhibited a maximum absorption wavelength of 458.2 nm and a molecular extinction coefficient of 42,800 in methanol. Thus, the compound exhibited excellent spectral absorption characteristics as a yellow-colored coupler.

SYNTHESIS EXAMPLE 4

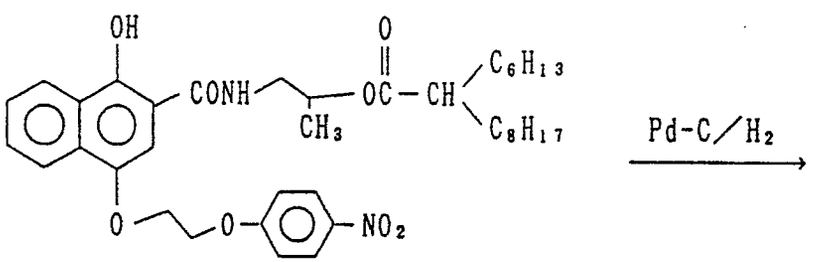
Synthesis of Exemplary Coupler (YC-86)



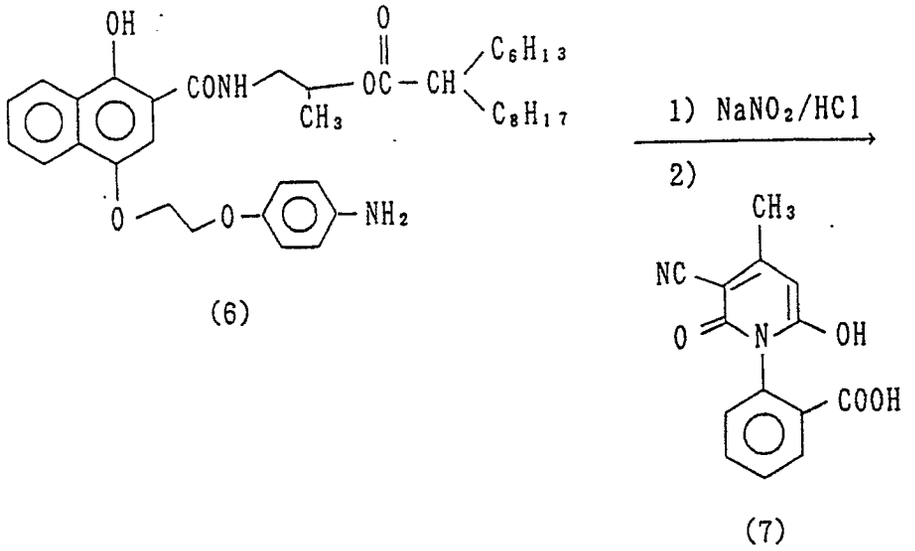
(1)



(3)

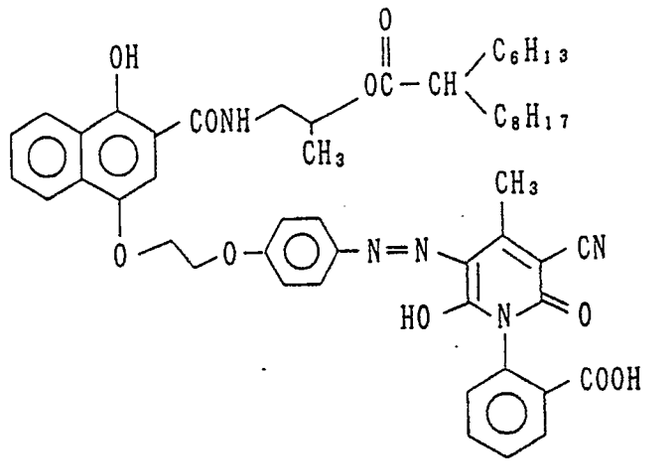


(5)



(6)

(7)



Exemplary Coupler (YC-86)

(1) Synthesis of Compound (3)

445.5 g of phenylester Compound (1) and 90.1 g of isopropanolamine (2) were heated in 600 ml of acetonitrile under reflux for 2 hours. After the system was cooled with water, the resulting crystal was filtered off, and then dried to obtain 342 g of Compound (3). (melting point (mp.) 162-165 °C).

(2) Synthesis of Compound (5)

341 g of hydroxyl Compound (3) and 231 g of 2-hexyldecanoyl chloride were heated in 880 ml of acetonitrile under reflux for 2 hours. After the system was cooled with water, the resulting crystal was filtered off, and then dried to obtain 437 g of nitro Compound (5) (mp. 97-100 °C).

(3) Synthesis of Compound (6)

370 g of nitro Compound (5), 6 g of 10% Pd-C catalyst, and 1 ℓ of ethyl acetate were charged into an autoclave. The material was then hydrogenated at a temperature of 50 °C for 3 hours. After the completion of reduction, the catalyst was filtered off, and the filtrate was then concentrated under reduced pressure to obtain a residue which was then crystallized from n-hexane. The crystal was filtered off, and then dried to

obtain 327 g of amine Compound (6) (mp. 95-97° C).

(4) Synthesis of Exemplary Coupler YC-86

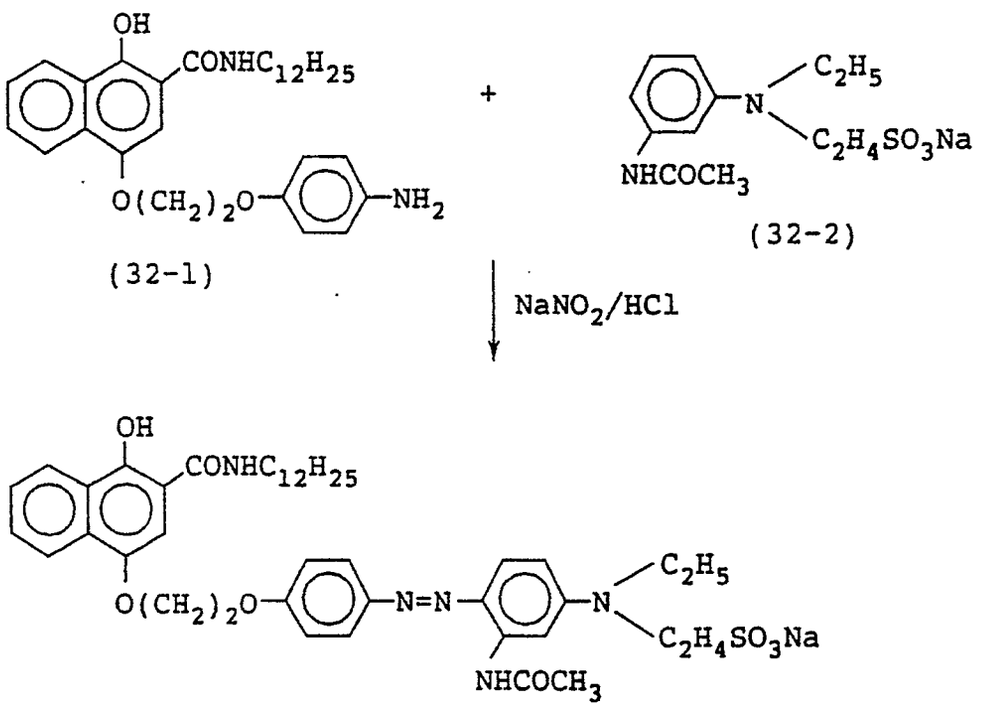
20.8 g of amine Compound (6) was dissolved in 60 ℓ of dimethylformamide. 7.6 ml of concentrated hydrochloric acid was added to the solution. An aqueous solution of 2.7 g of sodium nitrite in 10 ml of water was added dropwise to the system in 20 minutes. The system was further stirred for 30 minutes to prepare a diazo solution.

On the other hand, 9.7 g of pyridone Compound (7) and 13 g of sodium acetate were dissolved in a mixture of 30 ml of water and 30 ml of dimethylformamide at an elevated temperature. The system was then cooled with water. The diazo solution was gradually added to the system with stirring at a temperature of 10° C or lower. The system was further stirred for 15 minutes. The system was then extracted with ethyl acetate. The system was washed with water three times. The organic phase was concentrated under reduced pressure. The residue was crystallized from methanol-ethylacetate. The resulting crystal was filtered off, and dried to obtain 21.2 g of Exemplary Coupler YC-86 (mp. 117-119° C).

SYNTHESIS EXAMPLE 5

Synthesis of Exemplary Coupler (YC-32)

The compound (YC-32) is prepared by the following reaction process.



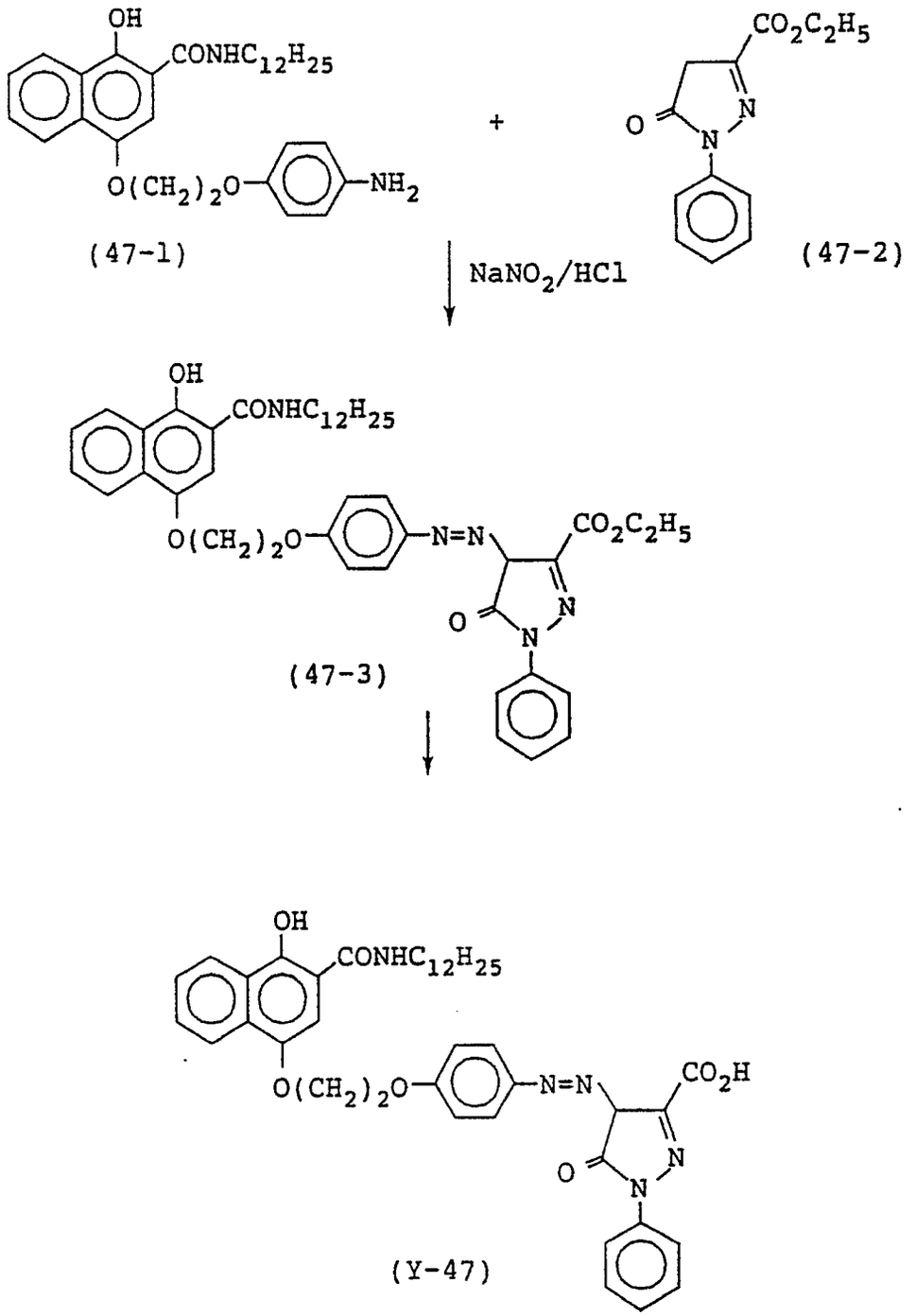
Into a mixture solution of 30 ml of N,N-dimethylformamide and 50 ml of methylcellosolve, was dissolved 9.1 g of a compound (32-1) and cooled to 5° C. After an addition of 4 ml of hydrochloric acid solution to the solution, 4 ml of an aqueous solution containing 1.28 g of sodium nitrite was added dropwise thereto under stirring. Under the temperature of about 5° C, the reaction was continued for 20 minutes.

6.30 g of the compound (32-2) and 12 g of sodium acetate were dissolved into a mixture of 20 ml of methylcellosolve and 10 ml of water, and cooled to 10° C. To the solution thus obtained, the diazonium salt solution obtained according to the previous processes was added dropwise. After dropwise addition, the mixture was stirred for 10 minutes, and 300 ml of water was added over about 20 minutes. The precipitated crystal was recovered by filtration to obtain 11.3 g of (YC-32).

SYNTHESIS EXAMPLE 6

Synthesis of Exemplary Coupler (YC-47)

Compound (YC-47) is prepared by the following reaction process.



Into a mixture solution of 150 ml of N,N-dimethylformamide and 90 ml of methylcellosolve, was dissolved 30.4 g of a compound (47-1) and was added 13 ml of hydrochloric acid. The solution thus obtained was cooled to 10 °C. 15 ml of an aqueous solution containing 4.3 g of sodium nitrite was added dropwise thereto under stirring, over 30 minutes. Under the temperature of 10 °C, the solution was stirred for 20 minutes. (Diazonium salt solution)

13.9 g of the compound (47-2) and 24.6 g of sodium acetate were dissolved into a mixture of 60 ml of

methylcellosolve and 30 ml of water, and cooled to 10 °C. To the solution thus obtained, the diazonium salt solution obtained in the above process was added dropwise over 1 hour. After dropwise addition, stirring finished to deposit oily precipitation. Supernatant liquid is removed by a decantation, and then 300 ml of acetonitrile was added to the residue and stirred. The precipitated crystal was filtered to obtain 26.2 g of the compound (47-3).

7 g of sodium hydroxide was dissolved in a mixture of 80 ml of water and 80 ml of methanol. 26.2 g of (47-3) was dissolved thereto and heated 45 °C with stirring.

After 15 minutes, the compound (47-3) was dissolved and then continued the reaction for 1 hour. After cooling to the room temperature, a mixture of 16.5 ml of hydrochloric acid and 50 ml of water was added dropwise. After 30 minutes stirring, precipitated crystal was filtered to obtain 25.0 g of the compound (YC-47).

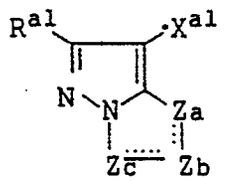
The synthesis of yellow-colored cyan couplers represented by the general formulae (CII) to (CIV) can be accomplished by any suitable method as described in JP-B-58-6939, JP-A-1-197563, JP-A-1-319744 and Japanese Patent Application Hei-1-316951 and those described for the method for synthesis of couplers of the general formula (CI) in the above cited patents.

In the present invention, yellow-colored cyan couplers as disclosed in the above cited JP-A-61-221748 and JP-A-1-319744 and yellow-colored cyan couplers represented by the general formulae (CI) to (CIV) can be used. The couplers represented by the general formulae (CI) to (CIV) are better than those described in the above cited JP-A-61-221748 and JP-A-1-319744 in view of their coupling activity and molecular extinction coefficient. Among the general formulae (CI) to (CIV), the couplers of general formulae (CI) and (CII) are better than those of general formulae (CIII) and (CIV). The yellow colored cyan couplers represented by the general formula (CI) are most preferable.

The yellow colored cyan coupler is preferably incorporated in a light-sensitive silver halide emulsion layer or in an adjacent layer thereto, particularly a red-sensitive emulsion layer, in a light-sensitive material. The total amount of the yellow-colored cyan coupler to be incorporated in the light-sensitive material can be from 0.005 to 0.30 g/m^2, preferably 0.02 to 0.20 g/m^2, more preferably 0.03 to 0.15 g/m^2.

The incorporation of the yellow-colored cyan coupler can be accomplished in the same manner as conventional couplers as described hereinafter.

The photographic material of the present invention contains a pyrazoloazole coupler represented by general formula (A).



(A)

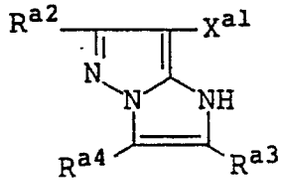
wherein R^{a1} represents a hydrogen atom or substituent; X^{a1} represents a hydrogen atom or a group capable of being separated therefrom upon a coupling reaction with an oxidation product of an aromatic primary amine developing agent; Za, Zb and Zc each represents a methine group, substituted methine group, =N- group or -NH- group; one of the Za-Zb and Zb-Zc bonds is a double bond and the other is a single bond; if the Zb-Zc bond is a carbon-carbon double bond, it may be a part of an aromatic ring; R^{a1} or X^{a1} may form a dimer or higher polymer; and if Za, Zb or Zc is a substituted methine, it may form a dimer or higher polymer.

The compound represented by the general formula (A) will be further described hereinafter.

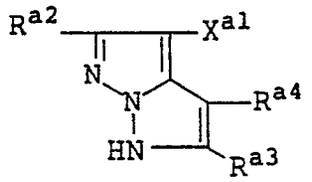
In the general formula (A), the term "polymer" means a group containing two or more groups represented by the general formulae (A) per molecule. A dimer and higher polymer couplers are included in the meaning of polymer. The polymer coupler may be a homopolymer comprising only a monomer unit containing a portion represented by the general formula (A) (preferably a monomer unit containing a vinyl group, hereinafter referred to as "vinyl monomer unit") or may form a copolymer with a noncoloring ethylenically unsaturated monomer which does not undergo a coupling reaction with an oxidation product of an aromatic primary amine developing agent.

The compound represented by the general formula (A) is a 5-membered ring/5-membered ring condensed nitrogen-containing heterocyclic coupler. The coloring nucleus of the coupler exhibits aromaticity isoelectric with naphthalene. Thus, the coupler has a chemical structure commonly referred to as "azapentalene". Preferred among couplers represented by the general formula (A) are 1H-imidazo[1,2-b]-

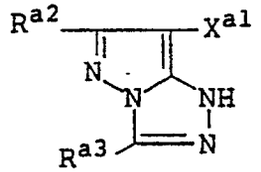
pyrazoles, 1H-pyrazolo[1,5-b]pyrazole, 1H-pyrazolo[5,1-c][1,2,4]triazole, 1H-pyrazolo[1,5-b][1,2,4]triazole, 1H-pyrazolo[1,5-d]tetrazole and 1H-pyrazolo[1,5-a]benzimidazole represented by the general formulae (A-1), (A-2), (A-3), (A-4), (A-5) and (A-6), respectively. Preferred among these compounds are (A-1), (A-3) and (A-4). Particularly preferred among these compounds are (A-3) and (A-4).



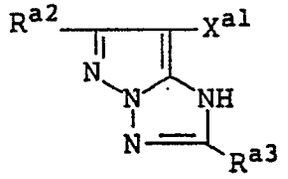
(A-1)



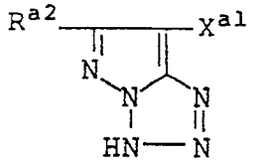
(A-2)



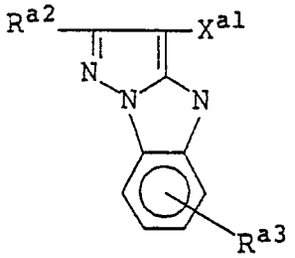
(A-3)



(A-4)



(A-5)



(A-6)

The substituents R^{a2}, R^{a3} and R^{a4} in the general formulae (A-1) to (A-6) each represents a hydrogen atom, a halogen atom, an alkyl group, an aryl group, a heterocyclic group, a cyano group, an alkoxy group, an aryloxy group, a heterocyclic oxy group, an acyloxy group, a carbamoyloxy group, a silyloxy group, a sulfonyloxy group, an acylamino group, an anilino group, a ureido group, an imido group, a sulfamoylamino group, an alkylthio group, an arylthio group, a heterocyclic thio group, an alkoxycarbonylamino group, an aryloxycarbonylamino group, a sulfonamido group, a carbamoyl group, an acyl group, a sulfamoyl group, a sulfonyl group, a sulfinyl group, an alkoxycarbamoyl group or an aryloxycarbonyl group. X^{a1} represents a hydrogen atom, a halogen atom, a carboxyl group or a group which is connected to the carbon atom in the coupling position via an oxygen atom, a nitrogen atom or a sulfur atom to undergo coupling elimination.

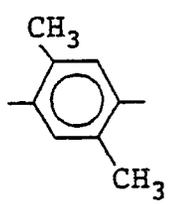
R^{a2}, R^{a3}, R^{a4} or X^{a1} may be a divalent group which forms a bis unit. If the portion represented by the general formula (A-1) to (A-6) is contained in a vinyl monomer unit, R^{a2}, R^{a3} or R^{a4} represents a bond or connecting group via which the portion represented by (A-1) to (A-6) is connected to the vinyl group.

More specifically, R^{a2}, R^{a3} and R^{a4} each represents a hydrogen atom, a halogen atom (e.g., chlorine, bromine), an alkyl group (e.g., methyl, propyl, t-butyl, trifluoromethyl, tridecyl, 3-(2,4-di-t-amylphenoxy)-propyl, 2-dodecyloxyethyl, 3-phenoxypropyl, 2-hexylsulfonyl ethyl, cyclopentyl, benzyl), an aryl group (e.g., phenyl, 4-t-butylphenyl, 2,4-di-t-amylphenyl, 4-tetradecanamidophenyl, perfluorophenyl), a heterocyclic group (e.g., 2-furyl, 2-thienyl, 2-pyrimidinyl, 2-benzothiazolyl), a cyano group, an alkoxy group (e.g., methoxy, ethoxy, 2-methoxyethoxy, 2-dodecyloxyethoxy, 2-methanesulfonylethoxy),an aryloxy group (e.g., phenoxy, 2-methylphenoxy, 4-t-butylphenoxy), a heterocyclic oxy group (e.g., 2-benzimidazolyloxy), an acyloxy group (e.g., acetoxy, hexadecanoyloxy), a carbamoyloxy group (e.g., N-phenylcarbamoyloxy, N-ethylcarbamoyloxy), a silyloxy group (e.g., trimethylsilyloxy), a sulfonyloxy group (e.g., dodecylsulfonyloxy), an acylamino group (e.g., acetamido, benzamido, tetradecanamido, α-(2,4-di-t-amylphenoxy)-butanamido, γ-(3-t-butyl-4-hydroxyphenoxy)butanamido, α-{4-(4-hydroxyphenylsulfonyl)phenoxy}dedanamido, an anilino group (e.g., phenylamino, 2-chloroanilino, 2-chloro-5-tetradecanamidoanilino, 2-chloro-5-dodecyloxycarbonylanilino, N-acetylanilino, 2-chloro-5-{α-(3-t-butyl-4-hydroxyphenoxy)dodecanamido} anilino), a ureido group (e.g., phenylureido, methylureido, N,N-dibutylureido), an imido group (e.g., N-succinimido, 3-benzylhydantoinyl, 4-(2-ethylhexanoylamino)phthalimido, sulfamoylamino group (e.g., N,N-dipropylsulfamoylamino, N-methyl-N-decylsulfamoylamino), an alkylthio group (e.g., methylthio, octylthio, tetradecylthio, 2-phenoxyethylthio, 3-phenoxypropylthio, 3-(4-t-butylphenoxy)propylthio), an arylthio group (e.g., phenylthio, 2-butoxy-5-t-octylphenylthio, 3-pentadecylphenylthio, 2-carboxyphenylthio, 4-tetradecanamidophenylthio), a heterocyclic thio group (e.g., 2-benzothioazolylthio), an alkoxycarbonylamino group (e.g., methoxycarbonylamino, tetradecyloxycarbonylamino), an aryloxycarbonylamino group (e.g., phenoxycarbonylamino, 2,4-di-tert-butylphenoxycarbonylamino), a sulfonamido group (e.g., methanesulfonamido, hexadecanesulfonamido, benzenesulfonamido, p-toluenesulfonamido, octadecanesulfonamido, 2-methyloxy-5-t-butylbenzenesulfonamido), a carbamoyl group (e.g., N-ethylcarbamoyl, N,N-dibutylcarbamoyl,

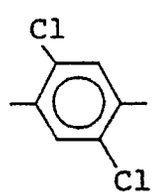
N-(2-dodecyloxyethyl)carbamoyl, N-methyl-N-dodecylcarbamoyl, N-{3-(2,4-di-tert-amylphenoxy)-propyl}carbamoyl), an acyl group (e.g., acetyl, (2,4-di-tert-amylphenoxy)acetyl, a benzoyl (e.g., perfluorobenzoyl), a sulfamoyl group (e.g., N-ethylsulfamoyl, N,N-dipropylsulfamoyl, N-(2-dodecyloxyethyl)-sulfamoyl, N-ethyl-N-dodecylsulfamoyl, N,N-diethylsulfamoyl), a sulfonyl group (e.g., methanesulfonyl, octanesulfonyl, benzenesulfonyl, toluenesulfonyl, perfluorophenylsulfonyl), a sulfinyl group (e.g., octylsulfinyl, dodecylsulfinyl, phenylsulfinyl), an alkoxycarbonyl group (e.g., methoxycarbonyl, butyloxycarbonyl, dodecyloxycarbonyl, octadecyloxycarbonyl), or an aryloxycarbonyl group (e.g., phenyloxycarbonyl, 3-pentadecylphenyloxycarbonyl).

X^{a1} represents a hydrogen atom, a halogen atom (e.g., fluorine, chlorine, bromine, iodine), a carboxyl group, a group which is connected to the compound via an oxygen atom (e.g., acetoxy, propanoyloxy, benzoyloxy, 2,4-dichlorobenzoyloxy, ethoxyoxaloyloxy, pyruvinyloxy, cinnamoyloxy, phenoxy, 4-cyanophenoxy, 4-methanesulfonamidophenoxy, 4-methanesulfonylphenoxy, α-naphthoxy, 3-pentadecylphenoxy, benzyloxycarbonyloxy, ethoxy, 2-cyanoethoxy, benzyloxy, 2-phenethyloxy, 2-phenoxyethoxy, 5-phenyltetrazolyloxy, 2-benzothiazolyloxy), a group which is connected to the compound via a nitrogen atom (e.g., benzenesulfonamido, N-ethyltoluenesulfonamido, heptafluorobutanamido, 2,3,4,5,6-pentafluorobenzamido, octanesulfonamido, p-cyanophenylureido, N,N-diethylsulfamoylamino, 1-piperidyl, 5,5-dimethyl-2,4-dioxo-3-oxazolidinyl, 1-benzyl-ethoxy-3-hydantoinyl, 2N-1,1-dioxo-3(2H)-oxo-1,2-benzoisothiazolyl, 2-oxo-1,2-dihydro-1-pyridinyl, imidazolyl, pyrazolyl, 3,5-diethyl-1,2,4-triazol-1-yl, 5- or 6-bromobenzotriazol-1-yl, 5-methyl-1,2,3-triazol-1-yl, benzimidazolyl, 3-benzyl-1-hydantoinyl, 1-benzyl-5-hexadecyloxy-3-hydantoinyl, 5-methyl-1-tetrazolyl, 4-methoxyphenylazo, 4-pivaloylaminophenylazo, 2-hydroxy-4-propanoylphenylazo), or a group which is connected to the compound via a sulfur atom (e.g., phenylthio, 2-carboxyphenylthio, 2-methoxy-5-t-octylphenylthio,4-methanesulfonylphenylthio, 4-octanesulfonamidophenylthio, 2-butoxyphenylthio, 2-(2-hexanesulfonylethyl)-5-tert-octylphenylthio, benzylthio, 2-cyanoethylthio, 1-ethoxycarbonyltridecylthio, 5-phenyl-2,3,4,5-tetrazolylthio, 2-benzothiazolylthio, 2-dodecylthio-5-thiophenylthio, 2-phenyl-3-dodecyl-1,2,4-triazolyl-5-thio).

If R^{a2}, R^{a3}, R^{a4} or X^{a1} is a divalent group which forms a bis unit, specific examples of such a divalent group include a substituted or unsubstituted alkylene group (e.g., methylene, ethylene, 1,10-decylene, $-CH_2CH_2-O-CH_2CH_2-$), a substituted or unsubstituted phenylene group (e.g., 1,4-phenylene, 1,3-phenylene,



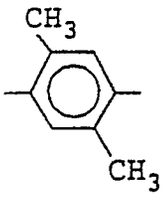
,



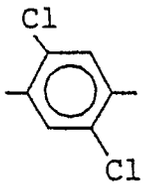
,

and a -NHCO-R^{a5}-CONH- group (in which R^{a5} represents a substituted or unsubstituted alkylene or phenylene group).

If the portion represented by the general formula (A-1) to (A-6) is contained in a vinyl monomer unit, examples of the connecting groups represented by R^{a2}, R^{a3} or R^{a4} include groups formed by combining groups selected from an alkylene group (substituted or unsubstituted alkylene group, e.g., methylene, ethylene, 1,10-decylene, $-CH_2CH_2OCH_2CH_2-$), a phenylene group (substituted or unsubstituted phenylene group, e.g., 1,4-phenylene, 1,3-phenylene,

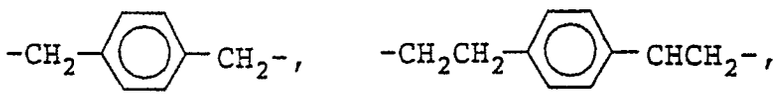


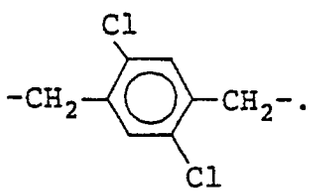
,



,

-NHCO-, -CONH-, -O-, -OCO-, and an aralkylene group (e.g.,



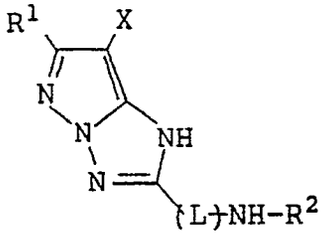


.

The vinyl group contained in the vinyl monomer unit may contain substituents rather than being the group represented by the general formula (A-1) to (A-6). Preferred examples of such substituents are a hydrogen atom, a chlorine atom, a C_{1-4} lower alkyl group and a substituted or unsubstituted aryl group.

Examples of the noncoloring ethylenically unsaturated monomer unit which does not undergo a coupling reaction with an oxidation product of an aromatic primary amine developing agent include acrylic acid, α-chloroacrylic acid, an α-arkacrylic acid (e.g., methacrylic acid), an amido or ester derived from these acrylic acids (e.g., acrylamido, n-butylacrylamido, t-butylacrylamido, diacetonacrylamido, methacrylamido, methyl acrylate, ethyl acrylate, n-propyl acrylate, n-butyl acrylate, t-butyl acrylate, iso-butyl acrylate, 2-ethylhexyl acrylate, n-octyl acrylate, lauryl acrylate, methyl methacrylate, ethyl methacrylate, n-butyl methacrylate, β-hydroxy methacrylate), methylene dibisacrylamide, a vinylester (e.g., vinyl acetate, vinyl propionate, vinyl laurate), acrylonitrile, methacrylonitrile, an aromatic vinyl compound (e.g., styrene and derivatives thereof, vinyltoluene, divinylbenzene, vinylacetophenone, sulfostyrene), itaconic acid, citraconic acid, crotonic acid, vinylidene chloride, a vinyl alkyl ether (e.g., vinyl ethyl ether), maleic acid, maleic anhydride, maleic ester, N-vinyl-2-pyrrolidone, N-vinylpyridine, and 2- and 4-vinylpyridine. Two or more of these noncoloring ethylenically unsaturated monomer units can be used.

Among the above mentioned particularly preferred compounds (A-3) and (A-4), the most preferably used are the compounds represented by the general formula (A-4), particularly those of the following general formula (M):



(M)

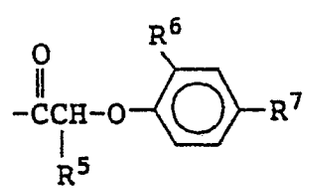
wherein R^1 represents an alkyl group, an alkoxy group or an aryloxy group; R^2 represents an acyl group or a sulfonyl group; -(L)- represents an alkylene or a phenylene group represented by $-(C(R^3)(R^4)-CH_2)-$, with the proviso that when -(L)- is an alkylene group, the carbon atom to which R^3 and R^4 are connected is connected to the coupler nucleus, and R^3 and R^4 each represents a hydrogen atom, an alkyl group or an aryl group, but are not hydrogen atoms at the same time; and X represents an aryloxy group, an alkoxy group, a 1-azolyl group, an alkylthio group or an arylthio group.

R^1, R^2 or X may be a divalent group which forms a bis unit. If the portion represented by the general formula (M) is contained in the vinyl monomer unit, any of R^1, R^2 and X represents a bond or a connecting group via which it is connected to the vinyl group.

The substituents R^1, R^2, -(L)- and X in the general formula (M) will be further described hereinafter.

R^1 is an alkyl group, alkoxy group or aryloxy group. Specifically, R^1 is a C_{1-30} straight-chain or branched alkyl group, C_{1-20} alkoxy group or C_{6-20} aryloxy group. More specifically, R^1 represents an alkyl group, such as methyl, ethyl, propyl, isopropyl, t-butyl, 2-ethylhexyl, dodecyl, 1-ethylpentyl, tridecyl, 2-methanesulfonylethyl, 3-(3-pentadecylphenoxy)propyl, 3-{4-{2-[4-(4-hydroxyphenylsulfonyl)phenoxy]-dodecanamide}phenyl}propyl, 2-ethoxytridecyl, trifluoromethyl, cyclohexyl, and 3-(2,4-di-t-amylphenoxy)-propyl; an alkoxy group such as methoxy, ethoxyisopropoxy, t-butoxy, 2-methoxyethoxy, 2-dodecylethoxy, 2-methanesulfonylethoxy, and 2-phenoxyethoxy; or an aryloxy group such as phenoxy, 2-napthyloxy, 2-methylphenoxy, 2-methoxyphenoxy, 4-methoxyphenoxy, 4-t-butylphenoxy, 3-nitrophenoxy, 3-acetamidophenoxy, and 2-benamidophenoxy. These groups may contain further substituents such as a halogen atom, an alkyl group, an aryl group, a heterocyclic group, a cyano group, a hydroxy group, a nitro group, a carboxy group, a sulfo group, an amino group, an alkoxy group, an aryloxy group, an acylamino group, an alkylamino group, an anilino group, a ureide group, a sulfamoylamino group, an alkylthio group, an arylthio group, an alkoxycarbonylamino group, a sulfonamide group, a carbamoyl group, a sulfamoyl group, a sulfonyl group, an alkoxycarbonyl group, a heterocyclic oxy group, an azo group, an acyloxy group, a carbamoyloxy group, a silyloxy group, an aryloxycarbonylamino group, an imide group, a heterocyclic thio group, a sulfinyl group, a phosphonyl group, an aryloxycarbonyl group, an acyl group, and an azolyl group. R^1 is preferably an alkyl group such as methyl, ethyl, isopropyl and t-butyl, an alkoxy group such as methoxy, ethoxy, isopropoxy, 2-methoxyethoxy, and 2-phenoxyethoxy, or an aryloxy group such as phenoxy, 2-methoxyphenoxy, 4-methoxyphenoxy, and 2-methylphenoxy.

R^2 represents an acyl group or sulfonyl group. Preferably, R^2 represents an acyl group such as C_{8-40} alkanoyl and aryloyl groups or sulfonyl group such as C_{10-40} alkylsulfonyl and arylsulfonyl groups. More preferably, R^2 represents (a) a straight-chain or branched alkanoyl group such as 2-ethylhexanoyl, decanoyl, tetradecanoyl, pentadecanoyl, stearoyl and isostearoyl, straight-chain alkanoyl group represented by the general formula (A_1):



(A_1)

wherein R^5 represents a hydrogen atom or alkyl group, and R^6 and R^7 each represents an alkyl group; (b) an aryloyl group such as 4-stearyloxybenzoyl, 3-(2-ethylhexanoylamino)benzoyl, 2,4-dioctyloxybenzoyl, 4-(4-dodecyloxybenzenesulfonamido)benzoyl, and 1-octyloxy-2-naphthoyl; (c) an alkylsulfonyl group such as dodecylsulfonyl and octadecylsulfonyl, or (d) an arylsulfonyl group such as 2-butyloxy-5-t-octylbenzenesulfonyl, 1-octyloxy-4-napthylsulfonyl, 2-octyloxy-5-t-octylbenzenesulfonyl, 2-(2-hexyloxyethoxy)-5-t-octylben-

zenesulfonyl, 2-(2-ethoxyethoxy)-5-(2-octyloxy-5-t-octylbenzenesulfonamide)benzenesulfonyl and 2-octyloxy-5-(2-octyloxy-5-t-octylbenzenesulfonamido)benzenesulfonyl. These groups may further contain the substituents described with reference to R^1. R^2 is preferably a substituted alkanoyl group represented by the general formula (A_1) or a substituted arylsulfonyl.

The group -(L)- represents an alkylene group represented by $-C(R^3)(R^4)-CH_2-$ wherein R^3 and R^4 each represents a hydrogen atom or an alkyl group such as methyl, ethyl, propyl, isopropyl, t-butyl and octyl or an aryl group such as phenyl, tolyl and 2-naphthyl or a phenylene group such as 1,2-phenylene, 1,3-phenylene, 1,4-phenylene, 4-methoxy-1,3-phenylene and 5-methyl-1,3-phenylene, with the proviso that R^3 and R^4 are not both hydrogen atoms at the same time. Preferably, -(L)- represents an alkylene group wherein R^3 and R^4 each is a hydrogen atom, a methyl group, a phenyl group or a phenylene group such as 1,3-phenylene and 1,4-phenylene.

X represents an aryloxy, alkoxy, 1-azolyl, alkylthio or arylthio group. Specifically, X represents an aryloxy group such as phenoxy, 4-methylphenoxy, 4-cyanophenoxy, 4-methanesulfonamidophenoxy, 4-acetamidophenoxy, 4-ethoxycarbonylphenoxy, 4-carboxyphenoxy, 3-carboxyphenoxy, 2-carboxyphenoxy, 4-[{1,1-dimethyl-1-(4-hydroxyphenyl)}methyl]phenoxy, 4-(4-hydroxybenzenesulfonyl)phenoxy, 4-methoxyphenoxy, 1-naphthoxy, 2-phenethyloxy, 5-phenyltetrazolyloxy and 2-benzothiazolyloxy, an alkoxy group such as methoxy, ethoxy, isopropoxy, t-butoxy, ethoxycarbonylmethoxy, 2-ethoxycarbonylethoxy, 2-cyanethoxy, 2-methanesulfonylethoxy, 2-benzenesulfonylethoxy, and 2-phenoxyethoxy, a 1-azolyl group such as 1-pyrazolyl, 1-imidazolyl, 3,5-dimethyl-1,2,4-triazol-1-yl, 5- or 6-bromobenzotriazol-1-yl, 5-methyl-1,2,3,4-tetrazol-1-yl, 1-benzimidazolyl, 4-chloropyrazol-1-yl, 4-nitro-pyrazol-1-yl, 4-ethoxycarbonyl-1-yl, 3- or 5-acetamidepyrazol-1-yl, and 2-acetamideimidazolyl-1-yl, an alkylthio group such as dodecylthio, and 1-carboxydodecylthio, or an arylthio group such as phenylthio, 2-naphthylthio, 2-butoxy-5-t-octylphenylthio, 2-pivaloylaminophenylthio, 4-dodecylphenylthio, 4-octyloxyphenylthio, 2-octyloxy-5-carboxyphenylthio, and 2-(3-carboxypropyloxy)-5-t-octylphenylthio. These groups may further contain the substituents s described with reference to R^1. X is preferably an aryloxy group, a 1-azolyl group or an arylthio group. X is more preferably a substituted phenoxy, substituted pyrazol-1-yl or substituted phenylthio group.

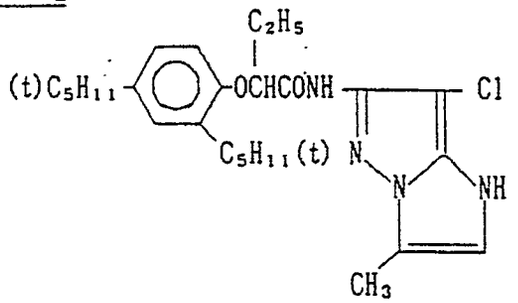
Examples of couplers represented by the general formulae (A-1) to (A-6) and (M) and synthesis methods thereof are described in the following literature references.

Compounds represented by the general formula (A-1) are described in JP-A-59-162548 and U.S. Patent 4,500,630. Compounds represented by the general formula (A-2) are described in JP-A-60-43659. Compounds represented by the general formula (A-3) are described in JP-B-47-27411, and U.S. Patent 3,725,067. Compounds represented by the general formula (A-4) are described in U.S. Patents 4,540,654, 4,705,863 and JP-A-61-65245, JP-A-62-209457 and JP-A-62-249155. Compounds represented by the general formulae (A-4) and (M) are described in JP-A-59-171956 and JP-A-60-172982. Compounds represented by the general formula (A-5) are described in JP-A-60-33552. Compounds represented by the general formula (A-6) are described in U.S. Patent 3,061,432.

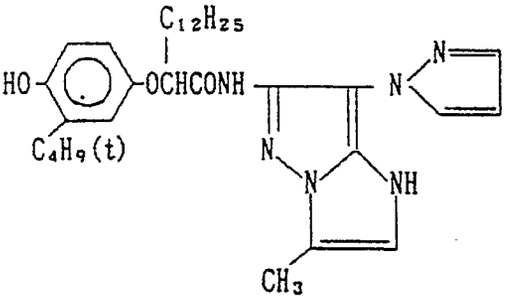
High coloring ballast groups as described in JP-A-58-42045, JP-A-59-214854, JP-A-59-177553, JP-A-59-177554 and JP-A-59-177557 can be appended to any of the compounds represented by the general formulae (A-1) to (A-6) and (M).

Specific examples of pyrazoloazole couplers to be used in the present invention, but the present invention should not be construed as being limited thereto.

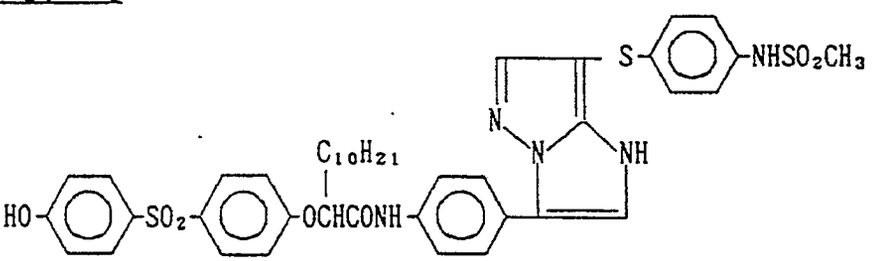
(A－1)－1



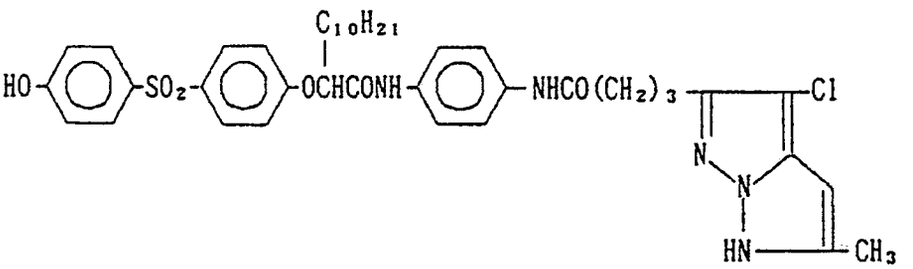
(A－1)－2

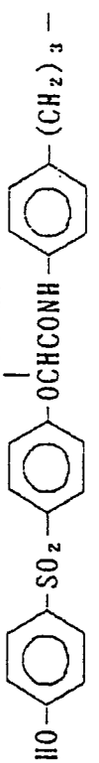
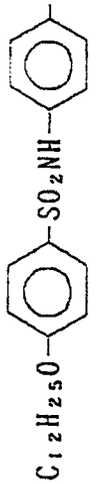
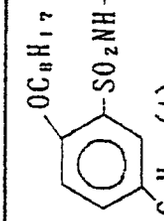


(A－1)－3

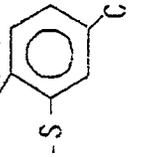
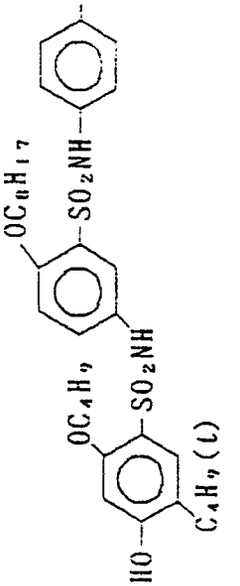
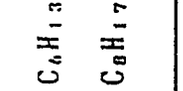


(A－2)－1

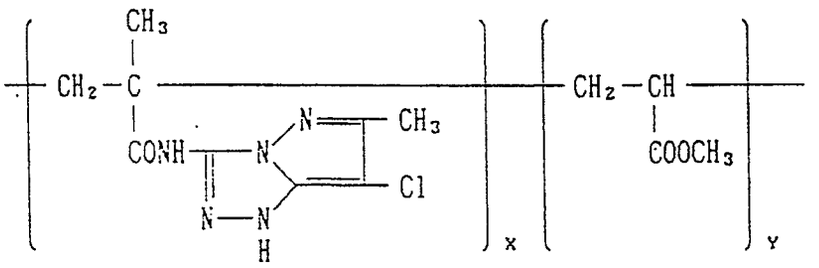


Coupler	R^{a2}	R^{a3}	X^{a1}
(A-3)-1	CH_3		Cl
〃 -2	CH_3		Cl
〃 -3			F
〃 -4	CH_3		Cl

Coupler	R^{a2}	R^{a3}	X^{a1}
(A-3)-5	CH_3, CH_3, CH	OC_8H_{17}, $SO_2CH_2CH_2CH_2-$, $C_8H_{17}(t)$	Cl
〃 -6	CH_3, CH_3, CH-	$C_{12}H_{25}O-$, $-SO_2NH-$, $O(CH_2)_2S-$	OC_4H_9, $C_8H_{17}(t)$, -S-
〃 -7	CH_3-	OC_8H_{17}, $-SO_2NH-$, $-CH_2S-$, $C_8H_{17}(t)$	OC_4H_9, $C_8H_{17}(t)$, -S-
〃 -8	$(CH_3)_3C-$	OC_4H_9, $SO_2(CH_2)_2-$, $C_8H_{17}(t)$	Cl

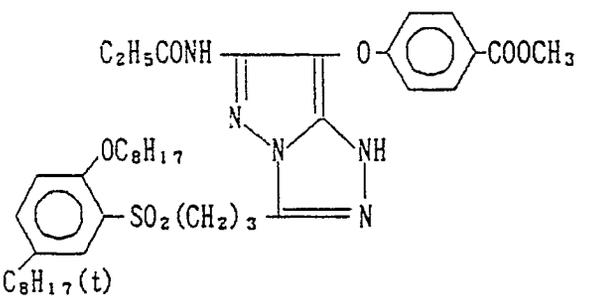
Coupler	R^{a2}	R^{a3}	X^{a1}
(A-3)-9	CH_3-		
〃 -10	CH_3-		Cl
〃 -11	CH_3-		Cl

〔A－3〕－12

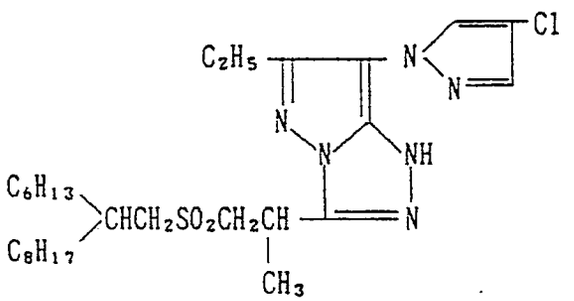


X：Y＝40：60 (by mole)

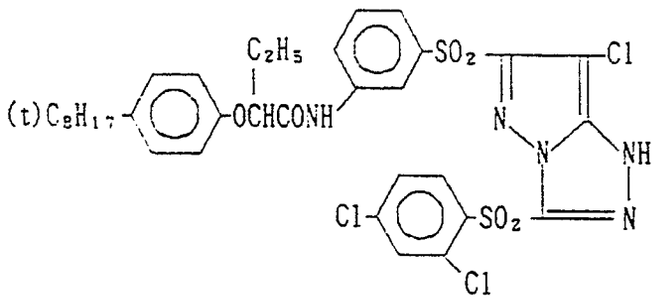
〔A－3〕－13



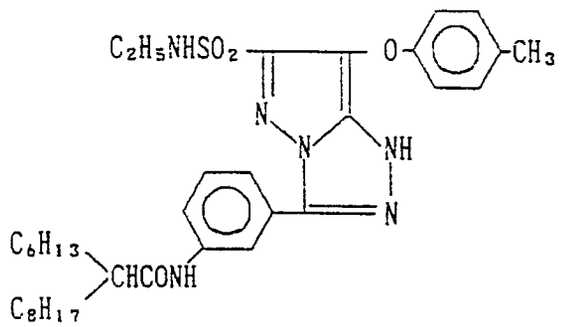
〔A－3〕－14



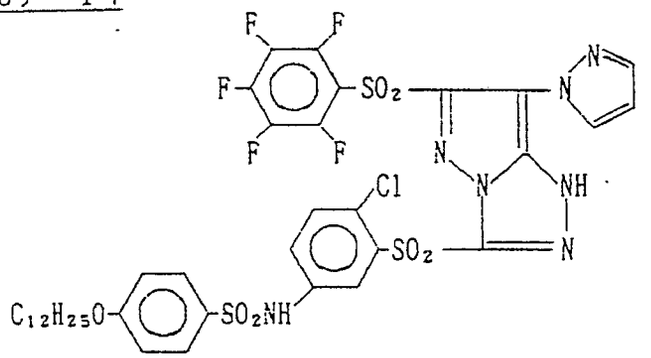
〔A－3〕－15



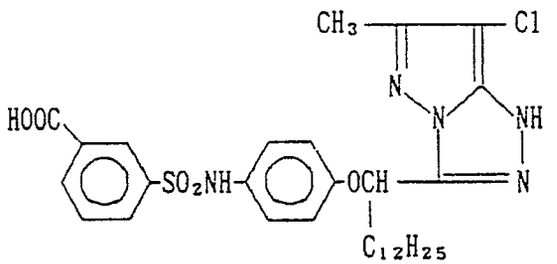
〔A－3〕－16



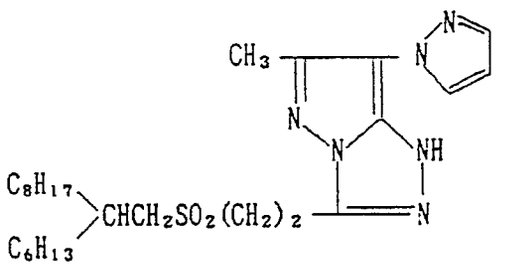
〔A－3〕－17



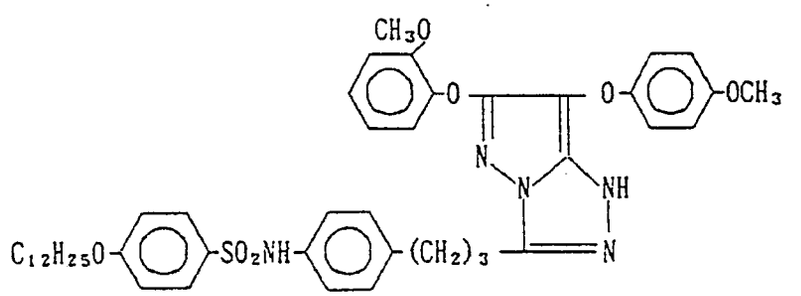
〔A－3〕－18

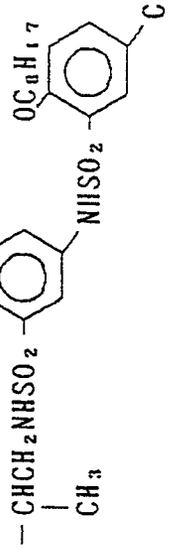
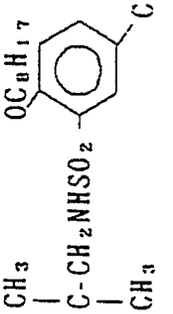
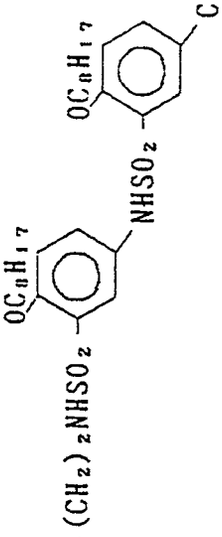
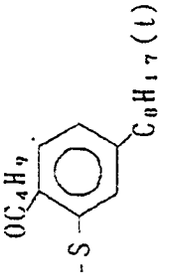


〔A－3〕－19

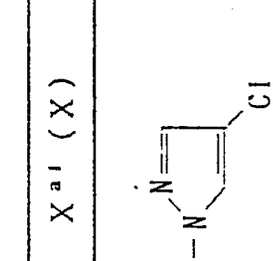
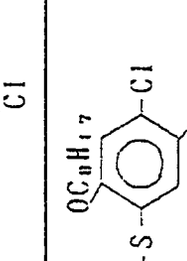
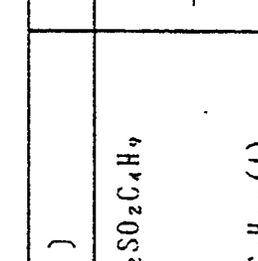
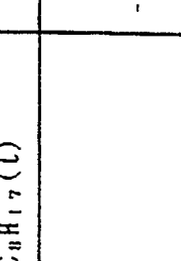
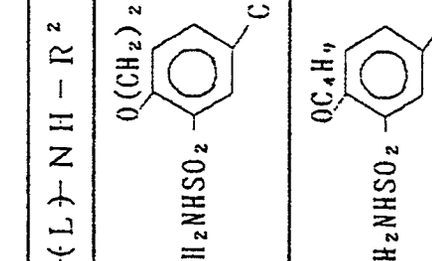
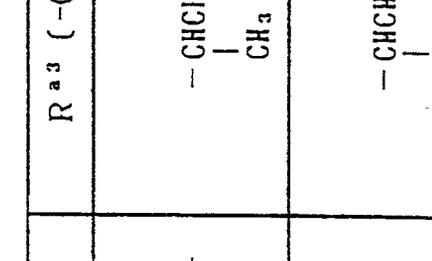


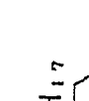
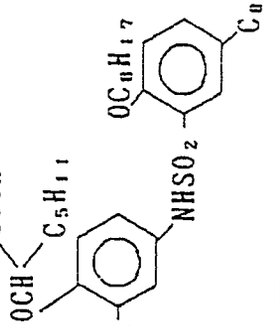
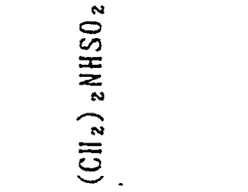
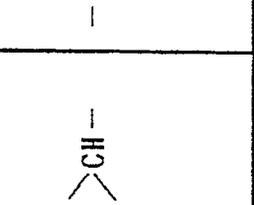
〔A－3〕－20



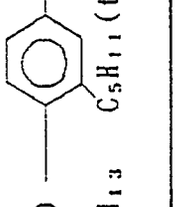
Coupler	$R^{a2}(R^1)$	$R^{a3}(-(L)-NH-R^2)$	$X^{a1}(X)$
(A-4)-1	CH_3		Cl
〃 -2	CH_3		F
〃 -3	C_2H_5O-		

Coupler	$R^{a2}(R^1)$	R^{a3} (-(L)-NH-R^2)	X^{a1} (X)
(A-4)-4	CH_3 CH_3 >CHO-		
" -5	$CH_3O(CH_2)_2O$-		
" -6	$CH_3NHCONH$-		Same as above

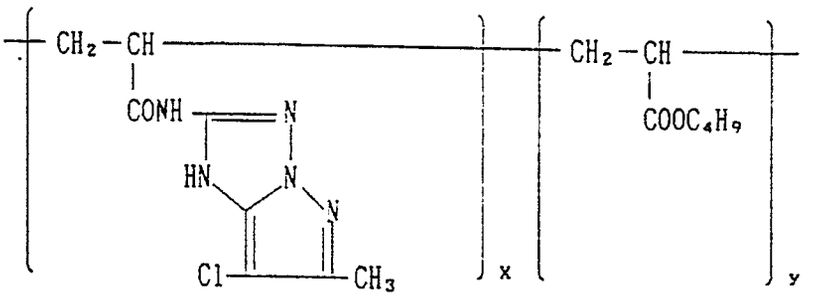
Coupler	R^{a2} (R')	R^{a3} (-(L)-NH-R^2)	X^{a1} (X)
(A-4)-7			
" -8	CF_3CH_2O-		
" -9			
" -10	$CH_3SO_2(CH_2)_2O-$		Same as above

Coupler	R^{a2} (R')	R^{a3} (-(L)-NH-R²)	X^{a1} (X)
(A-4)-11			
〃 -12	CH_3-		Cl
〃 -13	CH_3-		Cl

Coupler	R^{a2} (R^1)	R^{a3} ($-(L)-NH-R^2$)	X^{a1} (X)
(A-4)-14			
" -15			Same as above
" -16			
" -17			

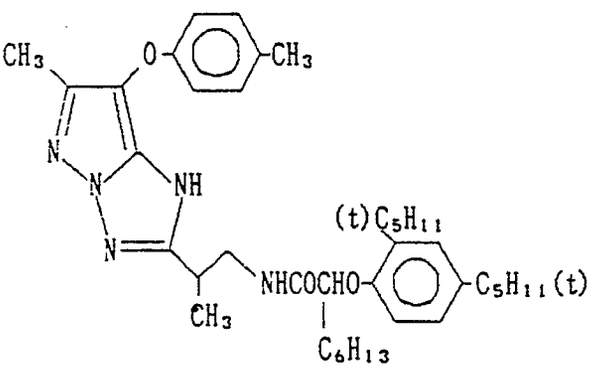
Coupler	R^{a2} (R^1)	R^{a3} (-(L)-NH-R^2)	X^{a1} (X)
(A-4)-18	CH_3-		Cl

(A－4)－19

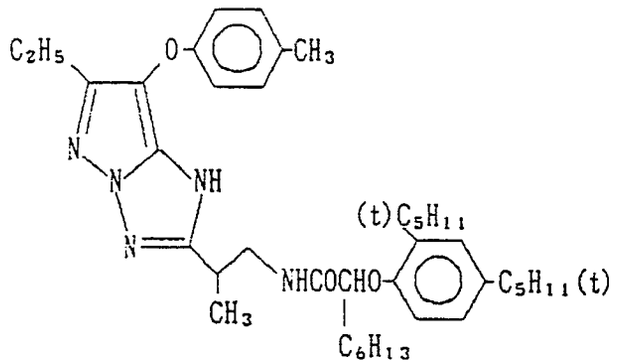


X：Y＝50：50 (by mole)

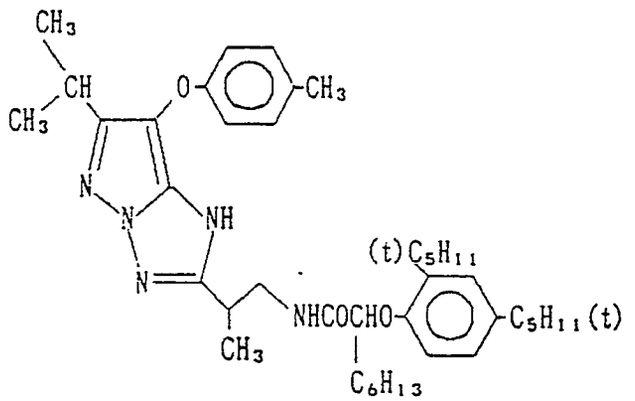
(A－4)－20



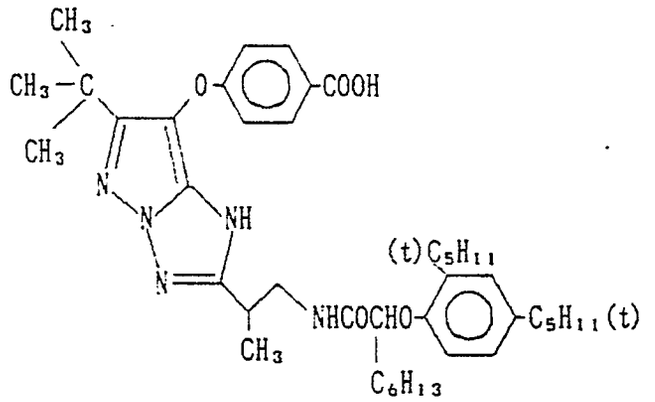
〔A－4〕－21



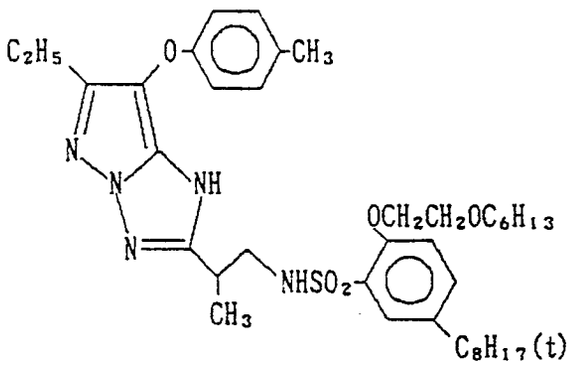
〔A－4〕－22



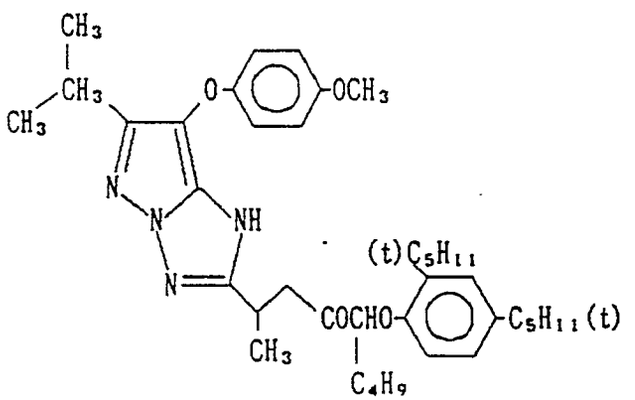
(A－4)－23



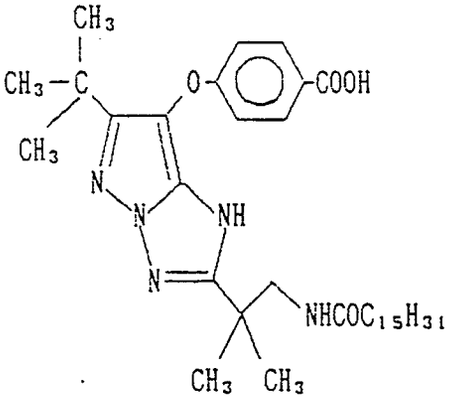
(A－4)－24



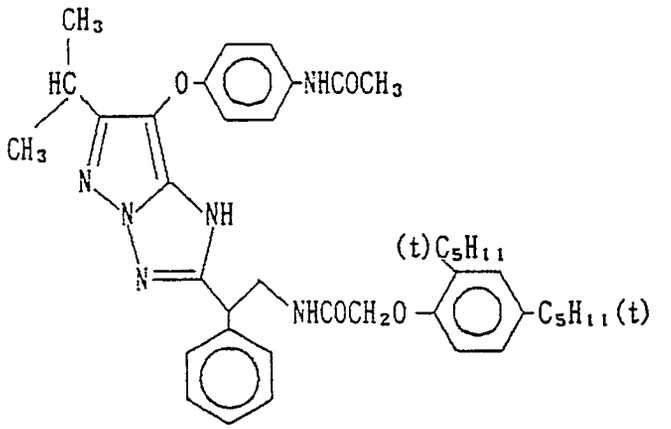
(A－4)－25



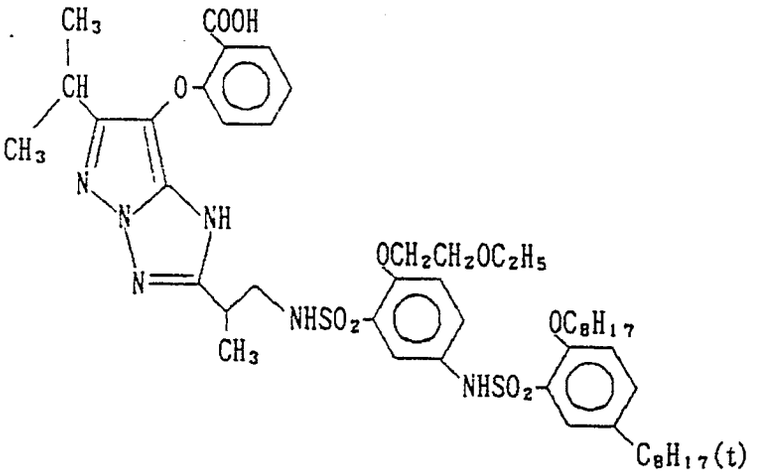
〔A－4〕－26



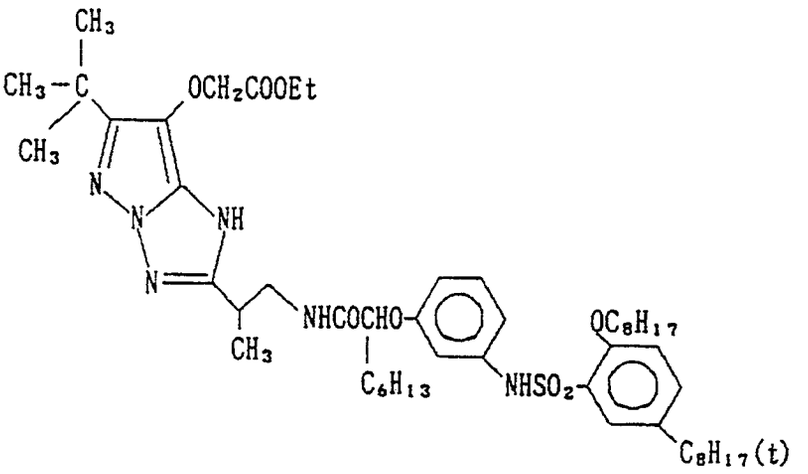
〔A－4〕－27



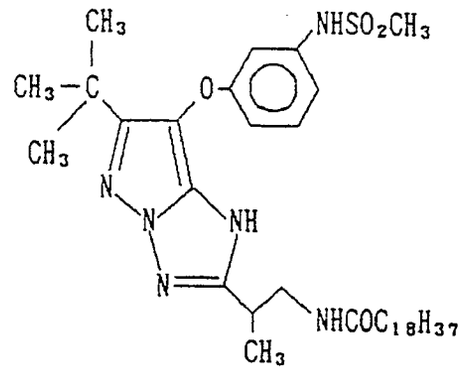
(A－4)－28



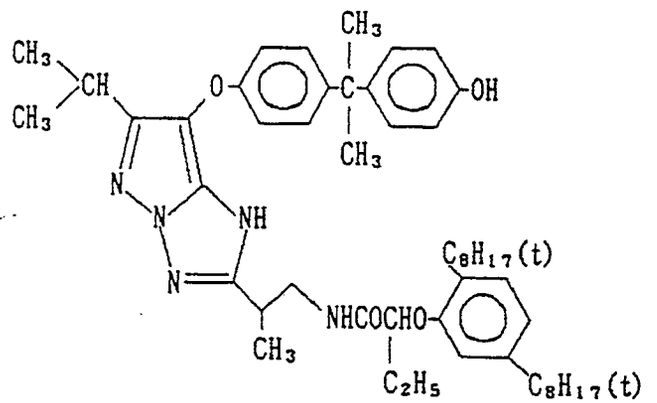
(A－4)－29



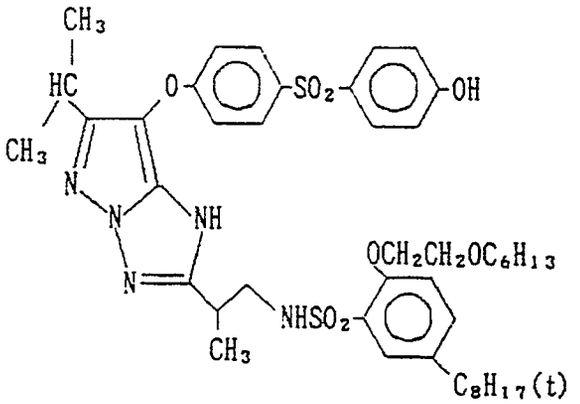
(A－4)－30



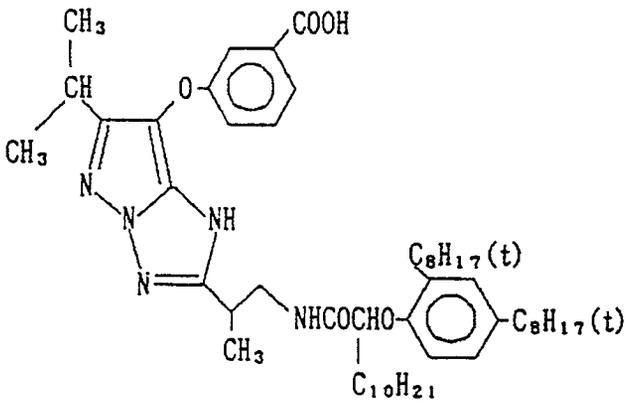
(A－4)－31



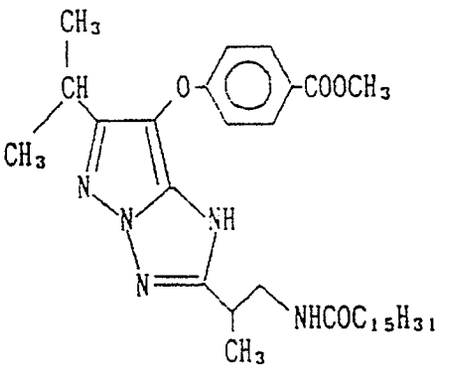
〔A－4〕－32



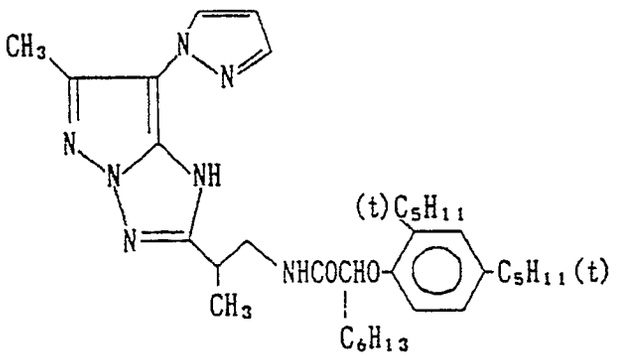
〔A－4〕－33



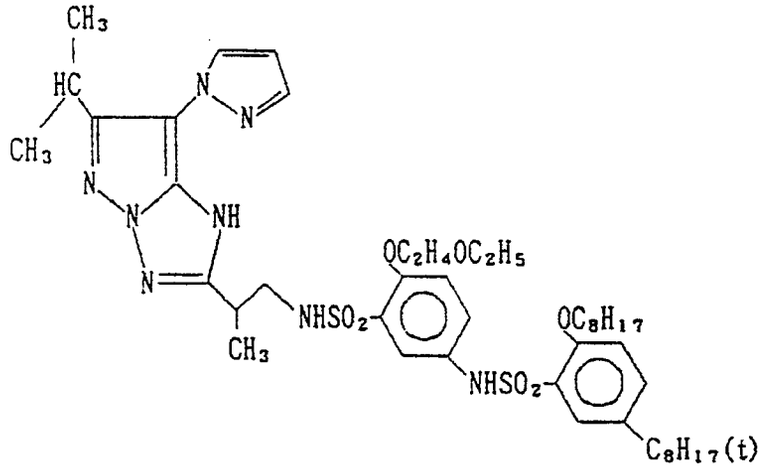
(A－4)－34



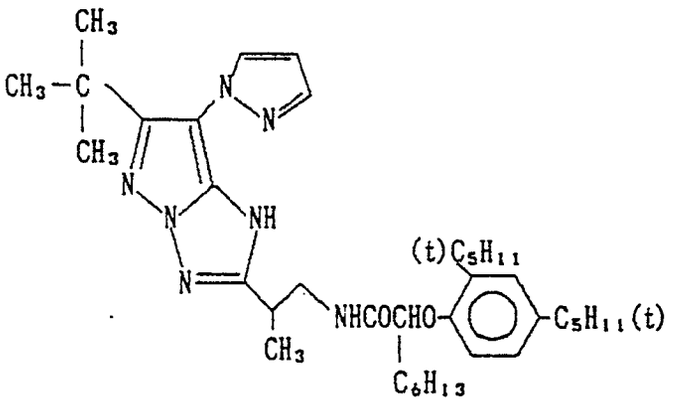
(A－4)－35



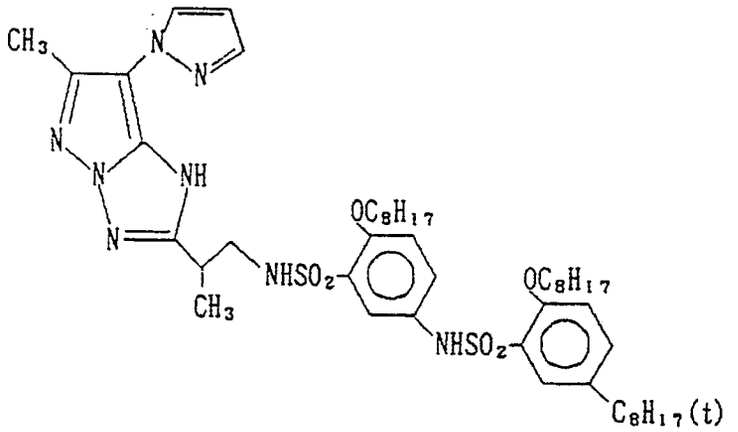
〔A－4〕－36



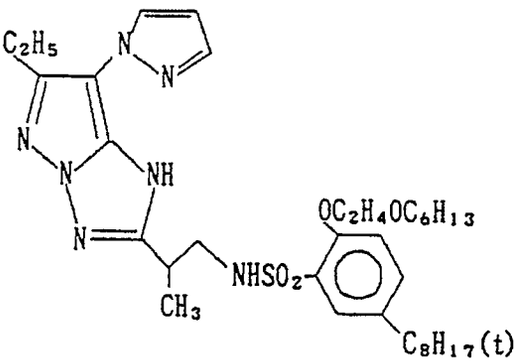
〔A－4〕－37



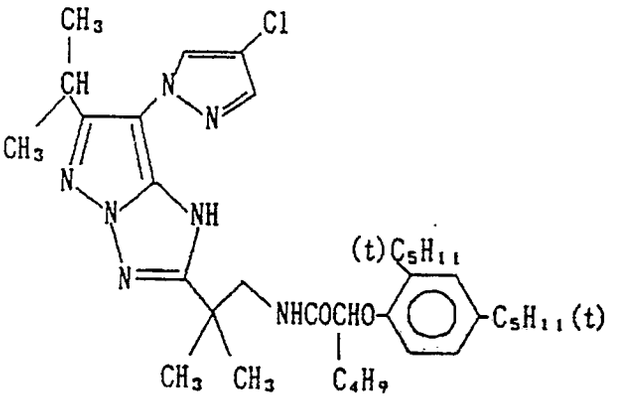
(A－4)－38



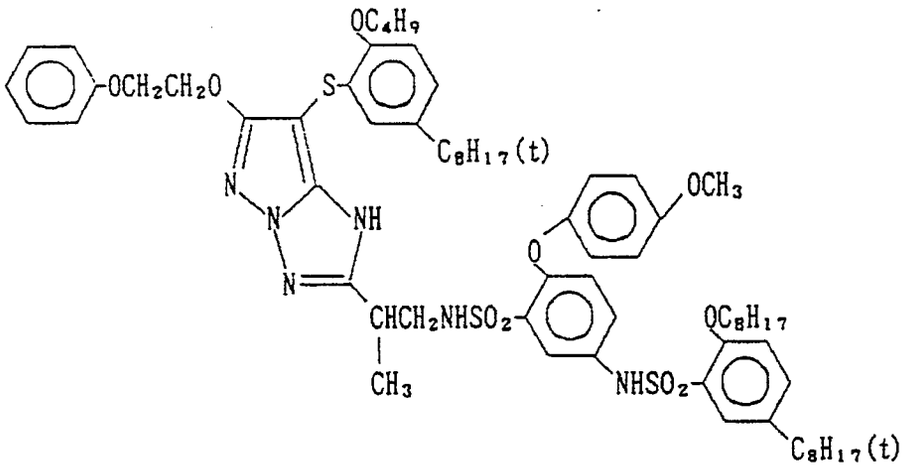
(A－4)－39



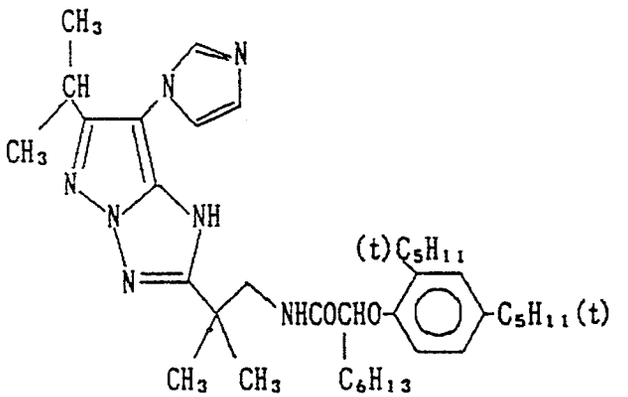
(A－4)－40



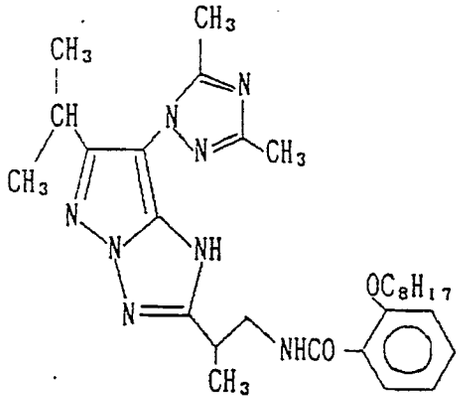
(A－4)－41



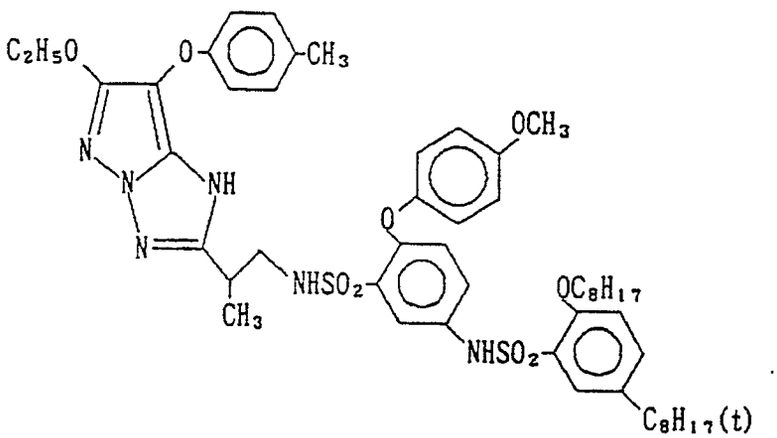
(A－4)－42



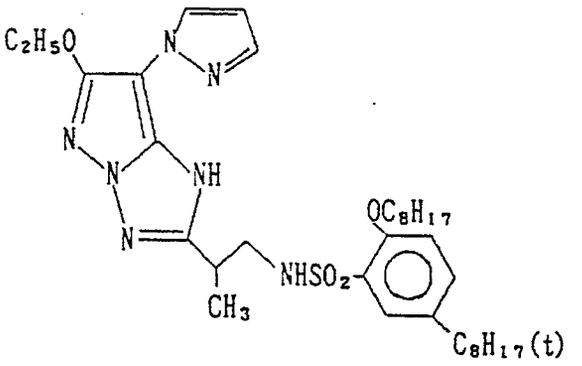
(A－4)－43



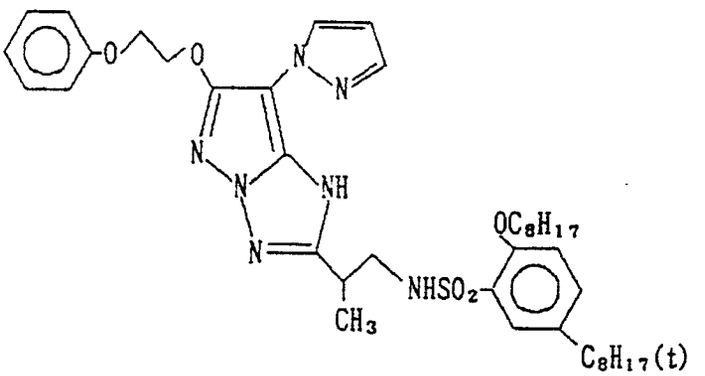
(A－4)－44



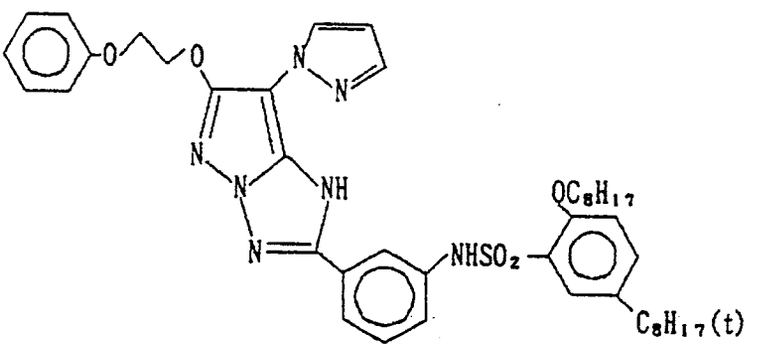
(A－4)－45



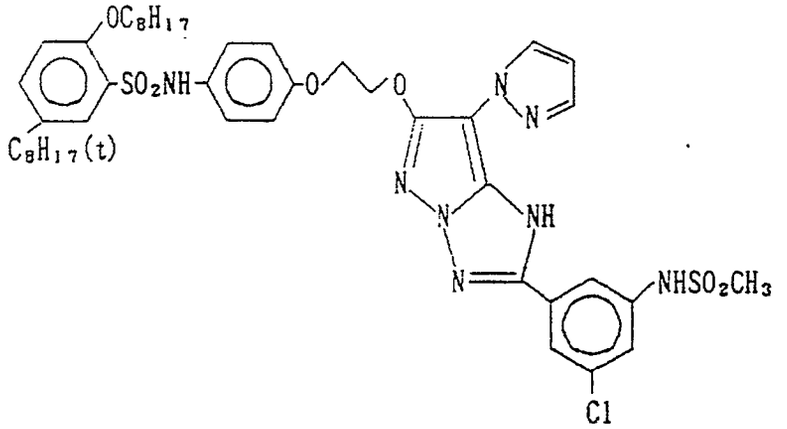
(A－4)－46



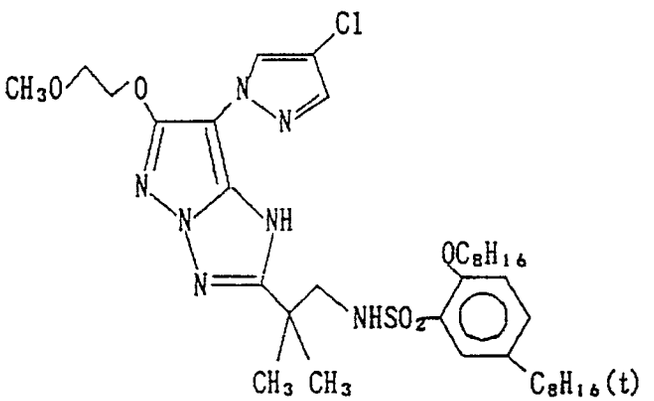
(A－4)－47



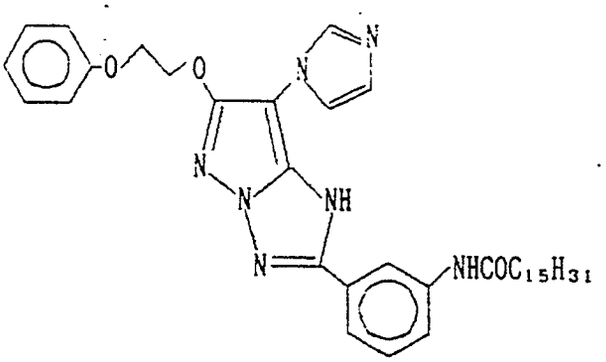
(A－4)－48



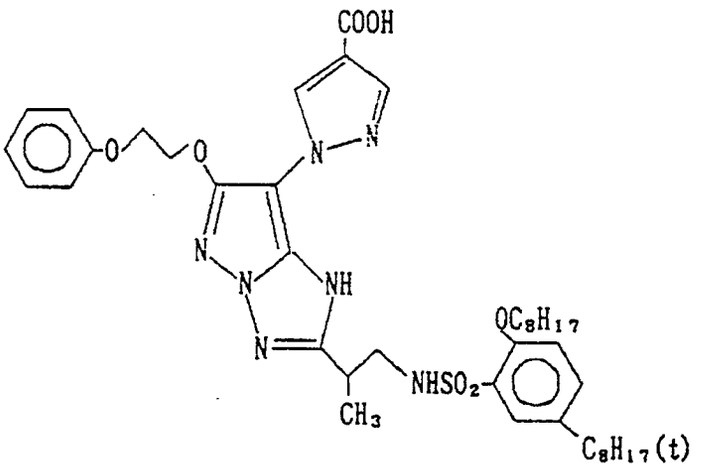
(A－4)－49



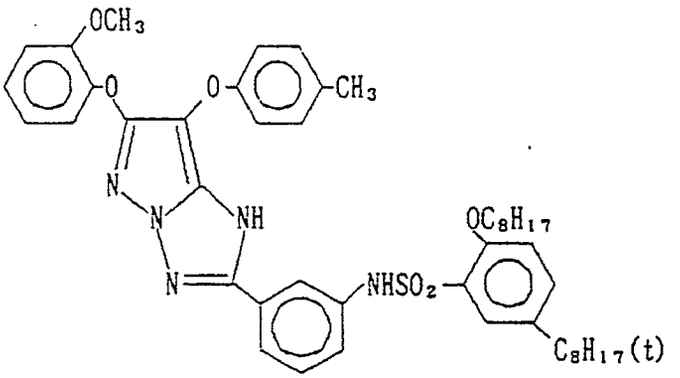
(A－4)－50


O
O
N
N
N
N
NH
N
NHCOC15H31

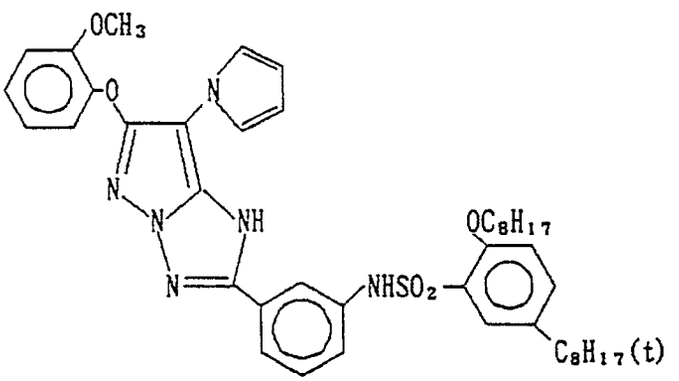
(A－4)－51


COOH
O
O
N
N
N
N
NH
N
OC8H17
NHSO2
CH3
C8H17(t)

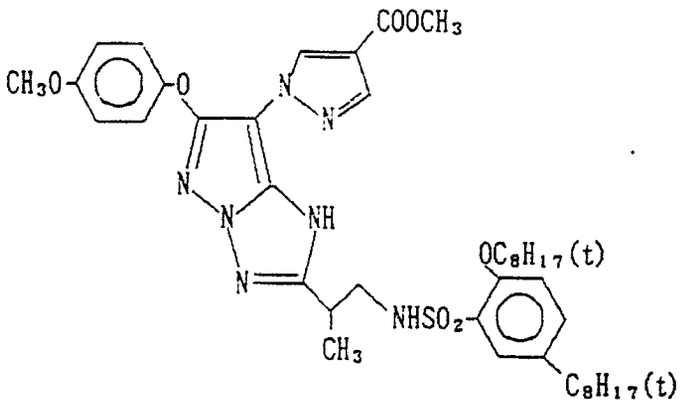
(A－4)－52



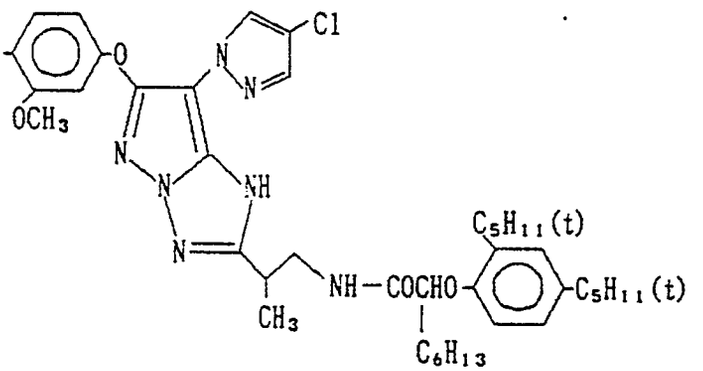
(A－4)－53



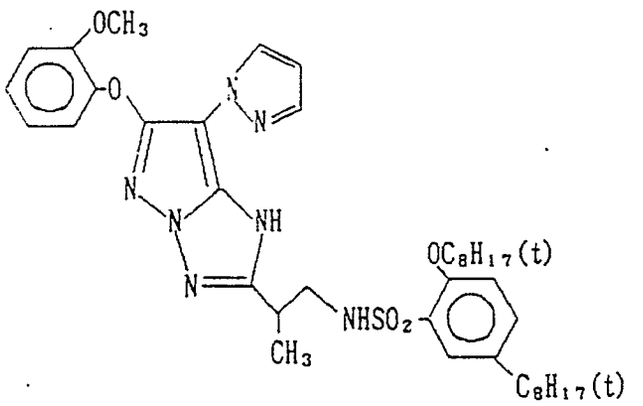
〔A－4〕－54



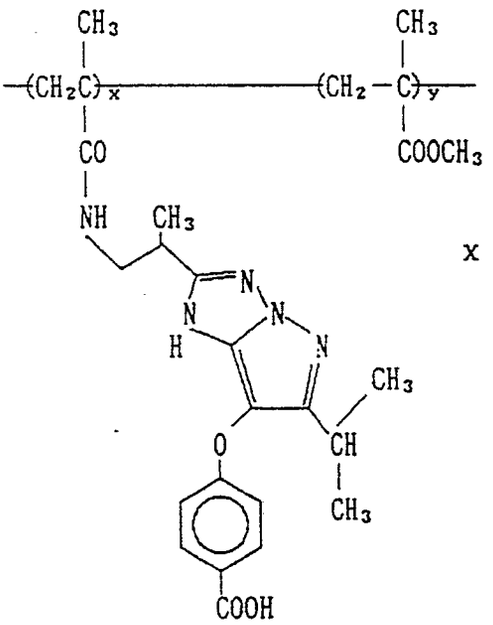
〔A－4〕－55



(A－4)－56



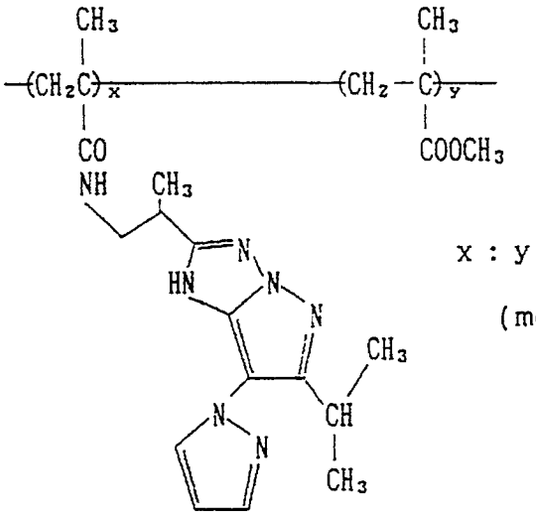
(A－4)－57



x : y = 40 : 60

(mole%)

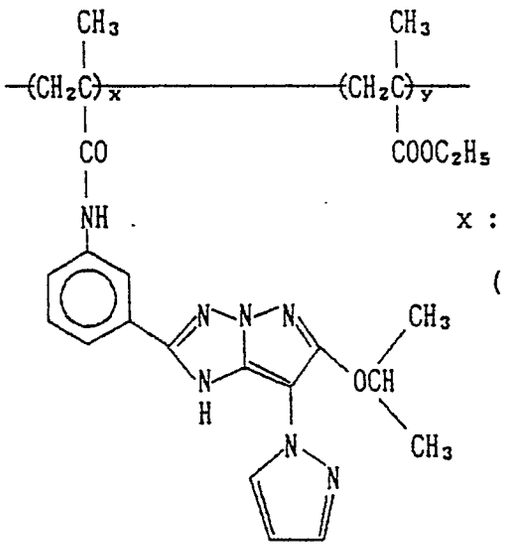
〔A－4〕－58



x : y＝30 : 70

(mole%)

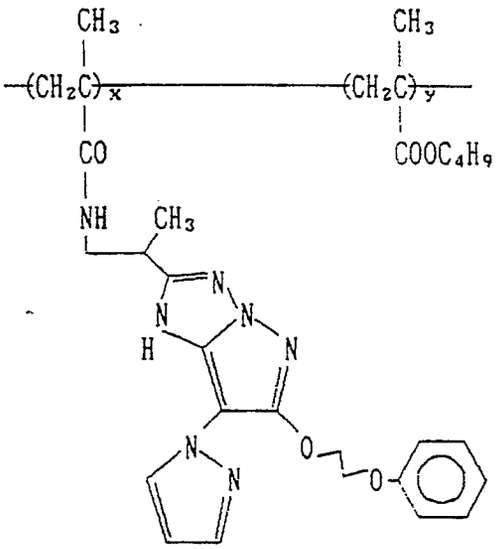
〔A－4〕－59



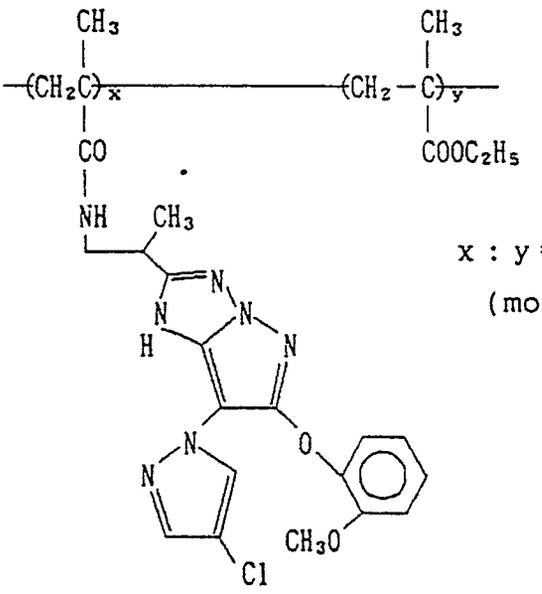
x : y＝50 : 50

(mole%)

〔A－4〕－60

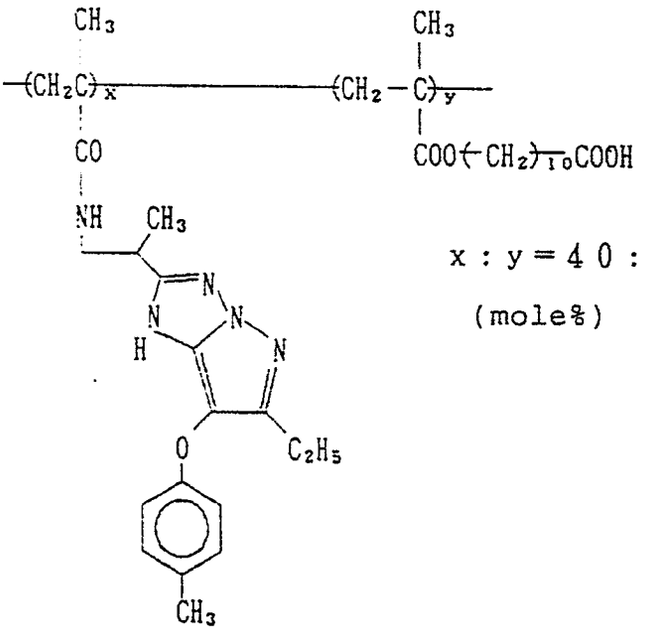


〔A－4〕－61



x : y = 45 : 55
(mole%)

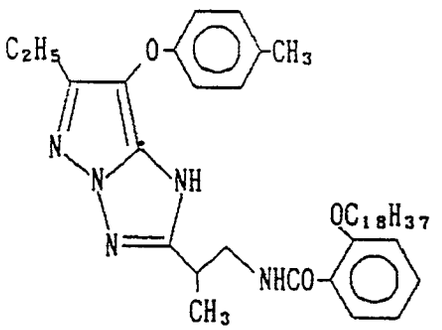
(A－4)－62



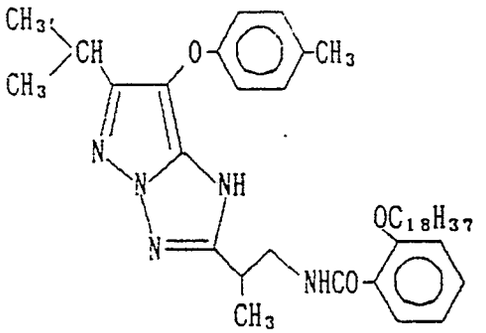
x : y = 40 : 60

(mole%)

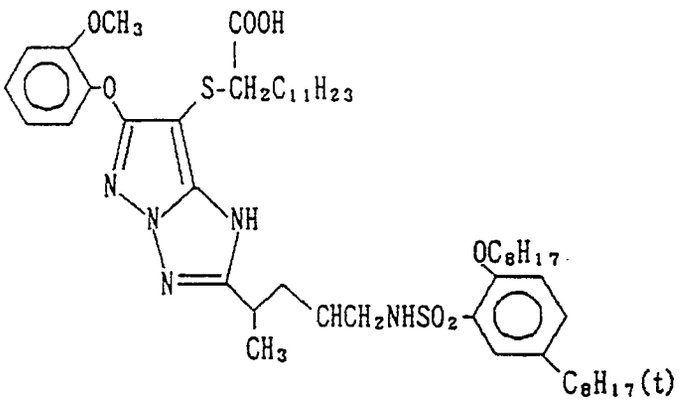
(A－4)－63



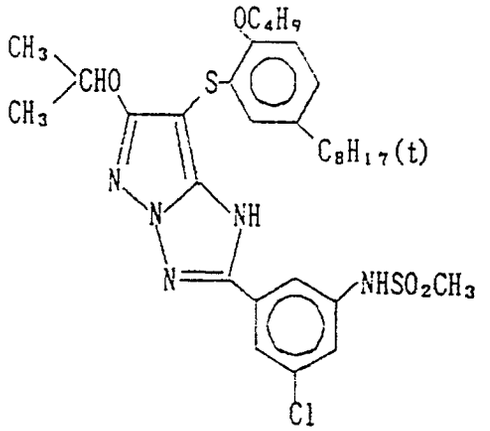
(A-4)-64



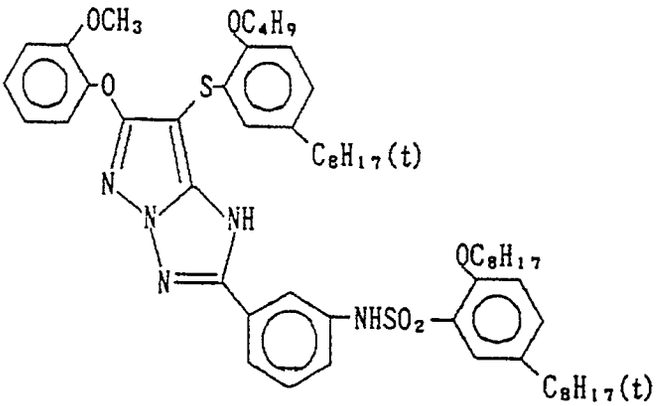
(A-4)-65



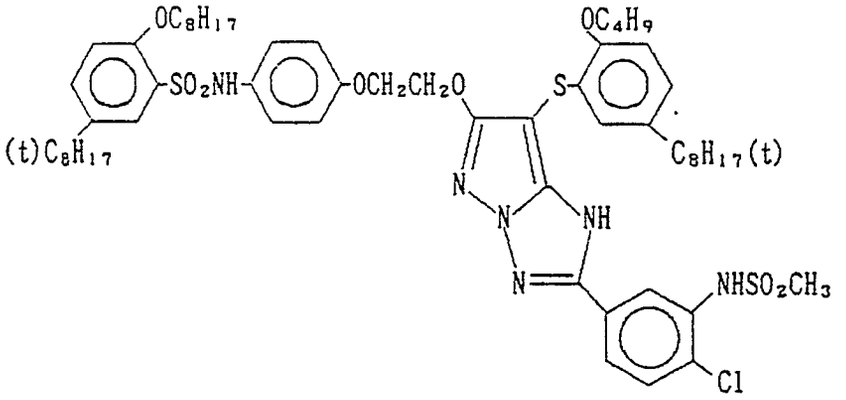
(A-4)-66



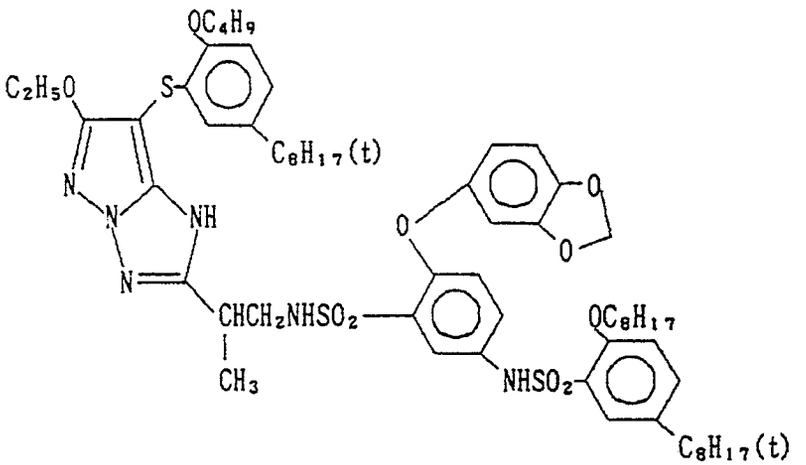
(A-4)-67



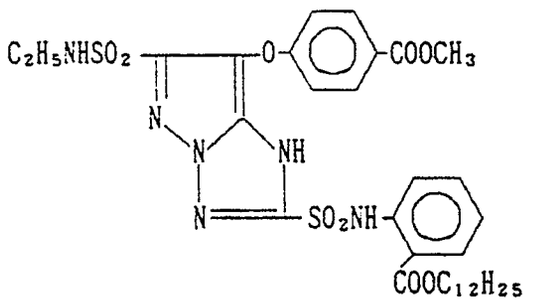
(A－4)－68



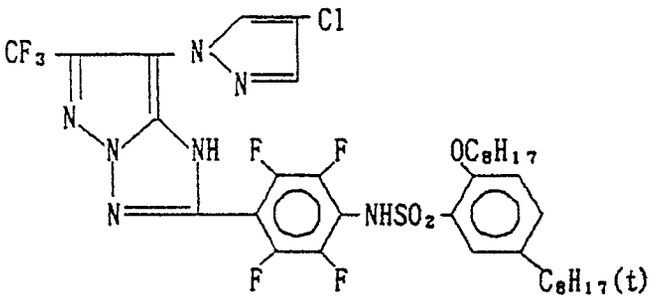
(A－4)－69



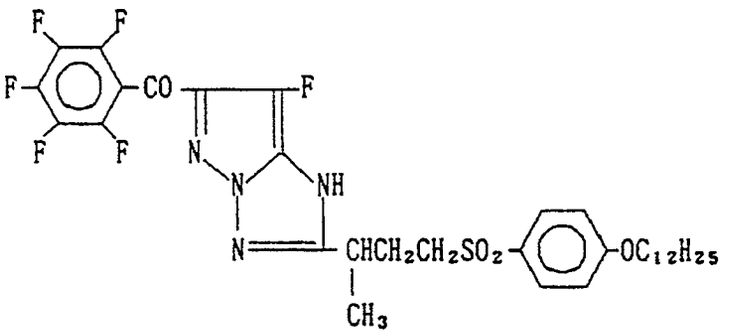
(A-4)-70



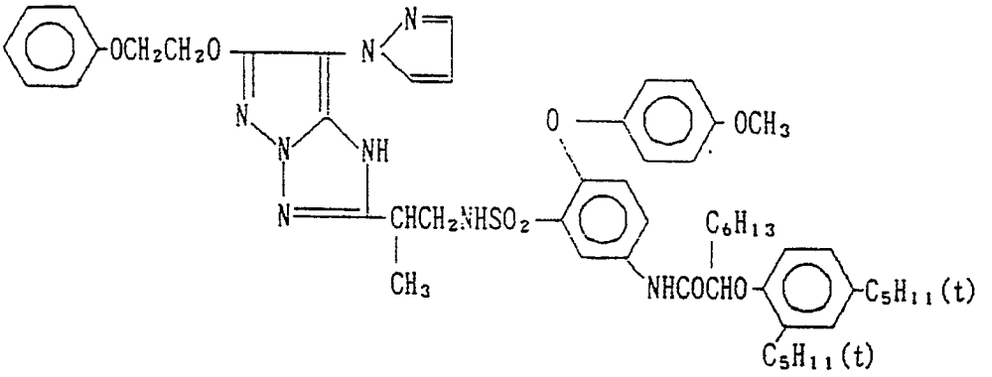
(A-4)-71



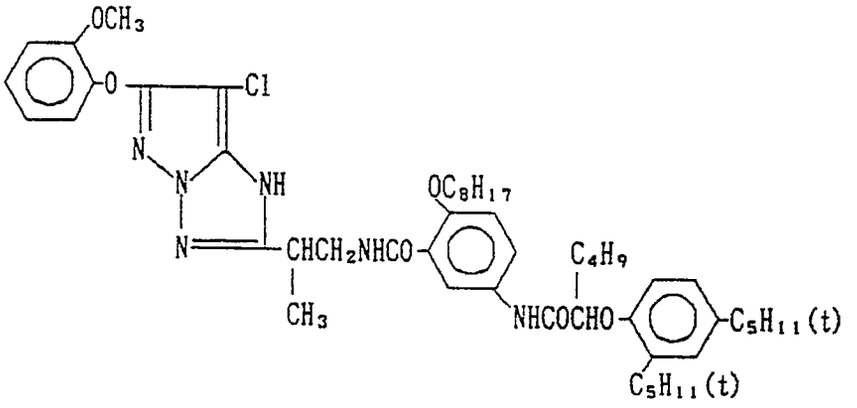
(A-4)-72



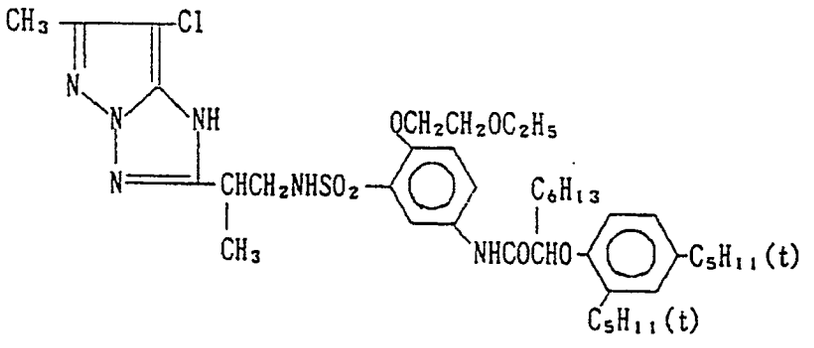
(A－4)－73



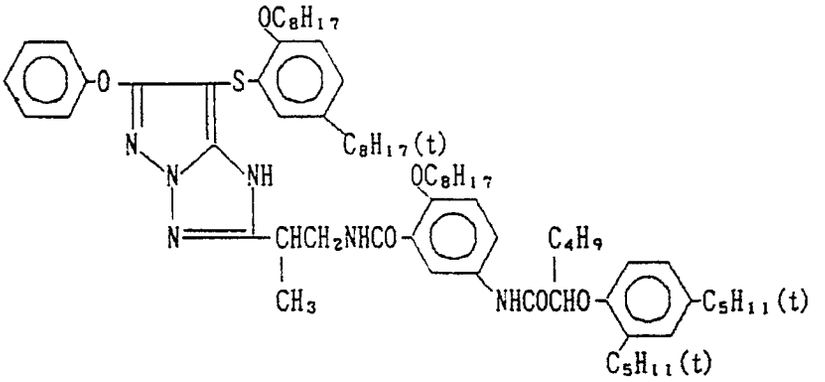
(A－4)－74



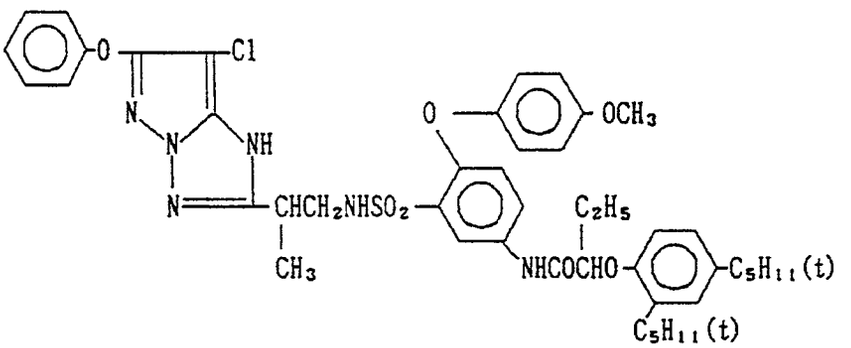
(A－4)－75



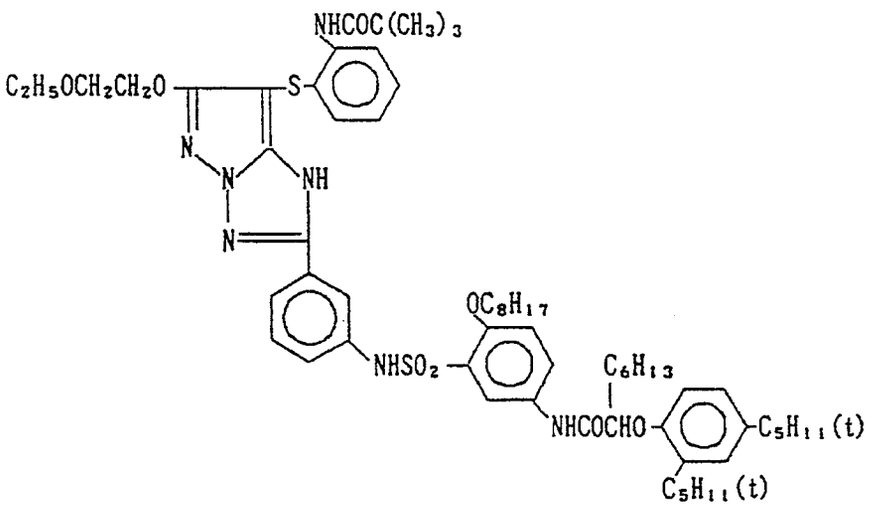
(A－4)－76



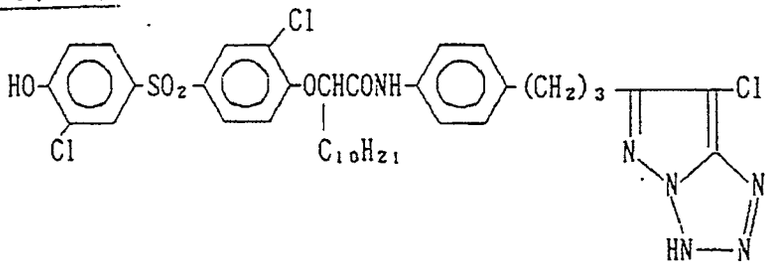
(A－4)－77



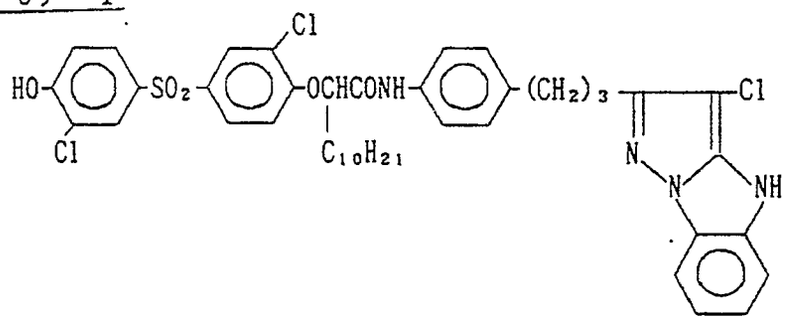
(A－4)－78



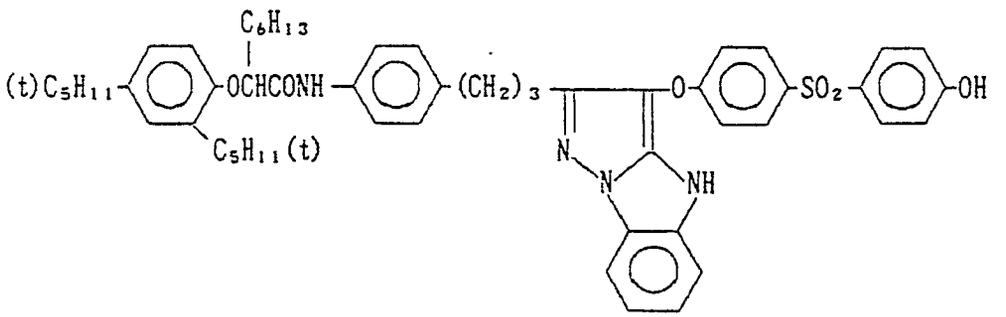
(A－5)－1



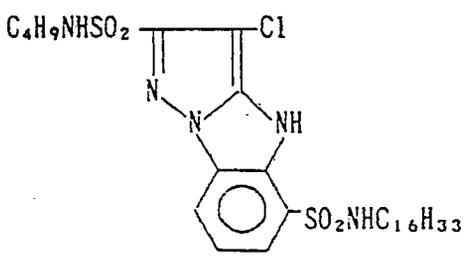
(A－6)－1



(A－6)－2



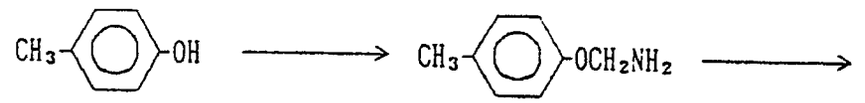
(A－6)－3



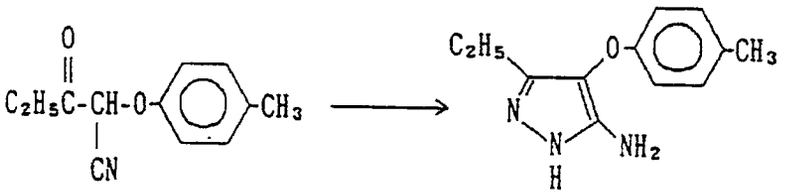
Synthesis examples are described hereinafter.

SYNTHESIS EXAMPLE 1

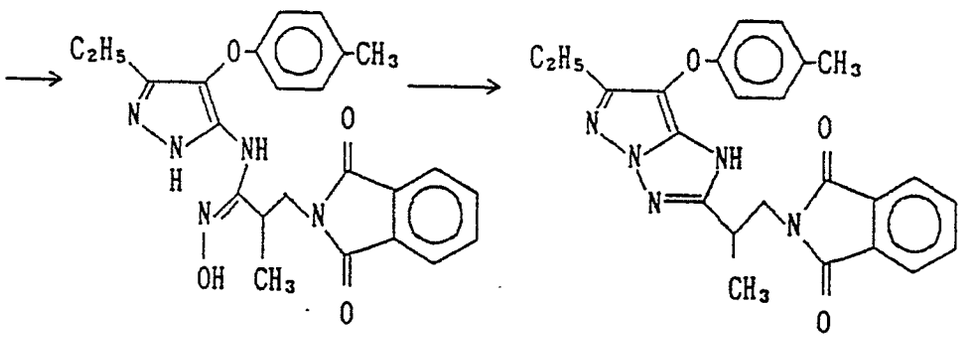
Synthesis of [A-4]-24



(1)



(2) (3)



(4) (5)

⟶ 〔A－4〕－24

103 g (0.75 mol) of potassium carbonate anhydride and 150 ml of acetonitrile were charged into a 500-ml three-necked flask equipped with a reflux condenser and a dropping funnel. The mixture was then stirred at room temperature. 54.0 g (0.5 mol) of p-cresol was added to the system. The system was then heated under reflux with stirring. 41.5 g (0.55 mol) of chloroacetonitrile was added dropwise to the system in about 5 minutes in such a manner that the reflux did not become vigorous. After the completion of dropwise addition, the system was further stirred under reflux for 2 hours. The system was then cooled with water to an internal temperature of about 30°C.

The system was then filtered with suction to remove inorganic matters therefrom. The filtrate was extracted with 150 ml of ethyl acetate, 50 ml of saturated brine and 100 ml of water. The resulting ethyl acetate phase was washed with a mixture of 50 ml of saturated brine and 100 ml of water three times, and then dried with sodium sulfate anhydride. The ethyl acetate was distilled off by a rotary evaporator. The resulting residue was distilled under reduced pressure to obtain 64.9 g (0.44 mol) of the desired Compound (1) (yield: 88%; b.p. 85-88°C/0.2 mmHg; m.p. 31-33°C)

102 g (1.0 mol) of methyl propionate was charged into a 300-ml three-necked flask equipped with a dropping funnel, a thermometer and a drying tube ($CaCl_2$). The system was stirred under cooling over an

ice bath. 56.1 g (0.50 mol) of potassium-t-butoxy was added to the system. 73.6 g (0.50 mol) of Compound (1) was then added dropwise to the system with stirring in a manner such that the internal temperature thereof did not exceed 10 °C. After completion of the dropwise addition, the system was further stirred for 3 hours while the internal temperature thereof was kept between 5 °C and 10 °C. The system was extracted with a mixture of 150 ml of water and 150 ml of hexane. The resulting aqueous phase was neutralized with 42.9 ml of concentrated hydrochloric acid and then extracted with 200 ml of ethyl acetate. The resulting ethyl acetate phase was washed with a mixture of 50 ml of saturated brine and 100 ml of water twice, and then dried with sodium sulfate anhydride. The ethyl acetate was distilled off by a rotary evaporator to obtain 46.1 g of Compound (2) in the form of crude product.

46.1 g of Compound (2) and 100 ml of isopropyl alcohol were charged into a 300-ml three-necked flask equipped with a reflux condenser and a dropping funnel. The system was heated under reflux with stirring. 19.7 g of a 80% aqueous solution of hydrazine hydrate was added dropwise to the system in a manner such that the reflux did not become vigorous. The system was heated under reflux with stirring for 3 hours. The system was then cooled with water to an internal temperature of about 30 °C. After the crystallization of Compound (3), the system was stirred at room temperature for 3 hours. 100 ml of water was added to the system. The system was further stirred for 3 hours. The system was filtered with suction to obtain 33.7 g (0.16 mol) of Compound (3) (yield: 31% from Compound (1); m.p. 174.0 - 176.5 °C)

300 ml of dimethylacetamide was added to 33.7 g (0.16 mol) of Compound (3). The mixture was then heated until Compound (3) was dissolved. The system was then cooled to a temperature of about 35 °C. 60 g (0.21 mol) of 2-methyl-3-phthalimidopropioimido methylhydrochloride was added to the system. The system was stirred at room temperature for about 24 hours. An aqueous solution of 22 g (0.21 mol) of hydroxyamine hydrochloride and an aqueous solution of 13 g (0.61 mol) of sodium acetate were added to the reaction solution. The reaction solution was then heated to 50 °C for about 1 hour. The system was gradually cooled to room temperature with stirring for about 10 hours. The system was then poured into 1.5 ℓ of ice with stirring. The resulting crystal was filtered off, thoroughly washed with water, and then dried to obtain 69 g (yield: 96%) of Compound (4).

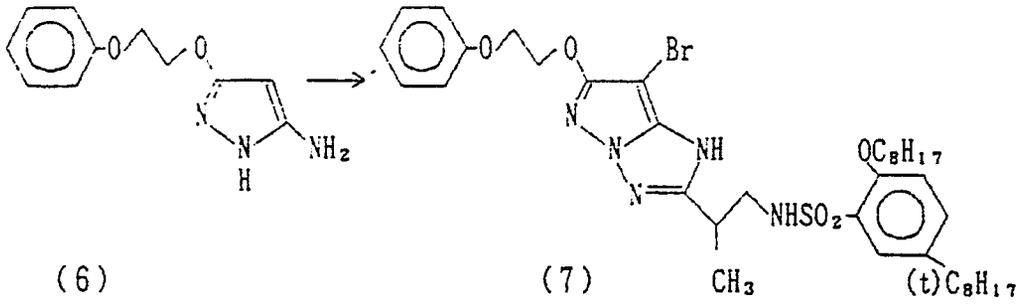
200 mol of dimethyl acetamide was added to 69 g (0.15 mol) of Compound (4). The mixture was stirred. Compound (4) was not completely dissolved. A solution of 28.6 g of paratoluenesulfonyl chloride in 80 ml of acetonitrile was added dropwise to the system over an ice bath over a period of about 30 minutes. After completion of the dropwise addition, the system was stirred for about 1 hour. 12.1 ml of pyridine was added to the system. The system was further stirred at room temperature for about 1 hour. The reaction solution was then poured into about 1.5 ℓ of iced water. The aqueous solution was removed by decantation. 1.5 ℓ of methanol was added to the resulting solid matters. The material was then crushed with stirring. The material was then filtered off to obtain Compound (4) in the form of colorless powder of tosylate.

The product was immediately dispersed in 1.3 ℓ of methanol. 12.1 ml of pyridine was then added to the system. The system was heated to the refluxing temperature. When the tosylate was dissolved, the heating was suspended. The system was gradually cooled to room temperature with stirring. The system was stirred at room temperature for about 2 days. The methanol was then distilled off under reduced pressure so that the system was concentrated to about 300 ml. The reaction solution was then poured into about 1.5 ℓ of water. The resulting powdered crystal was filtered off, and then dried to obtain 29.8 g (yield: 52%) of Compound (5).

2.5 g (0.062 mol) of 80% hydrazine hydrate was added to a solution of 15 g (0.039 mol) of Compound (5) in 150 ml of isopropyl alcohol. The system was heated under reflux for about 8 hours. The system was cooled to room temperature. Chloroform and saturated brine were added to the system. The resulting phthal hydrazide was filtered off with suction. The filtrate was extracted with chloroform three times. The extract was withdrawn, washed with saturated brine, dried with magnesium sulfate, filtered, and then evaporated to obtain an amine in the form of crystal. The amine thus obtained was dissolved in 100 ml of a 1 : 1 mixture of dimethyl acetamide and acetonitrile. 4.7 ml of triethyl amine was added to the solution. A solution of 16.9 g (0.039 mol) of 2-hexyloxyethoxy-4-octylbenzenesulfonyl chloride in 40 ml of acetonitrile was added dropwise to the system with stirring under cooling with ice. After completion of the dropwise addition, the system was stirred for about 1 hour, extracted, dried, and then evaporated to obtain an oily matter which was then purified through a silica gel column (elute: 4 : 1 mixture of n-hexane and ethyl acetate) to obtain 19.5 g of Exemplary Coupler [A-4]-24 (yield: 72%; m.p. 112 - 114 °C).

SYNTHESIS EXAMPLE 2

Synthesis of [A-4]-46



(6) (7)

⟶ [A-4]-46

Compound (6) was prepared by the synthesis method as described in JP-A-64-13071 or 64-13072. The conversion of Compound (6) to Compound (7) was accomplished by the synthesis example described in JP-A-62-209457.

20 g (0.5 mol) of 60% hydrogenated sodium was added to 300 ml of N,N-dimethylindazolinone. The mixture was stirred under cooling with ice. 34 g (0.5 mol) of pyrazole was added to the system in two or three batches. The system was stirred until the generation of hydrogen stopped. 76 g (0.1 mol) of Compound (7) was added to the system. The system was heated to a temperature of 120 to 125 °C for 6 hours. The system was then subjected to ordinary post-treatment. The resulting crude product was purified through a silica gel column chromatography to obtain 49 g of [A-4]-46 (yield: 65%).

The compound represented by general formula (A) may be incorporated in the red-sensitive emulsion layer and/or green-sensitive emulsion layer and/or its adjacent layers. The total amount of the compound of formula (A) to be incorporated is from 0.01 to 2.00 g/m^2, preferably 0.05 to 1.5 g/m^2, and more preferably 0.1 to 1.0 g/m^2.

The incorporation of the compounds of general formula (A) in the light-sensitive material can be effected in accordance with the method for incorporation of couplers as described later. The weight proportion of the high boiling organic solvent used as a dispersing solvent for the compounds of general formula (A) is from 0 to 4.0, preferably 0 to 2.0, more preferably 0.1 to 1.5, and particularly 0.1 to 1.0.

The yellow-colored cyan coupler of the present invention and the coupler represented by the general formula (A) may be incorporated in the same silver halide light-sensitive layer or its adjacent layers or separately incorporated in different silver halide light-sensitive layers or adjacent light-insensitive layers. Preferably, the yellow-colored cyan coupler is incorporated in the red-sensitive emulsion layer and/or its adjacent light-insensitive layers, and the coupler represented by the general formula (A) is incorporated in the green-sensitive emulsion layer and/or red-sensitive emulsion layer and/or their adjacent light-insensitive layers. Most preferably, the yellow-colored cyan coupler is incorporated in the red-sensitive emulsion layer, and the coupler represented by the general formula (A) is incorporated in the green-sensitive emulsion layer.

The silver halide color photographic material of the present invention preferably comprises at least one layer containing at least one compound represented by the general formula (I):

$$A - \{(L1)_a - (B)_m\}_p - (L2)_n - DI \qquad (I)$$

wherein A represents a group which is capable of undergoing a reaction with an oxidation product of an aromatic primary amine developing agent to cause cleavage of A from $\{(L1)_a - (B)_m\}_p - (L2)_n - DI$; L1 represents a group which causes cleavage of the bond between L1 and the group to its right as viewed in general formula (I) after cleavage of the bond between L1 and A; B represents a group which undergoes a reaction with an oxidation product of a developing agent to cause cleavage of the bond between B and the group to its right as viewed in general formula (I); L2 represents a group which causes cleavage of the bond between L2 and DI after cleavage of the bond of L2 to the group to its left as viewed in general formula (I);

DI represents a development inhibitor; a, m and n each represents an integer 0 or 1; and p represents an integer from 0 to 2, with the proviso that when p is 2, the two $\{(L1)_a - (B)_m\}$ groups may be the same or different.

The reaction processes by which the compound represented by the general formula (I) releases DI upon development are shown by the following equations:

$$A - (L1)_a - (B)_m - (L2)_n - DI \xrightarrow{QDI^+}$$

$$(L1)_a - (B)_m - (L2)_n - DI \longrightarrow$$

$$(B)_m - (L2)_n - DI \xrightarrow{QDI^+} (L2)_n - DI \longrightarrow DI$$

wherein p is 1 and A, L1, a, B, m, L2, n and DI are as defined in the general formula (I); and QDI^+ represents an oxidation product of a developing agent.

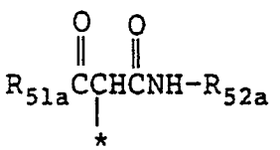
The compounds represented by the general formula (I) will be further described hereinafter.

In the general formula (I), A represents a coupler group or a redox group.

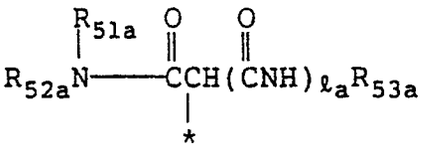
Examples of the coupler group represented by A include a yellow coupler group (e.g., open-chain ketomethylene coupler group such as acylacetanilide and malondianilide), a magenta coupler group (e.g., 5-pyrazolone, pyrazolotriazole or imidazopyrazole coupler residue), a cyan coupler group (e.g., phenol coupler group, naphthol coupler group, imidazole coupler group as described in European Patent Disclosure 249,453, pyrazolopyrimidine coupler group as described in European Patent Disclosure 304,001), and a colorless coupler group (e.g., indanone coupler group, acetophenone coupler group). Other examples of the coupler group represented by A include the heterocyclic coupler groups disclosed in U.S. Patents 4,315,070, 4,183,752, 4,174,969, 3,961,959, and 4,171,223, and JP-A-52-82423.

The redox group represented by A is a group which undergoes cross oxidation with an oxidation product of a developing agent. Examples of such a redox group include hydroquinones, catechols, pyrogallols, 1,4-naphthohydroquinones, 1,2-naphthohydroquinones, sulfonamidophenols, hydrazides, and sulfonamidonaphthols. Specific examples of these groups are described in JP-A-61-230135, JP-A-62-251746, and JP-A-61-278852, U.S. Patents 3,364,022, 3,379,529, 3,639,417, and 4,684,604, and Journal of Organic Chemistry, 29, 588 (1964).

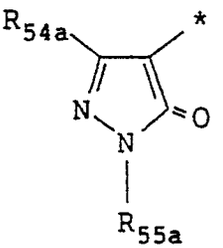
Preferred examples of A are the coupler group represented by the general formulae (Cp-1a), (Cp-2a), (Cp-3a), (Cp-4a), (Cp-5a), (Cp-6a), (Cp-7a), (Cp-8a), (Cp-9a), and (Cp-10a). These couplers exhibit an advantageously high coupling speed.



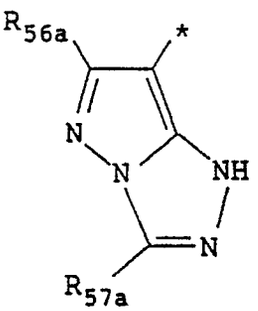
(Cp-1a)



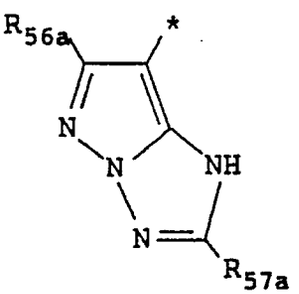
(Cp-2a)



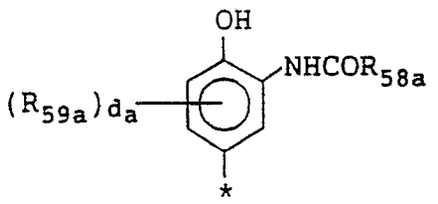
(Cp-3a)



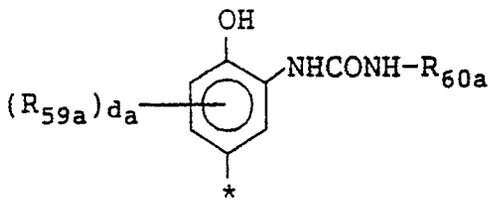
(Cp-4a)



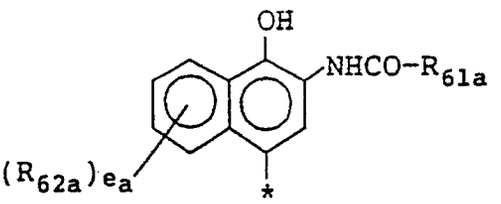
(Cp-5a)



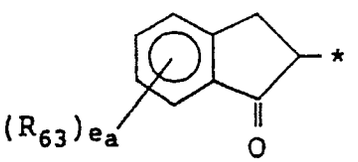
(Cp-6a)



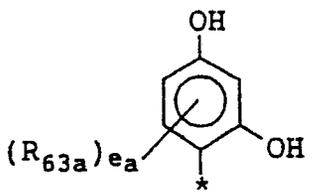
(Cp-7a)



(Cp-8a)



(Cp-9a)



(Cp-10a)

In these general formulae, the mark * is indicates the position at which A is connected to - $\{(L1)_a$ - (B)-$_m\}_p$ - $(L2)_n$ - DI.

In these general formulae, if R_{51a}, R_{52a}, R_{53a}, R_{54a}, R_{55a}, R_{56a}, R_{57a}, R_{58a}, R_{59a}, R_{60a}, R_{61a}, R_{62a} or R_{63a} contains a nondiffusing group, these R substituents are selected such that the total number of carbon atoms contained in these R substituents is from 8 to 40, preferably 10 to 30. If these R substituents do not contain mon-diffusing groups, the total number of carbon atoms contained in these R substituents is preferably 15 or less.

R_{51a} to R_{63a}, ℓ_a, d_a and e_a will be further described hereinafter. In the following definitions, R_{41a} represents an aliphatic group, an aromatic group or a heterocyclic group, R_{42a} represents an aromatic group

or a heterocyclic group, and R_{43a}, R_{44a} and R_{45a} each represents a hydrogen atom, an aliphatic group, an aromatic group or a heterocyclic group.

R_{51a} has the same meaning as R_{43a}. R_{52a} and R_{53a} each has the same meaning as R_{42a}. The suffix ℓ_a represents an integer 0 or 1. R_{54a} has the same meaning as R_{41a} or represents a $R_{41a}CON(R_{43a})$- group, a $(R_{41a})(R_{43a})N$- group, a $R_{41a}SO_2N(R_{43a})$- group, a $R_{41a}S$- group, a $R_{43a}O$- group, a $(R_{43a})(R_{45a})NCON(R_{44a})$- group or a N≡C- group. R_{55a} has the same meaning as R_{41a}. R_{56a} and R_{57a} have the same meaning as R_{43a} or represent a $R_{41a}S$- group, a $R_{43a}O$- group, a $R_{41a}CON(R_{43a})$- group or a $R_{41a}SO_2N(R_{43a})$- group. R_{58a} has the same meaning as R_{41a}. R_{59a} has the same meaning as R_{41a} or represents an $R_{41a}CON(R_{43a})$- group, an $R_{41a}OCON(R_{43a})$- group, an $R_{41a}SO_2N(R_{43a})$- group, an $(R_{43a})(R_{44a})NCON(R_{45a})$- group, an $R_{41a}O$- group, an $R_{41a}S$- group, a halogen atom, or an $(R_{41a})(R_{43a})N$- group. The suffix d_a represents an integer from 0 to 3. When d_a is plural, the two or three R_{59a} groups may be the same or different substituents or they may be divalent groups which are connected to each other to form a cyclic structure. Examples of such a cyclic structure include a pyridine ring and a pyrrole ring. R_{60a} has the same meaning as R_{41a}. R_{61a} has the same meaning as R_{41a}. R_{62a} has the same meaning as R_{41a} or represents a $R_{41a}OCONH$- group, a $R_{41a}SO_2NH$- group, a $(R_{43a})(R_{44a})NCON(R_{45a})$- group, a $(R_{43a})(R_{44a})NSO_2N(R_{45a})$- group, a $R_{43a}O$- group, a $R_{41a}S$-group, a halogen atom or a $(R_{41a})(R_{43a})N$- group. R_{63a} has the same meaning as R_{41a} or represents a $R_{43a}CON(R_{45a})$- group, a $(R_{43a})(R_{44a})NCO$- group, a $R_{41a}SO_2N(R_{44a})$- group, a $(R_{43a})(R_{44a})NSO_2$- group, a $R_{41a}SO_2$- group, a $R_{43a}OCO$-group, a $R_{43a}OSO_2$- group, a halogen atom, a nitro group, a cyano group or a $R_{43a}CO$- group. The suffix e_a represents an integer from 0 to 4. When there is a plurality of R_{62a} groups or R_{63a} groups, they may be the same or different.

In the foregoing definitions of the R groups, the aliphatic group is a C_{1-32}, preferably a C_{1-22} saturated or unsaturated, acyclic or cyclic, straight-chain or branched, substituted or unsubstituted aliphatic hydrocarbon group. Typical examples of such an aliphatic group include methyl, ethyl, propyl, isopropyl, butyl, (t)-butyl, (i)butyl, (t)amyl, hexyl, cyclohexyl, 2-ethylhexyl, octyl, 1,1,3,3-tetramethylbutyl, decyl, dodecyl, hexadecyl, and octadecyl groups.

The aromatic group is a C_{6-20} aromatic group, preferably a substituted or unsubstituted phenyl group or a substituted or unsubstituted naphthyl group.

The heterocyclic group is a C_{1-20}, preferably C_{1-7}, preferably 3- to 8-membered substituted or unsubstituted heterocyclic group containing one or more nitrogen, oxygen or sulfur atoms. Typical examples of such a heterocyclic group include 2-pyridyl, 2-furyl, 2-imidazolyl, 1-indolyl, 2,4-dioxo-1,3-imidazolidin-5-yl, 2-benzoxazolyl, 1,2,4-triazol-2-yl, and 1-pyrazolyl groups.

If the above mentioned aliphatic hydrocarbon group, aromatic group and heterocyclic group contain substituents, typical examples of such substituents include a halogen atom, a $R_{47a}O$- group, a $R_{46a}S$- group, a $R_{47a}CON(R_{48a})$- group, a $(R_{47a})(R_{48a})NCO$- group, a $R_{46a}OCON(R_{47a})$- group, a $R_{46a}SO_2N(R_{47a})$- group, a $(R_{47a})(R_{48a})NSO_2$- group, a $R_{46a}SO_2$- group, a $R_{47a}OCO$-group, a $(R_{47a})(R_{48a})NCON(R_{49a})$- group, groups having the same meaning as R_{46a}, a $R_{46a}COO$- group, a $R_{47a}OSO_2$- group, a cyano group, and a nitro group wherein R_{46a} representsan aliphatic, aromatic or heterocyclic group, and R_{47a}, R_{48a} and R_{49a} each represents an aliphatic group, an aromatic group, a heterocyclic group or hydrogen atom.

These aliphatic group, aromatic group and heterocyclic group are as defined above.

Preferred embodiments of R_{51a} to R_{63a}, ℓ_a, d_a and e_a will be described hereinafter.

In the general formula (Cp-1a), R_{51a} is preferably an aliphatic or aromatic group. In the general formula (Cp-2a), R_{51a} is preferably a hydrogen atom or an aliphatic group.

R_{52a} and R_{55a} each is preferably an aromatic group.

R_{53a} is preferably an aromatic or heterocyclic group.

In the general formula (Cp-3a), R_{54a} is preferably a $R_{41a}CONH$- group or a $(R_{41a})(R_{43a})N$- group. R_{56a} and R_{57a} each is preferably an aliphatic, aromatic, $R_{41a}O$-, or $R_{41a}S$- group. R_{58a} is preferably an aliphatic or aromatic group. In the general formula (Cp-6a), R_{59a} is preferably a chlorine atom, an aliphatic or $R_{41a}CONH$-group. The suffix d_a is preferably 1 or 2. R_{60a} is preferably an aromatic group.

In the general formula (Cp-7a), R_{59a} is preferably a $R_{41a}CONH$- group, and d_a is preferably 1. R_{61a} is preferably an aliphatic or aromatic group.

In the general formula (Cp-8a), e_a is preferably 0 or 1. R_{62a} is preferably a $R_{41a}OCONH$-, $R_{41a}CONH$- or $R_{41a}SO_2NH$- group. The preferred position at which these groups are connected to the coupler is the 5-position in the naphthol ring. In the general formula (Cp-9a), R_{63a} is preferably a $R_{41a}CONH$- group, a $R_{41a}SO_2NH$- group, a $(R_{41a})(R_{43a})NSO_2$- group, a $R_{41a}SO_2$- group, $(R_{41a})(R_{43a})NCO$- group, a nitro group or a cyano group.

In the general formula (Cp-10a), R_{63a} is preferably a $(R_{43a})(R_{43a})NCO$-, $R_{43a}OCO$- or $R_{43a}CO$- group.

In general formula (I), examples of the connecting groups represented by L1 and L2 include groups subjecting a cleavage reaction of a hemiacetal described in U.S. Patents 4,146,396, 4,652,516, and

4,698,297, timing groups subjecting an intramolecular nucleophilic substitution reaction to cause a cleavage reaction as described in U.S. Patent 4,248,962, timing groups subjecting an electron migration reaction to cause a cleavage reaction as described in U.S. Patent 4,409,323 and 4,421,845, groups subjecting the hydrolyzation reaction of an iminoketal to cause a cleavage reaction as described in U.S. Patent 4,546,073, and groups subjecting a hydrolyzation reaction of an ester to cause a cleavage reaction as described in West German Patent Disclosure 2,626,317. L1 and L2 each is connected to A or A-$(L1)_a$-$(B)_m$ via its hetero atom, preferably oxygen, sulfur or nitrogen atom.

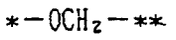
Preferred examples of the groups represented by L1 and L2 include the following:

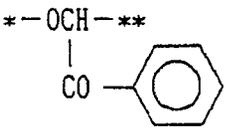
(1) Groups subjecting a cleavage reaction of hemiacetal

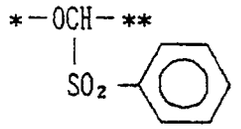
Examples of such groups are described in U.S. Patent 4,146,396, and JP-A-60-249148 and 60-249149. Such groups are represented by the general formula (T-1a) below. In the general formula (T-1a), the mark * indicates the left bonding position of L1 or L2 in the compound represented by the general formula (I), and the mark ** indicates the right bonding position of L1 or L2 in the compound represented by the general formula (I).

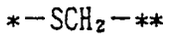
$$*\text{-}(W\text{-}C(R_{65})(R_{66})\text{-}_t** \qquad \text{(T-1a)}$$

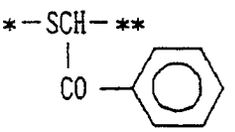
wherein W represents an oxygen atom, a sulfur atom or a -$N(R_{67})$- group; R_{65} and R_{66} each represents a hydrogen atom or substituent; R_{67} represents a substituent; and t represents an integer 1 or 2. When t is 2, the two -$(W\text{-}C(R_{65})(R_{66})$- groups may be the same or different. Typical examples of the substituents represented by R_{65} and R_{66} and typical examples of R_{67} include R_{69}, $R_{69}CO$-, $R_{69}SO_2$-, $(R_{69})(R_{70})NCO$-, and $(R_{69})(R_{70})NSO_2$-. R_{69} represents an aliphatic, aromatic or heterocylic group, and R_{70} represents an aliphatic group, an aromatic group, a heterocyclic group or a hydrogen atom. R_{65}, R_{66} and R_{67} may represent divalent groups which are connected to each other to form a cyclic structure. Specific examples of the group represented by the general formula (T-1a) include the following:

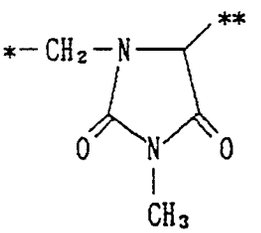


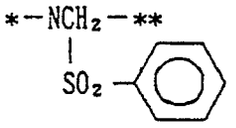


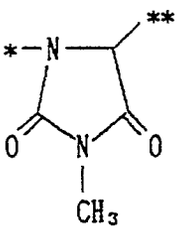


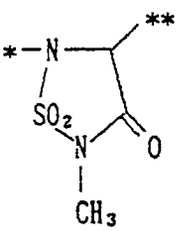










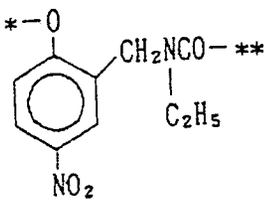


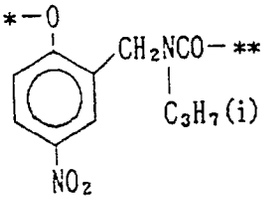
(2) Groups utilizing an intramolecular nucleophilic substitution reaction to cause cleavage reaction

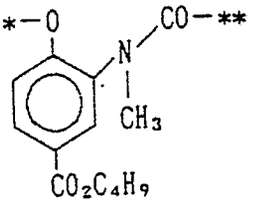
Examples of such groups include the timing groups as described in U.S. Patent 4,248,962. These timing groups are represented by the general formula (T-2a):

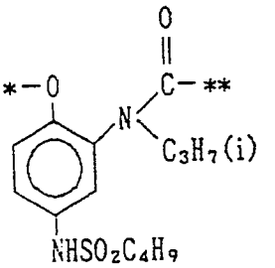
*-Nu-Link-E-** (T-2a)

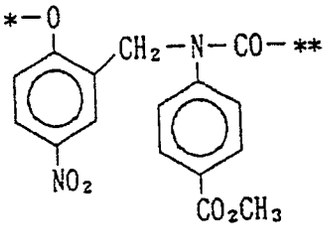
wherein the marks * and ** are as defined in the general formula (T-1a); Nu represents a nucleophilic group with nucleophilic seeds such as an oxygen atom or a sulfur atom; E represents an electrophilic group which can undergo a nucleophilic attack by Nu to cause cleavage of the bond at the mark **; and Link represents a connecting group which sterically connects Nu and E so that they can undergo an intramolecular nucleophilic substitution reaction. Specific examples of the group represented by the general formula (T-2a) include the following:

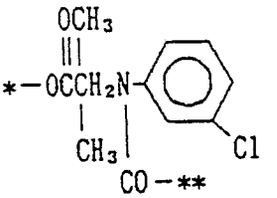










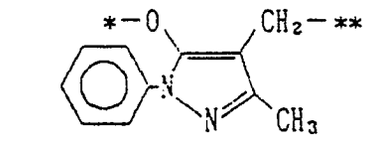


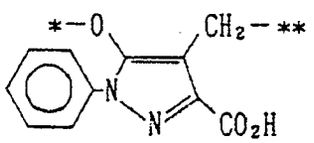
(3) Groups utilizing an electron migration reaction along a conjugated system to cause a cleavage reaction

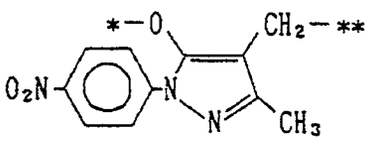
Examples of such groups are described in U.S. Patents 4,409,323, and 4,421,845 and are represented by the general formula (T-3a):

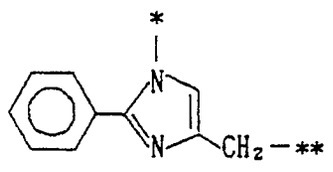
*-W-(V_1 = V_2)t-CH_2-** (T-3a)

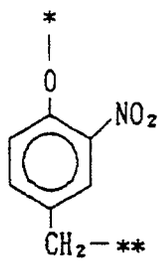
wherein V_1 and V_2 each represents -C(R_{65}) =, -C(R_{66}) = or a nitrogen atom; and the marks * and **, W, R_{65}, R_{66} and t are as defined in the general formula (T-1a). Specific examples of such a group include the following:

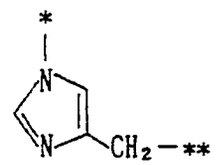


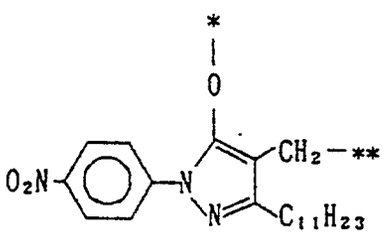












(4) Groups subjecting a hydrolyzation reaction of an ester to cause a cleavage reaction

Examples of such groups include the connecting groups described in West German Patent Disclosure 2,626,315. These groups are represented by the following general formulae wherein the marks * and ** are as defined in the general formula (T-1a):

*-O-CO-** (T-4a)

*-S-CS-** (T-5a)

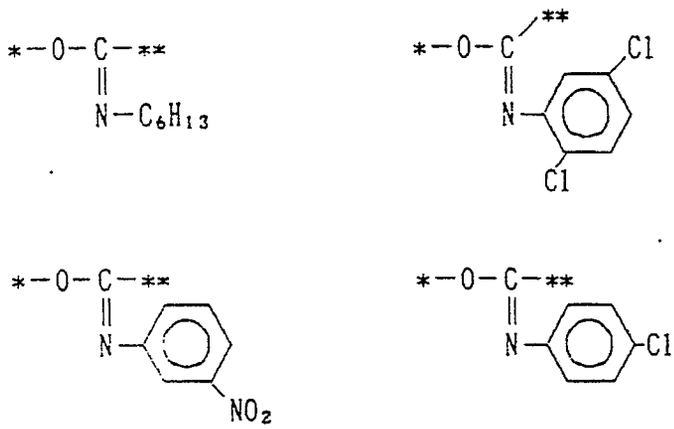
(5) Groups subjecting a cleavage reaction of an iminoketal

Examples of such groups include the connecting groups described in U.S. Patent 4,546,073. Such groups are represented by the following general formula (T-6a):

*-W-C(=NR_{68})-** (T-6a)

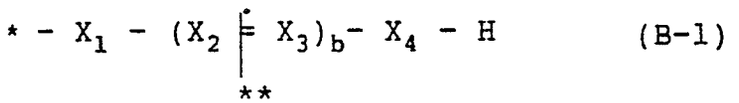
wherein the marks * and ** and W are as defined in the general formula (T-1a); and R_{68} has the same

meaning as R_{67} as defined in general formula (T-1a). Specific examples of the group represented by the general formula (T-6a) include the following:



In general formula (I), the group represented by B is one which becomes a redox group or a coupler after cleavage from A-$(L1)_a$. The group represented by B is a group which has the same meaning as described above with reference to the group represented by A. The group represented by B contains a group which is capable of undergoing a reaction with an oxidation product of a developing agent to eliminate the group connected to the right of B as viewed in the general formula (I). Examples of the group represented by B include groups represented by B as described in JP-A-63-6550, groups represented by COUP(B) as described in U.S. Patent 4,438,193, and groups represented by RED as described in U.S. Patent 4,618,571. B is preferably connected to A-$(L1)_a$ via its hetero atom, preferably oxygen atom or nitrogen atom.

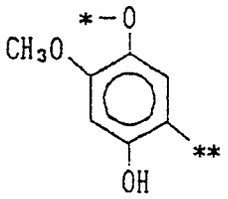
Preferred examples of the group represented by B include those represented by the following general formulae (B-1), (B-2), (B-3), and (B-4):

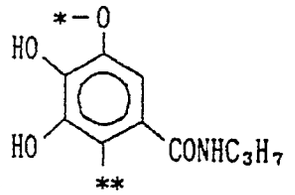


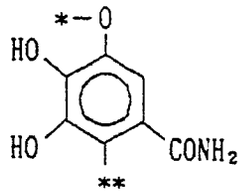
(B-1)

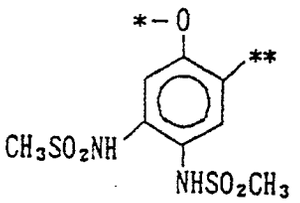
wherein the mark * indicates the position at which it is connected to the group to the left of B as viewed in the general formula (I); the mark ** indicates the position at which it is connected to the group to the right of B as viewed in the general formula (I); X_1 and X_4 each represents an oxygen atom or >N-SO_2R_{71} (in which R_{71} represents an aliphatic, aromatic or heterocyclic group); X_2 and X_3 each represents a methine group or a nitrogen atom; and b represents an integer from 1 to 3, with the proviso that at least one of the X_2 and X_3 groups represents a methine group containing a bonding position represented by the mark ** and that when b is 2 or 3, the plurality of X_2 and X_3 groups may be the same or different. When X_2 and X_3 are methine groups containing substituents, they may or may not be connected to each other to form a cyclic structure (e.g., benzene ring or pyridine ring). After the cleavage of the bond at the mark *, the group represented by the general formula (B-1) becomes a compound according to Kendall- Pelz' Law (T.H. James, "The Theory of the Photographic Process", 4th ed., Macmillan Publishing Co., Inc., page 299) which then undergoes reaction with an oxidation product of a developing agent to undergo oxidation.

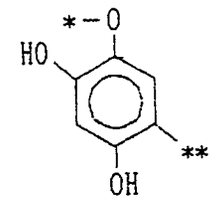
Specific examples of the group represented by (B-1) include the following:

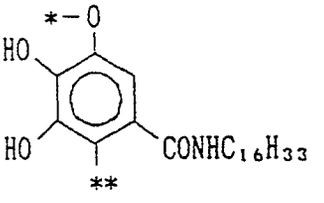


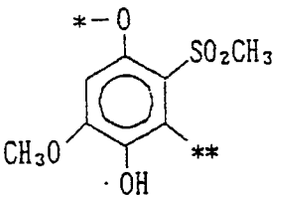


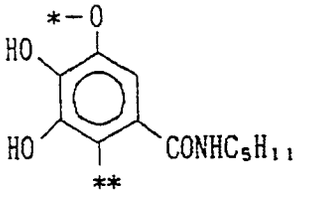


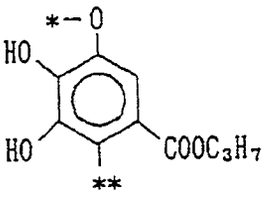


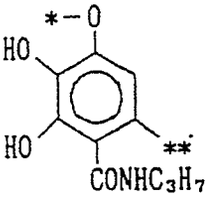




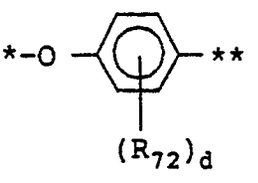




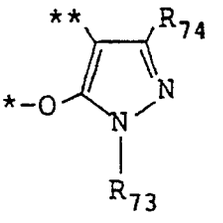




(B-2)



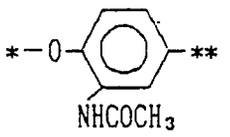
(B-3)

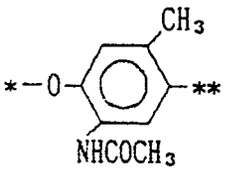


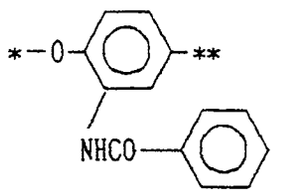
wherein the marks * and ** are as defined in the general formula (B-1); R_{72}, R_{73} and R_{74} each represents a group which allows the groups represented by (B-2) or (B-3) to serve as couplers containing a coupling-

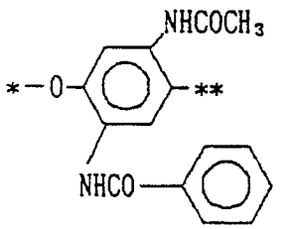
separatable group at the mark ** after cleavage at the mark *; and d represents an integer from 0 to 4. When d is 2 to 4, the R_{72} groups may be the same or different. The plurality of R_{72} groups may be connected to each other to form a cyclic structure (e.g., benzene ring). Examples of R_{72} include an acyamino, alkyl, anilino, amino and alkoxy group. Examples of R_{73} include a phenyl group and an alkyl group.

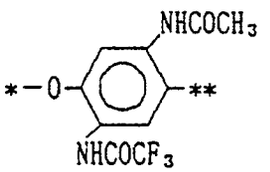
Specific examples of the groups represented by the general formulae (B-2) and (B-3) include the following:

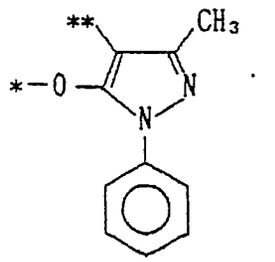


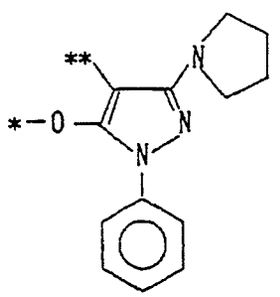




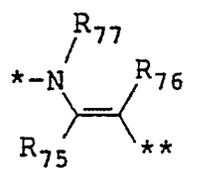






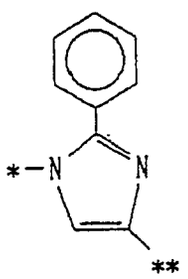


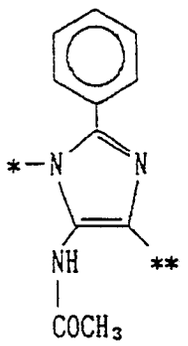
(B-4)

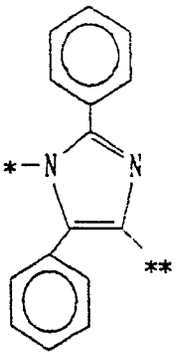


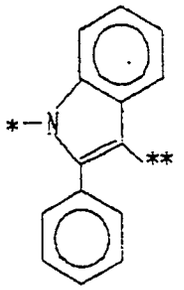
wherein the marks * and ** are as defined in the general formula (B-1); R_{75}, R_{76} and R_{77} each represents a substituent. R_{75}, R_{76} and R_{77} each represents hydrogen atom; an unsubstituted, substituted, straight chain, branched and cyclic alkyl group, for example, methyl, ethyl, i-propyl, t-butyl, octyl, allyl, cyclohexyl; an aryl group, for example, phenyl, toryl; an alkoxy group, for example, methoxy, ethoxy, ethoxyethoxy; an alkylthio group, for example, ethylthio, hexylthio; an aryloxy group, for example, phenoxy, 4-methoxyphenoxy; an arylthio group, for example, phenylthio; a carbonamido group, for example, methylcarbonylamido, phenylcarbonylamido; a carbamoyl group, for example, N,N-dimethylaminocarbonyl, phenylaminocarbonyl; a sulfonamido group, for example, methanesulfonamido, phenylsulfonamido; a sulfamoyl group, for example, N,N-diethylaminosulfonyl; and alkoxycarbonyl group, for example, ethoxycarbonyl, phenoxycarbonyl; an acyl group, for example, acetyl; a sulfonyl group, for example, butylsulfonyl, phenylsulfonyl; a halogen atom, for example, chlorine, bromine; and a cyano group. R_{75}, R_{76} and R_{77} may be the same or different. It is preferred that R_{75} and R_{77}, or R_{77} and R_{76} combine together to form a divalent group of forming a nitrogen-containing heterocyclic group, wherein R_{76} in a case of forming a ring together with R_{77} and R_{75}, or R_{75} in a case of forming a ring together with R_{77} and R_{76} represents the substituent disclosed above. The group represented by (B-4) becomes a coupler containing a coupling-separable group at the mark ** after cleavage at the mark *.

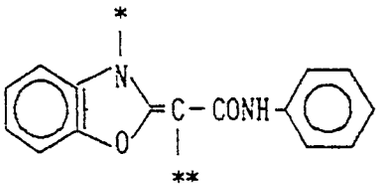
Specific examples of the group represented by the general formula (B-4) include the following:

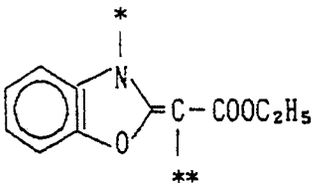


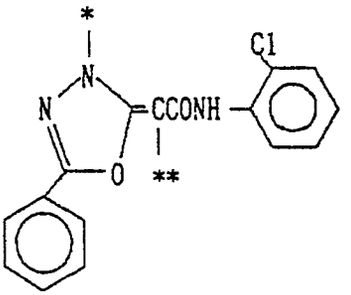








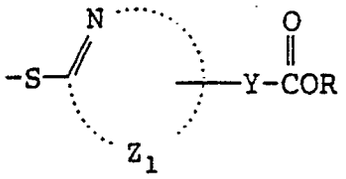




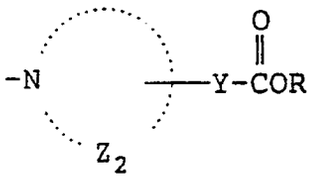
In the general formula (I), examples of the group represented by DI include a tetrazolylthio group, a thiadiazolylthio group, an oxadiazolylthio group, a triazolylthio group, a benzimidazolyl group, a benzthiazolylthio group, a tetrazolylseleno group, a benzoxazolylthio group, a benzotriazolyl group, a triazolyl group, and a benzoimidazolyl group. Examples of these groups are described in U.S. Patents 3,227,554, 3,384,657, 3,615,506, 3,617,291, 3,733,201, 3,933,500, 3,958,993, 3,961,959, 4,149,886, 4,259,437, 4,095,984, 4,477,563, and 4,782,012, and British Patent 1,450,479.

When the group represented by DI in general formula (I) is a group represented by either of the following general formulae (I-a) or (I-b), the effects of the present invention are particularly great:

(I-a)



(I-b)



wherein Y represents a divalent connecting group containing 8 or less carbon atoms or a bond; R represents a C_{1-6} aliphatic or heterocyclic group; Z_1 represents a nonmetallic atom group required to form a heterocyclic group of carbon and nitrogen atoms; and Z_2 represents a nonmetallic atom group required to form a heterocyclic group (a single or condensed ring) having a nitrogen atom.

Z_1 is preferably a nonmetallic atom group required to form a 5- to 7-membered substituted or condensed heterocyclic group, containing a -C=N- bond. Examples of such heterocyclic groups include triazole, tetrazole, oxadiazole, thiadiazole, benzimidazole, and benzthiazole. Particularly preferred among these groups are tetrazole, 1,3,4-thiadiazole, 1,3,4-oxadiazole, and 1,2,4-triazole.

Z_2 is preferably a nonmetallic atom group required to form a 5- to 7-membered substituted or unsubstituted heterocyclic group (a single or condensed ring) containing a least one nitrogen atom. Examples of such heterocyclic groups include imidazole, 1,2,4-triazole, benzotriazole, 1,2,3-triazole, pyrazole, indazole, imidazolin-2-thione, oxazolin-2-thione, 1,2,4-triazolin-3-thione, and 1,3,4-thiadiazolin-2-thione. Particularly preferred among these groups are 1,2,3-triazole and benzotriazole.

Examples of the substituents to the heterocyclic group represented by the general formula (I-a) or (I-b) other than Y-COOR include an aliphatic group (C_{1-6} aliphatic group, e.g., methyl, ethyl), a halogen atom (e.g., chlorine, fluorine, bromine), a heterocyclic group (C_{1-5} 3- to 6-membered heterocyclic group containing oxygen, sulfur or nitrogen atom as hetero atom, e.g., furyl, thienyl, imidazolyl), a nitro group, a cyano group, an aromatic group (C_{6-10} aromatic group, e.g., phenyl), an amino group, an alkylthio group (C_{1-10} alkylthio group, e.g., methylthio, ethylthio), and an acylamino group (C_{2-8} acylamino group, e.g., acetamido, benzamido).

The divalent group represented by Y is preferably an aliphatic or aromatic divalent connecting group which may contain an ether bond, a thioether bond, or a bonding group containing a hetero atom such as -NHCO-, $-SO_2-$, -CO-, and $-NHSO_2-$. Or Y may be a bond. Examples of the divalent group represented by Y include methylene, ethylene, propylene, $-CH(CH_3)-$, $-SCH_2-$, $-SCH(CH_3)-$, $-CH_2O-CH_2-$, $-SCH_2CH_2-$ and $-CH_2SCH_2-$.

In general formulae (I-a) and (I-b), the group represented by R is preferably a C_{1-6} aliphatic group which may be substituted. Examples of such aliphtic groups include methyl, ethyl, propyl, butyl, isopropyl, isobutyl, isoamyl, sec-amyl, and t-amyl. The R group may be substituted by various groups including an alkoxycarbonyl group (C_{2-6} alkoxycarbonyl group, e.g., methoxycarbonyl, propoxycarbonyl, butoxycarbonyl, isobutoxycarbonyl, isopropoxycarbonyl, pentyloxycarbonyl, isopentyloxycarbonyl, 2-methoxyethoxycarbonyl), a carbamoyl group (C_{0-6} carbamoyl group, e.g., N,N-diethylcarbamoyl, N-methyl-N-ethylcarbamoyl, pyrrolidinocarbonyl, piperidinocarbonyl), a halogen atom (e.g., chlorine, fluorine), a nitro group, a cyano group, an alkoxy group (C_{1-4} alkoxy group, e.g., methoxy, ethoxy, methoxyethoxy), a sulfamoyl group (C_{0-6} sulfamoyl group, e.g., N,N-diethylsulfamoyl, N-methyl-N-ethylsulfamoyl), an aryloxy group (C_{6-10} aryloxy group, e.g., 4-chlorophenoxy), an acyl group (C_{2-6} acyl group, e.g., acetyl, benzoyl), a sulfonyl

group (C_{1-6} sulfonyl group, e.g., methanesulfonyl, butanesulfonyl), a heterocyclic group (C_{1-5} 3- to 6-membered heterocyclic group containing hetero atoms such as nitrogen, oxygen and sulfur, e.g., 2-pyridyl, 3-pyridyl), and a phospholyl group (C_{2-5} phospholyl group, e.g., 0,0-diethylphospholyl).

Preferred embodiments of the compound represented by the general formula (I) will be described hereinafter.

In the general formula (I), p is preferably 0 or 1.

The compound represented by the general formula (I) is preferably nondiffusing. In particular, a nondiffusing group is preferably contained in A, L1 or B.

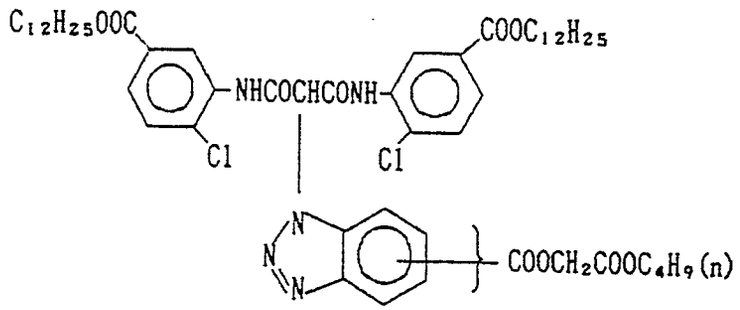
Particularly preferred among the compounds represented by the general formula (I) are those wherein A represents a coupler group.

Particularly preferred among the compounds represented by the general formula (I) are those (i) wherein a = 1, m = 0, p = 1, and n = 0, (ii) wherein a = 0, m = 1, p = 1, and n = 0, (iii) wherein a = 0, m = 0, p = 0, and n = 0, or (iv) wherein a = 1, m = 0, p = 1, and n = 1. These compounds are excellent in color reproducibility by inter image effect and sharpness by edge effect.

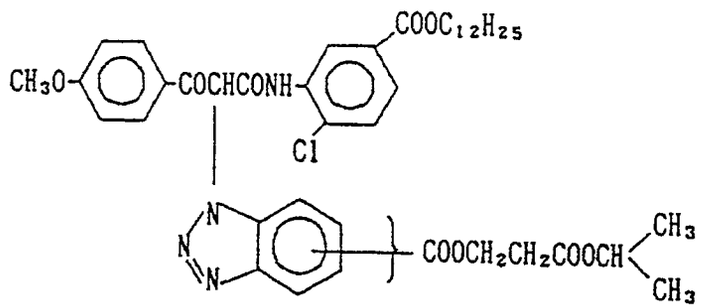
Examples of compounds represented by the general formula (I) and their synthesis methods are disclosed in known patents and literatures cited for the description of A, L_1, B, L_2 and DI in the general formula (I), and JP-A- 63-37346 and 61-156127.

Examples of the compounds of general formula (I) will be set forth below, but the present invention should not be construed as being limited thereto:

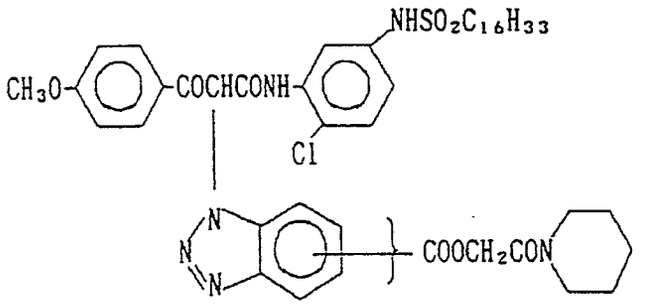
(D-1)



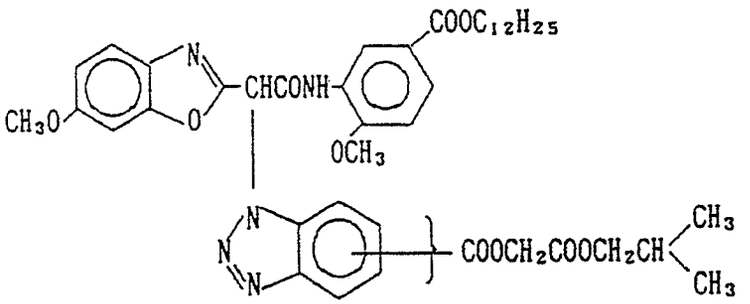
(D-2)



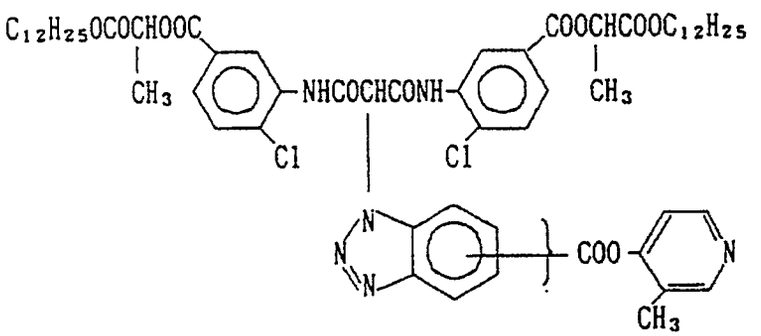
(D－3)



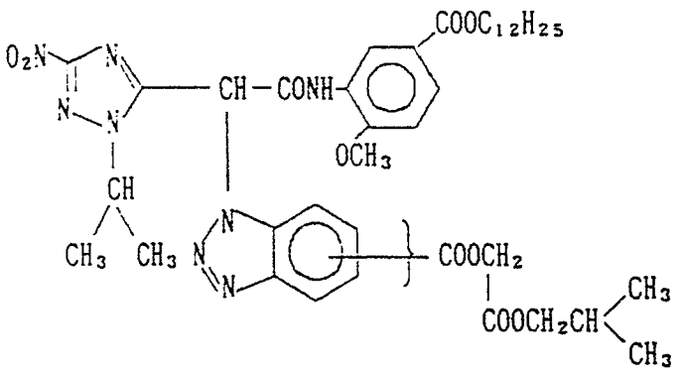
(D－4)



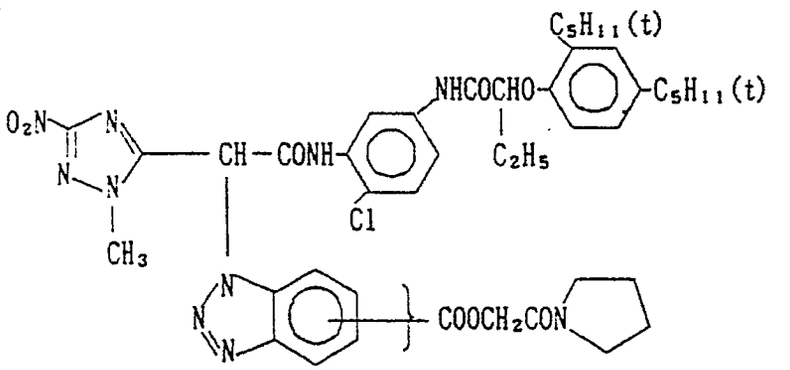
(D－5)



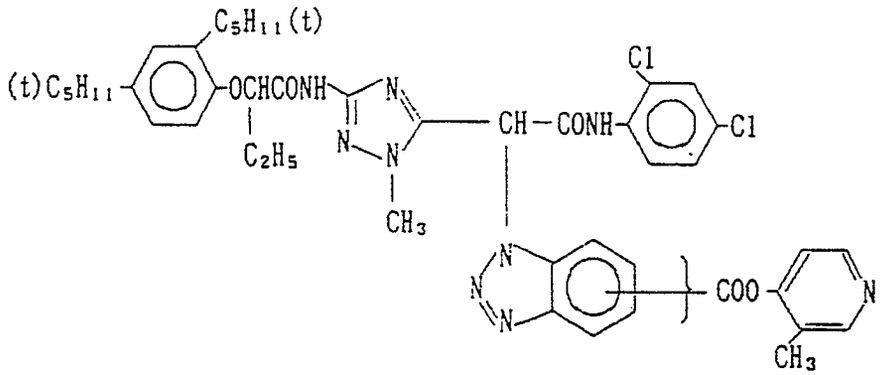
(D－6)



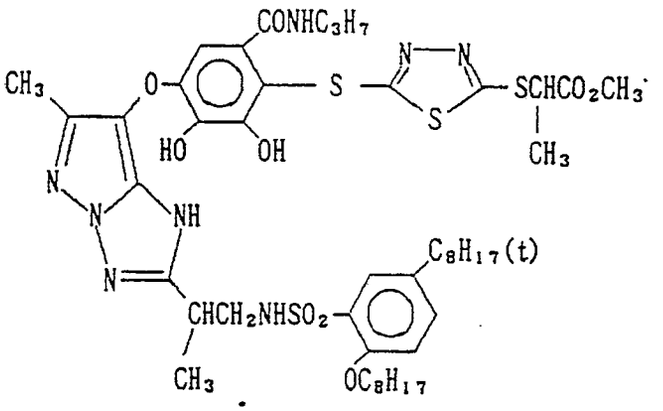
(D－7)



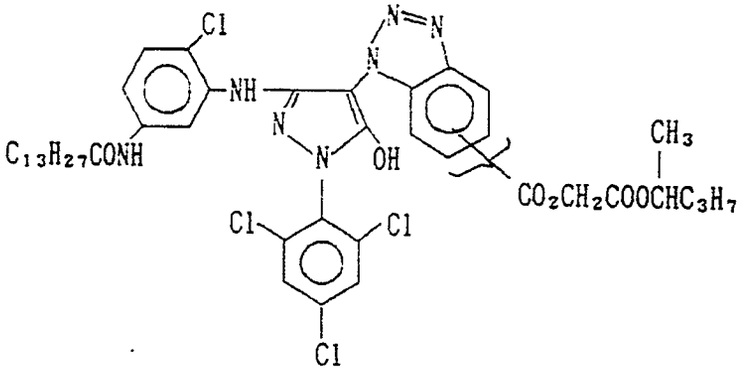
(D-8)



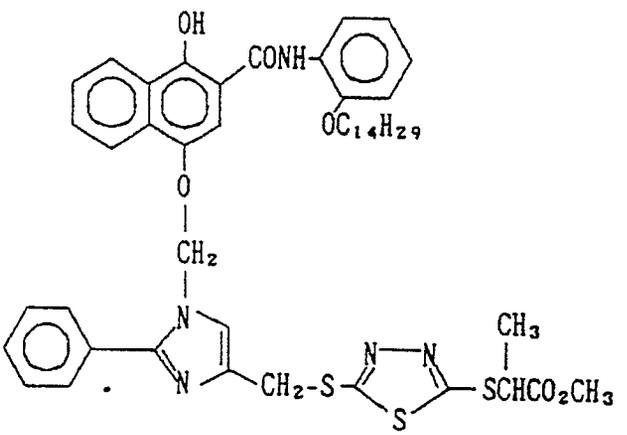
(D-9)



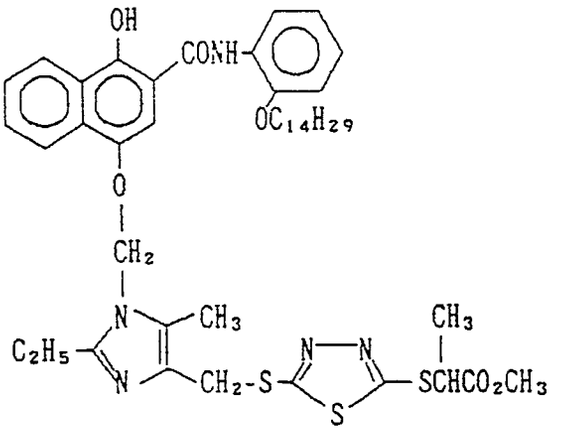
(D-10)



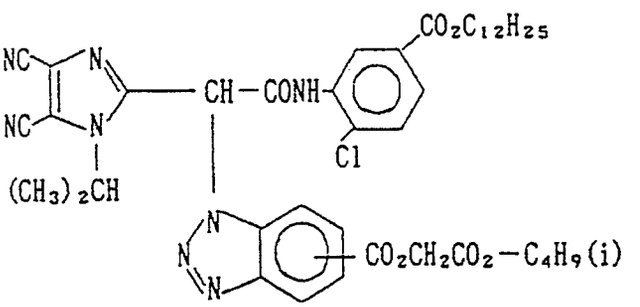
(D-11)



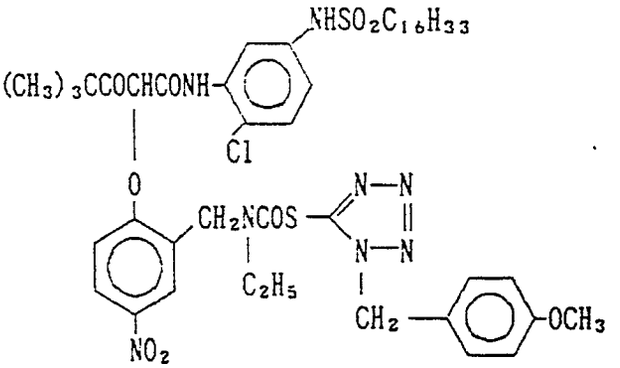
(D-12)



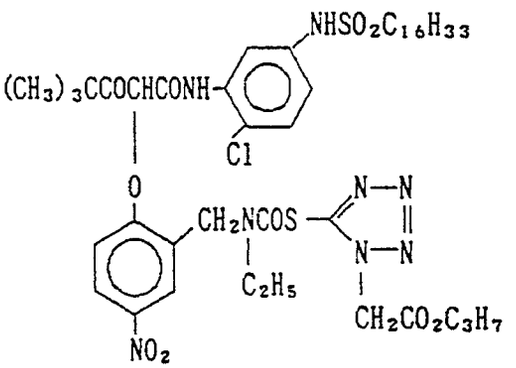
(D-13)



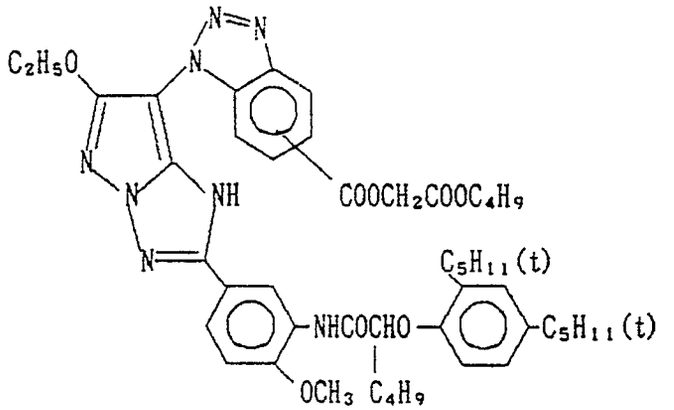
(D-14)



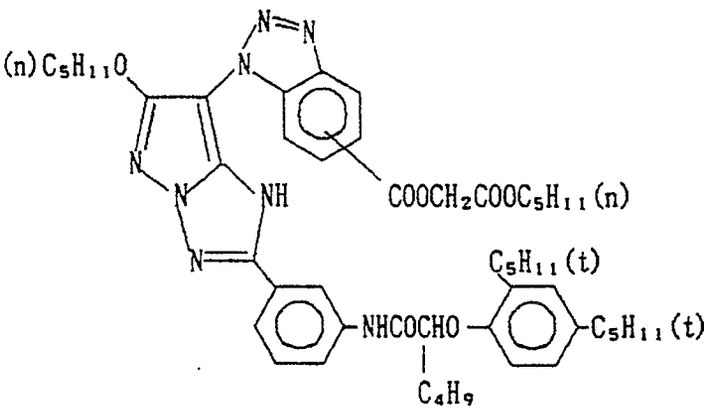
(D-15)



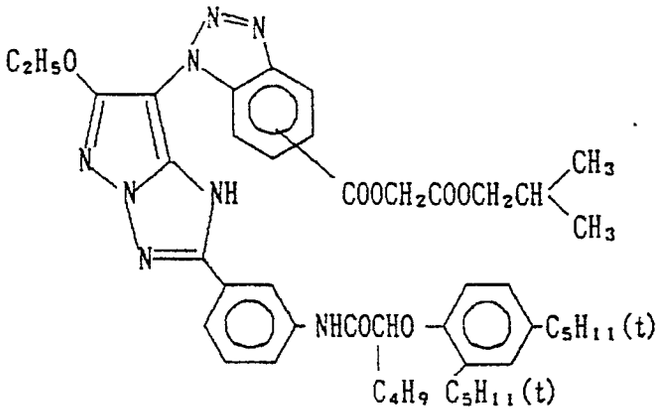
(D－16)



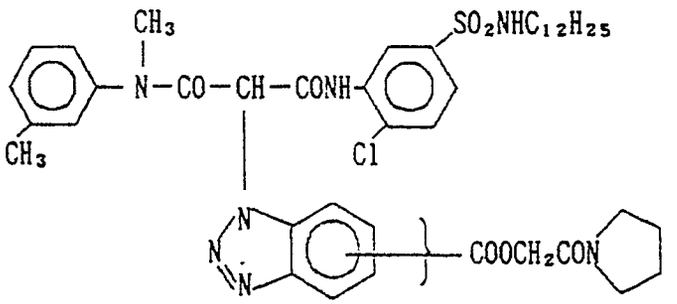
(D－17)



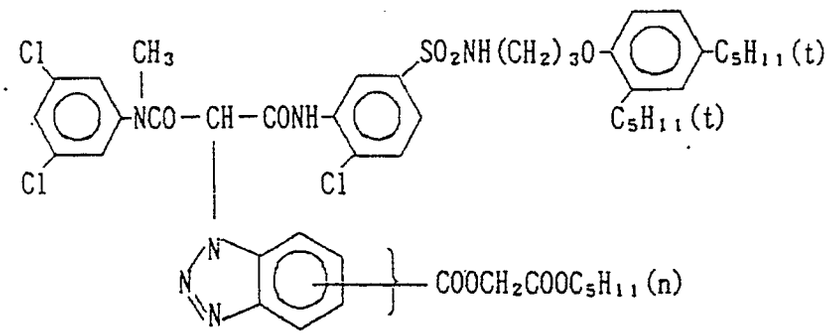
(D-18)



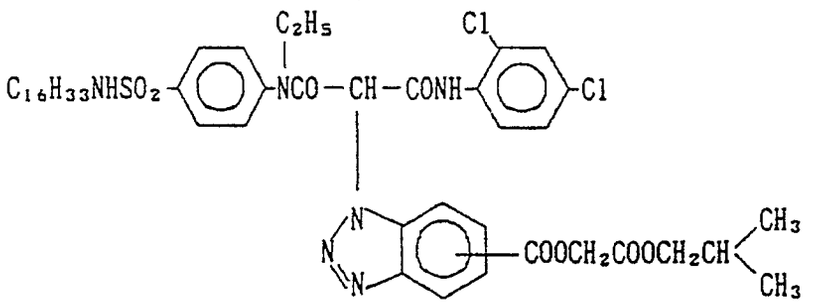
(D-19)



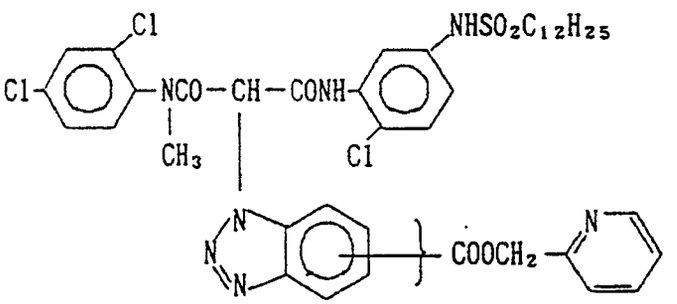
(D-20)



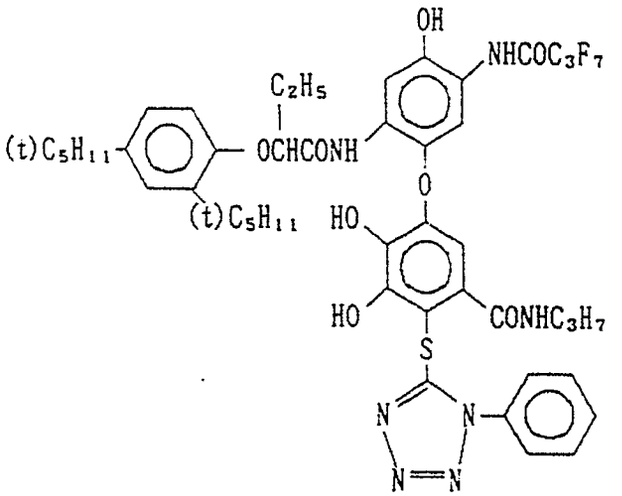
(D-21)



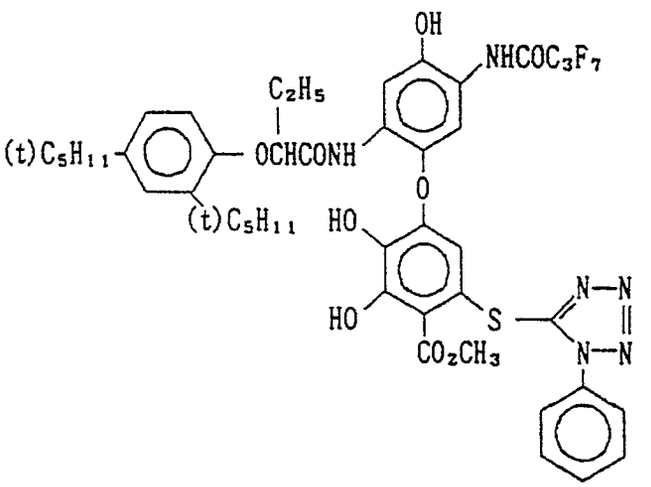
(D-22)



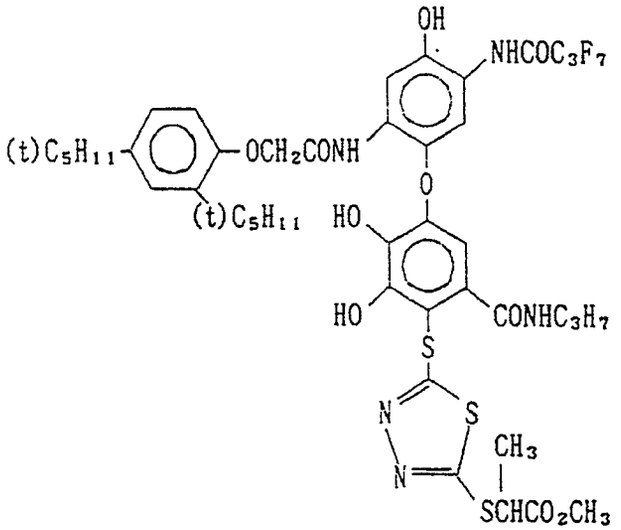
(D－23)



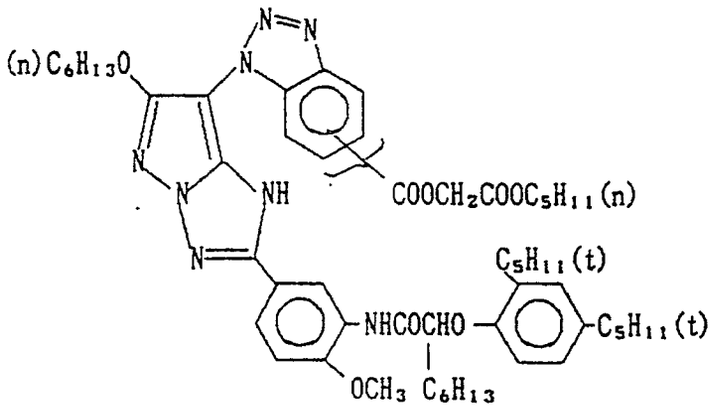
(D－24)



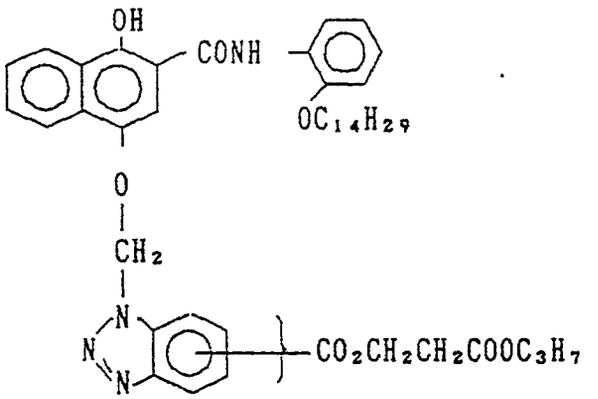
(D－25)



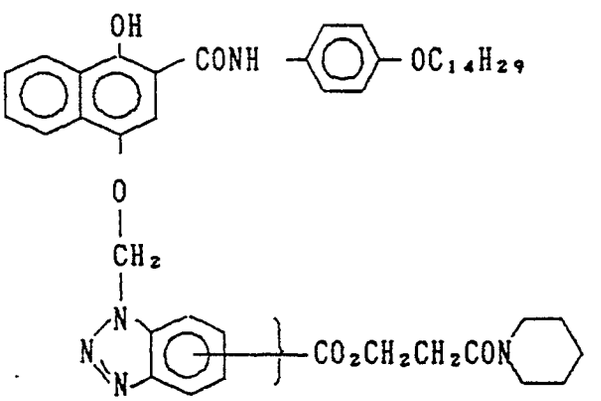
(D－26)



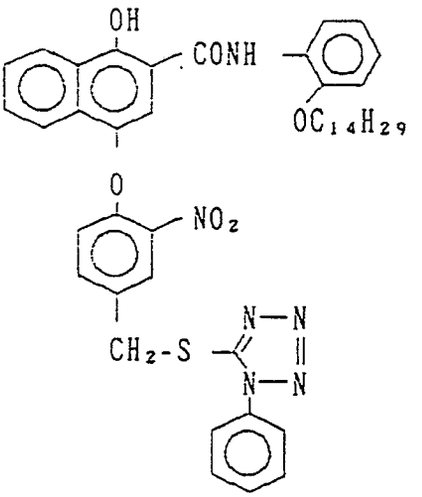
(D－27)



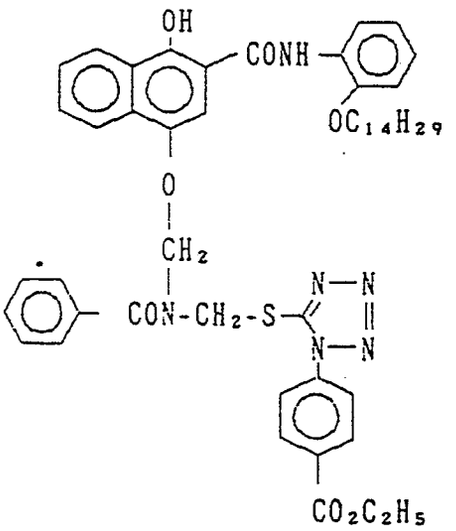
(D－28)



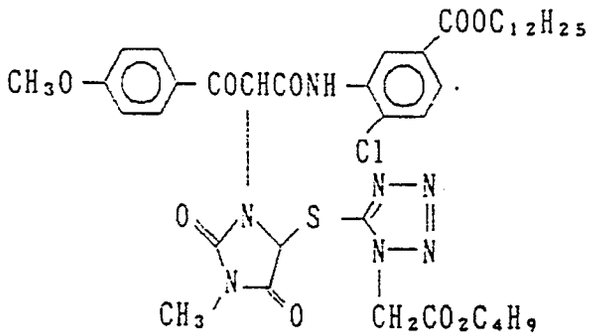
(D-29)



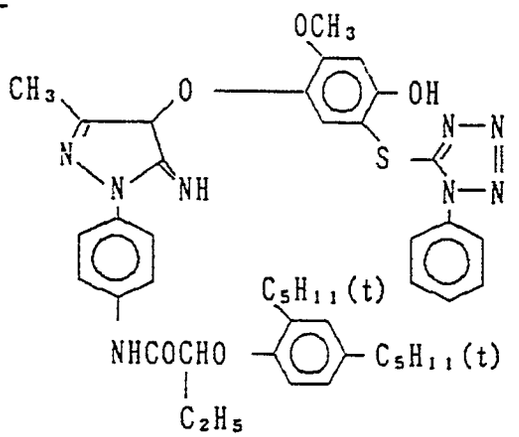
(D-30)



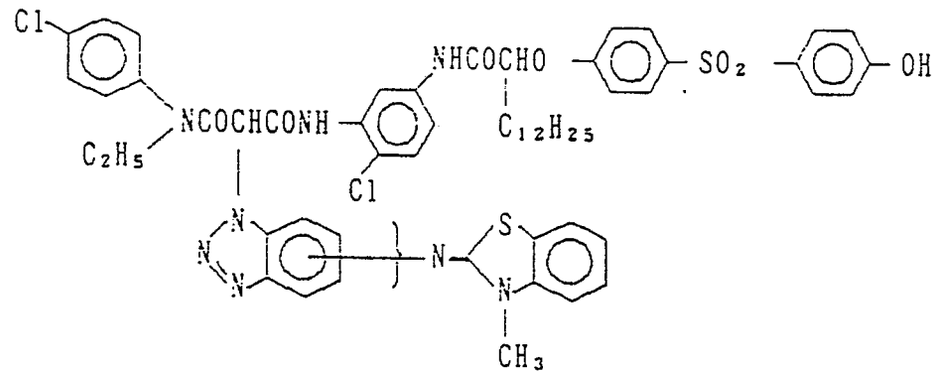
(D－31)



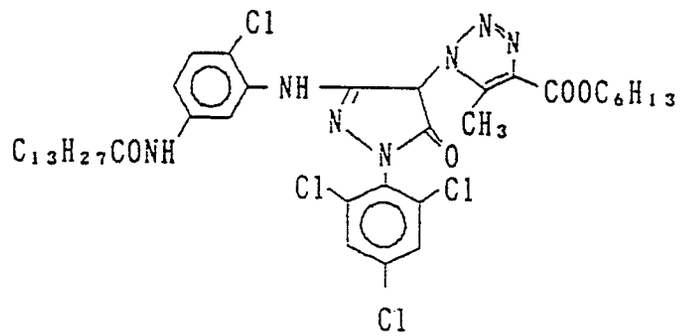
(D－32)



(D－33)



(D－34)



The compound represented by the general formula (I) is incorporated in any of the constituent layers of the silver halide color photographic material except the support. For example, the compounds of formula (I) can be incorporated into a light-sensitive layer or a light-insensitive layer, preferably a light-sensitive layer, a layer containing silver such as colloidal silver, and their adjacent layers. More preferably, the compound (I) is incorporated in a layer containing the above mentioned yellow colored cyan coupler of the present invention or the compound represented by the general formula (A) or their adjacent layers.

In order to further satisfy the required photographic characteristics for the present invention, two or more compounds represented by the general formula (I) can be incorporated in the same layer. Alternatively, the same compound can be incorporated in two or more different layers. The compound represented by the general formula (I) can be used in combination with other known DIR couplers and DIR compounds.

The amount of the compound (I) to be incorporated is normally from 1×10^{-2} to 50 mol%, preferably 5×10^{-2} to 30 mol%, and more preferably 1×10^{-1} to 20 mol%. If incorporated in a layer free of silver, the amount is normally from 1×10^{-7} to 5×10^{-4} mol/m^2, preferably 5×10^{-7} to 3×10^{-4} mol/m^2, and more preferably 1×10^{-6} to 1×10^{-4} mol/m^2.

The incorporation of the compound represented by the general formula (I) in the light-sensitive material can be accomplished by known addition or dispersion methods as described later on.

The present color photographic light-sensitive material for photographing can comprise at least one blue-sensitive layer, at least one green-sensitive layer and at least one red-sensitive layer on a support. The number of silver halide emulsion layers and light-insensitive layers and the order of arrangement of these layers are not specifically limited. In a typical embodiment, the present silver halide photographic material comprises light-sensitive layers consisting of a plurality of silver halide emulsion layers having substantially the same color sensitivity and different light sensitivities on a support. The light-sensitive layers having substantially the same color sensitivity are referred to as a light-sensitive layer unit and have a color sensitivity to any of blue light, green light and red light. In this multi-layer silver halide color photographic material, these light-sensitive layer units are normally arranged in the order of a red-sensitive layer unit, a

green-sensitive layer unit and a blue-sensitive layer unit as viewed from the support. However, the order can be optionally reversed depending on the purpose of application. Alternatively, two light-sensitive layers having the same color sensitivity can be arranged with a light-sensitive layer from a unit having a different color sensitivity interposed therebetween.

Light-insensitive layers can be provided between these silver halide light-sensitive layers and on the uppermost layer and lowermost layer.

These interlayers can comprise couplers, DIR compounds or the like as described in JP-A-61-43748, JP-A-59-113438, JP-A-59-113440, JP-A-61-20037 and JP-A-61-20038. These interlayers can further comprise a commonly used color mixing inhibitor.

The plurality of silver halide emulsion layers constituting each light-sensitive layer unit can be preferably arranged in a two-layer structure, i.e., a high sensitivity emulsion layer and a low sensitivity emulsion layer, as described in West German Patent 1,121,470 and British Patent 923,045. In general, these layers are preferably arranged in an order such that the light sensitivity becomes lower towards the support. Furthermore, a light-insensitive layer can be provided between these silver halide emulsion layers. As described in JP-A-57-112751, JP-A-62-200350, JP-A-62-206541, and JP-A-62-206543, a low sensitivity emulsion layer can be provided remote from the support while a high sensitivity emulsion layer can be provided nearer the support.

In one embodiment of a suitable layer arrangement, the arrangement can be, in order, a low sensitivity blue-sensitive layer (BL), a high sensitivity blue-sensitive layer (BH), a high sensitivity green-sensitive layer (GH), a low sensitivity green-sensitive layer (GL), a high sensitivity red-sensitive layer (RH), a low sensitivity red-sensitive layer (RL)/support. In another embodiment, the order can be BH, BL, GL, GH, RH, RL/support. In a further embodiment, the order can be BH, BL, GH, GL, RL, RH/support.

As described in JP-B-55-34932 (the term "JP-B" as used herein means an "examined Japanese patent publication"), a layer order of a blue-sensitive layer, GH, RH, GL, and RL/support can be arranged. Alternatively, as described in JP-A-56-25738 and 62-63936, a blue-sensitive layer, GL, RL, GH, RH/support can be arranged.

As described in JP-B-49-15495, a light-sensitive layer unit may have a layer arrangement such that the uppermost layer is a silver halide emulsion layer having the highest sensitivity, the middle layer is a silver halide emulsion layer having a lower sensitivity, and the lowermost layer is a silver halide emulsion layer having a lower sensitivity than that of the middle layer. In such a layer arrangment, the light sensitivity becomes lower towards the support. Even if the layer unit comprises three layers having different light sensitivities, a middle sensitivity emulsion layer, a high sensitivity emulsion layer and a low sensitivity emulsion layer can be arranged in this order remote from the support in a color-sensitive layer unit as described in JP-A-59-202464.

Alternatively, the order of a high sensitivity emulsion layer, a low sensitivity emulsion layer and a middle sensitivity emulsion layer or the order of a low sensitivity emulsion layer, a middle sensitivity emulsion layer and a high sensitivity emulsion layer can be used.

In the case where a layer unit comprises four or more layers, too, the order of arrangement of layers can be chosen and altered similarly.

In order to improve color reproducibility, a donor layer (CL) having an interimage effect and a different spectral sensitivity distribution from the main light-sensitive layer such as BL, GL and RL may be preferably provided adjacent or close to the main light-sensitive layer, as is disclosed in US Patents 4,663,271, 4,705,744 and 4,707,436, JP-A-62-160448 and JP-A-63-89850.

As described above, various layer structures and arrangements can be selected depending on the purpose of the light-sensitive material.

A suitable silver halide to be incorporated in the photographic emulsion layer in the present color light-sensitive photographic material is silver bromoiodide, silver chloroiodide or silver bromochloroiodide containing silver iodide in an amount of about 30 mol% or less. Particularly suitable is silver bromoiodide containing silver iodide in an amount of about 2 to about 10 mol%.

The silver halide grains in the photographic emulsions may be so-called regular grains having a regular crystal form, such as cube, octahedron and tetradecahedron, or those having an irregular crystal form such as a spherical or a tabular form, those having a crystal defect such as twinning plane, or those having a combination of these crystal forms.

The silver halide grains may be either fine grains having a projected area diameter of about 0.2 μm or less, or large grains having a projected area diameter of up to about 10 μm. The emulsion may be either a monodisperse emulsion or a polydisperse emulsion.

The preparation of the silver halide photographic emulsion which can be used in the present invention can be accomplished by any suitable method described in Research Disclosure (RD) No. 17643 (December

1978), pp. 22-23, "I. Emulsion Preparation and Types", and No. 18716 (November 1979), page 648; Glafkides, Chimie et Physique Photographique, Paul Montel (1967), G.F. Duffin, Photographic Emulsion Chemistry, Focal Press, (1966); and V.L. Zelikman et al., Making and Coating Photographic Emulsion, Focal Press, (1964).

Furthermore, the monodisperse emulsions as described in U.S. Patents 3,574,628 and 3,655,394 and British Patent 1,413,748 are preferably used in the present invention.

Tabular grains having an aspect ratio of about 3 or more can be used in the present invention. The preparation of such tabular grains is easily accomplished by any suitable method such as described in Gutoff, Photograpahic Science and Engineering, Vol. 14, pp. 248-257, 1970; U.S. Patents 4,434,226, 4,414,310, 4,433,048, and 4,439,520, and British Patent 2,112,157.

The individual silver halide crystals may have either a homogeneous structure or a heterogeneous structure comprising a core and an outer shell differing in halogen composition, or may have a layered structure. Furthermore, the grains may have fused thereto by epitaxy a silver halide having a different halogen composition or may be bonded to a compound other than silver halide, e.g., silver thiocyanate, lead oxide, etc. Mixtures of grains having various crystal forms may also be used.

The above mentioned emulsion may be either the surface latent image type in which latent images are formed mainly on the surface of grains, the internal latent image type in which latent images are formed in the inside of grains, or the type in which latent images are formed both on the surface and in the inside of grains. However, the above mentioned emulsion needs to be a negative type emulsion. If the above mentioned emulsion is the internal latent image type, it may be the core/shell internal latent image type emulsion described in JP-A-63-264740. The process for the preparation of such a core/shell internal latent image type emulsion is described in JP-A-59-133542. The thickness of the emulsion depends on the development process and is preferably from 3 to 40 nm, particularly 5 to 20 nm.

The silver halide emulsion to be used in the present invention is normally subjected to physical ripening, chemical ripening and spectral sensitization. Additives to be used in these steps are described in Research Disclosure Nos. 17643, 18716, and 307105 as tabulated below.

In the light-sensitive material of the present invention, two or more light-sensitive silver halide emulsions which are different in at least one characteristic, such as grain size, grain size distribution, halogen composition, grain shape and sensitivity, may be incorporated in the same layer.

Silver halide grains whose surface is fogged as described in U.S. Patent 4,082,553, silver halide grains whose interior is fogged as described in U.S. Patent 4,626,498 and JP-A-59-214852, and colloidal silver may be preferably incorporated in the light-sensitive silver halide emulsion layer and/or substantially light-insensitive hydrophilic colloidal layer. The silver halide grains whose interior and/or surface is fogged are silver halide grains which can be uniformly (nonimagewise) developed regardless of whether they are on the exposed or unexposed portion of the light-sensitive material. The process for the preparation of silver halide grains whose interior or surface is fogged is described in U.S. Patent 4,626,498 and JP-A-59-214852.

Silver halides which form the core of core/shell type silver halide grains whose interior is fogged may have the same halogen composition or different halogen compositions. The silver halide to be fogged on the surface or in the interior thereof, may be silver chloride, silver bromochloride, silver bromoiodide and silver bromochloroiodide. These fogged silver halide grains are not specifically limited in their size. The average grain size is preferably from 0.01 to 0.75 μm, particularly 0.05 to 0.6 μm. The grain shape is not specifically limited. The silver halide grains may have regular crystal forms or may be polydispersant, but are preferably monodispersant (that is, at least 95% of silver halide grains by weight or grain number have grain sizes which fall within ±40% of the average grain size).

In the present invention, finely divided light-insensitive silver halide grains are preferably used. Finely divided light-insensitive silver halide grains are finely divided silver halide grains which are not sensitive to light upon imagewise exposure for obtaining color images and are not substantially developed. Preferably, finely divided light-insensitive silver halide grains are not previously fogged.

The finely divided light-insensitive silver halide grains have a silver bromide content of 0 to 100 mol% and may optionally contain silver chloride and/or silver iodide, preferably 0.5 to 10 mol% silver iodide.

The finely divided light-insensitive silver halide grains preferably have an average grain diameter of 0.01 to 0.5 μm (as calculated in terms of average of diameters of projected area corresponding to sphere), more preferably 0.02 to 0.2 μm.

The preparation of the finely divided light-insensitive silver halide grains can be accomplished in the same manner as ordinary light-sensitive silver halide. In this case, the surface of the silver halide grains does not need to be optically sensitized. Also, the silver halide grains don not need to be spectrally sensitized. However, before being added to the coating solution, the silver halide emulsion preferably comprises a known stabilizer such as a triazole, azaindene, benzothiazolium or mercapto compound

incorporated therein. Colloidal silver may be preferably incorporated in the layer containing finely divided light-insensitive silver halide grains.

The amount of silver to be coated on the light-sensitive material of the present invention is preferably from 6.0 g/m^2 or less, more preferably 4.5 g/m^2 or less.

Known photographic additives which can be used in the present invention are also described in the above cited three references as shown in the following table.

Kind of additive	RD17643 [Dec.'78]	RD18716 [Nov. '79]	RD307105 [Nov. '89]
1. Chemical sensitizer	p. 23	p. 648 right column (RC)	p. 866
2. Sensitivity increasing agent		p.648 RC	
3. Spectral sensitizer and supersensitizer	pp.23-24	p.648 RC-p.649 RC	pp.866-868
4. Brightening agent	p. 24	p.647 RC	p.868
5. Antifoggant and stabilizer	pp. 24-25	p. 649 RC	pp.868-870
6. Light absorbent, filter dye, and ultraviolet absorbent	pp. 25-26	p. 649 RC-p. 650 left column (LC)	p.873
7. Stain inhibitor	p. 25 RC	p. 650 LC-RC	p.872
8. Dye image stabilizer	p. 25	p.650 LC	p.872
9. Hardening agent	p. 26	p. 651 LC	pp.874-875
10. Binder	p. 26	p. 650 LC	pp.873-874
11. Plasticizer and lubricant	p. 27	p. 650 RC	p.876
12. Coating aid and surface active agent	pp. 26-27	p. 650 RC	pp. 875-876
13. Antistatic agent	p. 27	p. 650 RC	pp. 876-877
14. Matting agent			pp. 878-879

In order to inhibit deterioration in photographic properties due to formaldehyde gas, a compound capable of reacting with and fixing formaldehyde such as disclosed in U.S. Patents 4,411,987 and 4,435,503

can be incorporated in the light-sensitive material.

Mercapto compounds described in U.S. Patents 4,740,454, and 4,788,132, and JP-A-62-18539, and JP-A-1-283551 may be preferably incorporated in the light-sensitive material of the present invention.

A compound which releases a fogging agent, a development accelerator, a silver halide solvent or precursors thereof regardless of the amount of developed silver produced by development disclosed in JP-A-1-106052 may be preferably incorporated in the light-sensitive material of the present invention.

A dye dispersed by the process described in International Patent Disclosure WO88/04794 and JP-A-1-502912 or a dye described in EP317,308A, U.S. Patent 4,420,555, and JP-A-1-259358 may be preferably incorporated in the light-sensitive material of the present invention.

Various color couplers can be used in the present invention. Specific examples of the color couplers are described in the patents identified in the above cited Research Disclosure No. 17643, VII-C to G and No. 307105, VII-C to G.

The yellow couplers which are preferably used in combination with the coupler represented by the general formula (CI) to (CIV) are those described in U.S. Patents 3,933,501, 4,022,620, 4,326,024, 4,401,752, 4,248,961, 3,973,968, 4,314,023, and 4,511,649, JP-B-58-10739, British Patents 1,425,020 and 1,476,760, and European Patent 249,473A.

The preferred magenta couplers include 5-pyrazolone compounds and pyrazoloazole compounds. Particularly preferred are those described in U.S. Patents 4,310,619, 4,351,897, 3,061,432, 3,725,064, 4,500,630, 4,540,654, and 4,556,630, European Patent 73,636, JP-A-60-33552, JP-A-60-43659, JP-A-61-72238, JP-A-60-35730, JP-A-55-118034, and JP-A-60-185951, RD Nos. 24220 (June 1984) and 24230 (June 1984), and WO(PCT)88/04795.

The cyan couplers include naphthol and phenol couplers. Preferred are those described in U.S. Patents 4,052,212, 4,146,396, 4,228,233, 4,296,200, 2,369,929, 2,801,171, 2,772,162, 2,895,826, 3,772,002, 3,758,308, 4,334,011, 4,327,173, 3,446,622, 4,333,999, 4,775,616, 4,451,559, 4,427,767, 4,690,889, 4,254,212, and 4,296,199, West German Patent Disclosure No. 3,329,729, European Patents 121,365A and 249,453A, and JP-A-61-42658. Furthermore, the pyrazoloazole couplers described in JP-A-64-553, JP-A-64-554, JP-A-64-555 and JP-A-64-556 and the imidazole couplers described in U.S. Patent 4,818,672 can be used.

Typical examples of polymerized dye-forming couplers are described in U.S. Patents 3,451,820, 4,367,282, 4,409,320, and 4,576,910, British Patent 2,102,173, and European Patent 341,188A.

Couplers which form a dye having moderate diffusibility preferably include those described in U.S. Patent 4,366,237, British Patent 2,125,570, European Patent 96,570, and West German Patent Publication No. 3,234,533.

In addition to the yellow-colored cyan couplers of the present invention, colored couplers for correction of undesired absorptions of the developed color preferably include those described in Research Disclosure No. 17643, VII-G, U.S. Patents 4,163,670, 4,004,929, and 4,138,258, JP-B-57-39413, and British Patent 1,146,368. Furthermore, couplers for correction of undesired absorptions of the developed color by a fluorescent dye released upon coupling described in U.S. Patent 4,774,181 and couplers containing as a separable group a dye precursor group capable of reacting with a developing agent to form a dye described in U.S. Patent 4,777,120, can be preferably used.

Couplers capable of releasing a photographically useful group upon coupling can also be used in the present invention. Preferred examples of DIR couplers which release a developing inhibitor are described in the patents cited in RD 17643, VII-F, and No. 307105, VII-F, JP-A-57-151944, JP-A-57-154234, JP-A-60-184248, and JP-A-63-37346, JP-A-63-37350 and U.S. Patents 4,248,962, and 4,782,012.

Bleach accelerator-releasing couplers such as described in RD Nos. 11449, and 24241, and JP-A-61-201247 are effective for reducing the time required for bleaching, particularly when incorporated in a light-sensitive material comprising the above mentioned tabular silver halide grains. Couplers capable of imagewise releasing a nucleating agent or a developing accelerator at the time of development preferably include those described in British Patents 2,097,140 and 2,131,188, and JP-A-59-157638 and JP-A-59-170840. Compounds which undergo a redox reaction with an oxidation product of a developing agent to release a fogging agent, development accelerator, silver halide solvent or the like such as described in JP-A-60-107029, JP-A-60-252340, JP-A-1-44940, and JP-A-1-45687 may be preferably used.

In addition to the foregoing couplers, the photographic material according to the present invention can further comprise competing couplers such as described in U.S. Patent 4,130,427, polyequivalent couplers such as described in U.S. Patents 4,283,472, 4,338,393, and 4,310,618, DIR redox compounds or DIR couplers or DIR coupler-releasing couplers such as described in JP-A-60-185950 and JP-A-62-24252, couplers capable of releasing a dye which returns to its original color after release such as described in European Patent 173,302A, couplers capable of releasing a ligand such as described in U.S. Patent

4,553,477, couplers capable of releasing a leuco dye such as described in JP-A-63-75747, and couplers capable of releasing a fluorescent dye such as described in U.S. Patent 4,774,181.

The incorporation of these couplers in the light-sensitive material can be accomplished by any suitable known dispersion method.

Examples of high boiling point solvents to be used in an oil-in-water dispersion process are described in U.S. Patent 2,322,027. Specific examples of high boiling point organic solvents which have a boiling point of 175° C or higher at normal pressure and which can be used in the oil-in-water dispersion process include phthalic esters (e.g., dibutyl phthalate, dicylcohexyl phthalate, di-2-ethylhexyl phthalate, decyl phthalate, bis-(2,4-di-t-amylphenyl)phthalate, bis(2,4-di-t-amylphenyl)isophthalate, bis(1,1-diethylpropyl) phthalate), phosphoric or phosphonic esters (e.g., triphenyl phosphate, tricresyl phosphate, 2-ethylhexyl diphenyl phosphate, tricyclohexyl phosphate, tri-2-ethylhexyl phosphate, tridecyl phosphate, tributoxy ethyl phosphate, trichloropropyl phosphate, di-2-ethylhexyl phenyl phosphonate), benzoic esters (e.g., 2-ethylhexyl benzoate, dodecyl benzoate, 2-ethylhexyl-p-hydroxy benzoate), amides (e.g., N,N-diethyldodecanamide, N,N-diethyllaurylamide, N-tetradecylpyrrolidone), alcohols or phenols (e.g., isostearyl alcohol, 2,4-di-tert-amylphenol), aliphatic carboxylic esters (e.g., bis(2-ethylhexyl)sebacate, dioctyl azerate, glycerol tributylate, isostearyl lactate, trioctyl citrate), aniline derivatives (N,N-dibutyl-2-butoxy-5-tert-octylaniline), and hydrocarbons (e.g., paraffin, dodecylbenzene, diisopropyl naphthalene). An auxiliary solvent can be used such as an organic solvent having a boiling point of about 30° C or higher, preferably 50° C to about 160° C. Typical examples of such an organic solvent are ethyl acetate, butyl acetate, ethyl propionate, methyl ethyl ketone, cyclohexanone, 2-ethoxyethyl acetate, and dimethylformamide.

The process and effects of a latex dispersion method and specific examples of latexes to be used in dipping are described in U.S. Patent 4,199,363, West German Patent Application (OLS) 2,541,274, and 2,541,230.

Various preservatives or antimolds such as 1,2-benzisothiazolin-3-one, n-butyl, p-hydroxybenzoate, phenol, 4-chloro-3,5-dimethylphenol, 2-phenoxyethanol, and 2-(4-thiazolyl)benzimidazole as described in JP-A-63-257747, JP-A-62-272248, and JP-A-1-80941 may be preferably incorporated in the present color light-sensitive material.

The present invention is applicable to various types of color light-sensitive materials, particularly preferably to color negative films for common use or motion picture, color reversal films for slide or television, color papers, color positive films and color reversal papers.

Suitable supports which can be used in the present invention are described in the above cited RD 17643 (page 28) and 18716 (right column on page 647 to left column on page 648).

In the present light-sensitive material, the total thickness of all the hydrophilic colloidal layers on the emulsion side is preferably from 28 μm or less, more preferably 23 μm or less, most preferably 18 μm or less and particularly 16 μm or less. The film swelling rate $T_{1/2}$ is preferably 30 seconds or less, more preferably 20 seconds or less. In the present invention, the film thickness is determined after being stored at a temperature of 25° C and a relative humidity of 55% over 2 days. The film swelling rate $T_{1/2}$ can be determined by a method known in the art, e.g., by means of a swellometer of the type as described in A. Green et al, Photographic Science Engineering, Vol. 19, No. 2, pp. 124-129. $T_{1/2}$ is defined as the time taken until half the saturated film thickness is reached wherein the saturated film thickness is 90% of the maximum swollen film thickness reached when the light-sensitive material is processed with a color developer at a temperature of 30° C over 195 seconds.

The film swelling rate $T_{1/2}$ can be adjusted by adding a film hardener to a gelatin binder or altering the ageing condition after coating. The percentage of swelling of the light-sensitive material is preferably from 150 to 400%. The percentage of swelling can be calculated from the maximum swollen film thickness determined as described above in accordance with the equation: (maximum swollen film thickness - film thickness)/film thickness.

In the light-sensitive material of the present invention, one or more hydrophilic colloidal layer as backing layers having a total dried thickness of 2 to 20 μm may be preferably provided on the side of the support opposite to the emulsion layer. The backing layers preferably contain the above mentioned additives, e.g., a light absorbent, filter dye, ultraviolet absorbent, antistatic agent, film hardener, binder, plasticizer, coating aid, surface active agent, etc. The percent of swelling of the backing layers is preferably from 150 to 500%.

The color photographic light-sensitive material according to the present invention can be developed in accordance with a conventional method as described in RD Nos 17643 (pp. 28-29) and 18716 (left column - right column on page 651).

The color developer to be used in the development of the present light-sensitive material is preferably an alkaline aqueous solution containing as a main component an aromatic primary amine color developing agent. For instance, the color developing agent can be an aminophenolic compound. In particular, p-

phenylenediamine compounds are preferably used. Typical examples of such p-phenylenediamine compounds include 3-methyl-4-amino-N,N-diethylaniline, 3-methyl-4-amino-N-ethyl-N-β-hydroxyethylaniline, 3-methyl-4-amino-N-ethyl-N-β-methanesulfonamideethylaniline, 3-methyl-4-amino-N-ethyl-N-β-methoxyethylaniline, and sulfates, hydrochlorides and p-toluenesulfonates thereof. These compounds can be used in combination of two or more thereof depending on the purpose of the application.

The color developer normally contains a pH buffer such as carbonate and phosphate of an alkaline metal or a development inhibitor or a fog inhibitor such as bromides, iodides, benzimidazoles, benzothiazoles and mercapto compounds. If desired, the color developer may further contain various preservatives, e.g., hydroxylamine, diethylhydroxylamine, sulfites, hydrazines (e.g., N,N-biscarboxymethyl hydrazine), phenylsemicarbazides, triethanolamine, and catecholsulfonic acids; organic solvents, e.g., ethylene glycol and diethylene glycol; development accelerators, e.g., benzyl alcohol, polyethylene glycol, quaternary ammonium salts, and amines; color-forming couplers; competing couplers; auxiliary developing agents, e.g., 1-phenyl-3-pyrazolidone; viscosity-imparting agents; various chelating agents exemplified by aminopolycarboxylic acids, aminopolyphosphoric acids, alkylphosphonic acids, and phosphonocarboxylic acids, e.g., ethylenediaminetetraacetic acid, nitrilotriacetic acid, diethylenetriaminepentaacetic acid, cyclohexanediaminetetraacetic acid, hydroxyethyliminodiacetic acid, 1-hydroxyethylidene-1,1-diphosphonic acid, nitrilo-N,N,N-trimethylenephosphonic acid, ethylenediamine-N,N,N',N'-tetramethylenephosphonic acid, and ethylenediamine-di(o-hydroxyphenylacetic acid), and salts thereof.

Reversal processing is usually carried out by black-and-white development followed by color development. Black-and-white developers to be used can contain one or more of known black-and-white developing agents, such as dihydroxybenzenes, e.g., hydroquinone, 3-pyrazolidones, e.g., 1-phenyl-3-pyrazolidone, and aminophenols, e.g., N-methyl-p-aminophenol. The color developer or black-and-white developer usually has a pH of from 9 to 12. The replenishment rate of the developer is usually 3 or less per m^2 of the light-sensitive material, depending on the type of the color photographic material to be processed. The replenishment rate may be reduced to 500 ml/m^2 or less by decreasing the bromide ion concentration in the replenisher. When the replenishment rate is reduced, it is preferable to reduce the area of the liquid surface in contact with the air in the processing tank to thereby prevent evaporation and air-oxidation of the liquid.

The area of the liquid surface in contact with the air can be represented by the opening ratio defined as follows:

Opening ratio = Area of liquid surface in contact with air (cm^3)/ volume of liquid (cm^3)

The opening ratio is preferably 0.1 or less, more preferably 0.001 to 0.05. The reduction of the opening ratio can be accomplished by providing a cover such as a floating cover on the surface of a photographic processing solution in the processing tank, or by a process which comprises the use of a mobile cover as described in JP-A-1-82033, or a slit development process as described in JP-A-63-216050. The reduction of the opening ratio can be applied not only to both the color development and black-and-white development but also to the subsequent steps such as bleach, blix, fixing, rinse and stabilization. The replenishment rate can also be reduced by a means for suppressing accumulation of the bromide ion in the developing solution.

The color development time is normally selected between 2 and 5 minutes. The color development time can be further reduced by carrying out color development at an elevated temperaure and a high pH value with a color developing solution containing a color developing agent in a high concentration.

The photographic emulsion layer which has been color-developed is normally subjected to bleaching and fixing to effect desilvering. Bleaching may be effected simultaneously with fixing (i.e., blix), or these two steps may be carried out separately. For speeding up processing, bleaching may be followed by blix. Further, an embodiment wherein two blix baths connected in series are used, an embodiment wherein blix is preceded by fixing, or an embodiment wherein blix is followed by bleaching may be selected arbitrarily according to the purpose.

Bleaching agents to be used include compounds of polyvalent metals, e.g., iron (III), peroxides, quinones, and nitro compounds. Typical examples of these bleaching agents are organic complex salts of iron (III) with aminopolycarboxylic acis, e.g., ethylenediaminetetraacetic acid, diethylenetriaminepentaacetic acid, cyclohexanediaminetetraacetic acid, methyliminodiacetic acid, 1,3-diaminopropanetetraacetic acid, and glycol ether diaminetetraacetic acid, or citric acid, tartaric acid, malic acid, etc. Of these, aminopolycarboxylic acid-iron (III) complex salts such as (ethylenediaminetetraacetato)iron (III) complex salts are preferred for speeding up processing and for conservation of the environment. In particular, aminopolycarboxylic acid-

iron (III) complex salts are useful in both a bleaching solution and a blix solution. The bleaching or blix solution comprising such an aminopolycarboxylic acid-iron (III) complex salt normally has a pH value of 4.0 to 8.0. For speeding up processing, it is possible to adopt a lower pH value.

The bleaching bath, blix bath or a prebath thereof can contain, if desired, a bleaching accelerator. Examples of useful bleaching accelerators include compounds containing a mercapto group or a disulfide group such as described in U.S. Patent 3,893,858, West German Patents 1,290,812, and 2,059,988 JP-A-53-32736, JP-A-53-57831, JP-A-53-37418, JP-A-53-72623, JP-A-95630, JP-A-53-95631, JP-A-53-104232, JP-A-53-124424, JP-A-53-141623, and JP-A-53-28426, and Research Disclosure No. 17129 (July 1978), thiazolidine derivatives such as described in JP-A-50-140129, thiourea derivatives such as described in U.S. Patent 3,706,561, iodides such as described in West German Patent 1,127,715 and JP-A-58-16235, polyoxyethylene compounds such as described in West German Patents 966,410 and 2,748,430, polyamine compounds such as described in JP-B-45-8836, the compounds as described in JP-A-49-40943, JP-A-49-59644, JP-A-53-94927, JP-A-54-35727, JP-A-55-26506, and JP-A-58-163940, and bromine ions. Preferred among these compounds are those containing a mercapto group or a disulfide group because of their great accelerating effects. In particular, the compounds disclosed in U.S. Patent 3,893,858, West German Patent 1,290,812, and JP-A-53-95630 are preferred. The compounds disclosed in U.S. Patent 4,552,834 are also preferred. These bleaching accelerators may be incorporated into the light-sensitive material. These bleaching accelerators are particularly effective for blix of color light-sensitive materials for photographing.

The bleaching solution or blix solution to be used in the present invention may preferably comprise an organic acid in addition to the above mentioned compounds for the purpose of inhibiting bleach stain. A particularly preferred organic acid is one having an acid dissociation constant (pKa) of 2 to 5. Specific examples of such an organic acid include acetic acid, propionic acid and hydroxyacetic acid.

Fixing agents to be used for fixation in a fixing solution or blix solution include thiosulfates, thiocyanates, thioethers, thioureas, and a large amount of iodides. The thiosulfates are normally used, with ammonium thiosulfate being applicable most often. These thiosulfates may be preferably used in combination with thiocyanates, thioether compounds, thiourea or the like. As preservatives of the fixing bath or blix bath there can be preferably used sulfites, bisulfites, carbonyl bisulfite adducts or sulfinic acid compounds such as described in European Patent 294769A. Further, various aminopolycarboxylic acids or organic phosphonic acids can be added to the fixing bath or blix bath for the purpose of stabilizing the solution.

In the present invention, the fixing solution or blix solution preferably comprises a compound having a pKa of 6.0 to 9.0, preferably an imidazole such as imidazole, 1-methylimidazole, 1-ethylimidazole and 2-methylimidazole, in an amount of 0.1 to 10 mol/ℓ.

The total desilvering time is preferably short so long as insufficient desilvering does not result. The total desilvering time is preferably from 1 to 3 minutes, more preferably 1 to 2 minutes. The desilvering temperature is from 25 to 50 °C, preferably 35 to 45 °C. In this preferred temperature range, the desilvering rate can be improved, and the occurrence of stain after processing can be effectively inhibited.

In the desilvering step, agitation is preferably intensified as much as possible. In particular, agitation can be intensified by various methods. For example, the processing solution may be jetted to the surface of the emulsion layer in the light-sensitive material as described in JP-A-62-183460 and JP-A-62-183461. The agitating effect can be improved by a rotary means as described in JP-A-62-183461. Furthermore, the agitating effect can be improved by moving the light-sensitive material with the emulsion surface in contact with a wiper blade provided in the bath so that turbulence occurs on the emulsion surface. Moreover, the agitation can be intensified by increasing the total circulated amount of processing solution. Such agitation improving methods can be effectively applied to the bleaching bath, blix bath or fixing bath. The improvement agitation effect expedites the supply of a bleaching agent, fixing agent or the like into the emulsion film, resulting in an improved desilvering rate. The above mentioned agitation improving method is more effective when a bleach accelerator is used. In this case, the agitation improving method can remarkably enhance the bleach accelerating effect or eliminate the effect of inhibiting fixation by the bleach accelerator.

An automatic developing machine which can be used in the present invention is preferably equipped with a light-sensitive material conveying means as described in JP-A-60-191257, JP-A-60-191258, and JP-A-60-191259. As described in the above cited JP-A-60-191257, such a conveying means can remarkably reduce the amount of the processing solution carried over from a bath to a succeeding bath, exhibiting a high effect of inhibiting the deterioration of properties of the processing solution. Such an effect is particularly effective for the reduction of the processing time at each step or the replenishment rate of the processing solution.

It is usual that the thus desilvered silver halide color photographic materials of the invention are subjected to washing and/or stabilization. The quantity of water to be used in the washing step can be

selected from a broad range depending on the characteristics of the light-sensitive material (for example, the kind of couplers, etc.), the end use of the light-sensitive material, the temperature of washing water, the number of washing tanks (number of stages), the replenishment system (e.g., counter-flow system or direct-flow system), and other various factors. Of these factors, the relationship between the number of washing tanks and the quantity of water in the multistage counter-flow system can be obtained according to the method described in Journal of the Society of Motion Picture and Television Engineers, Vol. 64, pp. 248-253 (May 1955). According to the multi-stage counter-flow system described in the above reference, although the requisite amount of water can be greatly reduced, bacteria would grow due to an increase of the retention time of water in the tank, and floating masses of bacteria stick to the light-sensitive material. In the present invention, in order to cope with this problem, the method of reducing calcium and magnesium ion concentrations described in JP-A-62-288838 can be used very effectively. Further, it is also effective to use isothiazolone compounds or thiabendazoles such as described in JP-A-57-8542, chlorine type bactericides, e.g., chlorinated sodium isocyanurate, benzotriazole, and bactericides described in Hiroshi Horiguchi, Bokinbobaizai no kagaku (Antibacterial and Antifungal Chemistry), Eisei Gijutsu Gakkai (ed.), Biseibutsu no mekkin, sakkin, bobigijutsu (Sterilizing and Antifungal Techniques of Microorganisms), and Nippon Bokin Bobi Gakkai (ed.), Bokin bobizai jiten (Antibacterial and Antifungal Agents Handbook) (1986).

The washing water has a pH value of from 4 to 9, preferably from 5 to 8. The temperature of the water and the washing time can be selected from broad ranges depending on the characteristics and end use of the light-sensitive material, but usually ranges from 15 to 45 °C in temperature and from 20 seconds to 10 minutes in time, preferably from 25 to 40 °C in temperature and from 30 seconds to 5 miniutes in time. The light-sensitive material of the invention may be directly processed with a stabilizer in place of the washing step. For the stabilization step, any of the known techniques as described in JP-A-57-8543, JP-A-58-14834, and JP-A-60-220345 can be used.

The aforesaid washing step may be followed by stabilization in some cases. For example, a stabilizing bath containing a dye stabilizer and a surface active agent may be used as a final bath for color light-sensitive materials for picture taking. Examples of such a dye stabilizer include aldehydes such as formaldehyde and glutaraldehyde, N-methylol compounds, hexamethylenetetramine, and aldehyde-sulfurous acid adducts. This stabilizing bath may also contain various chelating agents or bactericides.

The overflow accompanying replenishment of the washing bath and/or stabilizing bath can be reused in other steps such as desilvering.

In the processing using an automatic developing machine, if these processing solutions are concentrated due to evaporation, water may be preferably supplied to the system to make up for the concentration.

The silver halide color light-sensitive material may contain a color developing agent for the purpose of simplifying and expediting processing. Such a color developing agent is preferably used in the form of various precursors. Examples of such precursors include indoaniline compounds as described in U.S. Patent 3,342,597, Schiff's base type compounds as described in U.S. Patent 3,342,599, and Research Disclosure Nos. 14,850 and 15,159, aldol compounds described in Research Disclosure No. 13,924, metal complexes as described in U.S. Patent 3,719,492, and urethane compounds as described in JP-A-53-135628.

The silver halide color light-sensitive material may optionally comprise various 1-phenyl-3-pyrazolidones for the purpose of accelerating color development. Typical examples of such compounds are described in JP-A-56-64339, JP-A-57-144547, and JP-A-58-115438.

In the present invention, the various processing solutions are used at a temperature of 10 °C to 50 °C. The standard temperature is normally from 33 °C to 38 °C. However, a higher temperature can be used to accelerate processing, thereby reducing the processing time. On the contrary, a lower temperature can be used to improve the picture quality or the stability of the processing solutions.

The silver halide photographic material can also be applied to a heat-developable light-sensitive material as described in U.S. Patent 4,500,626, JP-A-60-133449, JP-A-59-218443, and JP-A-61-238056, and European Patent 210,660A2.

The present invention will be further described in the following examples, but the present invention should not be construed as being limited thereto.

EXAMPLE 1

A multilayer color light-sensitive material was prepared as Specimen 101 by coating on a undercoated cellulose triacetate film support various layers having the following compositions.

Composition of light-sensitive layer

The coated amount of silver halide and colloidal silver is represented in g/m^2 as calculated in terms of the amount of silver. The coated amount of coupler, additive and gelatin is represented in g/m^2. The coated amount of sensitizing dye is represented in mol per mol of silver halide contained in the same layer. The symbols indicating additives have the following meanings. The additives having a plurality of effects are represented by a symbol indicating one of the effects.

UV: ultraviolet absorbent; Solv: high boiling organic solvent; ExF: dye; ExS: sensitizing dye; ExC: cyan coupler; ExM: magenta coupler; ExY: yellow coupler; Cpd: additive; H: hardener; W: surface active agent

Specimen 101

1st Layer: Anti-halation layer	Coated Amount
Black colloidal silver	0.15
Gelatin	2.0
ExM-6	0.2
UV-1	0.03
UV-2	0.06
UV-3	0.07
Solv-1	0.3
Solv-2	0.08
ExF-1	0.01
ExF-2	0.01
ExF-3	0.005
Cpd-6	0.001

2nd Layer: Low sensitivity red-sensitive emulsion layer

Silver bromoiodide emulsion (AgI content: 4 mol%; uniform AgI type; grain diameter: 0.4 μm (as calculated in terms of sphere); grain diameter variation coefficient: 30% (as calculated in terms of sphere); tabular grain; diameter/thickness: 3.0)	0.37
Silver bromoiodide emulsion (AgI content: 6 mol%; internal high AgI type with core/shell ratio of 2 : 1; grain diameter: 0.45 μm (as calculated in terms of sphere); grain diameter variation coefficient: 23% (as calculated in terms of sphere); tabular grain; diameter/thickness: 2.0)	0.19
Gelatin	0.8
ExS-1	2.3×10^{-4}
ExS-2	1.4×10^{-4}
ExS-5	2.3×10^{-4}
ExS-7	4.2×10^{-6}
ExC-1	0.12
ExC-13	0.04
ExC-2(D-25)	0.03
ExC-3	0.009

3rd layer: Middle sensitivity red-sensitive emulsion layer

Silver bromoiodide emulsion (AgI content: 6 mol%; internal high AgI type with core/shell ratio of 2 : 1; grain diameter: 0.65 μm (as calculated in terms of sphere); grain diameter variation coefficient: 23% (as calculated in terms of sphere); tabular grain; diameter/thickness: 2.0)	0.65

Gelatin	1.0
ExS-1	2.3×10^{-4}
ExS-2	1.4×10^{-4}
ExS-5	2.3×10^{-4}
ExS-7	4.2×10^{-6}
ExC-1	0.25
ExC-13	0.05
ExC-2(D-25)	0.01
ExC-3	0.10
Solv-2	0.10

4th Layer: High sensitivity red-sensitive emulsion layer

Silver bromoiodide emulsion (AgI content: 9.3 mol%; poly-structural grain with core/intermediate shell ratio of 3 : 4 : 2; AgI content: 24, 0, 6 mol% towards surface; grain diameter: 0.75 μm (as calculated in terms of sphere); grain diameter variation coefficient: 23% (as calculated in terms of sphere); tabular grain; diameter/thickness: 2.5)	0.5
Gelatin	1.4
ExS-1	1.9×10^{-4}
ExS-2	1.2×10^{-4}
ExS-5	1.9×10^{-4}
ExS-7	8.0×10^{-6}
ExC-1	0.08
ExC-4	0.09
Solv-1	0.08

Solv-2	0.20
Cpd-7	4.6×10^{-4}

5th Layer: Interlayer

Gelatin	0.6
Cpd-1	0.1
Polyethyl acrylate latex	0.08
Solv-1	0.08

6th Layer: Low sensitivity green-sensitive emulsion layer

Silver bromoiodide emulsion (AgI content: 4 mol%; uniform AgI type; grain diameter: 0.33 μm (as calculated in terms of sphere); grain diameter variation coefficient: 37% (as calculated in terms of sphere); tabular grain; diameter/thickness ratio: 2.0)	0.24
Gelatin	0.4
ExS-3	1.6×10^{-4}
ExS-4	4.8×10^{-4}
ExS-5	1×10^{-4}
ExM-5	0.10
ExM-14	0.08
ExM-7	0.03
ExY-8	0.01
Solv-1	0.08
Solv-4	0.01

7th Layer: Middle sensitivity green-sensitive emulsion layer

Silver bromoiodide emulsion (AgI content: 4 mol%; uniform AgI type; grain diameter: 0.55 µm (as calculated in terms of sphere); grain diameter variation coefficient: 15% (as calculated in terms of sphere); tabular grain; diameter/thickness ratio: 4.0)	0.36
Gelatin	0.6
ExS-3	2×10^{-4}
ExS-4	7×10^{-4}
ExS-5	1.4×10^{-4}
ExM-5	0.11
ExM-14	0.08
ExM-7	0.04
ExY-8	0.04
Solv-1	0.16
Solv-4	0.01

8th Layer: High sensitivity green-sensitive emulsion layer

Silver bromoiodide emulsion (AgI content: 8.8 mol%; polystructural grain with ratio of amount of silver of 3 : 4 : 2; AgI content: 24, 0, 3 mol% towards surface; grain diameter: 0.75 µm (as calculated in terms of sphere); grain diameter variation coefficient: 23% (as calculated in terms of sphere); diameter/thickness ratio: 1.6)	0.6
Gelatin	0.6
ExS-4	5.2×10^{-4}

ExS-5	1×10^{-4}
ExS-8	0.3×10^{-4}
ExM-5	0.05
ExM-14	0.04
ExM-6	0.03
ExY-8	0.02
ExC-1	0.01
ExC-4	0.01
Solv-1	0.23
Solv-2	0.05
Solv-4	0.01
Cpd-7	1×10^{-4}
Cpd-8	0.01

9th Layer: Interlayer

Gelatin	0.6
Cpd-1	0.04
Polyethyl acrylate latex	0.05
Solv-1	0.02
UV-4	0.03
UV-5	0.04

10th Layer: Donor layer having interimage effect on red-sensitive layer

Silver bromoiodide emulsion (AgI content: 8 mol%; internal high AgI type with core/ shell ratio of 2 : 1; grain diameter: 0.65 μm (as calculated in terms of sphere); grain diameter variation coefficient: 25%	0.72

(as calculated in terms of sphere); tabular grain; diameter/thickness ratio: 2.0)

Silver bromoiodide emulsion (AgI content: 4 mol%; uniform AgI type; grain diameter: 0.4 μm (as calculated in terms of sphere); grain diameter variation coefficient: 30% (as calculated in terms of sphere); tabular grain; diameter/thickness: 3.0)	0.21
Yellow colloidal silver	
Gelatin	1.0
ExS-3	6×10^{-4}
ExM-10(D-10)	0.19
Solv-1	0.30
Solv-6	0.03

11th Layer: Yellow filter layer

Yellow colloidal silver	0.06
Gelatin	0.8
Cpd-2	0.13
Solv-1	0.13
Cpd-1	0.07
Cpd-6	0.002
H-1	0.13

12th Layer: Low sensitivity blue-sensitive emulsion layer

Silver bromoiodide emulsion (AgI content: 4.5 mol%; uniform AgI type; grain diameter: 0.7 μm (as calculated in terms of sphere); grain diameter variation coefficient: 15% (as calculated in terms of sphere); tabular grain; diameter/thickness: 7.0)	0.45

Silver bromoiodide emulsion (AgI content: 3 mol%; uniform AgI type; grain diameter: 0.3 μm (as calculated in terms of sphere); grain diameter variation coefficient: 30% (as calculated in terms of sphere); tabular grain; diameter/thickness: 7.0)	0.25
Gelatin	2.1
ExS-6	9×10^{-4}
ExC-1	0.13
ExC-4	0.03
ExY-9	0.16
ExY-11	0.90
ExY-15	0.15
Solv-1	0.51

13th Layer: Interlayer

Gelatin	0.4
ExY-12	0.20
Solv-1	0.19

14th Layer: High Sensitivity blue-sensitive emulsion layer

Silver bromoiodide emulsion (AgI content: 10 mol%; internal high AgI type; grain diameter: 1.0 μm (as calculated in terms of sphere); grain diameter variation coefficient: 25% (as calculated in terms of sphere); polytwinning tabular grain; diameter/thickness ratio: 2.0)	0.4
Gelatin	0.5
ExS-6	1×10^{-4}
ExY-9	0.01

ExY-11	0.12
ExY-15	0.09
ExC-1	0.01
Solv-1	0.10

15th Layer: 1st protective layer

Emulsion of finely divided silver bromoiodide grains (AgI content: 2 mol%; uniform AgI type; grain diameter: 0.07 µm (as calculated in terms of sphere))	0.12
Gelatin	0.7
UV-4	0.11
UV-5	0.16
Solv-5	0.02
H-1	0.13
Cpd-5	0.10
Polyethyl acrylate latex	0.09

16th layer: 2nd protective layer

Emulsion of finely divided silver bromoiodide grains (AgI content: 2 mol%; uniform AgI type; grain diameter: 0.07 µm (as calculated in terms of sphere))	0.36
Gelatin	0.85
Polymethyl methacrylate grains	0.2
(diameter: 1.5 µm)	
Cpd-4	0.04
W-4	0.02

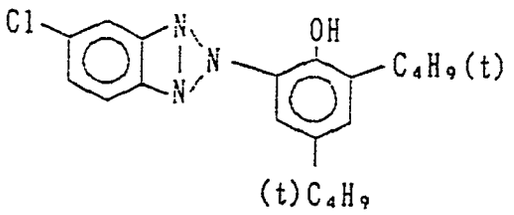
H-1	0.17

In addition to the above mentioned components, an emulsion stabilizer Cpd-3 (0.07 g/m^2), and surface

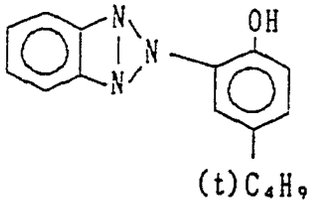
active agents W-1 (0.006 g/m^2), W-2 (0.16 g/m^2), W-3 (0.10 g/m^2) and W-5 (0.10 g/m^2) were added to each of these layers as a coating aid or an emulsion dispersant.

The structural formulae of the compounds incorporated in these layers are set forth below:

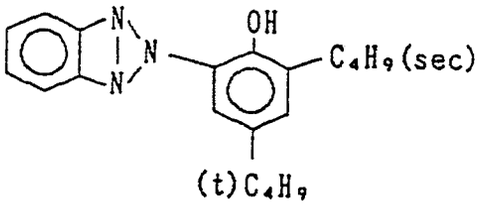
UV－1



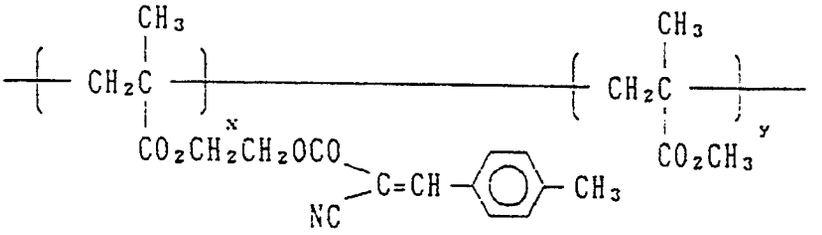
UV－2



UV－3

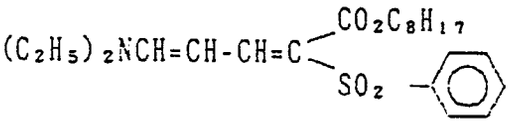


UV－4



x：y＝70：30（wt%）

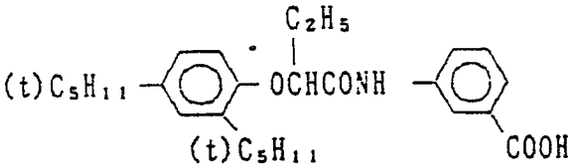
UV－5



Solv-1: Tricresyl phosphate

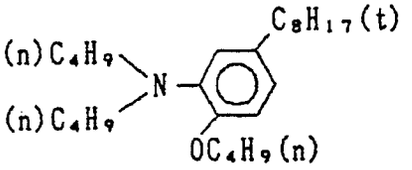
Solv-2: Dibutyl phthalate

Solv-4



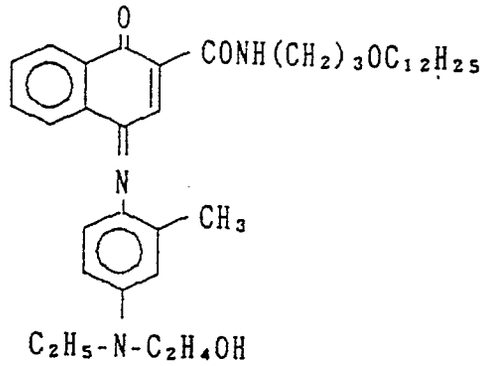
Solv-5: Trihexyl phosphate

Solv-6

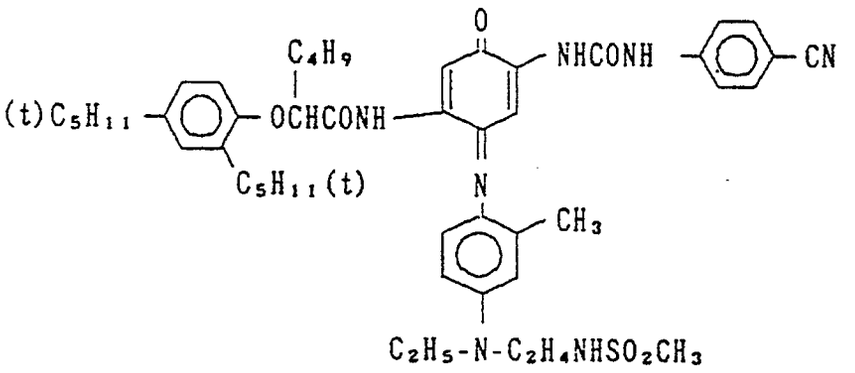


ExF-1

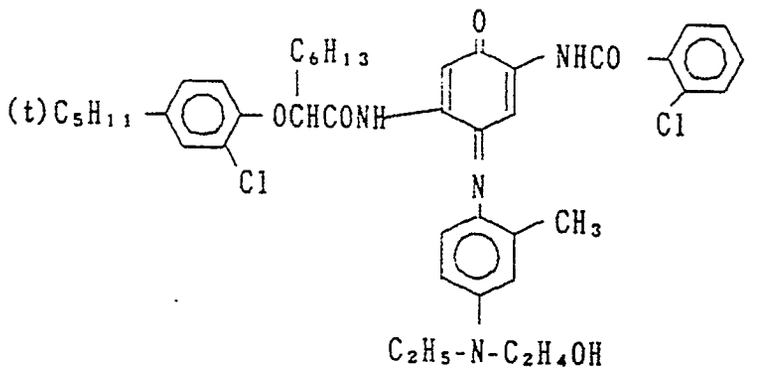
1 : 1 mixture (by weight) of:



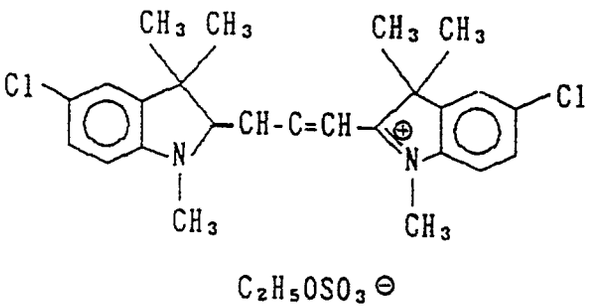
and



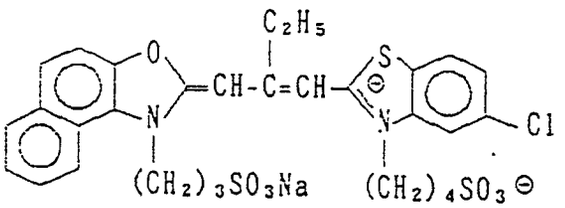
Ex F - 2



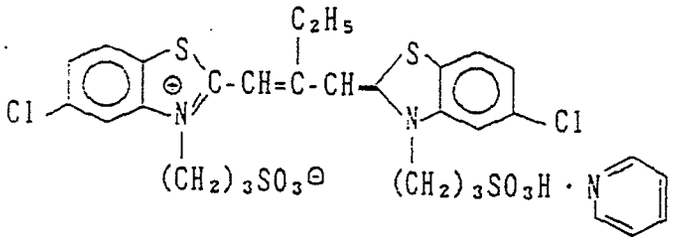
Ex F - 3



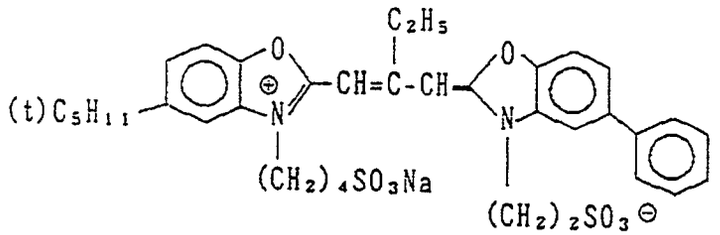
Ex S - 1



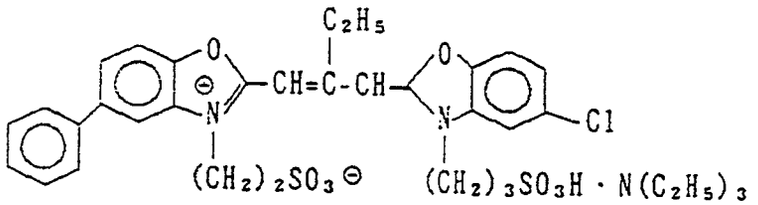
Ex S - 2



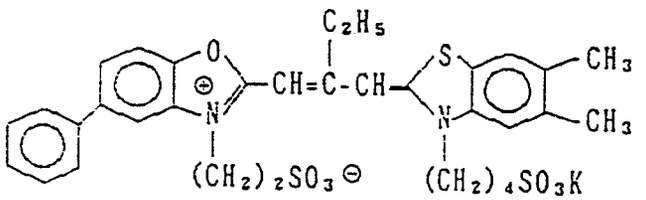
Ex S - 3



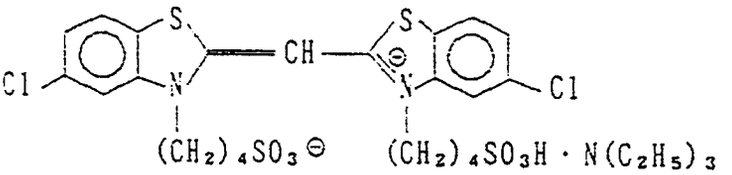
Ex S - 4



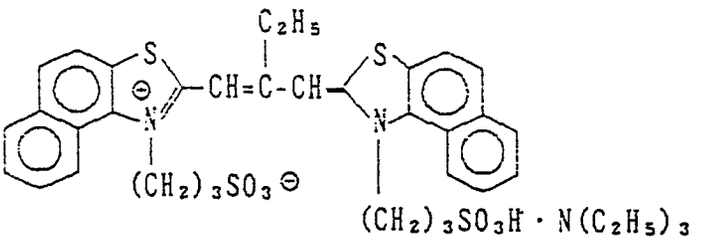
Ex S - 5



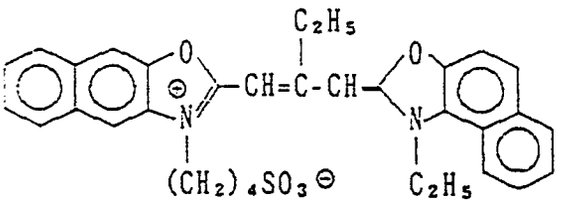
Ex S - 6



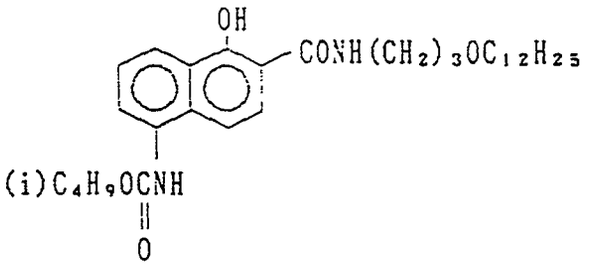
Ex S - 7



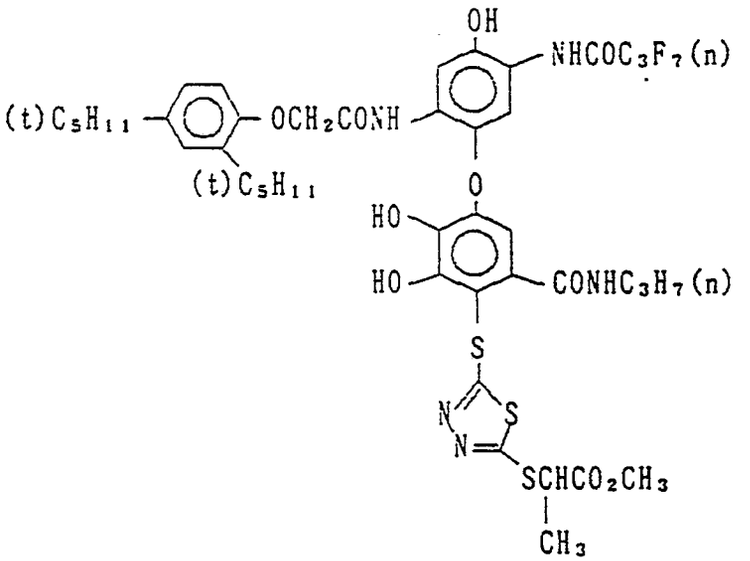
Ex S - 8



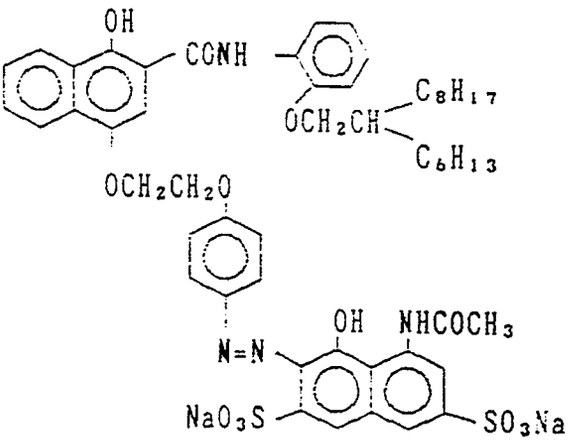
Ex C－1



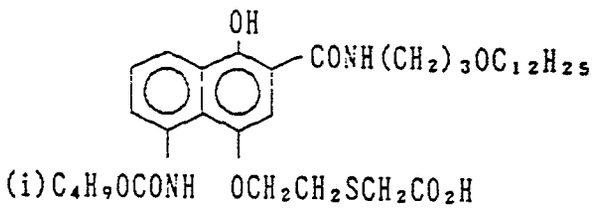
Ex C－2 (D－25)



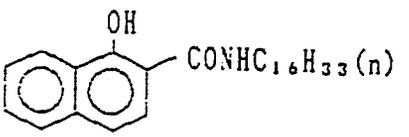
Ex C - 3



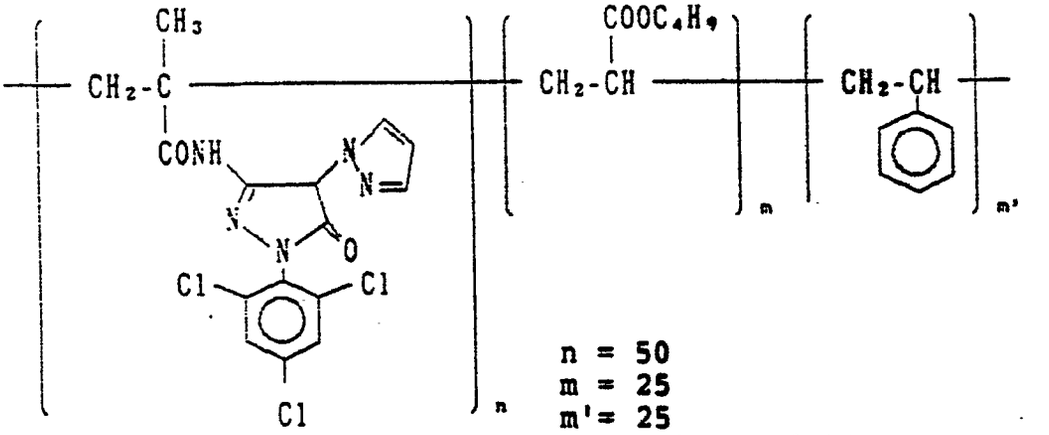
Ex C - 4



Ex C - 13

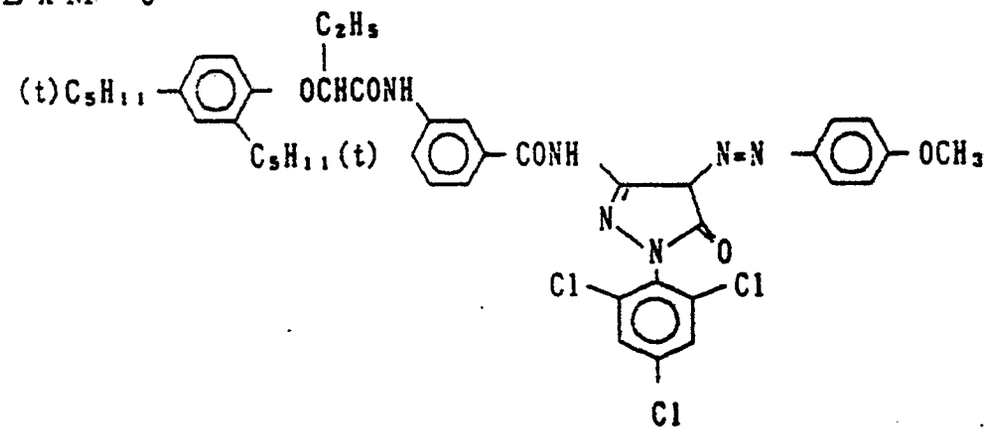


Ex M − 5

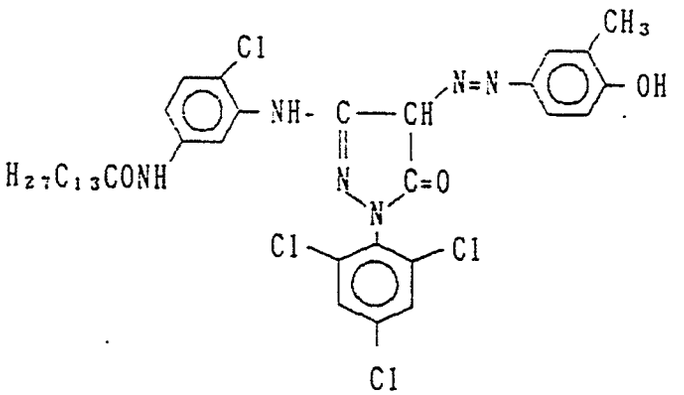


n = 50
m = 25
m'= 25
Molecular weight: approx. 20,000

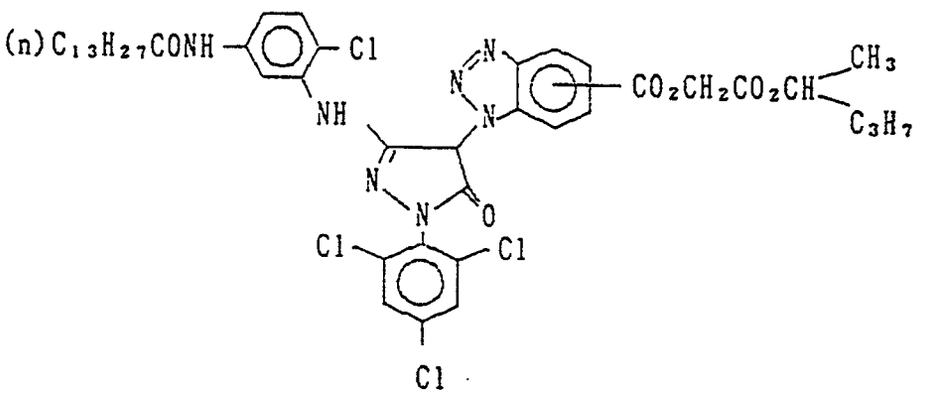
Ex M − 6



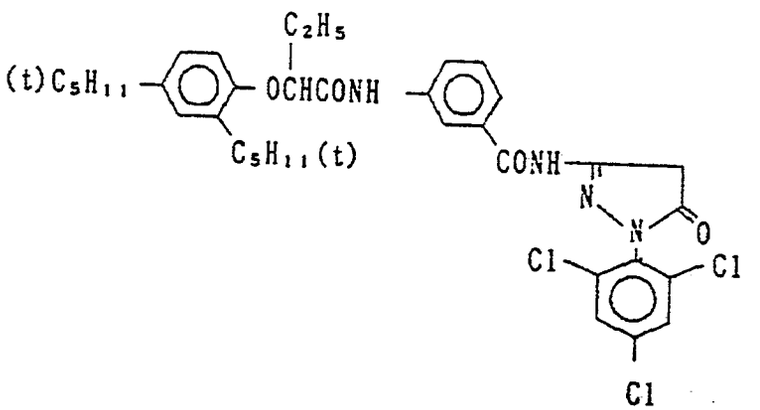
Ex M－7



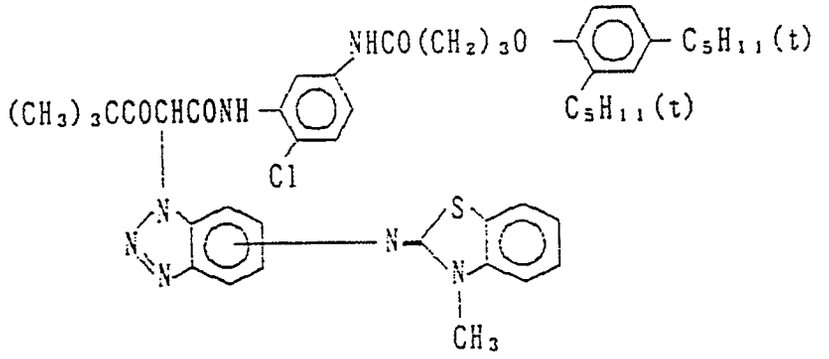
Ex M－10 (D－10)



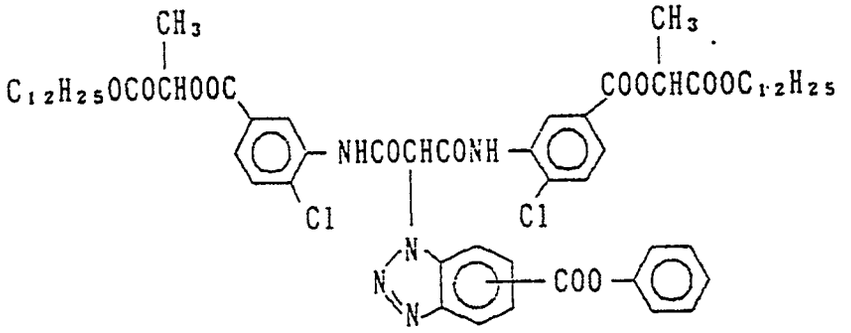
Ex M－14



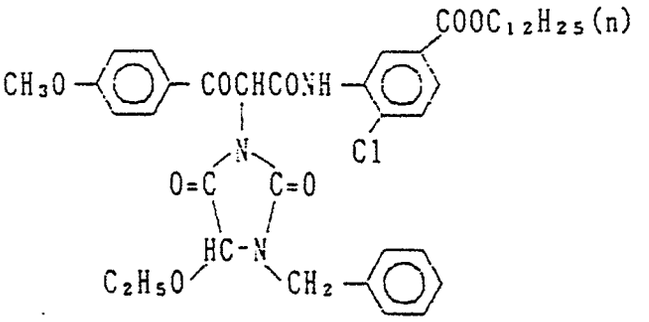
Ex Y－8



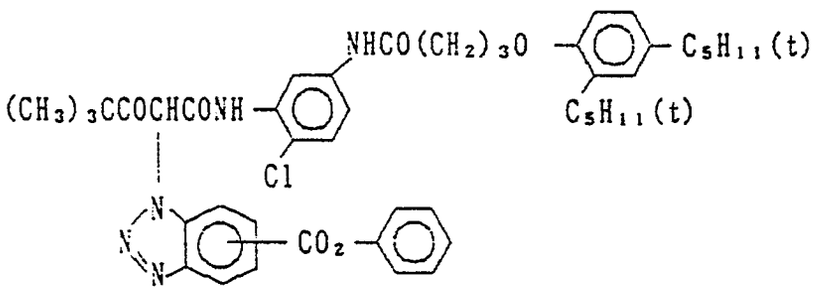
Ex Y－9



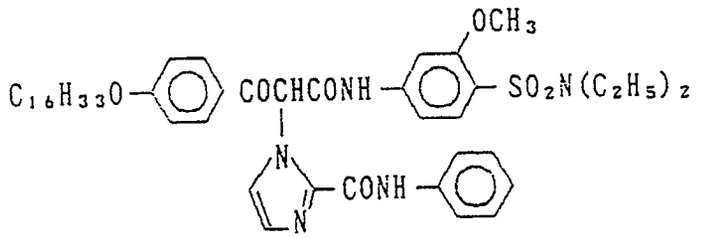
Ex Y-11



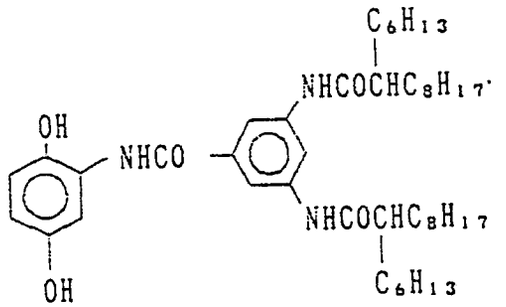
Ex Y-12



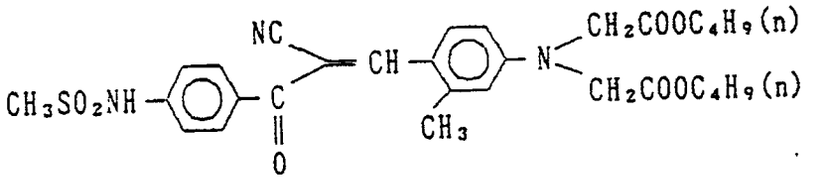
Ex Y-15



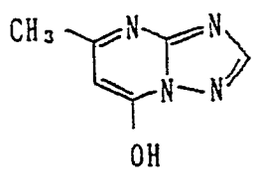
Cpd-1



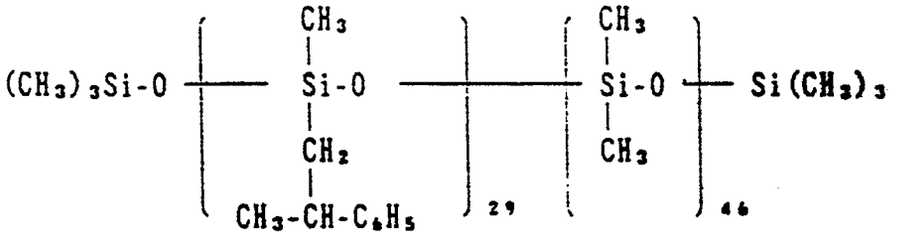
Cpd-2



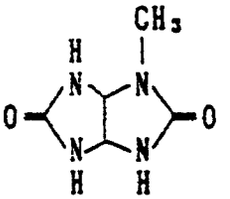
Cpd-3



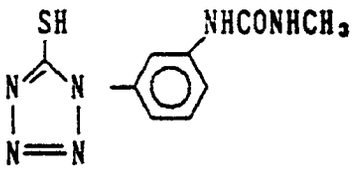
Cpd-4



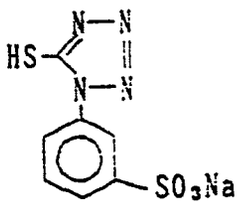
Cpd-5



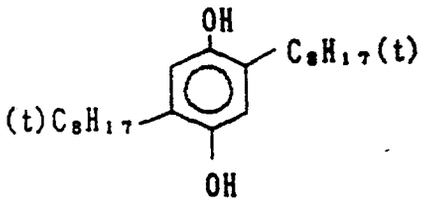
Cpd-6



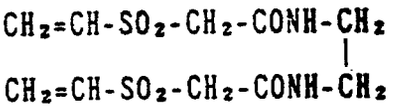
Cpd-7



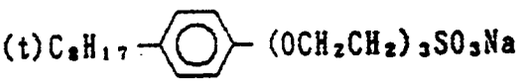
Cpd-8



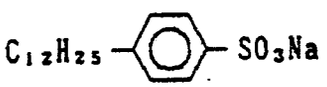
H-1



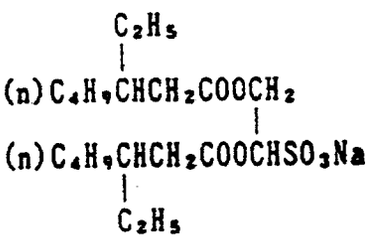
W-1



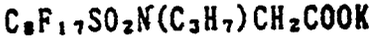
W-2



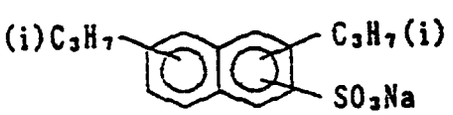
W-3



W-4



W-5



Various specimens were then prepared as follows:

Specimen 102

Specimen 102 was prepared in the same manner as in Specimen 101, except that the present yellow-

colored cyan coupler (Exemplary Compound (YC-5)) was incorporated in the 2nd, 3rd and 4th layers in the amounts of 0.04, 0.06 and 0.02 g/m^2, respectively.

Specimen 103

Specimen 103 was prepared in the same manner as in Specimen 101, except that ExM-14 incorporated in the 6th, 7th and 8th layers was replaced by Exemplary Compound [A-4]-46 in equimolecular amounts.

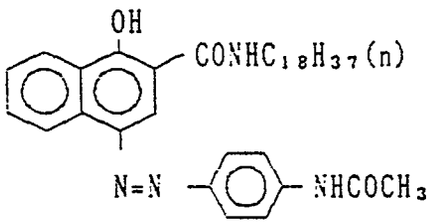
Specimen 104

Specimen 104 was prepared in the same manner as in Specimen 101, except that the yellow-colored cyan coupler used in Specimen 102 was incorporated in the 2nd, 3rd and 4th layers and Coupler [A-4]-46 of Specimen 103 was incorporated in the 6th, 7th and 8th layers.

Specimen 105

Specimen 105 was prepared in the same manner as in Specimen 104, except that the yellow-colored cyan coupler (YC-5) used in the 2nd, 3rd and 4th layers of Specimen 104 was replaced by Exemplary Compound C-2 as described in JP-A-61-221748 in equimolar amounts.

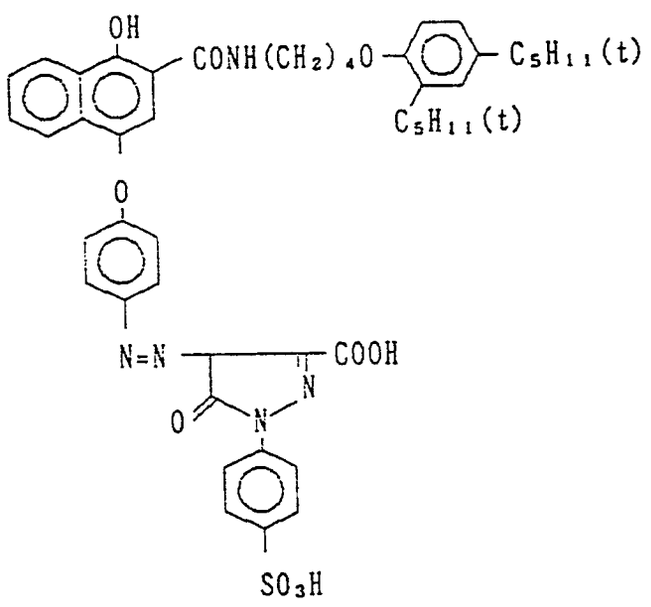
Exemplary Compound C-2 as described in JP-A-61-221748:



Specimen 106

Specimen 106 was prepared in the same manner as in Specimen 104, except that the yellow-colored cyan coupler (YC-5) used in the 2nd, 3rd and 4th layers in Specimen 104 was replaced by Exemplary Compound II-3 as described in JP-A-1-319744 in equimolar amounts.

Exemplary Compound II-3 described in JP-A-1-319744:



Specimens 107 and 108

Specimens 107 an 108 were prepared in the same manner as in Specimen 104, except the yellow-colored cyan coupler (YC-5) incorporated in the 2nd, 3rd and 4th layers in Specimen 104 was replaced by Couplers (YC-1) and (YC-10) in equimolar amounts, respectively, and Coupler [A-4]-46 incorporated in the 6th, 7th and 8th layers in Specimen 104 was replaced by Couplers [A-4]-24 and [A-4]-41 in equimolar amounts, respectively.

Specimens 109, 110 and 111

Specimens 109, 110 and 111 were prepared in the same manner as in Specimen 104, except that the yellow-colored cyan coupler (YC-5) incorporated in the 2nd, 3rd and 4th layers in Specimen 104 was replaced by Couplers (YC-37), (YC-32) and (YC-47) in equimolar amounts, respectively.

Specimen 112

Specimen 112 was prepared in the same manner as in Specimen 104, except that the yellow colored cyan coupler (YC-5) incorporated in the 2nd, 3rd and 4th layers in Specimen 104 was replaced by a 1 : 1 mixture (molar ratio) of Couplers (YC-3) and (YC-16) in equimolar amounts, and the coupler [A-4]-46 incorporated in the 6th, 7th and 8th layers in Specimen 104 was replaced by a 2 : 1 mixture (molar ratio) of Couplers [A-4]-39 and [A-3]-14 in equimolar amounts.

Specimen 113

Specimen 113 was prepared in the same manner as in Specimen 104, except that the yellow-colored cyan coupler (YC-5) incorporated in the 2nd, 3rd and 4th layers in Specimen 104 was replaced by Couplers (YC-32), (YC-13) and (YC-5) in equimolar amounts, respectively, and the coupler [A-4]-46 incorporated in the 6th, 7th and 8th layers in Specimen 104 was replaced by Couplers [A-4]-1, [A-4]-9 and [A-4]-51 in equimolar amounts, respectively.

The specimens thus prepared and the couplers incorporated therein are tabulated below.

Table 1

Specimen No.	Yellow-colored cyan coupler	Coupler [A]
101 (comparative)	---	---
102 (")	2nd-4th layers (YC-5)	---
103 (")	---	6th-8th layers (A-4)-46
104(present invention)	2nd-4th layers (YC-5)	6th-8th layers (A-4)-46
105 (comparative)	2nd-4th layers (C-2) (JP-A-61-221748)	6th-8th layers (A-4)-46
106 (present invention)	2nd-4th layers (YC-55) (II-3, JP-A-1-319744)	6th-8th layers (A-4)-46
107 (")	2nd-4th layers (YC-1)	6th-8th layers (A-4)-24
108 (")	2nd-4th layers (YC-10)	6th-8th layers (A-4)-41
109 (")	2nd-4th layers (YC-37)	6th-8th layers (A-4)-46
110	2nd-4th layers (YC-32)	6th-8th layers (A-4)-46
111	2nd-4th layers (YC-47)	6th-8th layers (A-4)-46
112 (")	2d-4th layers (YC/3)/ (YC-16)=1/1	6th-8th layers (A-4)-39/ (A-3)-14=2/1

Table 1 (cont'd)

Specimen No.	Yellow-colored cyan coupler	Coupler (A)
113 (present invention)	2nd layer (YC-32)	6th layer (A-4)-1
	3rd layer (YC-13)	7th layer (A-4)-9
	4th layer (YC-5)	8th layer (A-4)-51

These specimens were cut into 35-mm wide strips, subjected to the following tests and exposure, subjected

to the following processings, and then evaluated for properties from their characteristic curves. The specimens thus prepared were stored at a temperature of 25 °C and a relative humidity of 55% for 10 days before the tests.

(1) These specimens were wedgewise exposed to light through a red separation filter (1/100 sec., 20CMS), and then processed. The density measured by blue light was determined at the exposure which gives a density of (minimum density + 1.0) on the characteristic curve obtained by the measurement by red light. Color stain (ΔD_{γ}) was obtained by subtracting the minimum density by blue light from this value. The smaller this value is, the smaller is color stain and the more advantageous is the color reproduction.

(2) These specimens were wedgewise exposed to white light (color temperature of light source: 4,800 °K), and then processed. The logarithm of the reciprocal of the exposure which gives a density of (minimum density + 0.2) was determined as sensitivity on the characteristic curves obtained by the density measurement by red light and green light. The difference from the value of Specimen 101 as reference value was then determined (ΔS_R, ΔS_G). The greater these values are, the higher is sensitivity.

(3) Two batches of these specimens were stored at a temperature of 50 °C and a relative humidity of 40% for 7 days and at a temperature of 5 °C and a relative humidity of 55% for 7 days, respectively. These specimens were then wedgewise exposed to white light. These specimens were processed at the same time. The logarithm of the reciprocal of the exposure which gives a density of (minimum density + 0.2) was determined on the characteristic curves obtained by the measurement by red light and green light. The difference from the value obtained from the specimens which had been stored at a temperature of 5 °C and a relative humidity of 55% for 7 days was determined (ΔT_R, ΔT_G). The smaller this value is, the better is the preservability of the specimen.

(4) Two batches of these specimens were wedgewise exposed to white light, stored at a temperature of 50 °C and a relative humidity of 30% for 7 days and at a temperature of 5 °C and a relative humidity of 55% for 7 days, respectively, processed at the same time, and then measured by red light and green light to obtain the respective characteristic curves. The logarithm of the reciprocal of the exposure which gives a density of (minimum density + 0.2) on these characteristic curves were determined. The difference from the value obtained from the specimens which had been stored at a temperature of 5 °C and a relative humidity of 55% for 7 days was determined (ΔL_R, ΔL_G).

The results of these tests are set forth in Table 2.

Table 2

Specimen No.	Color stain	Sensitivity		Preservability of light-sensitive material		Latent image stability	
	ΔD_Y	ΔS_R	ΔS_G	ΔT_R	ΔT_G	ΔL_R	ΔL_G
101 (comparative)	0.14	(reference)		0.07	0.06	0.05	0.08
102 (")	0.03	0.04	0.00	0.06	0.06	0.04	0.08
103 (")	0.13	0.00	0.06	0.07	0.05	0.05	0.07
104 (present invention)	0.02	0.05	0.07	0.02	0.01	0.00	0.02
105 (comparative)	0.09	0.00	0.07	0.10	0.05	0.07	0.07
106 (present invention)	0.07	0.02	0.07	0.06	0.05	0.04	0.05
107 (present invention)	0.02	0.05	0.06	0.02	0.02	0.00	0.02
108 (")	0.02	0.05	0.07	0.02	0.01	0.00	0.00
109 (")	0.04	0.04	0.07	0.03	0.01	0.02	0.02
110	0.04	0.04	0.07	0.03	0.01	0.02	0.02
111	0.07	0.02	0.07	0.06	0.05	0.04	0.05
112 (")	0.02	0.05	0.06	0.02	0.02	0.00	0.03
113 (")	0.01	0.06	0.08	0.00	0.02	0.00	0.01

Development was effected in accordance with the following steps and the following processing solutions by means of an automatic processing machine for motion picture film. Specimen 101 which had been imagewise exposed to light was processed until the accumulated replenishment of the color developer reached three times the capacity of the running solution tank before the other specimens were processed.

Processing step

Step	Time	Temperature	Replenishment rate*	Tank capacity
Color development	3 min. 00 sec.	37.5 °C	20 ml	10 ℓ
Bleach	40 sec.	38.0°C	4.5 ml	5 ℓ
Blix	40 sec.	38.0°C	-	5 ℓ
Fixing	40 sec.	38.0°C	30 ml	5 ℓ
Washing (1)	30 sec.	38.0°C	-	5 ℓ
Washing (2)	30 sec.	38.0°C	30 ml	5 ℓ
Stabilization	30 sec.	38.0°C	20 ml	5 ℓ
Drying	1 min.	55°C		

* Determined per 35-mm width and 1-m length

The washing step was effected in a counter-current process wherein the washing water flows backward. The overflow from the washing tank (1) was all introduced into the fixing bath.

The overflow solution from the bleaching bath and the fixing bath were all introduced into the blix bath.

The amount of the developer brought over to the bleaching step, and the amount of the fixing solution brought over to the washing step were 2.5 ml and 2.0 ml per m of 35-mm wide light-sensitive material, respectively. The time for crossover was 5 seconds in all the steps. This crossover time is included in the processing time of the previous step.

The bleaching bath, blix bath and fixing bath each had an opening ratio of 0.02.

The agitation in the automatic developing machine was accomplished by means of a magnet pump available from Iwaki K.K. Specifically, the processing solution was jetted through a nozzle having a diameter of 1.2 mm from the outer side of the rack to the inner side of the rack to collide with the emulsion surface of the light-sensitive material at a distance of about 10 mm.

The size, flow rate and number of nozzles of the pumps used at these baths are set forth below.

Step	Pump	Flow rate (ℓ/min.)	Number of nozzles
Color development	MD-20	15	54
Bleach	MD-20	15	54
Fixing	MD-20	15	54
Washing(1)	MD-10	8	36
Washing(2)	MD-10	8	36
Stabilizing	MD-10	8	36

At the various baths, the evaporation loss was made up for by the replenishment with water every day.

At the bleaching bath, the bleaching solution was aerated only during the processing of the specimens.

The various processing solutions had the following compositions:

Developer

	Running Solution	Replenisher
Diethylenetriamine-pentaacetic acid	2.0 g	2.0 g
1-Hydroxyethylidene-1,1-diphosphonic acid	3.0 g	3.2 g
Sodium sulfite	4.0 g	5.8 g
Potassium carbonate	40.0 g	40.0 g
Potassium bromide	1.3 g	0.4 g
Potassium iodide	1.5 mg	--
Hydroxylamine sulfate	2.4 g	3.6 g
2-Methyl-4-[N-ethyl-N-(β-hydroxyethyl)amino] aniline sulfate	4.5 g	6.4 g
Water to make	1.0 ℓ	1.0 ℓ
pH (adjusted with 50% potassium hydroxide)	10.05	10.15

Bleaching solution

	Running Solution	Replenisher
Ferric ammonium 1,3-propylenediamine-tetraacetate monohydrate	110 g	220 g
Ammonium bromide	70 g	140 g
Ammonium nitrate	20 g	40 g
Acetic acid	30 g	60 g
Hydroxyacetic acid	60 g	120 g
Water to make	1,000 ml	1,000 ml
pH (adjusted with 27% aqueous ammonia)	3.8	2.5

Fixing solution

	Running Solution	Replenisher
Diammonium ethylene-diaminetetraacetate	18 g	54 g
Ammonium sulfite	20.0 g	60 g
Aqueous solution of ammonium thiosulfate (700 g/ℓ)	280.0 ml	840 ml
Imidazole	25 g	75 g
Water to make	1.0 ℓ	1.0 ℓ
pH	7.4	7.45

Blix solution (running solution)

A 1 : 6 mixture of the above mentioned bleaching solution and fixing solution was used as the running solution. The overflow solution from the above mentioned bleaching bath and fixing bath were all introduced into the blix bath.

Washing solution (the running solution was used also as replenisher)

Tap water was passed through a mixed bed column packed with an H-type strongly acidic cation exchange resin (Amberlite IR-120B available from Rohm & Haas) and an OH-type strongly basic anion exchange resin (Amberlite IRA-400 available from the same company) so that the calcium and magnesium ion concentrations were each reduced to 3 mg/ℓ or less. Dichlorinated sodium isocyanurate and sodium

sulfate were then added to the solution in amounts of 20 mg/ℓ and 150 mg/ℓ, respectively. The washing solution thus obtained had a pH value of 6.5 to 7.5.

Stabilizing Solution

37% Formaldehyde	1.2 ml
Surface active agent $[C_{10}H_{21}-O-(CH_2CH_2O)_{10}-H]$	0.4 g
Disodium ethylenediaminetetraacetate dihydrate	0.05 g
Water to make	1 ℓ
pH	5.0 to 7.0

The processing was effected with the above mentioned processing solutions under the above mentioned conditions. In order to make up for the evaporation loss, the same water replenisher as used for the washing solution was used.

Table 2 above shows that the use of the yellow-colored cyan couplers of the present invention provides improved inhibition of color stain in cyan images. It is also shown that the yellow-colored cyan couplers of the present invention provide better results than do the comparative couplers. It is further shown that the use of the yellow-colored cyan couplers provides a high sensitivity and remarkable improvements in the preservability of the light-sensitive material and the stability of latent images as compared to the use of the comparative yellow-colored cyan couplers.

Further, it is shown that the use of the couplers represented by the general formula (A) provides a high sensitivity and improvements in preservability of light-sensitive material and stability of latent images.

Surprisingly, the combination of the yellow-colored cyan coupler of the present invention and the coupler represented by the general formula (A) provides better improvements in preservability of light-sensitive material and stability of latent images over the separate use of these couplers.

Among the yellow-colored cyan couplers of the present invention, the couplers which release 6-hydroxy-2-pyridon-5-ylazo group are best in the the properties tested in the present example. The couplers which release 2-acylaminophenylazo group and 2-sulfonamidephenylazo group are ranked next. The couplers which release pyrazolon-4-ylazo group are ranked next.

EXAMPLE 2

A multilayer color light-sensitive material Specimen 201 was prepared by coating on an undercoated cellulose triacetate film support the following layers.

(Composition of light-sensitive material)

The figures indicate the coated amount of each component in g/m^2. The coated amount of silver halide and colloidal silver are represented in terms of the amount of silver. The coated amount of sensitizing dye is represented in mol amount per mol of silver halide contained in the same layer.

1st layer: Anti-halation layer	
Black colloidal silver	0.20
Gelatin	1.50
UV-1	0.20
CC-1	0.05
CM-1	0.05
Solv-1	0.20
2nd layer: Interlayer	
Gelatin	1.50

UV-1	0.01
Solv-1	0.01

3rd layer: Low sensitivity red-sensitive emulsion layer

Monodisperse emulsion of silver bromoiodide (mean silver bromoiodide content: 7.5 mol%; mean grain diameter: 0.46 μm)	1.00
Monodisperse emulsion of silver bromoiodide (mean silver bromoiodide content: 2.0 mol%; mean grain diameter: 0.32 μm)	0.50
Gelatin	1.50
ExS-1	2.5×10^{-4}
ExS-2	2.5×10^{-4}
ExS-3	5.0×10^{-5}
ExC-1	1.00
ExC-4	0.05
CC-1	0.05
ExD-1	0.002
Solv-1	0.50

4th layer: High sensitivity red-sensitive emulsion layer

Monodisperse emulsion of silver bromoiodide (mean silver bromoiodide content: 6.0 mol%; mean grain diameter: 0.78 μm)	2.00
Gelatin	1.50
ExS-1	2.0×10^{-4}
ExS-2	2.0×10^{-4}
ExS-3	1.0×10^{-5}

ExC-2	0.015
ExC-3	0.25
CC-1	0.015
ExD-2	0.05
Solv-1	0.50

5th layer: Interlayer

Gelatin	0.50

6th layer: Low sensitivity green-sensitive emulsion layer

Monodisperse emulsion of silver bromoiodide (mean silver bromoiodide content: 7.5 mol%; mean grain diameter: 0.46 μm)	1.00
Gelatin	1.80
ExS-4	5.0×10^{-4}
ExS-5	1.0×10^{-4}
ExM-1	0.56
CM-1	0.05
ExD-3	0.015
ExD-4	0.02
Solv-2	0.65

7th layer: Interlayer

Gelatin	0.80
Solv-1	0.20

8th layer: High sensitivity green-sensitive emulsion layer

Monodisperse emulsion of silver bromoiodide (mean silver	1.30

bromoiodide content: 6.0 mol%; mean grain diameter: 0.78 μm)	
Gelatin	1.00
ExS-6	1.5×10^{-4}
ExS-7	2.5×10^{-4}
ExS-8	5.0×10^{-5}
ExM-2	0.05
ExM-3	0.15
CM-2	0.05
ExD-3	0.01
Solv-3	0.50

9th layer: Yellow filter layer

Yellow colloidal silver	0.10
Gelatin	0.80
Cpd-1	0.10
Solv-3	0.10

10th layer: Low sensitivity blue-sensitive emulsion layer

Monodisperse emulsion of silver bromoiodide (mean silver bromoiodide content: 7.5 mol%; mean grain diameter: 0.46 μm)	0.25
Monodisperse emulsion of silver bromoiodide (mean silver bromoiodide content: 2.0 mol%; mean grain diameter: 0.32 μm)	0.25
Gelatin	1.00
ExS-10	7.0×10^{-4}
ExY-1	0.50

ExY-2	0.10
ExD-2	0.01
Solv-3	0.15

11th layer: High sensitivity blue-sensitive emulsion layer

Monodisperse emulsion of silver bromoiodide (mean silver bromoiodide content: 8.0 mol%; mean grain diameter: 0.95 μm)	0.50
Monodisperse emulsion of silver bromoiodide (mean silver bromoiodide content: 7.5 mol%; mean grain diameter: 0.46 μm)	0.20
Gelatin	1.10
ExS-9	1.0×10^{-4}
ExS-10	3.0×10^{-4}
ExY-1	0.30
ExY-2	0.05
Solv-3	0.07

12th layer: 1st protective layer

Monodisperse emulsion of silver bromoiodide (mean silver bromoiodide content: 2.0 mol%; "mean grain diameter: 0.08 μm)	0.40
Gelatin	1.00
UV-1	0.10
UV-2	0.05
Cpd-2	0.50
Cpd-3	0.20
Solv-1	0.10

Solv-4	0.10

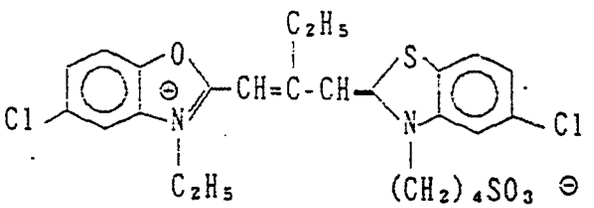
13th layer: 2nd protective layer

Gelatin	0.60
Alkali-soluble matting agent (average grain diameter: 2 μm)	0.10
Lubricant	0.04
ExF-1	0.005
ExF-2	0.01
W-1	0.005

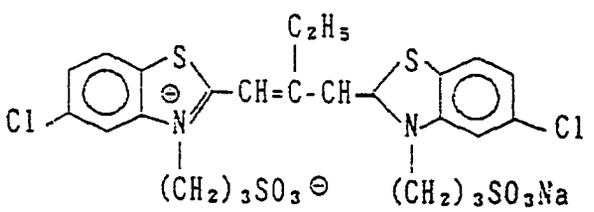
In addition to these compounds, a coating aid W-2, a dispersion aid W-3, film hardeners H-1 and H-2, an antiseptic agent Cpd-4, a stabilizer Cpd-5, and fog inhibitors Cpd-6 and Cpd-7 were added to each of these layers.

The chemical structures of these compounds are set forth below.

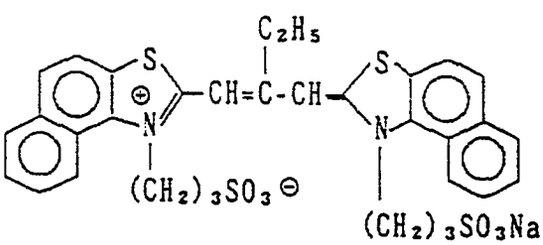
ExS-1



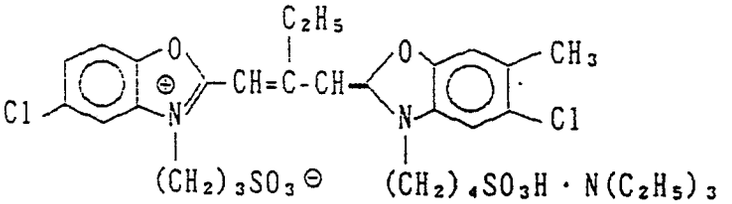
ExS-2



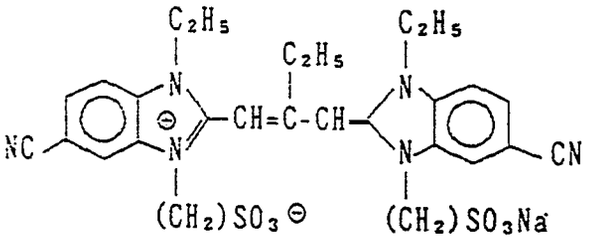
ExS-3



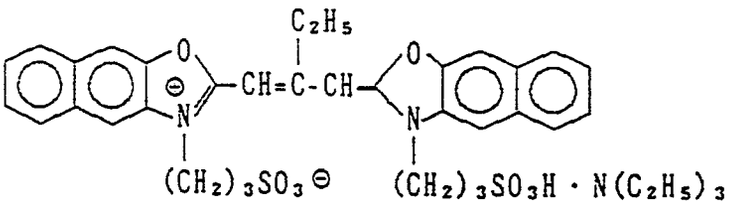
Ex S－4



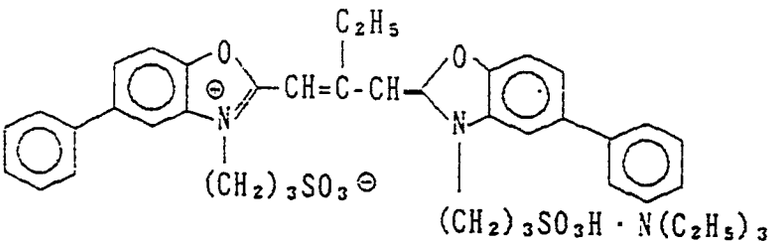
Ex S－5



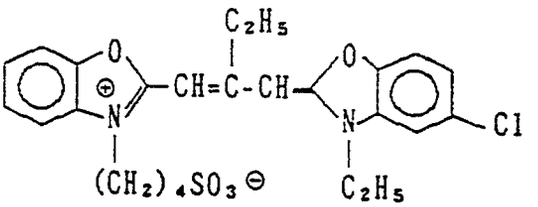
Ex S－6



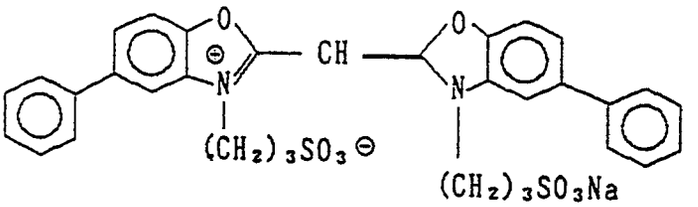
Ex S－7



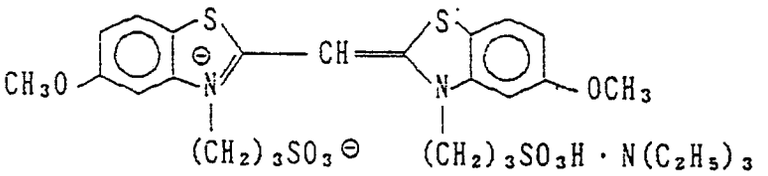
Ex S－8



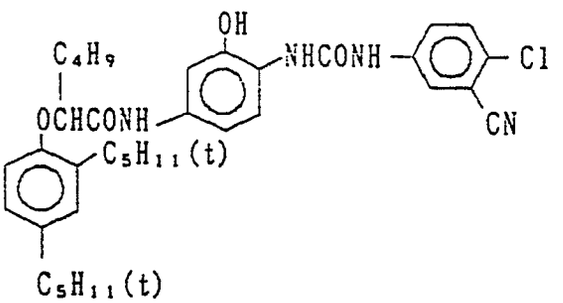
Ex S－9



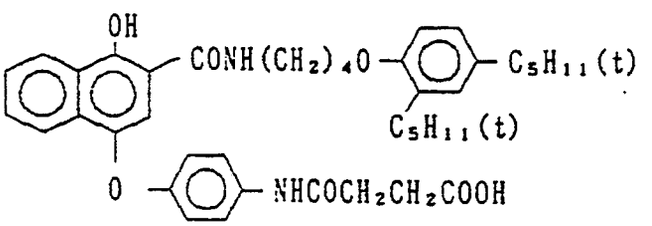
Ex S－10



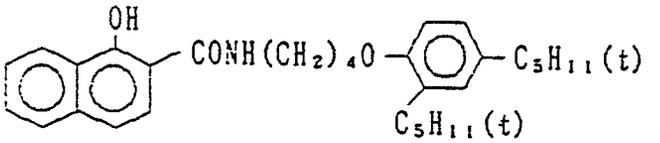
Ex C－1



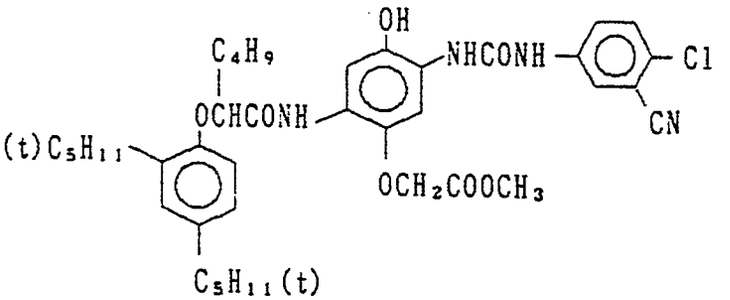
Ex C－2



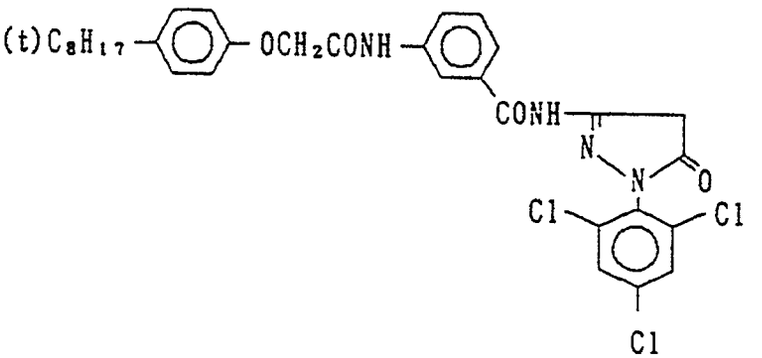
Ex C－3



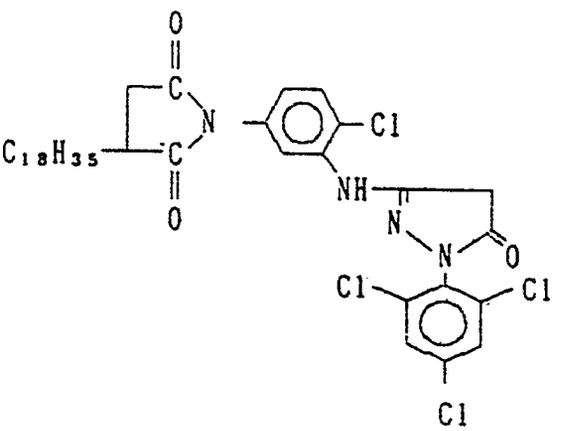
Ex C－4



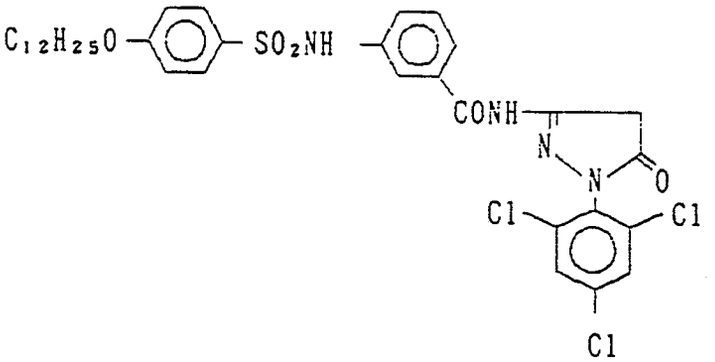
Ex M－1



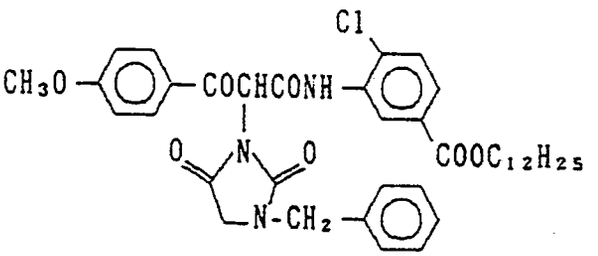
Ex M－2



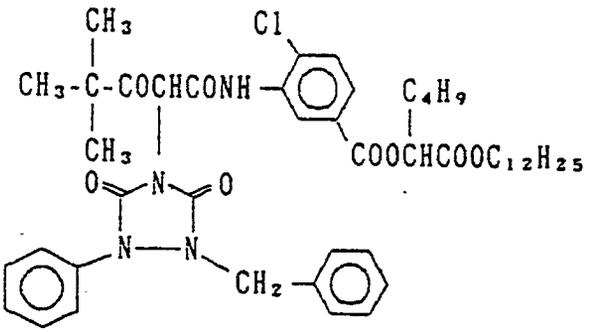
Ex M-3



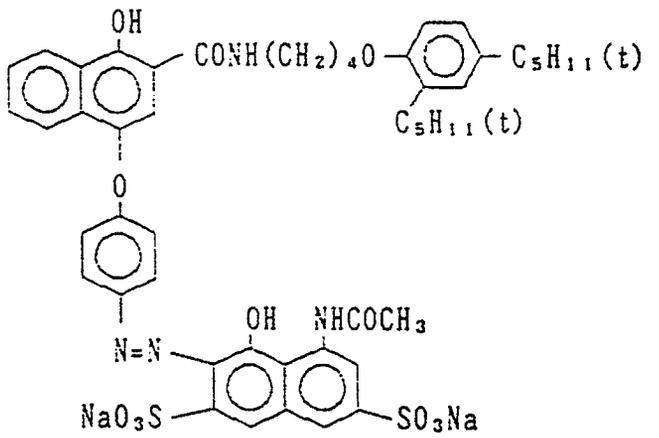
Ex Y-1



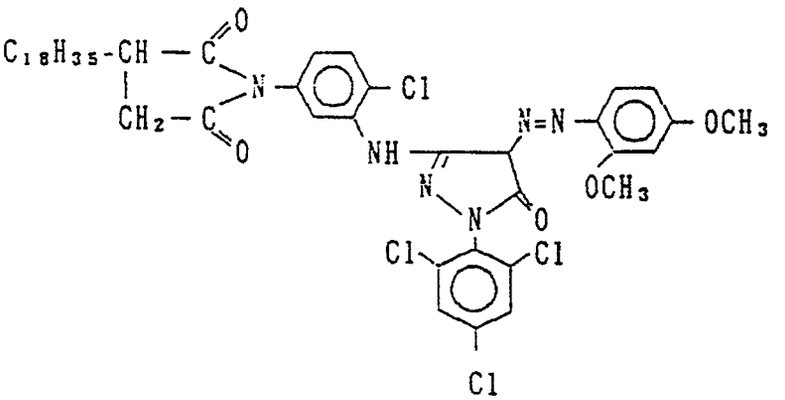
Ex Y-2



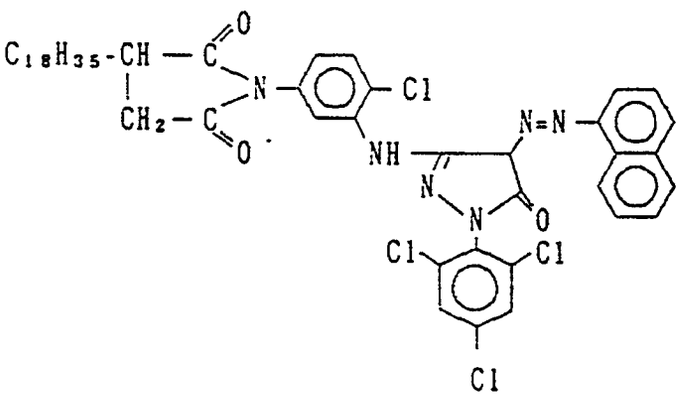
CC－1



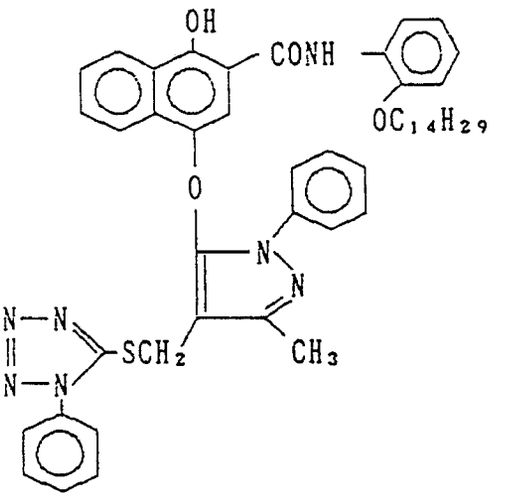
CM－1



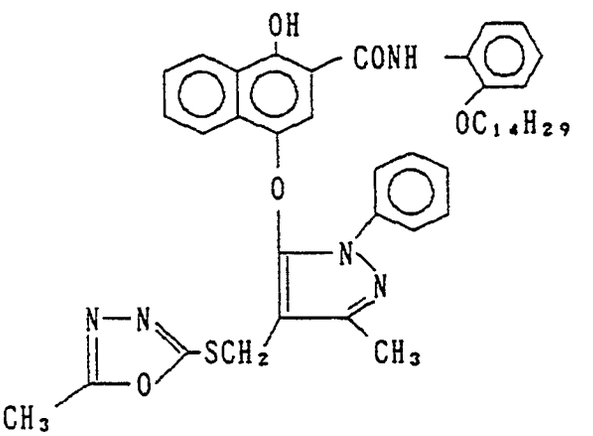
CM－2



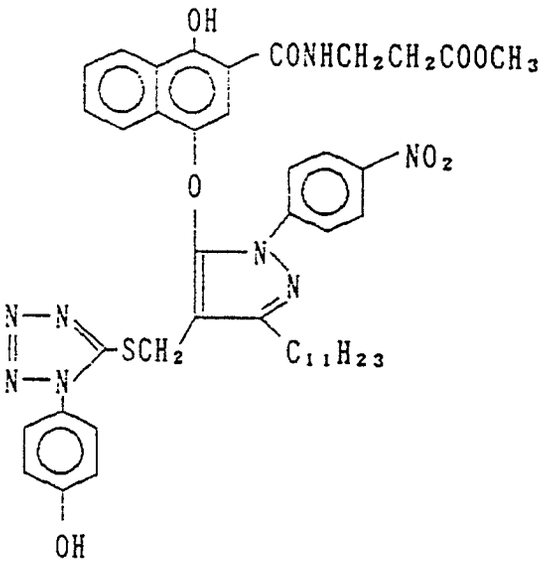
Ex D-1


OH
CONH
OC14H29
O
N
N
N—N
N—N
SCH2
CH3

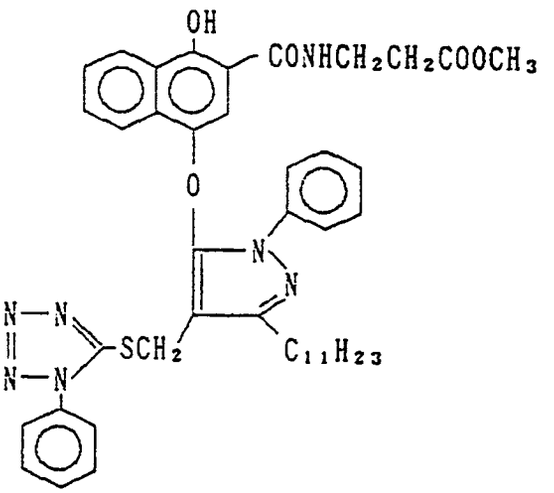
Ex D-2


OH
CONH
OC14H29
O
N
N
N—N
O
SCH2
CH3
CH3

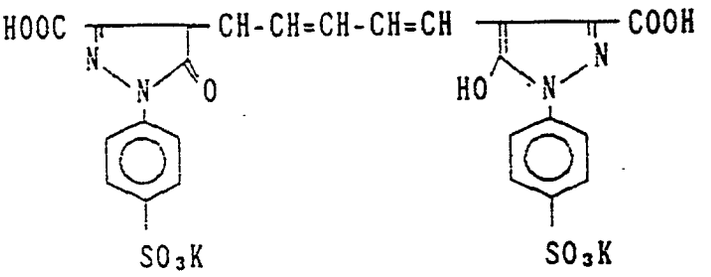
Ex D－3



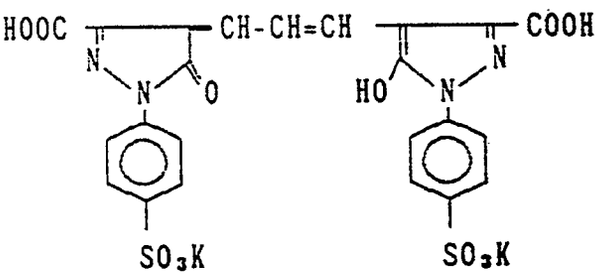
Ex D－4



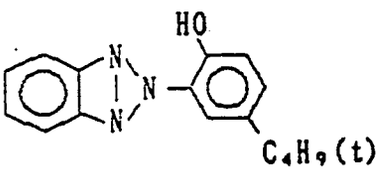
E x F − 1



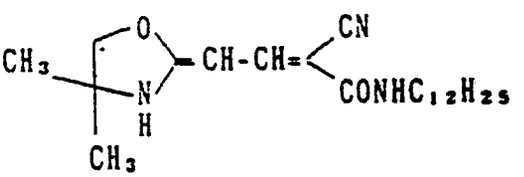
E x F − 2



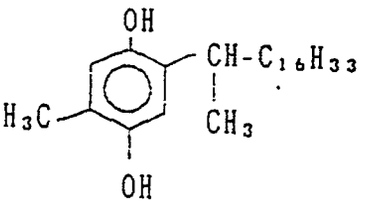
U V − 1



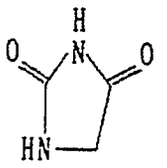
U V − 2



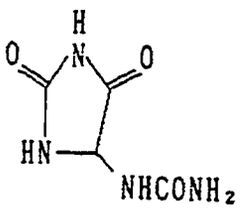
Cpd-1



Cpd-2

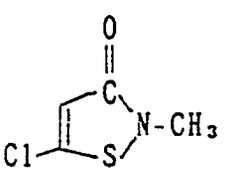


Cpd-3

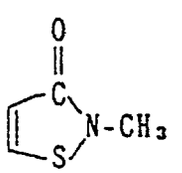


Cpd-4

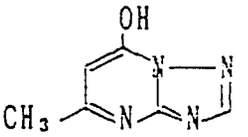
1 : 1 mixture (weight ratio) of:



and

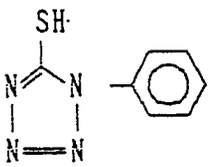


Cpd-5

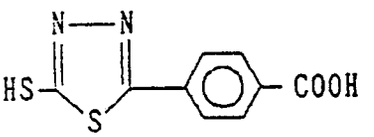


Cpd-6

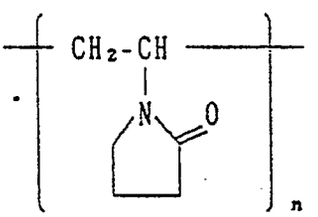
1 : 1 mixture (molar ratio) of:



and

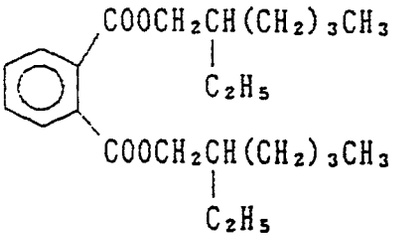


Cpd-7

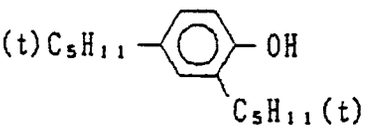


(n ≒ 60)

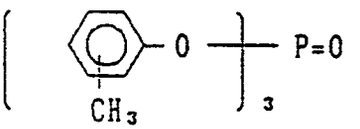
Solv-1



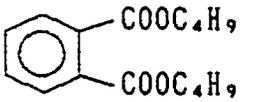
Solv-2



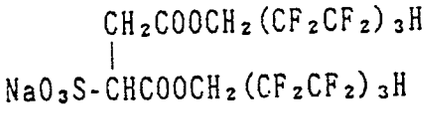
Solv-3



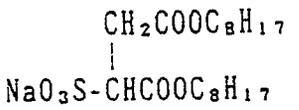
Solv-4



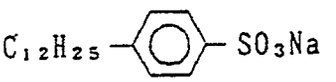
W − 1



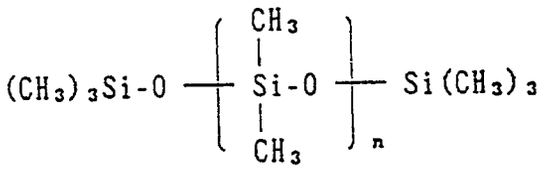
W − 2



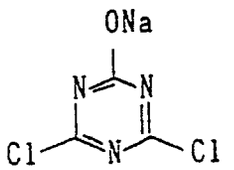
W − 3



Lubricant



H − 1



H − 2 $\left[(CH_2{=}CHSO_2CH_2)_3CCH_2SO_2(CH_2)_3 \right]_2N(CH_2)_2SO_3K$

Preparation of Specimen 202

Specimen 202 was prepared in the same manner as Specimen 201, except that the yellow-colored cyan coupler (YC-18) was coated on the 3rd and 4th layers in an amount of 0.02 g/m^2, ExM-1 incorporated in the 6th layer was replaced by [A-3]-11 in an equimolar amount, and ExM-2 incorporated in the 8th layer was replaced by [A-3]-14 in an equimolar amount.

Preparation of Specimens 203 to 209

Specimens 203 to 209 were prepared in the same manner as Specimen 202, except that the yellow-colored cyan couplers as set forth in Table 3 were incorporated in the 3rd and 4th layers and the couplers incorporated in the 6th layer and 8th layer were replaced by the couplers represented by the general formula (A) in equimolar amounts, respectively.

Preparation of Specimen 210

Specimen 210 was prepared in the same manner as Specimen 202 except that the amount of ExC-1 incorporated in the 3rd layer was reduced to 0.80 g/m^2 and the loss was made up for by an equimolar

amount of [A-3]-17. Additionally, the yellow-colored cyan couplers and the couplers represented by the general formula (A) were altered as set forth in Table 3.

Preparation of Specimen 211

Specimen 211 was prepared in the same manner as Specimen 101, except that the amount of CM-1 incorporated in the 6th layer and CM-2 incorporated in the 8th layer were each halved and the yellow-colored cyan coupler incorporated in the red-sensitive emulsion layer and the coupler represented by the general formula (A) were altered as set forth in Table 3.

TABLE 1

Sample No.	Yellow-Colored Cyan Coupler		Coupler, Formula [A]	
	3rd Layer	4th Layer	6th Layer	8th Layer
201(Comp.Ex.)	—	—	—	—
202(Invention)	(YC-18)	(YC-13)	[A-3]-11	[A-3]-14
203(Invention)	(YC-3)	(YC-3)	[A-3]-11	[A-4]-21
204(Invention)	(YC-7)	(YC-7)	[A-3]-1	[A-3]-1
205(Invention)	(YC-15)	(YC-20)	[A-4]-24	[A-3]-14
206(Invention)	(YC-16)	(YC-2)	[A-4]-36	[A-3]-39
207(Invention)	(YC-40)	(YC-25)	[A-4]-67	[A-4]-56
208(Invention)	(YC-53)	(YC-22)	[A-4]-17	[A-4]-51
209(Invention)	(YC-47)/(YC-48)=3/1	(YC-5)	[A-4]-41	[A-4]-36/[A-4]-61=4/1
210(Invention)	—	(YC-7)*	[A-4]-37	—
211(Invention)	(YC-18)	(YC-13)	[A-4]-67	[A-4]-56

Specimens 201 to 211 thus prepared were then measured for color stain (ΔD_γ) in cyan images in accordance with the method described in Example 1. These specimens were exposed to light through a green separation filter. The density value measured by blue light was determined at the exposure which gives a density of (minimum density + 1.0) on the characteristic curve obtained by the density measurement by green light. Color stain (ΔD_G) in magenta images was determined by substracting from this value the minimum density measured by blue light. The smaller the absolute value is, the smaller is the color stain and the more advantageous is the color reproduction, as in Example 1. These specimens were also evaluated for preservability of the light-sensitive material as in Example 1. The results are set forth in Table 4.

Table 4

Specimen No.	Color stain		Preservability of light-sensitive material	
	ΔD_γ	ΔD_G	ΔT_R	ΔT_G
201 (comparative)	0.16	0.10	0.08	0.09
202 (present invention)	0.02	-0.04	0.02	0.03
203 (")	0.02	-0.03	0.02	0.02
204 (")	0.02	-0.04	0.02	0.03
205 (")	0.02	-0.03	0.02	0.03
206 (")	0.02	-0.04	0.02	0.02
207 (")	0.03	-0.03	0.03	0.02
208 (")	0.05	-0.04	0.05	0.04
209 (")	0.03	-0.04	0.04	0.02
210 (")	0.02	-0.03	0.03	0.04
211 (")	0.02	0.01	0.02	0.02

The processing step used in this example will be set forth below. The processing was effected by means of an automatic developing machine. Specimen 201 which had been imagewise exposed to light was processed until the accumulated replenishment of the color developer reached three times the capacity of the running solution tank before the other specimens were processed.

Processing step

Step	Time	Temperature	Replenishment rate*	Tank capacity
Color development	3 min. 15 sec.	38°C	15 ml	20 ℓ
Bleach	6 min. 30 sec.	38°C	10 ml	40 ℓ
Washing	2 min. 10 sec.	35°C	10 ml	20 ℓ
Fixing	4 min. 20 sec.	38°C	20 ml	30 ℓ
Washing (1)	1 min. 05 sec.	35 °C	-**	10 ℓ
Washing (2)	1 min. 00 sec.	35 °C	20 ml	10 ℓ
Stabilization	1 min. 05 sec.	38 °C	10 ml	10 ℓ
Drying	4 min. 20 sec.	55 °C		

* Determined per 35-mm width and 1-m length

** The washing step was effected in a counter-current process wherein the washing water flows backward.

The various processing solutions had the following compositions:

Color developer

	Running Solution	Replenisher
Diethylenetriamine-pentaacetic acid	1.0 g	1.1 g
1-Hydroxyethylidene-1,1-diphosphonic acid	3.0 g	3.2 g
Sodium sulfite	4.0 g	4.9 g
Potassium carbonate	30.0 g	30.0 g

Potassium bromide	1.4 g	--
Potassium iodide	1.5 mg	--
Hydroxylamine sulfate	2.4 g	3.6 g
4-(N-ethyl-N-(β-hydroxyethyl) amino)-2-methylaniline sulfate	4.5 g	7.2 g
Water to make	1.0 ℓ	1.0 ℓ
pH	10.05	10.10

Bleaching solution

	Running Solution	Replenisher
Ferric sodium ethylenediamine-tetraacetate trihydrate TB1.00" 3.60" 4.80"	100 g	140 g
Disodium ethylenediamine-tetraacetate	10.0 g	11.0 g
Ammonium bromide	140.0 g	180.0 g
Ammonium nitrate	30.0 g	40.0 g
27% Aqueous ammonia	6.5 ml	2.5 ml
Water to make	1.0 ℓ	1.0 ℓ
pH	6.0	5.5

Fixing solution

	Running Solution	Replenisher
Disodium ethylenediamine-tetraacetate	0.5 g	1.0 g
Sodium sulfite	7.0	12.0
Sodium bisulfite	5.0	9.5

Aqueous solution of ammonium thiosulfate (700 g/l)	170.0 ml	240.0 ml
Water to make	1.0 ℓ	1.0 ℓ
pH	6.7	6.6

Washing solution (The running solution was used also as replenisher)

Tap water was passed through a mixed bed column packed with an H-type strongly acidic cation exchange resin (Amberlite IR-120B available from Rohm & Haas) and an OH-type strongly basic anion exchange resin (Amberlite IRA-400 available from the same company) so that the calcium and magnesium ion concentrations were each reduced to 3 mg/ℓ or less. Dichlorinated sodium isocyanurate and sodium sulfate were then added to the solution in amounts of 20 mg/ℓ and 150 mg/ℓ, respectively.

The washing solution thus obtained had a pH value of 6.5 to 7.5.

Stabilizing solution

	Tank Solution	Replenisher
37% Formaldehyde	2.0 ml	3.0 ml
Polyoxyethylene-p-monononyl-phenyl ether (average polymerization degree: 10)	0.3 g	0.45 g
Disodium ethylenedimaine-tetraacetate	0.05 g	0.08 g
Water to make	1.0 ℓ	1.0 ℓ
pH	5.0-8.0	5.0-8.0

Table 4 shows that the incorporation of the yellow-colored cyan couplers of the present invention in the red-sensitive emulsion layer provides a reduction in color stain in cyan images, higher color reproducibility and improved preservability of light-sensitive material.

Another advantage was found in that the incorporation of the couplers represented by the general formula (A) in the green-sensitive emulsion layer provides improved color reproduction of magenta dyes as well as reduction of the amount of yellow-colored magenta couplers to be used and improvements in inhibition of color stain. It is obvious that the couplers represented by the general formula (A) are excellent in preservability of light-sensitive material.

EXAMPLE 3

Specimen 301 was prepared in the same manner as Specimen 112, except that the amount of ExM-7 incorporated in the 6th and 7th layers and ExM-8 incorporated in the 8th layer were each halved. Specimen

301 thus prepared was cut into 35-mm wide strips.

Specimens were also prepared in the same manner as Specimens 301, 101 and 112, except that the amount of chemical sensitizers and other additives, i.e., sensitizing dye and Cpd-7, incorporated during the preparation of emulsions were altered so that these specimens exhibited the same sensitivity and gradation without altering the coated amount of silver.

These specimens were cut into 35-mm wide strips, and then used to photograph a Macbeth chart, various chromaticity diagrams and various patterns under the same exposure conditions. These specimens were then processed in the same manner as in Example 1.

The Macbeth charts, chromaticity diagrams and patterns were printed on a color paper (Super FA, available from Fuji Photo Film) through these color negative films by a printer processor PP-400 (Fuji Photo Film). The color paper was then processed in accordance with CP-40 processing (Fuji Photo Film).

As a result, Specimens 301 and 101 took almost the same period of time for exposure. Specimen 112 took a slightly longer time for exposure than Specimen 301. With respect to color reproducibility, on the other hand, Specimens 301 and 112 were better than Specimen 101, particularly in the hue of yellow to red.

These results show that the combined use of the yellow-colored cyan couplers and the couplers of the general formula (A) enables a reduction in the amount of the yellow-colored magenta coupler incorporated in the green-sensitive emulsion layer and provides color prints with an excellent color reproducibility without increasing the exposure time in the printer.

EXAMPLE 4

Specimen 401 was prepared in the same manner as Specimen 201 in Example 2 in JP-A-1-269935. Specimen 402 was prepared in the same manner as in Specimen 401, except that EX-3 incorporated in the 2nd, 3rd and 4th layers was replaced by the yellow-colored cyan coupler (YC-7) in equimolar amounts, respectively.

These specimens were subjected to exposure, develoment and evaluation for properties in the same manner as in Example 1.

As a result, it was confirmed that Specimen 402 comprising the yellow-colored cyan coupler of the present invention provides better results in sensitivity of cyan density, preservability of light-sensitive material and stability of latent images than does Specimen 401. This shows that the yellow-colored cyan couplers of the present invention are excellent.

EXAMPLE 5

A multilayer color light-sensitive material Specimen 501 was prepared by coating on an undercoated cellulose triacetate film support the following layers.

Composition of light-sensitive material

The figures indicate the coated amount of each component in g/m^2. The coated amount of silver halide and colloidal silver are represented in terms of the amount of silver. The coated amount of sensitizing dye is represented in a mol amount per mol of silver halide contained in the same layer.

(Specimen 501)

1st layer: anti-halation layer	Coated Amount
Black colloidal silver	0.18
Gelatin	1.40
2nd layer: interlayer	
2,5-Di-t-pentadecyl hydroquinone	0.18
EX-1	0.070
EX-3	0.025
EX-12	2.0×10^{-3}
U-1	0.060
U-2	0.080
U-3	0.10
HBS-1	0.10
HBS-2	0.020

Gelatin	1.04
3rd layer: 1st red-sensitive emulsion layer	
Emulsion A	0.25
Emulsion B	0.25
Sensitizing Dye I	6.9×10^{-5}
Sensitizing Dye II	1.8×10^{-5}
Sensitizing Dye III	3.1×10^{-4}
EX-2	0.34
EX-10 (D-25)	0.020
U-1	0.070
U-2	0.050
U-3	0.070
HBS-1	0.060
Gelatin	0.87
4th layer: 2nd red-sensitive emulsion layer	
Emulsion G	1.00
Sensitizing Dye I	5.1×10^{-5}
Sensitizing Dye II	1.4×10^{-5}
Sensitizing Dye III	2.3×10^{-4}
EX-2	0.40
EX-3	0.060
EX-10 (D-25)	0.015
U-1	0.070
U-2	0.050
U-3	0.070

Gelatin	1.30
5th layer: 3rd red-sensitive emulsion layer	
Emulsion D	1.60
Sensitizing Dye I	5.4×10^{-5}
Sensitizing Dye II	1.4×10^{-5}
Sensitizing Dye III	2.4×10^{-4}
EX-2	0.097
EX-3	0.013
EX-4	0.080
HBS-1	0.22
HBS-2	0.10
Gelatin	1.63
6th layer: Interlayer	
EX-5	0.040
HBS-1	0.020
Gelatin	0.80
7th layer: 1st green-sensitive emulsion layer	
Emulsion A	0.15
Emulsion B	0.15
Sensitizing Dye IV	3.0×10^{-5}
Sensitizing Dye V	1.0×10^{-4}
Sensitizing Dye VI	3.8×10^{-4}
EX-1	0.021
EX-6	0.26
EX-7	0.030

EX-8	0.025
HBS-1	0.20
HBS-3	0.010
Gelatin	0.63

8th layer: 2nd green-sensitive emulsion layer

Emulsion C	0.45
Sensitizing Dye IV	2.1×10^{-5}
Sensitizing Dye V	7.0×10^{-5}
Sensitizing Dye VI	2.6×10^{-4}
EX-6	0.094
EX-7	0.026
EX-8	0.018
HBS-1	0.20
HBS-3	8.0×10^{-3}
Gelatin	0.50

9th layer: 3rd green-sensitive emulsion layer

Emulsion E	1.20
Sensitizing Dye IV	3.5×10^{-5}
Sensitizing Dye V	8.0×10^{-5}
Sensitizing Dye VI	3.0×10^{-4}
EX-1	0.025
EX-11	0.10
EX-13	0.015
HBS-1	0.30
HBS-2	0.10

Gelatin	1.54
10th layer: Yellow filter layer	
Yellow colloidal silver	0.050
EX-5	0.080
HBS-1	0.030
Gelatin	0.50
11th layer: 1st blue-sensitive emulsion layer	
Emulsion A	0.080
Emulsion B	0.070
Emulsion F	0.070
Sensitizing Dye VII	3.5×10^{-4}
EX-8	0.042
EX-9	0.72
HBS-1	0.28
Gelatin	1.10
12th layer: 2nd blue-sensitive emulsion layer	
Emulsion G	0.45
Sensitizing Dye VII	2.1×10^{-4}
EX-9	0.15
EX-10 (D-25)	7.0×10^{-3}
HBS-1	0.050
Gelatin	0.78
13th layer: 3rd blue-sensitive emulsion layer	
Emulsion H	0.77
Sensitizing Dye VII	2.2×10^{-4}

EX-9	0.20
HBS-1	0.070
Gelatin	0.69
14th layer: 1st protective layer	
Emulsion I	0.20
U-4	0.11
U-5	0.17
HBS-1	5.0×10^{-2}
Gelatin	0.50
15th layer: 2nd protective layer	
H-1	0.30
B-1 (diameter: 1.7 µm)	5.0×10^{-2}
B-2 (diameter: 1.7 µm)	0.10
B-3	0.10
S-1	0.20
Gelatin	0.70

In order to improve preservability, processability, pressure resistance, anti-fungal and bacterial properties, anti-static properties and coatability, all these layers further comprised W-1, W-2, W-3, B-4, B-5, F-1, F-2, F-3, F-4, F-5, F-6, F-7, F-8, F-9, F-10, F-1, F-12, F-13, F-14, iron salts, lead salts, gold salts, platinum salts, iridium salts, and rhodium salts.

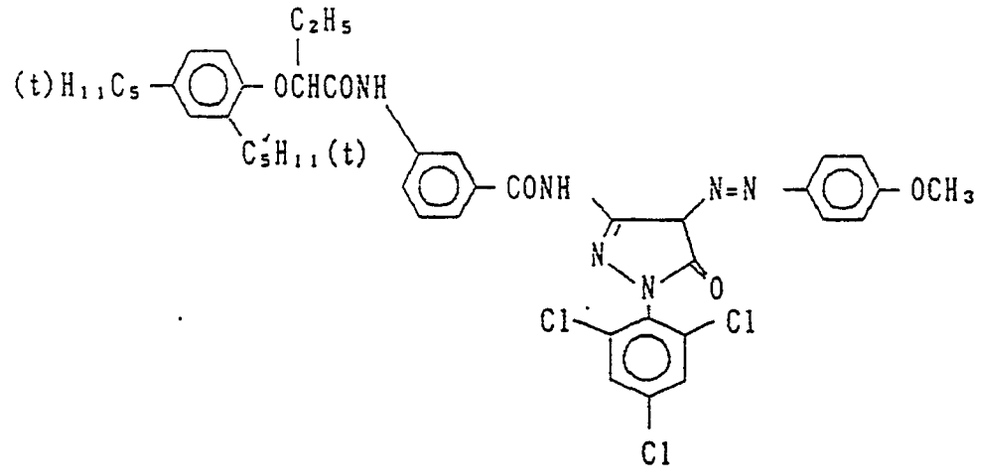
The silver halide emulsions used are set forth in Table 5.

Table 5

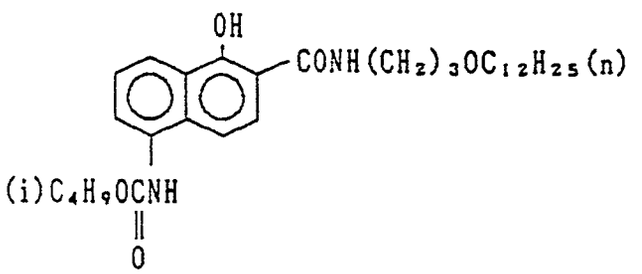
Emulsion	% Mean AgI content	Mean grain diameter (μm)	% Grain diameter variation coefficient	dia./thick. ratio	Ratio of amount of Ag (AgI content %)
A	4.0	0.45	27	1	Core/shell= 1/3(13/1); double structure
B	8.9	0.70	14	1	Core/shell= 3/7(25/2); double structure
C	10	0.75	30	2	Core/shell= 1/2(24/3); double structure
D	16	1.05	35	2	Core/shell= 4/6(40/0): double structure
E	10	1.05	35	3	Core/shell= 1/2(24/3); double structure
F	4.0	0.25	28	1	Core/shell= 1/3(13/1); double structure
G	14.0	0.75	25	2	Core/shell= 1/2(42/0); double structure
H	14.5	1.30	25	3	Core/shell= 37/63(34/3); double structure
I	1	0.07	15	1	Uniform grain

The chemical structures of the compounds incorporated in these layers are set forth below.

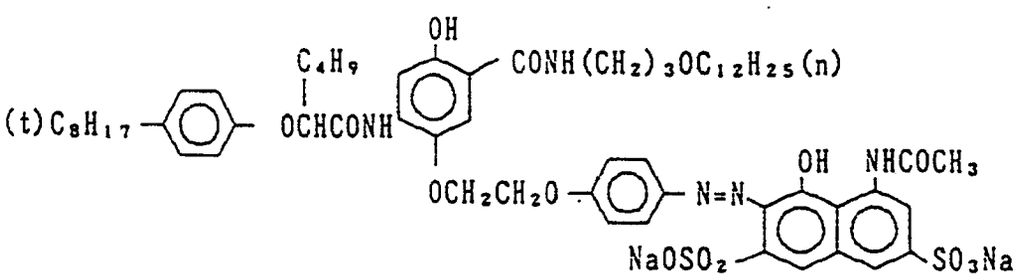
EX－1



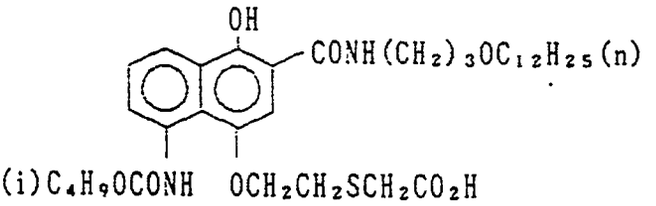
EX－2



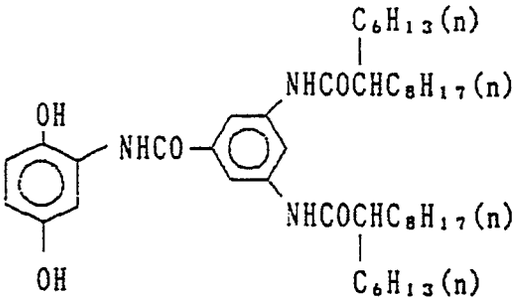
EX－3



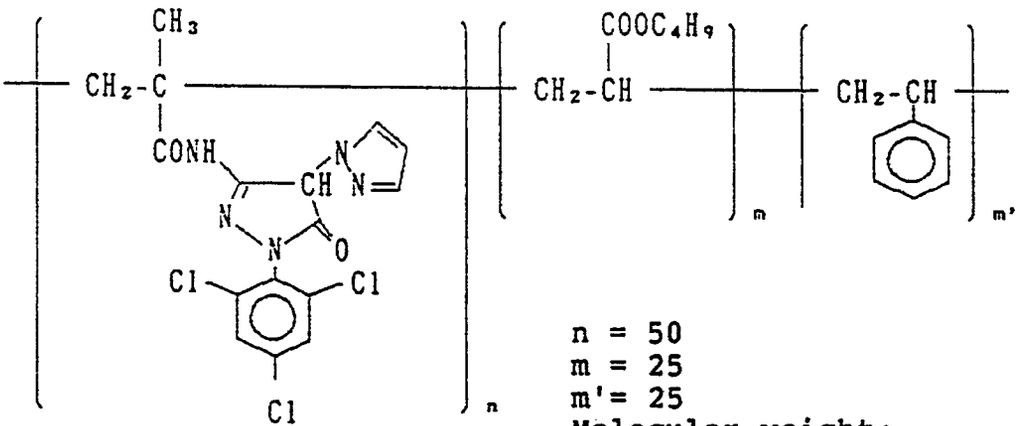
EX－4



EX－5

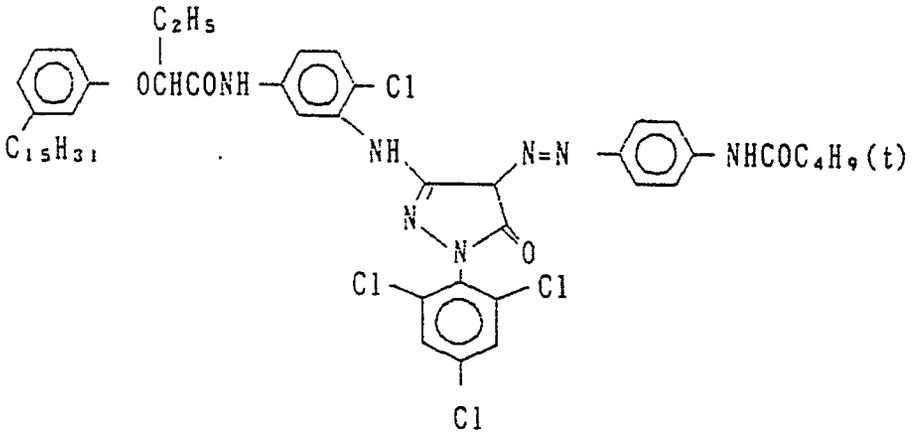


EX－6

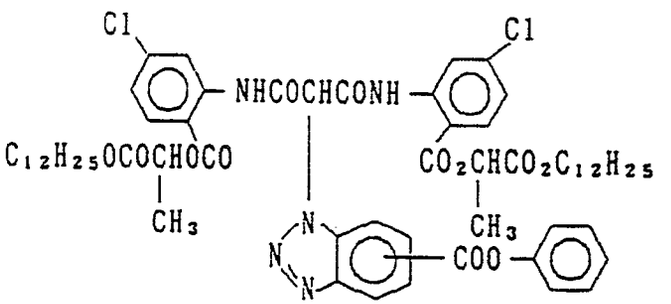


n = 50
m = 25
m'= 25
Molecular weight:
approx. 20,000

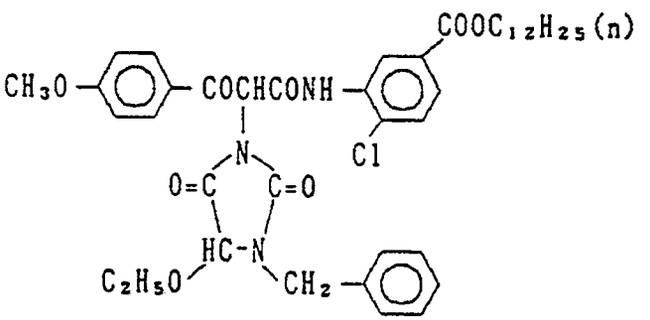
EX-7



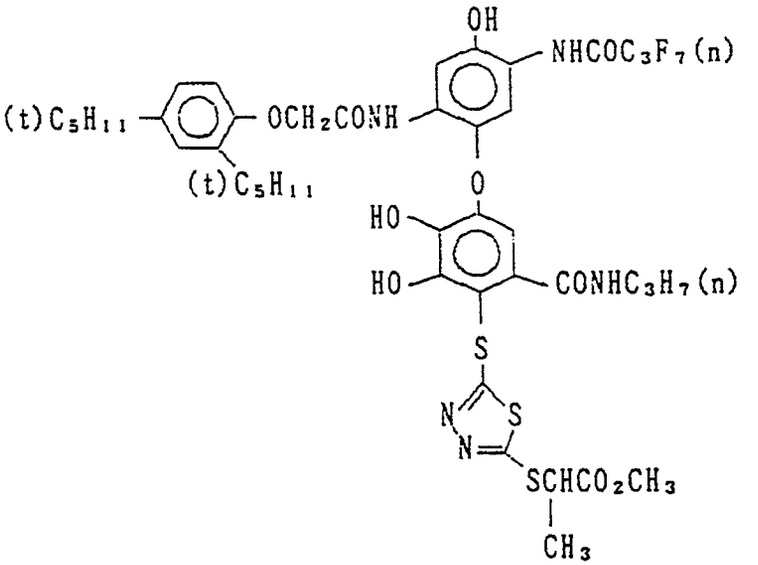
EX-8



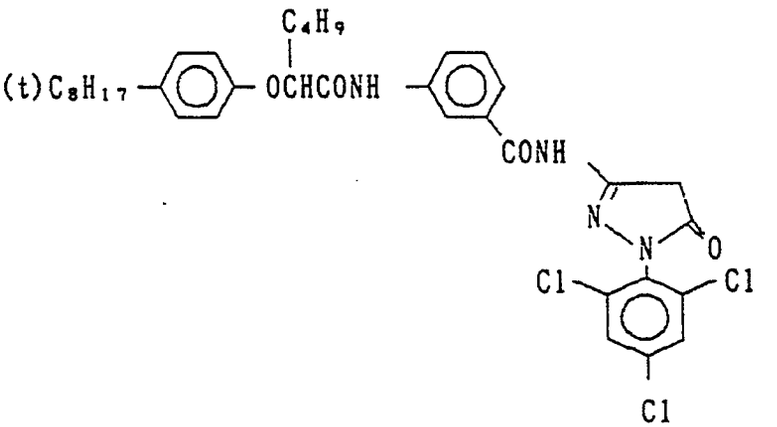
EX-9



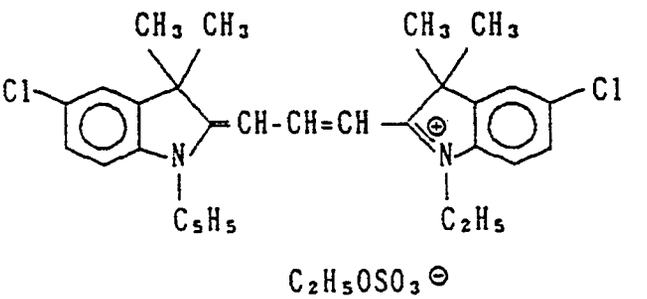
EX-10 (D-25)



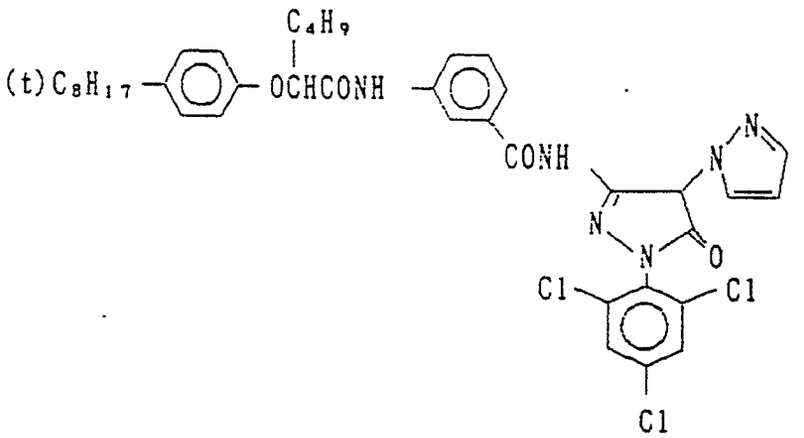
EX-11



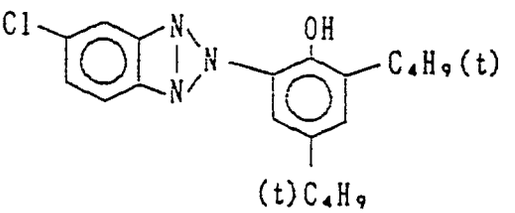
EX-12



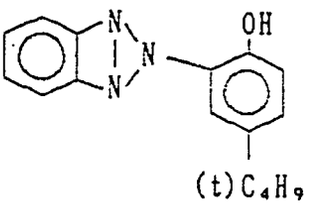
EX－13



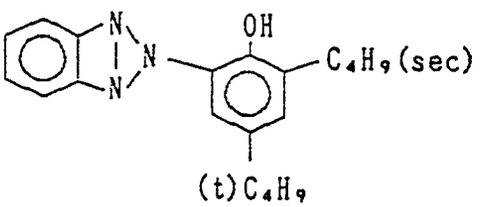
U－1



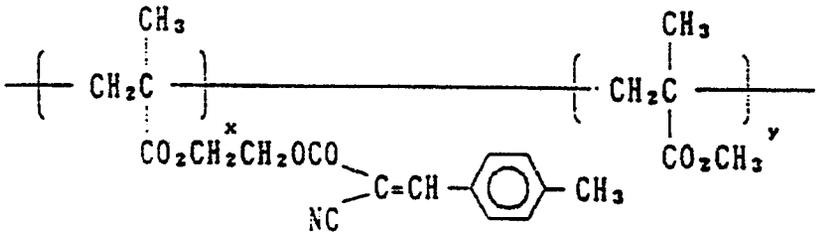
U－2



U－3

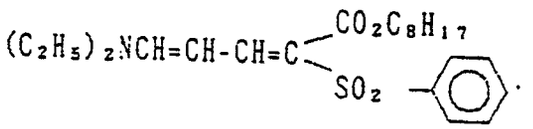


U − 4



x : y = 7 0 : 3 0 (w t %)

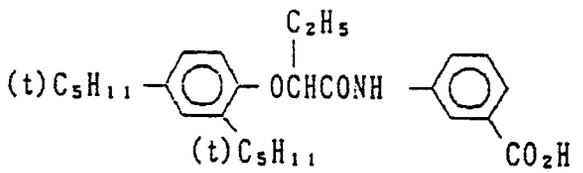
U − 5



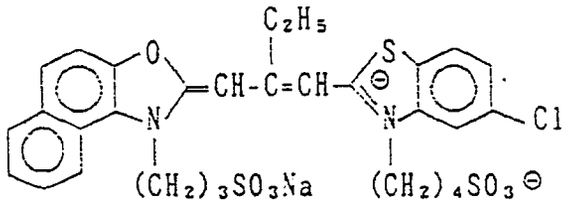
HBS-1: Tricresyl phosphate

HBS-2: Di-n-butyl phthalate

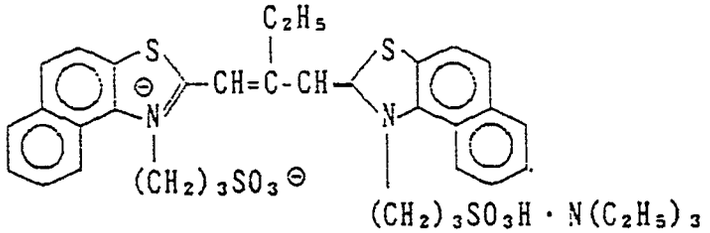
HBS-3



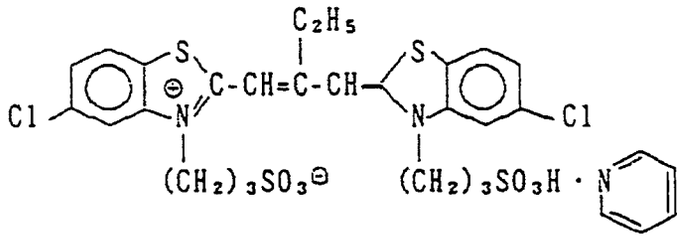
Sensitizing Dye I



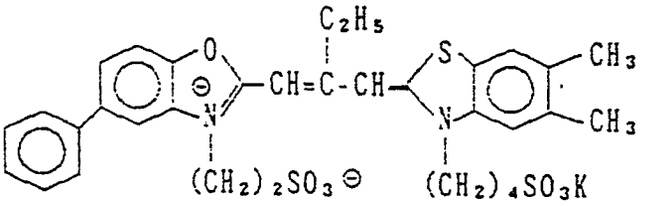
Sensitizing Dye II



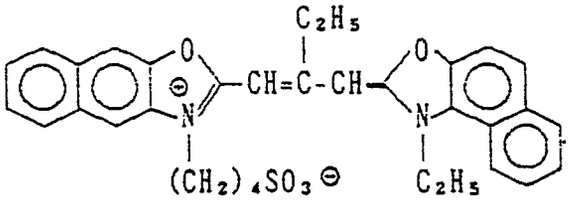
Sensitizing Dye III



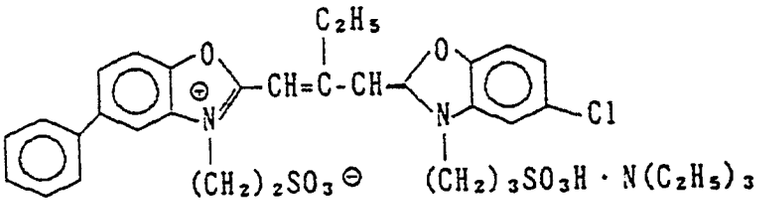
Sensitizing Dye IV



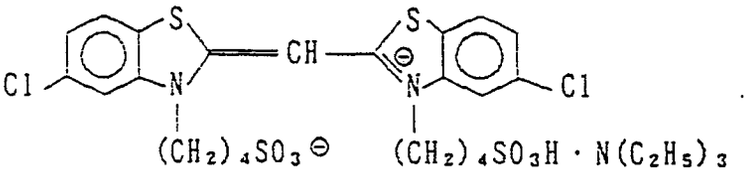
Sensitizing Dye V



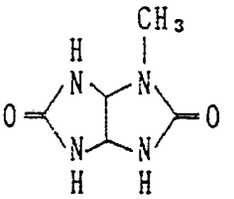
Sensitizing Dye VI



Sensitizing Dye VII

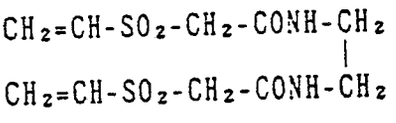


S − 1

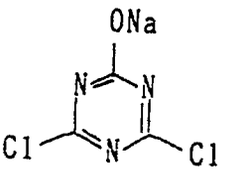


H-1

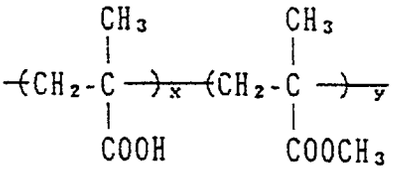
3 : 1 mixture (molar ratio) of :



and

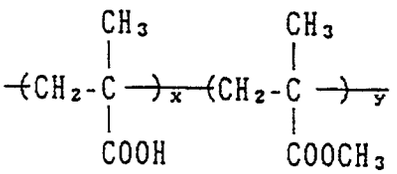


B − 1



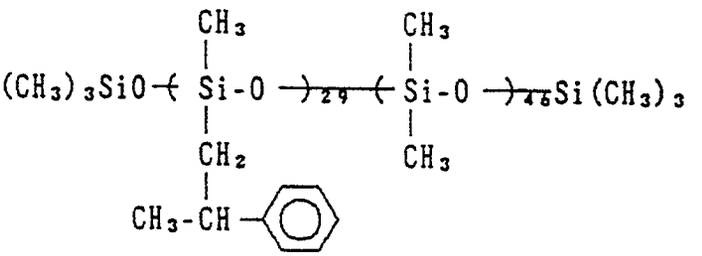
x / y = 10 / 90

B − 2

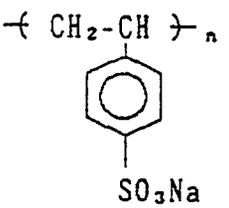


x / y = 40 / 60

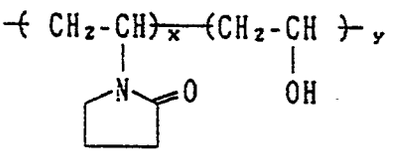
B − 3



B − 4

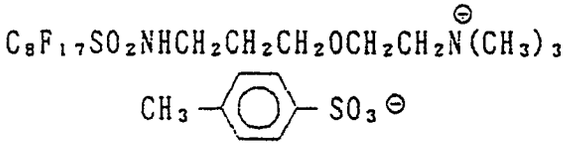


B − 5

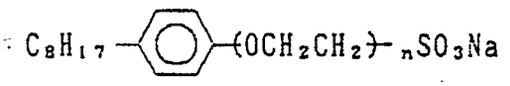


x / y = 70 / 30

W − 1

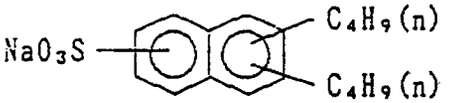


W − 2

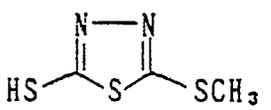


n = 2 ~ 4

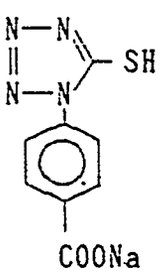
W − 3



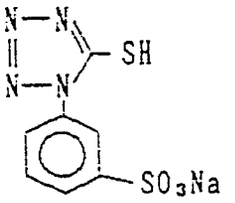
F − 1



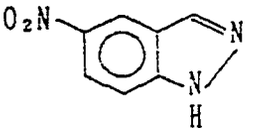
F − 2



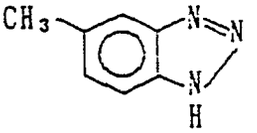
F − 3



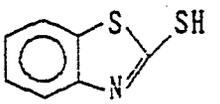
F − 4



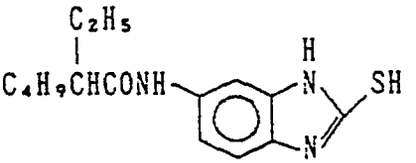
F − 5



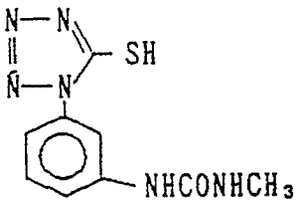
F − 6



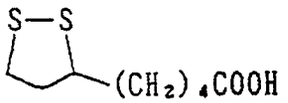
F − 7



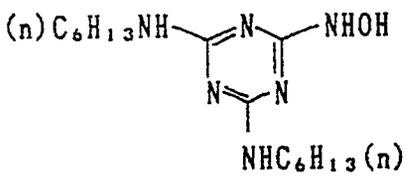
F − 8



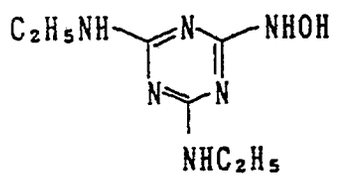
F − 9



F − 1 0

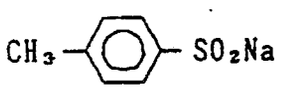


F − 1 1

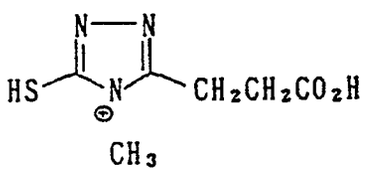


F − 1 2

F − 1 3



F − 1 4



Specimen 502 was prepared in the same manner as Specimen 501, except that the yellow-colored cyan coupler (YC-85) was coated on the 3rd, 4th and 5th layers in the amounts of 0.06 g/m^2, 0.05 g/m^2, and 0.02 g/m^2, respectively, EX-6 incorporated in the 7th layer was replaced by Coupler [A-4]-48 in an equimolar amount (as calculated in terms of EX-6), EX-6 incorporated in the 8th layer was replaced by Coupler [A-4]-48 in half the equimolar amount, EX-11 incorporated in the 9th layer was replaced by Coupler [A-4]-51 in an equimolar amount, and HBS-3 incorporated in the 7th and 8th layer was omitted.

Specimens 503 to 510 were prepared in the same manner as Specimen 502 exept that the yellow-colored cyan coupler YC-85 incorporated in the red-sensitive emulsion layer in the 3rd, 4th and 5th layers, Couplers [A-4]-48 and [A-4]-51 incorporated in the green-sensitive emulsion layer in the 7th, 8th and 9th layers, and EX-8 and EX-10(D-25) incorporated in the 3rd, 4th, 7th, 8th, 11th, and 12th layers were replaced by the compounds set forth in Tables 6 and 7 in equimolar amounts, respectively.

Table 6

Specimen No.	Red-sensitive emulsion layer Yellow-colored cyan coupler and Compound (I)			Green-sensitive emulsion layer Coupler (A) and Compound (I)			Blue-sensitive emulsion layer Compound (I)	
	3rd layer	4th layer	5th layer	7th layer	8th layer	9th layer	11th layer	12th layer
501 (comparative)	--	--	--	--	--	--	EX-8	EX-10
	EX-10 (D-25)	EX-10 (D-25)	--	EX-8	EX-8	--		(D-25)
502 (present invention)	YC-85 (D-25)	YC-85 (D-25)	YC-85 ---	[A-4]-48 EX-8	[A-4]-48 EX-8	[A-4]-51 --	EX-8	EX-10 (D-25)
503 (")	YC-86 (D-25)	YC-86 (D-25)	YC-86 ---	[A-4]-74 EX-8	[A-4]-74 EX-8	[A-4]-53 ---	EX-8	EX-10 (D-25)
504 (")	YC-85	YC-85	YC-85	[A-4]-77	[A-4]-76	[A-4]-75 ([A-4]-73)*	EX-8	EX-10 (D-25)
	(D-25)	(D-25)	--	EX-8	EX-8	---		
505 (")	YC-87/YC-39 =2/1 (molar ratio)	YC-88/YC-27 =2/1 (molar ratio)	YC-90/YC-29 =1/1 (molar ratio)	[A-4]-68 EX-8	[A-4]-53 EX-8	[A-4]-47 --	EX-8	EX-10 (D-25)
506 (")	YC-86	YC-28/YC-89 =1/1 (molar ratio)	YC-25	[A-4]-35/ [A-4]-69=1/2 (molar ratio)	[A-3]-18	[A-3]-19	EX-8	EX-10 (D-25)
	(D-25)				EX-8	---		

* Replaced in the equimolecular amount with EX-13

Table 7

Specimen No.	Red-sensitive emulsion layer Yellow-colored cyan coupler and Compound (I)			Green-sensitive emulsion layer Coupler (A) and Compound (I)			Blue-sensitive emulsion layer Compound (I)	
	3rd layer	4th layer	5th layer	7th layer	8th layer	9th layer	11th layer	12th layer
507 (")	YC-85 D-11	YC-85 D-27	YC-85 ---	(A-4)-48 D-18	(A-4)-48 D-10	(A-4)-51 ---	D-13	D-2
508 (")	YC-85 D-24	YC-85 D-30	YC-85 --	(A-4)-48 D-9	(A-4)-48 D-34	(A-4)-51 --	D-33	D-22
509 (")	YC-85 D-12	YC-85 D-29	YC-85 ---	(A-4)-48 D-16	(A-4)-48 D-4	(A-4)-51 --	D-15	D-7
510 (comparative)	YC-85 --	YC-85 --	YC-85 --	(A-4)-48 ---	(A-4)-48 ---	(A-4)-51 ---	---	---

These specimens were cut and then evaluated for properties in the manner as described below. The processing was effected in the same manner as in Example 1 with the same processing solutions used in Example 1.

(5-1) Color stain (ΔD_γ)

Color stain (ΔD_{γ}) was determined in the same manner as in (1) in Example 1.

(5-2) Sharpness (MTF value)

The specimens were exposed to light through an MTF pattern. The MTF value was then determined. The MTF values of the cyan and magenta dyes are set forth in Table 7.

(5-3) Dye image fastness (D%)

The specimens were exposed to white light, and then processed to obtain color images. Characteristic curves were obtained from the color images by blue light, green light and red light. These specimens were stored at a temperature of 80° C and a relative humidity of 70% for 7 days. After the test, these specimens were again measured for density, and the density was determined at the exposure which gives a density of (minimum density developed before test + 1.5). The percentage of the density (D%) was determined with respect to the value obtained before test. The nearer to 100 the value is, the more fast is the color image.

The results of the tests (5-1) to (5-3) are set forth in Table 8.

Table 8

Specimen No.	Color stain (ΔD_{γ})	MTF value [25cycle/mm] G	MTF value [25cycle/mm] R	Color image fastness (D%) B	Color image fastness (D%) G	Color image fastness (D%) R
501 (comparative)	0.15	77	58	80	93	95
502 (present invention)	0.02	79	62	80	97	97
503 (")	0.02	79	63	80	97	98
504 (")	0.02	79	63	80	97	98
505 (")	0.02	79	63	80	97	98
506 (")	0.02	79	63	80	97	98
507 (")	0.02	82	63	91	99	98
508 (")	0.02	82	63	92	99	98
509 (")	0.02	82	63	92	99	98
510 (comparative)	0.05	64	46	87	95	95

Table 8 shows that the combination of the yellow-colored cyan couplers and the couplers of the general formula (A), with the additional use of the compounds of the general formula (I), provides improvements in sharpness as well as in color image fastness, particularly in yellow dye images, but also in magenta dye images. This provides improvements in inhibition of deterioration of the three colors, i.e., yellow, magenta and cyan images. Thus, the comparison between Specimens 507 to 509 and Specimens 502 to 506 shows that the three color images of substantially the same level exhibit relatively uniform fastness.

The comparison with Specimens 502 to 509 shows that the combination the yellow-colored cyan couplers and the compounds of formula (I) provides the unexpected effect that improvements are obtained also in color stain as compared to Specimen 510 which comprises only the yellow-colored cyan coupler.

The combination of the yellow-colored cyan couplers and the pyrazoloazole couplers of the general formula (A) provides improvements in color reproducibility and high sensitivity as well as improvements in

preservability of light-sensitive material and stability of latent images. The use of the pyrazoloazole couplers reduces the amount of the yellow-colored magenta couplers to be used in combination with the conventional 5-pyrazolone couplers.

Thus, the present invention provides a silver halide color photographic material which exhibits an excellent color reproducibility and preservability.

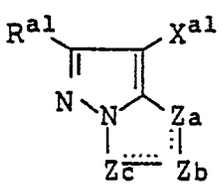
In addition, the use of the compounds represented by the general formula (I) provides improvements in color image fastness. This improves the inhibition of deterioration of yellow, magenta and cyan images. Thus, all the three color images exhibit uniform fastness. This further improves the color reproducibility.

Thus, the present invention also provides a silver halide color photographic material which exhibits improved color image fastness.

While the invention has been described in detail and with reference to specific embodiments thereof, it will be apparent to one skilled in the art that various changes and modifications can be made therein without departing from the spirit and scope thereof.

Claims

1. A silver halide color photographic material comprising a support having thereon (i) at least a silver halide emulsion layer, (ii) a yellow-colored cyan coupler which is capable of undergoing reaction with an oxidation product of an aromatic primary amine developing agent to release a group containing a water-soluble 6-hydroxy-2-pyridon-5-ylazo group, a water soluble 2-acylaminophenylazo group, a water soluble 2-sulfonamidophenylazo group, a water soluble 5-aminopyrazol-4-ylazo group or a water soluble pyrazolon-4-ylazo group and (iii) a coupler represented by the general formula (A):



(A)

wherein R^{a1} represents a hydrogen atom or substituent; X^{a1} represents a hydrogen atom or a group capable of being separated from the compound of formula (A) upon a coupling reaction with an oxidation product of an aromatic primary amine developing agent; Za, Zb and Zc each represents a methine group, substituted methine group, =N- group or -NH- group; one of the Za-Zb and Zb-Zc bonds is a double bond and the other is a single bond; if the Zb-Zc bond is a carbon-carbon double bond, it may be a part of an aromatic ring; R^{a1} or X^{a1} may form a dimer or higher polymer; and if Za, Zb or Zc is a substituted methine, it may form a dimer or higher polymer.

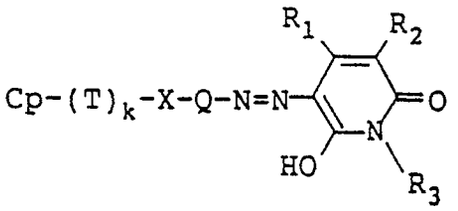
2. The silver halide color photographic material as claimed in claim 1, further comprising a compound represented by the general formula (I):

$$A - \{(L1)_a - (B)_m\}_p - (L2)_n - DI \qquad (I)$$

wherein A represents a group which is capable of undergoing a reaction with an oxidation product of an aromatic primary amine developing agent to cause cleavage of A from $\{(L1)_a - (B)_m\}_p - (L2)_n - DI$; L1 represents a group which causes cleavage of the bond between L1 and the group to its right as viewed in general formula (I) after cleavage of the bond between L1 and A; B represents a group which undergoes a reaction with an oxidation product of a developing agent to cause cleavage of the bond between B and the group to its right as viewed in general formula (I); L2 represents a group which causes cleavage of the bond between L2 and DI after cleavage of the bond of L2 to the group to its left as viewed in general formula (I); DI represents a development inhibitor; a, m and n each represents an integer 0 or 1; and p represents an integer 0 to 2, with the proviso that if p is 2, the two $\{(L1)_a - (B)_m\}$ groups are the same or different.

3. The silver halide color photographic material as claimed in claim 1, wherein the cyan coupler is

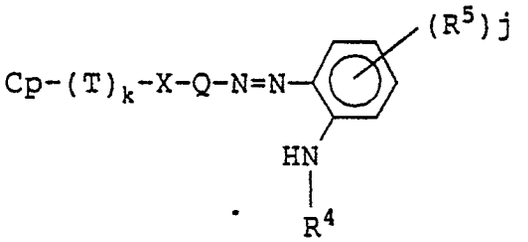
represented by general formula (CI):



(CI)

wherein Cp represents a cyan coupler group, T represents a timing group, k represents an integer 0 or 1, X represents a divalent connecting group containing N, O or S by which $(T)_k$ and Q are connected to each other, Q represents an arylene group or a divalent heterocyclic group, R_1 and R_2 each independently represents a hydrogen atom, a carboxyl group, a sulfo group, a cyano group, an alkyl group, a cycloalkyl group, an aryl group, a heterocyclic group, a carbamoyl group, a sulfamoyl group, a carbonamido group, a sulfonamido group or an alkylsulfonyl group, and R_3 represents a hydrogen atom, an alkyl group, a cycloalkyl group, an aryl group or a heterocyclic group, with the proviso that at least one of T, X, Q, R_1, R_2 and R_3 contains a water-soluble group.

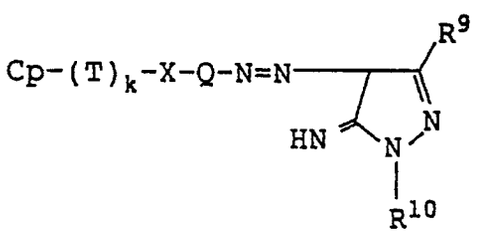
4. The silver halide color photographic material as claimed in claim 1, wherein the cyan coupler is represented by general formula (CII):



(CII)

wherein Cp represents a cyan coupler group, T represents a timing group, k represents an integer 0 or 1, X represents a divalent connecting group containing N, O or S by which $(T)_k$ and Q are connected to each other, Q represents an arylene group or a divalent heterocyclic group, R^4 represents an acyl group or sulfonyl group, R^5 represents a substitutable group, and j represents an integer from 0 to 4.

5. The silver halide color photographic material as claimed in claim 1, wherein the cyan coupler is represented by general formula (CIII):

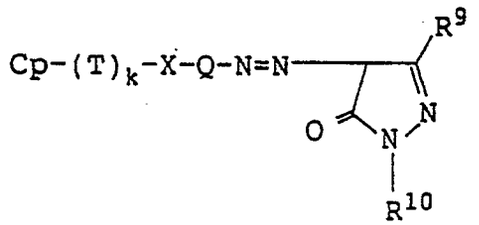


(CIII)

wherein Cp represents a cyan coupler group, T represents a timing group, k represents an integer 0 or 1, X represents a divalent connecting group containing N, O or S by which $(T)_k$ and Q are connected to each other, Q represents an arylene group or a divalent heterocyclic group, R^9 represents a hydrogen atom, a carboxyl group, a sulfo group, a cyano group, an alkyl group, a cycloalkyl group, an aryl group, an alkoxy group, a cycloalkyloxy group, an aryloxy group, a heterocyclic group, a carbamoyl group, a

sulfamoyl group, a carbonamide group, a sulfonamide group or an alkylsulfonyl group, and R^{10} represents a hydrogen atom, an alkyl group, a cycloalkyl group, an aryl group or a heterocyclic group, with the proviso that at least one of T, X, Q, R^9, and R^{10} contains a water-soluble group.

6. The silver halide color photographic material as claimed in claim 1, wherein the cyan coupler is represented by general formula (CIV):

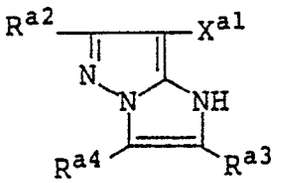


(CIV)

wherein Cp represents a cyan coupler group, T represents a timing group, k represents an integer 0 or 1, X represents a divalent connecting group containing N, O or S by which $(T)_k$ and Q are connected to each other, Q represents an arylene group or a divalent heterocyclic group, R^9 represents a hydrogen atom, a carboxyl group, a sulfo group, a cyano group, an alkyl group, a cycloalkyl group, an aryl group, an alkoxy group, a cycloalkyloxy group, an aryloxy group, a heterocyclic group, a carbamoyl group, a sulfamoyl group, a carbonamide group, a sulfonamide group or an alkylsulfonyl group, and R^{10} represents a hydrogen atom, an alkyl group, a cycloalkyl group, an aryl group or a heterocyclic group, with the proviso that at least one of T, X, Q, R^9, and R^{10} contains a water-soluble group.

7. The silver halide color photographic material as claimed in claim 1, wherein the cyan coupler is incorporated in a red-sensitive emulsion layer.

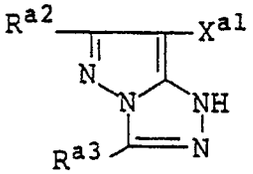
8. The silver halide color photographic material as claimed in claim 1, wherein the coupler of general formula (A) is represented by general formula (A-1):



(A-1)

wherein R^{a2}, R^{a3} and R^{a4} each represents a hydrogen atom, a halogen atom, an alkyl group, an aryl group, a heterocyclic group, a cyano group, an alkoxy group, an aryloxy group, a heterocyclic oxy group, an acyloxy group, a carbamoyloxy group, a silyloxy group, a sulfonyloxy group, an acylamino group, an anilino group, a ureido group, an imido group, a sulfamoylamino group, an alkylthio group, an arylthio group, a heterocyclic thio group, an alkoxycarbonylamino group, an aryloxycarbonylamino group, a sulfonamido group, a carbamoyl group, an acyl group, a sulfamoyl group, a sulfonyl group, a sulfinyl group, an alkoxycarbamoyl group or an aryloxycarbonyl group, and X^{a1} represents a hydrogen atom, a halogen atom, a carboxyl group or a group which is connected to the carbon atom in the coupling position via an oxygen atom, a nitrogen atom or a sulfur atom to undergo coupling elimination.

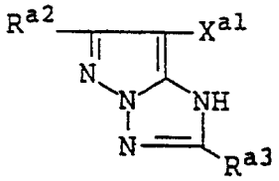
9. The silver halide color photographic material as claimed in claim 1, wherein the coupler of general formula (A) is represented by general formula (A-3):



(A-3)

wherein R^{a2} and R^{a3} each represents a hydrogen atom, a halogen atom, an alkyl group, an aryl group, a heterocyclic group, a cyano group, an alkoxy group, an aryloxy group, a heterocyclic oxy group, an acyloxy group, a carbamoyloxy group, a silyloxy group, a sulfonyloxy group, an acylamino group, an anilino group, a ureido group, an imido group, a sulfamoylamino group, an alkylthio group, an arylthio group, a heterocyclic thio group, an alkoxycarbonylamino group, an aryloxycarbonylamino group, a sulfonamido group, a carbamoyl group, an acyl group, a sulfamoyl group, a sulfonyl group, a sulfinyl group, an alkoxycarbamoyl group or an aryloxycarbonyl group, and X^{a1} represents a hydrogen atom, a halogen atom, a carboxyl group or a group which is connected to the carbon atom in the coupling position via an oxygen atom, a nitrogen atom or a sulfur atom to undergo coupling elimination.

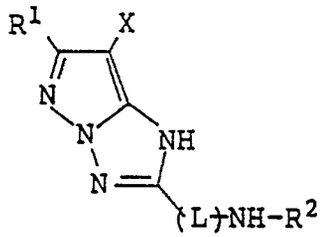
10. The silver halide color photographic material as claimed in claim 1, wherein the coupler of general formula (A) is represented by general formula (A-4):



(A-4)

wherein R^{a2} and R^{a3} each represents a hydrogen atom, a halogen atom, an alkyl group, an aryl group, a heterocyclic group, a cyano group, an alkoxy group, an aryloxy group, a heterocyclic oxy group, an acyloxy group, a carbamoyloxy group, a silyloxy group, a sulfonyloxy group, an acylamino group, an anilino group, a ureido group, an imido group, a sulfamoylamino group, an alkylthio group, an arylthio group, a heterocyclic thio group, an alkoxycarbonylamino group, an aryloxycarbonylamino group, a sulfonamido group, a carbamoyl group, an acyl group, a sulfamoyl group, a sulfonyl group, a sulfinyl group, an alkoxycarbamoyl group or an aryloxycarbonyl group, and X^{a1} represents a hydrogen atom, a halogen atom, a carboxyl group or a group which is connected to the carbon atom in the coupling position via an oxygen atom, a nitrogen atom or a sulfur atom to undergo coupling elimination.

11. The silver halide color photographic material as claimed in claim 1, wherein the coupler of general formula (A) is represented by formula (M):

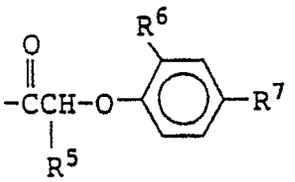


(M)

wherein R^1 represents an alkyl group, an alkoxy group or an aryloxy group; R^2 represents an acyl group or a sulfonyl group; -(L)- represents an alkylene or a phenylene group represented by $-(C(R^3)(R^4)-CH_2)-$, with the proviso that when -(L)- is an alkylene group, the carbon atom to which R^3 and R^4 are connected is connected to the coupler nucleus, and R^3 and R^4 each represents a hydrogen atom, an

alkyl group or an aryl group, but are not hydrogen atoms at the same time; and X represents an aryloxy group, an alkoxy group, a 1-azolyl group, an alkylthio group or an arylthio group.

12. The silver halide color photographic material as claimed in claim 11, wherein R^2 of the coupler of general formula (M) is represented by formula (A_1):



(A_1)

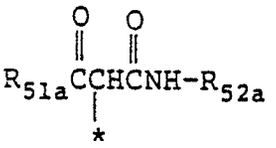
wherein R^5 represents a hydrogen atom or alkyl group, and R^6 and R^7 each represents an alkyl group, an aryloyl group, an alkylsulfonyl group, or an arylsulfonyl group.

13. The silver halide color photographic material as claimed in claim 1, wherein the cyan coupler is in a red-sensitive emulsion layer or in a light-insensitive layer adjacent thereto.

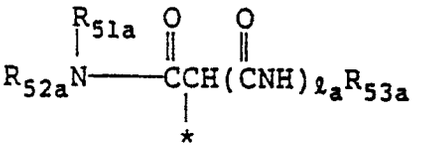
14. The silver halide color photographic material as claimed in claim 13, wherein the coupler of general formula (A) is incorporated in a green-sensitive emulsion layer or in a light-insensitive layer adjacent thereto.

15. The silver halide color photographic material as claimed in claim 1, wherein the coupler of general formula (A) is in a red-sensitive emulsion layer or in a green-sensitive layer or in a light-insensitive layer adjacent to said red-sensitive layer or to said green-sensitive layer.

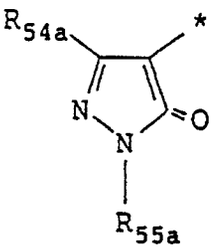
16. The silver halide color photographic material as claimed in claim 2, wherein A is a coupler represented by one of the following general formulae (Cp-1a) to (Cp-10a):



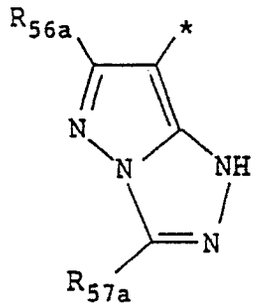
(Cp-1a)



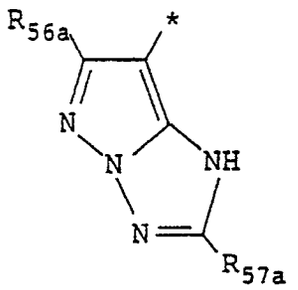
(Cp-2a)



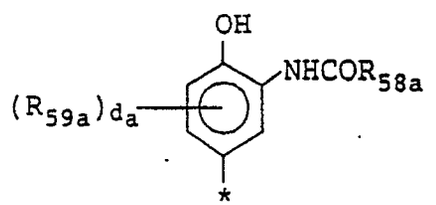
(Cp-3a)



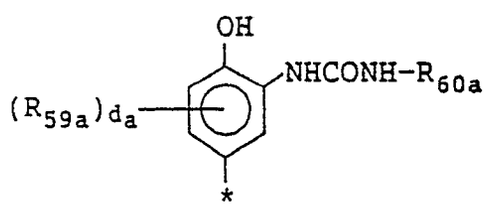
(Cp-4a)



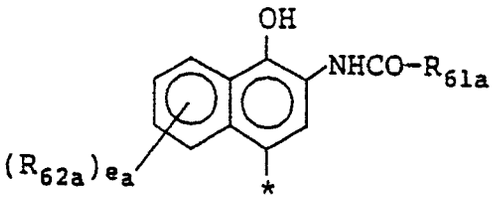
(Cp-5a)



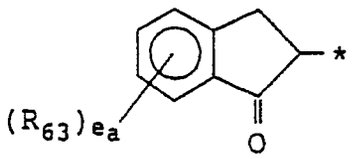
(Cp-6a)



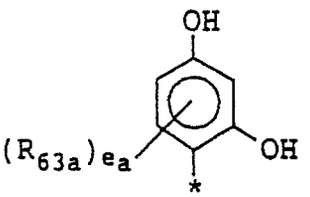
(Cp-7a)



(Cp-8a)



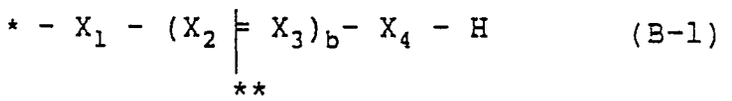
(Cp-9a)



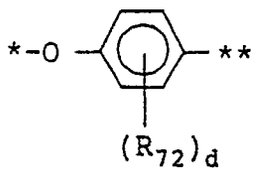
(Cp-10a)

wherein R_{51a} represents a hydrogen atom, an aliphatic group, an aromatic group or a heterocyclic group, R_{52a} and R_{53a} each represents an aromatic group or a heterocyclic group, R_{54a} represents an aliphatic group, an aromatic group, a heterocyclic group, a $R_{41a}CON(R_{43a})$- group, a $(R_{41a})(R_{43a})N$-group, a $R_{41a}SO_2N(R_{43a})$- group, a $R_{41a}S$- group, a $R_{43a}O$- group, a $(R_{43a})(R_{45a})NCON(R_{44a})$-group or a $N{\equiv}C$- group, R_{55a} represents an aliphatic group, an aromatic group or a heterocyclic group, R_{56a} and R_{57a} each represents a hydrogen atom, an aliphatic group, an aromatic group, a heterocyclic group, a $R_{41a}S$-group, a $R_{43a}O$- group, a $R_{41a}CON(R_{43a})$- group or $R_{41a}SO_2N(R_{43a})$- group, R_{58a} represents an aliphatic group, an aromatic group or a heterocyclic group, R_{59a} represents an aliphatic group, an aromatic group, a heterocyclic group, an $R_{41a}CON(R_{43a})$- group, an $R_{41a}OCON(R_{43a})$- group, an $R_{41a}SO_2N(R_{43a})$- group, an $(R_{43a})(R_{44a})NCON(R_{45a})$- group, an $R_{41a}O$- group, an $R_{41a}S$-group, a halogen atom, or an $(R_{41a})(R_{43a})N$- group, the suffix d_a represents an integer from 0 to 3, with proviso that when d_a is plural, the two or three R_{59a} groups may be the same or different substituents or they may be divalent groups which are connected to each other to form a cyclic structure, R_{60a} represents an aliphatic group, an aromatic group or a heterocyclic group, R_{61a} represents an aliphatic group, an aromatic group or a heterocyclic group, R_{62a} represents an aliphatic group, an aromatic group, a heterocyclic group, a $R_{41a}OCONH$-group, a $R_{41a}SO_2NH$- group, a $(R_{43a})(R_{44a})NCON(R_{45a})$-group, a $(R_{43a})(R_{44a})NSO_2N(R_{45a})$- group, a $R_{43a}O$- group, a $R_{41a}S$- group, a halogen atom or a $(R_{41a})(R_{43a})N$-group, R_{63a} represents an aliphatic group, an aromatic group, a heterocyclic group, a $R_{43a}CON(R_{45a})$-group, a $(R_{43a})(R_{44a})NCO$- group, a $R_{41a}SO_2N(R_{44a})$- group, a $(R_{43a})(R_{44a})NSO_2$- group, a $R_{41a}SO_2$-group, a $R_{43a}OCO$-group, a $R_{43a}OSO_2$- group, a halogen atom, a nitro group, a cyano group or $R_{43a}CO$-group, and the suffix e_a represents an integer from 0 to 4, with proviso that when there is a plurality of R_{62a} groups, they may be the same or different, wherein R_{41} represents an aliphatic, aromatic or heterocyclic group, R_{42} represents an aromatic or heterocyclic group, and R_{43}, R_{44} and R_{45} each represents a hydrogen atom, an aliphatic group, aromatic group or a heterocyclic group.

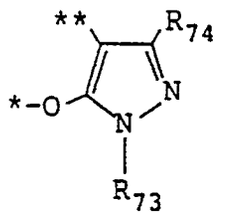
17. The silver halide color photographic material as claimed in claim 2, wherein p is 1 or 2, and B is represented by any of the following general formulae:



(B-1)



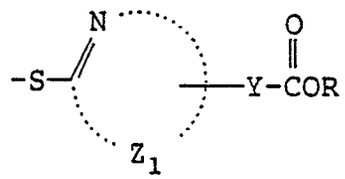
(B-2)



(B-3)

wherein the mark * indicates the position at which it is connected to the group to the left of B of general formula (I), the mark ** indicates the position at which it is connected to the group to the right of B of general formula (I), X_1 and X_4 each represents an oxygen atom or >N-SO_2R_{71}, X_2 and X_3 each represents a methine group or a nitrogen atom,and b represents an integer from 1 to 3, with the proviso that at least one of the X_2 and X_3 groups represents a methine group containing a bonding position represented by the mark ** and that when b is 2 or 3, the plurality of X_2 and X_3 groups may be the same or different,R_{72}, R_{73} and R_{74} each represents a group which allows the groups represented by (B-2) or (B-3) to serve as couplers containing a coupling-separatable group at the mark ** after cleavage at the mark *; and d represents an integer from 0 to 4, with the proviso that when d is 2 to 4, the R_{72} groups may be the same or different.

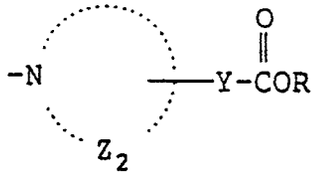
18. The silver halide color photographic material as claimed in claim 2, wherein DI is a group represented by the following general formula (I-a):



(I-a)

wherein Y represents a divalent connecting group containing 8 or less carbon atoms or a bond; R represents a C_{1-6} aliphatic or heterocyclic group; Z_1 represents a nonmetallic atom group required to form a heterocyclic group of carbon and nitrogen atoms; and Z_2 represents a nonmetallic atom group required to form a heterocyclic group (a single or condensed ring) having a nitrogen atom.

19. The silver halide color photographic material as claimed in claim 2, wherein DI is a group represented by the following general formula (I-b)



(I-b)

wherein Y represents a divalent connecting group containing 8 or less carbon atoms or a bond; R represents a C_{1-6} aliphatic or heterocyclic group; Z_1 represents a nonmetallic atom group required to form a heterocyclic group of carbon and nitrogen atoms; and Z_2 represents a nonmetallic atom group required to form a heterocyclic group (a single or condensed ring) having a nitrogen atom.

20. The silver halide color photographic material as claimed in claim 1, wherein the cyan coupler is one capable of releasing a 6-hydroxy-2-pyridon-5-ylazo group.

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 91 10 7617

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
Y,D	DE-A-3 815 469 (AGFA-GEVAERT) * page 2, line 63 - page 3, line 15 * * page 6, line 19 - page 11, line 70 @ page 13, line 40 - page 18, line 4 @ page 19, lines 47 - 50 * * page 21, lines 12 - 25 *	1-3,5-15, 20	G 03 C 7/32 G 03 C 7/333 G 03 C 7/305
Y	JP-A-6 330 424 (FUJI) * page 28, left-hand column *compound (1) *	1-3,5-7, 13,20	
Y	US-A-4 824 772 (ICHIJIMA ET AL.) * column 2, line 28 - column 3, line 41 * * column 19, line 46 - column 20, line 35 @ column 43 *compound (47) @ column 47 *compound (52) * * column 49 *compounds (57),(58) *	1,2,8-12, 14-19	
Y	EP-A-0 318 992 (FUJI) * page 29 *compound (T-144) * * page 39, line 17 *	1,2,8-12, 14-19	
A	PATENT ABSTRACTS OF JAPAN vol. 6, no. 217 (P-152)(1095) 30 October 1982, & JP-A-57 122434 (KONISHIROKU) 30 July 1982, * the whole document *	15	

TECHNICAL FIELDS SEARCHED (Int. Cl.5)
G 03 C

The present search report has been drawn up for all claims

Place of search	Date of completion of search	Examiner
The Hague	09 July 91	MAGRIZOS S.

CATEGORY OF CITED DOCUMENTS

X: particularly relevant if taken alone
Y: particularly relevant if combined with another document of the same catagory
A: technological background
O: non-written disclosure
P: intermediate document
T: theory or principle underlying the invention

E: earlier patent document, but published on, or after the filing date
D: document cited in the application
L: document cited for other reasons

&: member of the same patent family, corresponding document