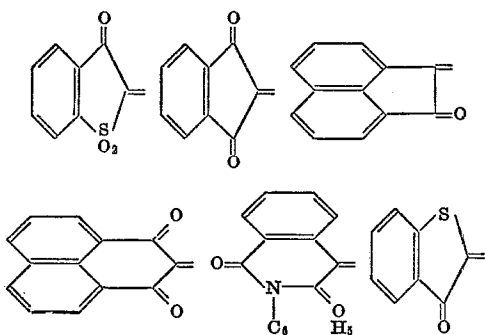




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acid, those of the isoxazolone, oxindole, 2-thio-2,5-thiazolidine dione or 2,4 - imidazolinedione series, or the ketomethylene compounds characterized by the following structural formulas:



$R^4$ =a hydrogen atom, an alkyl group having up to 4 carbon atoms, hydroxyl, alkoxy having up to 4 carbon atoms such as methoxy or ethoxy, or aryl such as phenyl;

$R^5$ =(1) a saturated or unsaturated aliphatic group having preferably up to 6 carbon atoms which may be substituted, e.g. with phenyl, hydroxyl, halogen such as chlorine, bromine or iodine, with amino, carboxyl, sulfo or (2) aryl such as phenyl or naphthyl;

X=oxygen or sulfur;

$n=0, 1, \text{ or } 2$ ;

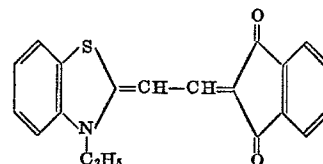
Q=the members required to complete a 5-membered or 6-membered heterocyclic group which may contain a condensed benzene or naphthalene ring and further substituents. Suitable heterocyclic groups are those customarily found in cyanine chemistry, for example those of the thiazole series (e.g. thiazole, 4 - methylthiazole, 5-methylthiazole, 4,5 - dimethylthiazole, 4-phenylthiazole, 5-phenylthiazole, 4,5-diphenylthiazole, benzothiazole, 4-chlorobenzothiazole 5-chlorobenzothiazole, 6-chlorobenzothiazole, 7-chlorobenzothiazole, 6-bromobenzothiazole, 5 - iodobenzothiazole, 6-iodobenzothiazole, 4-methylbenzothiazole, 5-methylbenzothiazole, 6-methylbenzothiazole, 5,6 - dimethylbenzothiazole, 4-phenylbenzothiazole, 5-phenylbenzothiazole, 6-phenylbenzothiazole, 5 - hydroxybenzothiazole, 6 - hydroxybenzothiazole, 4 - methoxybenzothiazole, 5 - methoxybenzothiazole, 6-methoxybenzothiazole, 5-ethoxybenzothiazole, 6-ethoxybenzothiazole, 5,6-dimethoxybenzothiazole, 5,6-methylene-dioxybenzothiazole, 5-diethylaminobenzothiazole, 6 - diethylaminobenzothiazole, 5-carboxybenzothiazole, 5-sulphobenzothiazole, tetrahydrobenzothiazole, 7 - oxotetrahydrobenzothiazole, naphtho[1,2-d]thiazole, naphtho[2,1-d]thiazole, 5-methoxynaphtho[2,1-d]thiazole, 5 - ethoxynaphtho[2,1-d]thiazole, 7 - methoxynaphtho[2,1 - d]thiazole, 8-methoxynaphtho[1,2 - d]thiazole, etc.), those of the selenazole series, (e.g. 4 - methylselenazole, 4-phenylselenazole, benzoselenazole, 5 - chlorobenzoselenazole, 5,6 - dimethylbenzoselenazole, 5 - hydroxybenzoselenazole, 5 - methoxybenzoselenazole, tetrahydrobenzoselenazole, naphtho[1,2 - d]selenazole or naphtho[2,1 - d]selenazole), those of the oxazole series (e.g. oxazole, 4 - methylloxazole, 4 - phenyloxazole, 4,5-diphenyloxazole, benzoxazole, 5 - chlorobenzoxazole, 6-chlorobenzoxazole, 5,6 - dimethylbenzoxazole, 5-phenylbenzoxazole, 5 - hydroxybenzoxazole, 5-methoxybenzoxazole, 5 - ethoxybenzoxazole, 6 - dialkylaminobenzoxazole, 5 - carboxybenzoxazole, 5 - sulphobenzoxazole, 5 - sulphonamidobenzoxazole, 5 -  $\beta$ -carboxyvinylbenzoxazole, naphtho[1,2 - d]oxazole, naphtho[2,1 - d]oxazole or naphtho[2,3 - d]oxazole), those of the imidazole series (e.g. 1 - methylimidazole, 1 - ethyl - 4 - phenylimidazole, 1 - butyl - 4,5-dimethylimidazole, 1 - methylbenzimidazole, 1 - butyl-4-methylbenzimidazole, 1 - ethyl - 5,6 - dichlorobenzimidazole, 1 - ethyl - 5 - trifluoromethylbenzimidazole, 1-methyl-

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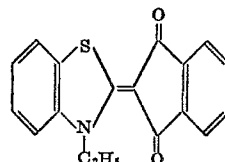
naphtho[1,2 - d]imidazole or 1 - ethylnaphtho[2,3-d]imidazole), those of the 3,3 - dialkylindolenine series (e.g. 3,3 - dimethylindolenine, 3,3,5 - trimethylindolenine, 3,3 - dimethyl - 5 - methoxyindolenine, etc.), those of the pyridine series (e.g. pyridine, 3 - methylpyridine, 4 - methylpyridine, 6 - methylpyridine, 3,4-dimethylpyridine, 3,5 - dimethylpyridine, 3,6 - dimethylpyridine, 4,6 - dimethylpyridine, 4 - chloropyridine, 5 - chloropyridine, 6 - chloropyridine, 3 - hydroxypyridine, 4 - hydroxypyridine, 6 - hydroxypyridine, 3-phenylpyridine, 4 - phenylpyridine, 2,3 - dimethylpyridine, 2,6 - dimethylpyridine, 2 - hydroxypyridine, quinoline, 3 - methylquinoline, 5 - methylquinoline, 7-methylquinoline, 8 - methylquinoline, 6 - chloroquinoline, 8 - chloroquinoline, 6 - methoxyquinoline, 6-ethoxyquinoline, 6 - hydroxyquinoline, 8 - hydroxyquinoline, 5 - oxo - 5,6,7,8 - tetrahydroquinoline, isoquinoline or 3,4 - dihydroisoquinoline), those of the thiazoline series (e.g. thiazoline, 4 - methylthiazoline, etc.) and those of the pyrrole, tetrahydropyridine, thiadiazole, oxadiazole, pyrimidine, triazine or benzothiazine series. The heterocyclic rings may carry any substituents, e.g., they may be substituted with other alkyl groups preferably containing up to 3 carbon atoms, such as methyl or ethyl, with halogens such as chlorine, iodine or bromine, with trifluoromethyl or hydroxyl or with alkoxy having preferably up to 3 carbon atoms such as methoxy or ethoxy, with hydroxyalkyl, alkylthio or aryl such as phenyl or aralkyl such as benzyl or amino or substituted amino and the like.

[N - alkylbenzothiazole]-dimethine[1,3 - dialkylthio-barbituric acid]merocyanines are examples of suitable compounds and the following are more specific examples:

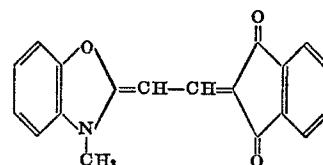
(1)



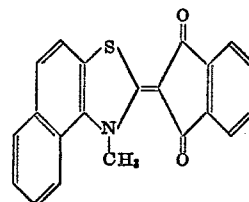
(2)



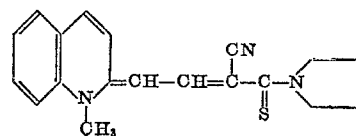
(3)



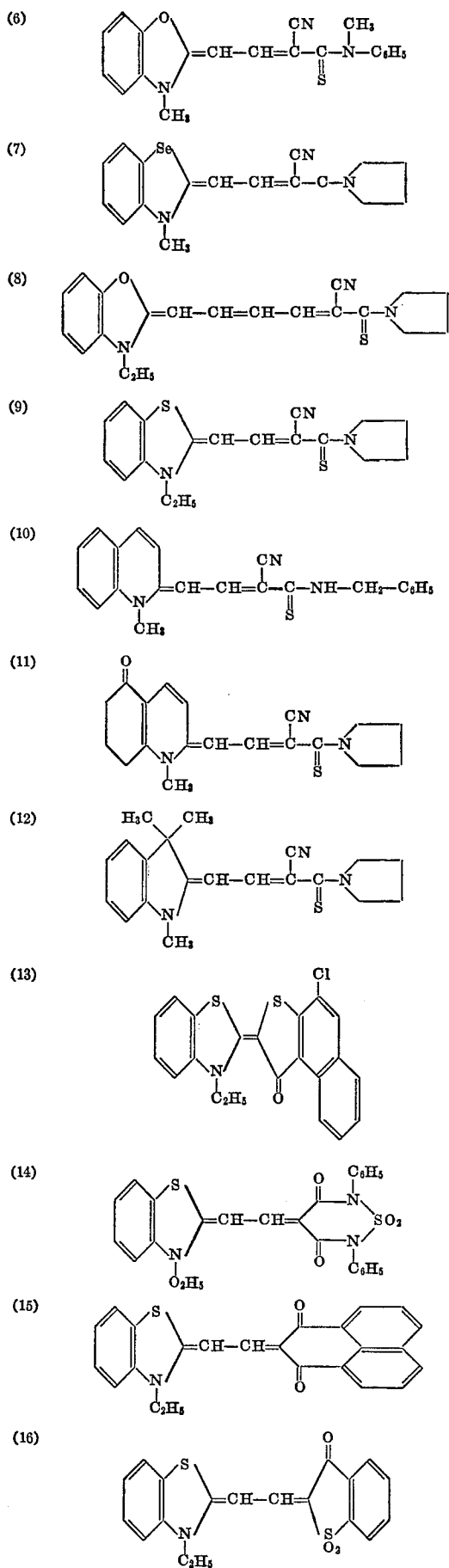
(4)



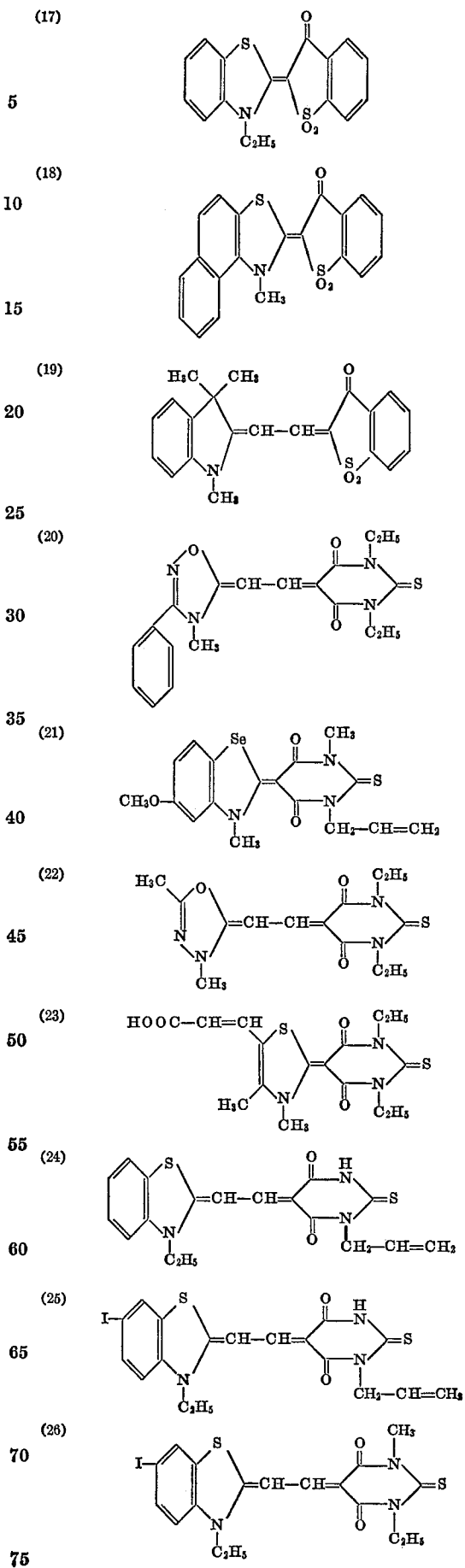
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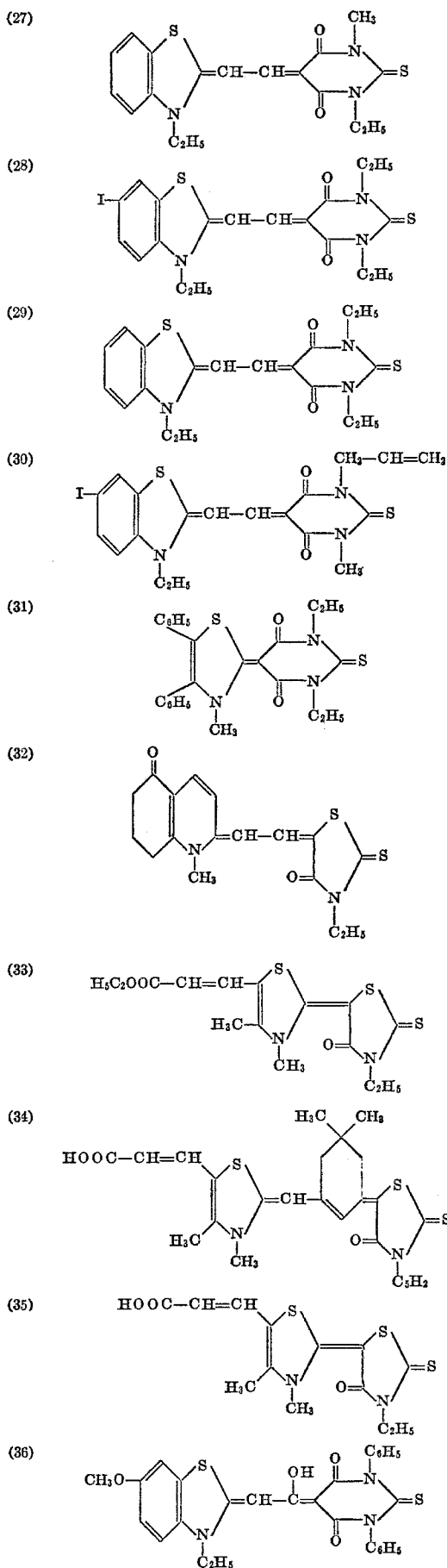
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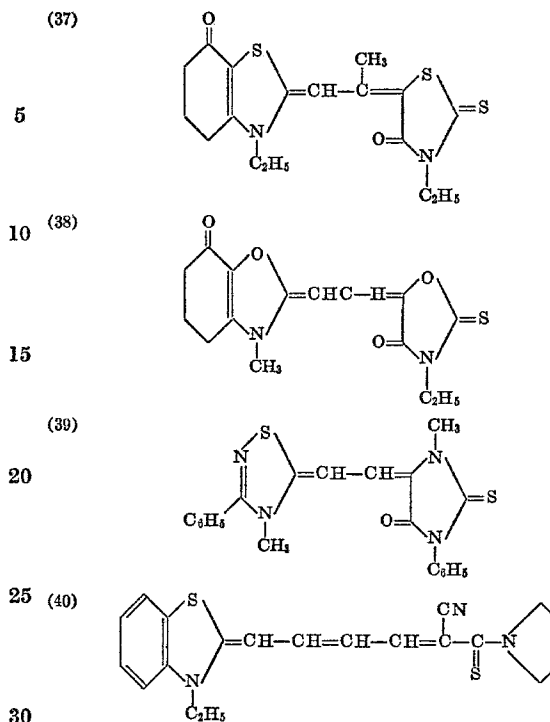
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The merocyanines according to the invention may be prepared by known methods (see F. M. Hamer "The Cyanine Dyes and Related Compounds" (1964). Preparation of the open chain cyanothioacetamide merocyanines has been described in British patent specification No. 1,219,330.

The image-producing compounds must meet the following conditions:

(1) They must react in the presence of the sensitizers defined above upon exposure to form non-transferable reaction products.

(2) They must be transferable at temperatures of between 80 and 200° C. to the image receiving layer.

(3) They must be capable of reacting with the compounds of the image receiving layer to form colored reaction products.

The following reducing agents, for example are particularly suitable image-producing compounds:

(1) Phenols and naphthols, especially benzene compounds of the naphthalene series containing at least two aromatic hydroxyl groups which may be partly etherified such as 1-hydroxy-4-alkoxy-naphthalenes, or which are substituted with a hydroxyl group and an amino or substituted amino group, substituted being in the para- or ortho-position in the case of benzene derivatives. Examples of such compounds are listed in Table 1 below.

TABLE 1

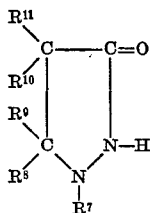
- 1-hydroxy-4-methoxynaphthalene,  
 1-hydroxy-4-ethoxynaphthalene,  
 1-hydroxy-2-methyl-4-methoxynaphthalene,  
 4,4'-dimethoxynaphthalene-1,1'-dihydroxy-2,2'-binaphthyl,  
 1,4-dihydroxynaphthalene,  
 1-hydroxy-4-aminonaphthalene,  
 1,2,3-trihydroxy-5-acetylbenzene,  
 methyl 3,4,5-trihydroxybenzoate,  
 ethyl 3,4,5-trihydroxybenzoate,  
 1,2,3,4-tetrahydro-8-hydroxyquinoline,  
 1-(2'-methylsulphonamidoethyl)-1,2,3,4-tetrahydro-6-hydroxyquinoline,  
 4-methylaminophenol,  
 4-isopropylideneaminophenol,  
 4-aminophenol,  
 4-hydroxyanilinomethanesulfonic acid,

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4-hydroxy-3-methylanilinomethanephosphonic acid,  
1-hydroxy-4-propoxynaphthalene.

Also suitable as image-producing compounds are the aminophenol developers described in German patent specifications 1,159,758, 1,200,679, 1,203,129 and 1,203,605.

(2) Pyrazolidone-(3) derivatives of the following formula:



wherein

R<sup>7</sup> represents hydrogen, alkyl or aryl which may be substituted for example with lower alkyl or alkoxy groups or with halogen such as chlorine, bromine, or iodine, and

R<sup>8</sup>, R<sup>9</sup>, R<sup>10</sup> and R<sup>11</sup> represent hydrogen, alkyl, aryl or a substituted alkyl or aryl group.

The compounds listed in the following table have been found to be suitable.

TABLE 2

1-phenyl-3-pyrazolidone,  
1-m-tolyl-3-pyrazolidone,  
1-p-tolyl-3-pyrazolidone,  
1-phenyl-4-methyl-3-pyrazolidone,  
1-phenyl-5-methyl-3-pyrazolidone,  
1,4-dimethyl-3-pyrazolidone,  
4-methyl-3-pyrazolidone,  
4,4-dimethyl-3-pyrazolidone,  
1-phenyl-4,4-dimethyl-3-pyrazolidone,  
1-(m-chlorophenyl)-4-methyl-3-pyrazolidone,  
1-(p-chlorophenyl)-4-methyl-3-pyrazolidone,  
1-(m-chlorophenyl)-3-pyrazolidone,  
1-(p-chlorophenyl)-3-pyrazolidone,  
1-(p-tolyl)-4-methyl-3-pyrazolidone,  
1-(o-tolyl)-4-methyl-3-pyrazolidone,  
1-(p-tolyl)-3-pyrazolidone,  
1-(m-tolyl)-3-pyrazolidone,  
1-(m-tolyl)-4,4-dimethyl-3-pyrazolidone,  
1-(2'-trifluoroethyl)-4,4-dimethyl-3-pyrazolidone,  
5-methyl-3-pyrazolidone.

The compounds listed above may be prepared by the processes described in British patent specification Nos. 679,677 and 679,678, the "phenimines" which may be obtained by reacting acrylonitrile derivatives with the corresponding hydrazine compounds being saponified to 3-pyrazolidones.

3-pyrazolidones may also be prepared by the process described in British patent specification No. 703,669, the end products being obtained by direct condensation of esters of acrylic acid or derivatives thereof with hydrazines. This process is particularly suitable for reactions with hydrazine itself. The 3-pyrazolidones obtained in this way which have an oily consistency can be obtained as crystalline compounds by converting them into salts, e.g. hydrochlorides, sulfates, or 1,5-naphthalene disulfonates. The preparation of 4,4-dialkyl-3-pyrazolidones has been described in U.S. Pat. 2,772,282. In the process described there, 2,2-dialkyl-β-chloropropionic acid chlorides are reacted with hydrazines.

The 3-pyrazolidones may be used as free bases or in the form of their salts.

(3) N,N-dialkylphenylenediamine derivatives, especially those in which the alkyl groups contain up to 3 carbon atoms and the phenylene nucleus may be substituted with alkyl or alkoxy groups.

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The free primary amino groups may be blocked, for example in the form of a Schiff's base by reaction with aldehyde, especially benzaldehyde, or with a sulfomethyl group which may be introduced by a Mannich reaction. Phenylenediamine derivatives which have a blocked primary amino group are particularly suitable because stable layers can easily be prepared with such compounds.

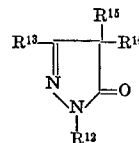
The compounds listed in the following table have been found to be suitable.

TABLE 3

N,N-diethyl-p-phenylenediaminesulfate,  
N,N'-dibenzylidene-p-phenylenediamine,  
N,N-diethyl-N'-sulfomethyl-p-phenylenediamine,  
N,N-dimethyl-N'-sulfomethyl-p-phenylenediamine,  
3-methyl-4-sulfomethylamino-N,N-diethylaniline  
N-benzylidene-N',N'-diethyl-p-phenylenediamine,  
3-methoxy-4-sulfomethylamino-N,N-diethylaniline.

The image producing substances of the above mentioned type are known per se. Methods of their preparation may be found in German patent specification Nos. 1,159,758 and 1,203,129, and in the literature.

(4) Pyrazolin-5-one derivatives: Those pyrazolin-5-one derivatives which contain at least one hydrogen in the 4-position or a 4-aminophenyl-amino group are preferred, such as those of the formula:



wherein

R<sup>12</sup>=(1) hydrogen, (2) a saturated or olefinically unsaturated aliphatic group preferably containing up to 6 carbon atoms which may be substituted, e.g. with phenyl as in the benzyl group, with cyano, with halogen, e.g. fluorine, with amino in which case the amino group may itself be substituted, e.g. alkylated amino groups, especially dialkylamino, the alkyl groups in the alkylamino group preferably containing up to 3 carbon atoms, (3) aryl, especially a group of the phenyl series, the aryl ring being itself optionally substituted, e.g. with alkyl or alkoxy preferably containing up to 3 carbon atoms, with nitro, halogen such as fluorine, chlorine or bromine, with amino or substituted amino groups, e.g. alkylated amino groups, (4) a heterocyclic group, e.g. benzothiazolyl or (5) cycloalkyl such as cyclohexyl or cyclopentyl;

R<sup>13</sup>=(1) hydrogen, (2) a saturated or olefinically unsaturated aliphatic group preferably containing up to 6 carbon atoms, the aliphatic group optionally carrying further substituents, e.g. phenyl as in the case of a benzyl or phenylethyl, halogen such as fluorine, chlorine or bromine, alkoxy, alkoxy, (3) aryl, especially a group of the phenyl series, the aryl ring being optionally itself substituted, e.g. with alkyl or alkoxy having preferably up to 3 carbon atoms, or with nitro or halogen such as chlorine or bromine, (4) a heterocyclic group, e.g. pyridyl, (5) cycloalkyl such as cyclohexyl or cyclopentyl, (6) hydroxyl which may be etherified, especially with short chained aliphatic radicals having up to 3 carbon atoms, (7) amino which may be substituted, e.g. with alkyl, preferably having up to 3 carbon atoms, or (8) alkoxy having up to 5 carbon atoms;

R<sup>14</sup>=(1) hydrogen, (2) a saturated or olefinically unsaturated aliphatic group having preferably up to 6 carbon atoms which may be substituted, e.g. with phenyl as in the case of benzyl or phenylethyl groups, or with halogen such as chlorine or bromine, nitrile, alkoxy, alkoxy, alkoxy or anilino, (3) aryl,

especially a group of the phenyl series, in which the aryl ring may itself be substituted, e.g. with alkyl or alkoxy having preferably up to 3 carbon atoms, nitro or nitrile, (4) amino in which the amino groups may be substituted, e.g. with alkyl having preferably up to 3 carbon atoms, cycloalkyl, phenyl or acyl, especially acyl groups of short chained aliphatic carboxylic acid, (5) alkoxy having preferably up to 3 carbon atoms; R<sup>15</sup>=hydrogen or a 4-aminophenylamino group or 4-di-alkylaminophenylamino group.

Furthermore, R<sup>13</sup> and R<sup>14</sup> may together represent the ring members required to complete a 5-membered or 6-membered carbocyclic or heterocyclic ring.

Suitable compounds are listed in the following table where the first 33 compounds have H for R<sup>15</sup>.

TABLE 4

Pyrazolin-5-one No.	R <sup>12</sup>	R <sup>13</sup>	R <sup>14</sup>
1	C <sub>6</sub> H <sub>5</sub>	NH <sub>2</sub>	H
2	H	CH <sub>3</sub>	H
3	C <sub>6</sub> H <sub>5</sub>	CH <sub>3</sub>	H
4	Cyclohexyl	CH <sub>3</sub>	H
5	p-Tolyl	CH <sub>3</sub>	H
6	m-Chlorophenyl	CH <sub>3</sub>	H
7	2,5-dichlorophenyl	CH <sub>3</sub>	H
8	3-nitrophenyl	CH <sub>3</sub>	H
9	4-nitrophenyl	CH <sub>3</sub>	H
10	C <sub>6</sub> H <sub>5</sub>	-COOC <sub>2</sub> H <sub>5</sub>	H
11	1-phenylethyl	C <sub>6</sub> H <sub>5</sub>	H
12	C <sub>6</sub> H <sub>5</sub>	-CH <sub>2</sub> COOC <sub>2</sub> H <sub>5</sub>	H
13	C <sub>6</sub> H <sub>5</sub>	Pyridyl-4	H
14	2-diethylaminoethyl	CH <sub>3</sub>	H
15	2,4,6-trichlorophenyl	CH <sub>3</sub>	H
16	2,4,6-trichlorophenyl	CH <sub>3</sub>	H
17	2-cyanoethyl	C <sub>6</sub> H <sub>5</sub>	H
18	3,5-dimethylphenyl	-COOC <sub>2</sub> H <sub>5</sub>	H
19	C <sub>6</sub> H <sub>5</sub>	OH	H
20	3-nitrophenyl	CH <sub>3</sub>	CH <sub>3</sub>
21	3-aminophenyl	CH <sub>3</sub>	CH <sub>3</sub>
22	3-nitrophenyl	CH <sub>3</sub>	Benzyl
23	2-ethoxyphenyl	C <sub>6</sub> H <sub>5</sub>	CH <sub>3</sub>
24	C <sub>6</sub> H <sub>5</sub>	C <sub>6</sub> H <sub>5</sub>	CH <sub>3</sub>
25	C <sub>6</sub> H <sub>5</sub>	C <sub>6</sub> H <sub>5</sub>	C <sub>2</sub> H <sub>5</sub>
26	C <sub>6</sub> H <sub>5</sub>	-CH <sub>2</sub> -CH <sub>2</sub>	-C <sub>2</sub> H <sub>4</sub> -CH <sub>2</sub> -
27	C <sub>6</sub> H <sub>5</sub>	CH <sub>3</sub>	-CH <sub>2</sub> CONHC <sub>6</sub> H <sub>5</sub>
28	C <sub>6</sub> H <sub>5</sub>	-COOC <sub>2</sub> H <sub>5</sub>	CH <sub>3</sub>
29	C <sub>6</sub> H <sub>5</sub>	-CH <sub>2</sub> -CH <sub>2</sub>	-CH <sub>2</sub> -S-C
30	3-nitrophenyl	CH <sub>3</sub>	C <sub>2</sub> H <sub>5</sub>
31	C <sub>6</sub> H <sub>5</sub>	CH <sub>3</sub>	-NHCOCH <sub>3</sub>
32	C <sub>6</sub> H <sub>5</sub>	-COOC <sub>2</sub> H <sub>5</sub>	-OC <sub>2</sub> H <sub>5</sub>
33	C <sub>6</sub> H <sub>5</sub>	CH <sub>3</sub>	CH <sub>3</sub>
34	1-[benzothiazolyl-2]-3,4-dimethyl-4-[4-diethylaminophenylamino]-pyrazolin-5-one		
35	1-phenyl-3,4-dimethyl-4-[p-diethylaminophenylamino]-pyrazolin-5-one		

The pyrazolin-5-one derivatives are prepared by methods known from the literature. Reference may be made e.g. to the monograph by R. H. Wiley "Pyrazolones, Pyrazolidones and Derivatives" (1964) and German patent specification No. 1,155,675.

The light sensitive layers contain at least one of the sensitizers in quantities of 10 to 300 mg./m.<sup>2</sup> and one or more image producing compounds in quantities of 0.02 to 0.5 g./m.<sup>2</sup>. This range of concentration has been found to be suitable although amounts outside this range may, of course, be employed. The concentration depends mainly on the requirements of a particular reproduction process.

Particularly suitable combinations of sensitizers with the image producing reducing agents can be determined by simple tests. Suitable tests for this purpose will be described hereinafter. The choice of solvent and of the binding agent used for producing the light sensitive layer is also important for obtaining optimum results. Combinations of components particularly suitable for any given purpose can be determined by the usual tests known to the ordinary skilled person.

To prepare the light sensitive layer, the sensitizers and image producing compound may be suspended or dissolved in solvents and mixed with a binding agent and applied in this form to the layer support.

The usual natural or synthetic film-forming polymers are suitable as binding agents for the light sensitive layer, e.g. proteins, especially gelatin, cellulose derivatives, especially cellulose ethers, cellulose esters or carboxymethyl cellulose, alginic acid and its derivatives, starch ether or galactomannane, polyvinyl alcohol, polyvinyl pyrrolidone,

polyvinyl chloride, copolymers of vinyl chloride and vinyl acetate, polyvinyl acetate or completely or partly saponified polyvinyl acetate or copolymers of vinyl acetate, for example with olefines such as ethylene or propylene and copolymers of monomers of acrylic or methacrylic acid or derivatives thereof such as esters, amides of nitriles, etc. The light sensitive layers may be used as self-supporting layers or applied to a support. Suitable supports are e.g. paper, especially baryta-coated or polyolefine-coated, more particular polyethylene-coated paper and cellulose esters, e.g. cellulose triacetate, polyesters, especially those based on ethylene terephthalate, glass, etc.

The image receiving material advantageously consists of an image receiving layer applied to a suitable support. Substantially the same substances as those described above

for the light sensitive material are suitable as binding agent for the image receiving layer or as the layer support.

When choosing a binder for the light sensitive layer and the image receiving layer, care should be taken to ensure that the layers will not stick at elevated temperature. These difficulties, however, are well-known from other transfer processes, e.g. the silver salt diffusion process or heat development processes, and can easily be solved by making use of the experience gained in these known fields.

The image receiving layer contains compounds which should be insensitive, or as restricted as possible in their sensitivity, to visible light under the conditions of the process of the invention, and which react with the transferred image producing compounds to form colored products. Numerous compounds have been found suitable for this purpose. Chemically, these compounds belong to a wide variety of classes so that their systematic chemical classification is not possible. However, suitable compounds or suitable combinations of an image producing compound arranged in the light sensitive layer and of the reactant for the image forming reaction in the image receiving layer can be sufficiently clearly defined by simple laboratory tests customarily employed in the art. Thus, for example, the two reactants must react when briefly heated for a few seconds to a temperature of between about 80 and 200° C. to form a stable dye. A second test must then be carried out to choose suitable image producing compounds. The purpose of this test is to show whether the image producing compound will react sufficiently rapidly in the presence of the merocyanine dye on exposure to light, so that, when the mixture is heated after it has been

exposed, it will not produce a colored compound with the reactant in the image receiving layer.

The following classes of compounds are examples of suitable compounds in the image receiving layer for reaction with the image producing compound transferred from the light sensitive layer.

(1) Heavy metal compounds, especially compounds of metals of Groups IIIa to Va and Groups Ib, IIb and VIb and VIII of the Periodic Table, e.g. compounds of the following heavy metals: cadmium, mercury, iron, cobalt, nickel, copper, silver, gold, bismuth or thallium. Salts of these metals with long chained aliphatic, carboxylic acids are especially suitable e.g. nickel stearate, cobalt palmitate, iron stearate, and the addition compound of bismuth nitrate with amines such as triethanolamine. It is found to be especially suitable to use silver compounds which are substantially insensitive to light under the conditions of the copying process according to the invention, e.g. the silver salts described in U.S. Pat. No. 3,330,663, i.e. silver salts of aliphatic carboxylic acids having a thioether group or silver salts of long-chained fatty acids such as silver behenate, silver palmitate or silver stearate, etc. containing 8 to 24 carbon atoms. When the above-mentioned heavy metal compounds are used, brown to black copies are obtained. The image consists of the particular metal and/or a reaction product of transferred image producing compound.

(2) The image receiving layer may also contain oxidizing agents and dye components which react imagewise with the transferred image producing compound by oxidative coupling to form dyes, e.g. the known color couplers of color photography which couple with oxidized phenylene diamine derivatives to produce dyes, or compounds which when in their oxidized form react under oxidizing conditions e.g. with pyrazolin-5-one compounds to yield colored coupling products. Suitable reactants for these reactions are e.g. oxidation products of p-phenylene diamines or their derivatives which react with pyrazolin-5-one compounds to form the azomethine dyes known in conventional color photography.

(3) Diazonium salts which react with the transferred reducing agent, e.g. with aminophenols, aminonaphthols, phenylenediamine derivatives or pyrazolin-5-one compounds, to form a colored product. This reaction is similar in principle to the one employed in the known photographic diazo type process.

(4) Leucophthalocyanines are also suitable for use as reactants for the reaction which produces the image dye. Leucophthalocyanines which have not or could not be prepared from finished phthalocyanines are known as phthalocyanine precursors. This term is used, for example, in the article by B. R. A. Brooks, J. G. Burt, B. F. Skiles and M. S. Whelen, *J. Org. Chem.* 24, page 383 (1959). In the relevant chapter in Ullmanns Encyclopädie der Technischen Chemie, 3rd edition, volume 13, the term "phthalocyanometal complexes" is used for the same type of materials for which in the present context the term "leucophthalocyanines" is used. The last mentioned expression is explained e.g. in U.S. Pat. No. 2,772,285. Although that patent refers only to leuco copper phthalocyanine, its explanation is also applicable analogously to the corresponding complexes with other metals which form phthalocyanines. Leucophthalocyanines according to this definition are colorless or only slightly colored products in which the phthalocyanine structure is already completely formed, and which can be converted into phthalocyanines by a reduction process. In this reduction process, constituents which the leucophthalocyanine molecule contains in addition to phthalocyanine may also be split off. Such leucophthalocyanines may be prepared e.g. by first preparing a phthalocyanine, e.g. a phthalocyanine which is free from metal or CuPc (Pc=phthalocyanine), NiPc, CoPc or ZnPc and then treating the phthalocyanine with additional ligands under oxidizing conditions, or by heating a reaction mixture which is in itself suitable for the preparation

of a phthalocyanine to a temperature slightly below that required for the preparation of the phthalocyanine, or by carrying out the reaction without the reduction potential required for formation of the phthalocyanine.

Leucophthalocyanine which contain metal are more suitable for this reaction because those which are free from metal are relatively unstable. The highly stable and only slightly colored leuco cobalt phthalocyanines are especially suitable.

Especially to be mentioned are the leuco cobalt phthalocyanines which are described in *Angewandte Chemie*, 68, page 145 (1956), e.g. the phthalocyanine cobalt ethylene diamine complex.

Instead of ethylene diamine, other diamines or polyamine may also be used as ligands, for example propylene diamine-(1,2), and -(1,3), monoethylpropylene diamine-(1,3), hydroxyethylethylene diamine, N-methyl-N- $\beta$ -hydroxyethylpropylene diamine, N,N'-diethylethylene diamine, N,N-di-( $\beta$ -aminoethyl)-ethylene diamine, N,N'-di-( $\beta$ -aminoethyl)-ethylene diamine or N,N-di-[ $\beta$ -aminoethyl-aminoethyl]-amine or also monoamines such as 3-(2'-ethylhexyloxy)-propylamine-(1) or stearylamine. The solubility properties of the leuco-CoPc depend on the type of amine used in the molecule.

In another embodiment of the process of the present invention, reactants can be completely omitted from the image-receiving layer. This applies e.g. to image producing compounds for example phenols or naphthols which yield sufficiently colored compounds when heated alone or in the presence of atmospheric oxygen. In this case, plain, uncoated paper may be used as image receiving material.

In addition to the image producing reactants properly speaking, the image receiving layers may contain other additives which advantageously influence the color tone, contrast, stability, etc., of the copy. Image receiving layers of this type are already known and have been described, for example, in German auslegeschrift Nos. 895,101; 1,003,577; 1,159,758; 1,004,043 and 1,165,410, in Dutch patent specification No. 277,086, in U.S. Pat. 3,335,006 and in Belgian patent specification Nos. 614,064 and 609,057.

The image receiving layers may also contain white pigments, e.g. zinc oxide, silicon oxide or titanium dioxide as fillers, for improving the whites and for controlling the tendency of the layers to stick, and they may contain terpene resins and organic acids for improving the stability in storage. Image receiving layers of this type have been described in U.S. Pat. Nos. 3,074,809 and 3,107,174.

The color tone of the images produced can be altered e.g. with compounds of the 1-(2H)-phthalazinone series. Toners of this type have been described in U.S. Pat. Nos. 3,080,254 and 3,446,648. Additives which accelerate the reduction reaction in the image receiving layer have also been found to be advantageous. Suitable compounds for this purpose are e.g. sterically hindered phenols such as 2,6-di-tertiary butyl-p-cresol. Compounds of this type have been described in U.S. Pat. 3,218,166. Furthermore, the image tone and image density can be improved with certain metal salts, e.g. copper-II stearate. Metal ion image intensifiers and their application have been described in German auslegeschrift No. 1,572,209.

The usual sources of light used in reproduction work, such as mercury lamps, iodine quartz lamps or incandescent lamps, may be used for exposing the light sensitive layers according to the invention. The spectral sensitivity of the light sensitive material depends on the nature of the dye used or the combination of dye and reducing image producing compound.

Exposure may be carried out either in contact or optically or by reflection.

Transfer of the image producing compounds from the unexposed areas of the light sensitive layers to the image receiving layer is carried out by heating at temperatures of between 80 and 200° C. Heating may be effected e.g.

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by passing the exposed light sensitive layer in contact with the image receiving layer over hot plates or rollers or by irradiation with infra-red light. The most advantageous temperature and heating time depend, of course, on the nature of the image producing compound and can be determined by a few simple tests.

According to one variation of the material according to the invention, the image receiving layer and the light sensitive layer may be combined on one support. In this case, it is necessary to use a transparent support on which the image receiving layer, e.g. a layer containing silver behenate dispersed in a copolymer of styrene and isobutylene, is applied first, and the light sensitive layer, e.g. an ethylcellulose layer containing the sensitizer and reducing compound, is applied on the image receiving layer.

The sensitivity of these light sensitive layers may advantageously be increased or extended to other spectral regions to an extent depending on the absorption of the compounds according to the invention by combining the dyes used according to the invention with photoreducible dyes, e.g. those mentioned in U.S. Pat. 3,094,417, such as erythrosine.

## EXAMPLE 1

## Light sensitive material

The following casting solution is applied to a layer support of glassine paper:

30 mg. of dye No. 28,  
50 mg. of 1-hydroxy-4-methoxynaphthalene,  
2.5 g. of ethyl cellulose as a 5% solution in butanone-2  
and  
150 ml. of butanone-2.

The layer is dried in the usual manner.

The image receiving layer is prepared by grinding the following components for 6 hours in a ball mill:

2.1 g. of a mixture of 1 mol silver behenate and 1 mol behenic acid,  
1.66 g. of terpene resin,  
0.86 g. of 1-(2H)-phthalazinone,  
4.8 g. of zinc oxide,  
0.56 g. of silica gel,  
0.37 g. of 2,6-di-tert.butyl-4-methylphenol,  
0.034 g. of tetrachlorophthalic acid anhydride,  
15 g. of an 8% ethylmethacrylate solution in pentanone-3,  
80 g. of a 1.5% polyvinyl acetate solution in butyl acetate,  
and  
30 g. of butylacetate,

and applying the resulting mixture to paper and drying. When dry, the layer contains about 0.2 of silver per m.<sup>2</sup> in the form of silver behenate.

## Processing

The light sensitive material is exposed behind a  $\sqrt{2}$  step wedge to a 1000 watt iodine quartz lamp from a distance of 30 cm. for 5 minutes.

The exposed layer is then brought into contact with the image receiving layer and heated to a temperature of 125° C. for 10 seconds or treated in an ordinary commercial heat development apparatus.

The results are listed in Table 5 below along with those obtained using the other dyes instead of dye No. 28.

TABLE 5

Dye No.:	Steps $\sqrt{2}$
1	9
2	3
3	3
4	6
5	3
6	3
7	7
8	9
9	7
10	6

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TABLE 5—Continued

Dye No.:	Steps $\sqrt{2}$
11	6
12	8
13	5
14	2
15	9
16	10
17	1-2
18	6
19	3
20	2
21	3
22	4
23	5
24	7
25	4
26	8
27	10
28	12
29	11
30	12
31	7
32	1
33	4
34	2
35	3
36	5
37	6
38	1
39	3

## EXAMPLE 2

## Light sensitive material

The following casting solution is applied to a layer support of glassine paper:

30 mg. of dye No. 27  
50 mg. of 1-hydroxy-4-methoxynaphthalene,  
2.5 g. of ethylcellulose  
150 ml. of ethyl acetate.

The layer is dried in the usual manner.

## Processing

The light sensitive material is exposed through a positive transparent original to a 1000 watt normal incandescent lamp (tungsten filament) at a distance of 5 to 10 cm. for 30 seconds.

Instead of a transparent original, a text printed on ordinary paper may be used. The exposure to reflected light required in this case takes 15 to 25 seconds under otherwise the same conditions.

The exposed layer is then brought into contact with the image receiving layer described in Example 1 and the layers are heated to a temperature of 125 to 140° C. for 5 to 20 seconds or treated in an ordinary commercial heat development apparatus.

A sharp, positive black copy of the original is obtained.

Other combinations may be used instead of dye No. 27 and instead of the image producing compound mentioned above. The results are summarized in Table 6 below.

TABLE 6

Dye No.	Image producing compound	Quantity in mg.	Color of the copy
16	1-hydroxy-5-methoxynaphthalene	50	Grey black.
29	Compound 3 of Table 4	100	Do.
16	Compound 33 of Table 4	100	Do.
1	Compound 26 of Table 4	100	Do.
28	Compound 20 of Table 4	100	Do.
29	1,2,3,4-tetrahydro-8-hydroxyquinoline	50	Black brown.
1	1-phenyl-3-pyrazolidone	50	Dark brown.
1	1-phenyl-5-methyl-3-pyrazolidone	50	Black brown.
16	do	50	Do.
29	do	50	Do.
1	N-benzylidene-N,N'-diethyl-p-phenylenediamine	50	Brown.

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Other silver compounds may be used instead of the silver behenate used in the image receiving layer in this case, e.g. silver stearate or silver salts of octadecylmercaptoacetic acid, 2-octadecylmercapto-5-carboxymethylmercapto-1,3,4-thiadiazole (as described in U.S. Pat. 3,330,663) etc. The choice of suitable compounds depends on the purpose for which they are to be used and the required color of the image.

## EXAMPLE 3

## Light sensitive material

A light sensitive layer is prepared from a solution of 30 mg. of dye No. 1, 100 mg. of N,N-diethyl-N'-sulfomethyl-p-phenylenediamine, 100 mg. of sodium acetate, 100 ml. of ethanol, and 50 ml. of a 1.5% solution of a cellulose ether in ethanol by casting the solution on paper and drying.

## Image receiving material

A layer is prepared from a solution 1.5 g. of 1-phenyl-3-methylpyrazolone-(5), 1 g. of sodium bromate, 35 ml. of water and 15 ml. of a 5% aqueous polyvinyl alcohol solution, by casting the solution on paper and drying.

## Processing

Processing is carried out as in Example 2. A red positive is obtained.

## EXAMPLE 4

## Light sensitive material

A light sensitive layer is prepared as in Example 3 from 30 mg. of dye No. 28, 100 mg. of N,N-diethyl-N'-sulfomethyl-p-phenylenediamine, 100 mg. of sodium acetate, 100 ml. of ethanol and 50 ml. of a 1.5% solution of a cellulose ether in ethanol.

## Image receiving material

A layer cast on a paper support is prepared from 150 mg. of N,N-diethyl-p-aminophenyl-diazonium chloride zinc chloride complex, 10 ml. of water and 5 ml. of a 1.5% aqueous solution of a cellulose ether.

## Processing

Processing is carried out as in Example 2. A brown positive is obtained.

## EXAMPLE 5

## Light sensitive material

A light sensitive layer is prepared from a solution of 30 mg. of dye No. 29, 100 mg. of 1-phenyl-3-methyl-pyrazolin-5-one, 2.5 g. of ethylcellulose and 150 ml. of ethyl acetate.

The solution is applied to a layer support of glassine paper and dried in the usual manner.

## Image receiving layer

The following casting solution is applied to a layer support of baryta paper:

5 g. of 4-dimethylaminobenzene diazonium tetrafluoroborate, 1 g. of saponin, 1 g. of polyoxyethylenehydroxyethylcellulose and 100 ml. of water.

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## Processing

Processing is carried out as described in Example 2. A positive red image of the original on a grey background is obtained.

## EXAMPLE 6

Light sensitive material as in Example 1.

## Image receiving material

50 mg. of a leuco cobalt phthalocyanine stearylamine complex prepared by the method described below are dissolved in 40 g. of a 1.5% polyvinylacetate solution in acetone and 26 g. of a 4% cellulose acetate solution in acetone, and the solution is cast on paper and dried.

## Processing

Process is carried out as described in Example 2. A blue positive of the original is obtained.

The leuco CoPc used was prepared as follows:

50 g. of a crude product prepared according to Example 1 of German patent specification No. 855,710 were converted into the nitrate by treatment with concentrated nitric acid as described in German patent specification No. 839,939. 16 g. of the dried nitrate were boiled in 50 ml. of cleaning petrol with 15 g. of stearylamine for 20 minutes, the mixture was diluted with 750 ml. of cleaning petrol, the resulting solution was filtered at 100° C. and stirred cold for several hours and the product which crystallized was removed by suction filtration and dried. The filtered off reaction product, of which 27 g. were obtained, was dissolved in boiling ethanol, the solution was stirred cold and the crystalline product was removed by suction filtration and dried. 12 g. of an orange coloured substance were obtained.

Instead of 1-hydroxy-4-methoxynaphthalene, 50 mg. of 1-phenyl-5-methyl-3-pyrazolidone may be used as image producing compound with the same result.

## EXAMPLE 7

Light sensitive material as in Example 1.

## Image receiving material

An image receiving material is prepared from

5 g. of iron(III) chloride, 2 g. of nitrilotriacetic acid and 30 ml. of a 5% aqueous solution of polyvinyl alcohol.

The solution is neutralized with ammonia and cast on paper.

Processing is carried out as described in Example 2. A cyan positive is obtained.

## EXAMPLE 8

Light sensitive material as in Example 1.

## Image receiving material

1 g. of bismuth nitrate is shaken for 6 hours in a ball mill with 40 g. of a 1.5% solution of polyvinyl acetate in acetone and 26 g. of a 4% solution of acetylcellulose in acetone.

The mixture is cast on paper and dried.

## Processing

Processing is carried out as described in Example 2. A brown positive is obtained. Instead of bismuth nitrate, 0.6 g. of thallium(I) chloride or 0.8 g. of mercury(II) bromide may be used with equal success in the image receiving layer.

## EXAMPLE 9

Light sensitive material as in Example 1. Processing is carried out as described in Example 2, but the image receiving material used is ordinary writing paper. A positive cyan image of the original is obtained.

## EXAMPLE 10

Light sensitive material as in Example 1.

## Image receiving material

Ammonia is added to a solution of 5 g. of copper(II) chloride in 75 ml. of H<sub>2</sub>O until the precipitate which forms redissolves, and 30 ml. of 5% aqueous polyvinyl alcohol are then added and the solution is cast on paper and dried.

Processing is carried out as described in Example 2. A grey green positive image of the original is obtained.

## EXAMPLE 11

When used in combination with other dyes, e.g. erythrosine which has its sensitivity at 540 nm., the dyes listed in the following table extend the sensitivity of the light sensitive material to the blue or red region of the spectrum.

## Light sensitive material

The following casting solution is applied to a layer support of polyethylene terephthalate and dried:

30 mg. of erythrosine,  
30 mg. of a dye shown in Table 7 below,  
50 mg. of 1-hydroxy-4-methoxynaphthalene,  
2.5 g. of ethylcellulose and  
150 ml. of ethyl acetate.

## Processing

A set of interference filters was used to determine the spectral sensitivity of the light sensitive material. The filters are permeable to the following wavelengths: 350 nm., 390 nm., 405 nm., 435 nm., 480 nm., 505 nm., 515 nm., 540 nm., 550 nm., 570 nm., 590 nm., 605 nm.

The light sensitive material is exposed behind the interference filters to a 1000 watt iodine quartz lamp from a distance of 30 cm. for a time varying between 5 and 30 minutes according to the dye used. The exposed material is then brought into contact with the image receiving layer described in Example 1 and processed in an ordinary commercial heat development apparatus.

TABLE 7

Dye No.:	Sensitivity at the following wavelengths
Erythrosine—	540 nm.
Erythrosine+1	480, 505, 515, 540 nm.
Erythrosine+7	490, 505, 515, 540 nm.
Erythrosine+8	480, 505, 515, 540, 550, 570 nm.
Erythrosine+15	480, 505, 515, 540, 550 nm.
Erythrosine+16	480, 505, 515, 540, 550 nm.
Erythrosine+28	480, 505, 515, 540 nm.
Erythrosine+30	480, 505, 515, 540 nm.

## EXAMPLE 12

## Light sensitive material

The following casting solution is applied to a layer support of polyethyleneterephthalate and dried:

30 mg. of erythrosine,  
30 mg. of a dye as shown in Table 8 below,  
50 mg. of 1-hydroxy-4-methoxynaphthalene,  
2.5 g. of ethylcellulose and  
150 ml. of ethylacetate.

Processing is carried out as described in Example 1.

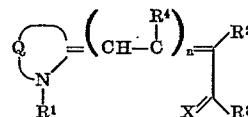
A higher sensitivity is obtained by combining the two dyes, as indicated by the step numbers shown in Table 8 below.

TABLE 8

Dye:	Steps $\sqrt{2}$
Erythrosine alone	7
With dye 1	10
With dye 15	11
With dye 16	12

We claim:

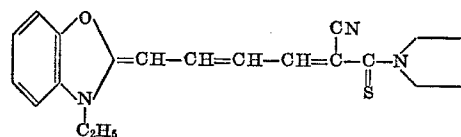
1. In the process for the production of an image by (a) imagewise exposing a light-sensitive layer which contains a sensitizer sensitizing the layer to light and an image-producing compound transferrable to an image-receiving layer at temperatures between 80 to 200° C. the image-producing compound in the exposed areas being converted in the presence of the sensitizer into a non-transferrable compound, and (b) transferring the unconverted image-producing compound to an image-receiving layer and causing it to react in the image-receiving layer to form a colored product, the improvement according to which the sensitizer is at least one dye of the formula:



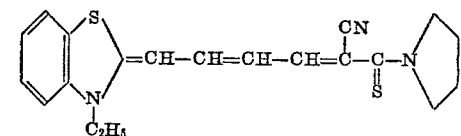
wherein

- R<sup>1</sup>=(1) a saturated or unsaturated aliphatic group, (2) cycloalkyl or (3) aryl;  
R<sup>2</sup>=CN, CO—R<sup>5</sup>, CO—N(R<sup>5</sup>)<sub>2</sub> or COOR<sup>5</sup>;  
R<sup>3</sup>=R<sup>5</sup>, O—R<sup>5</sup> or N(R<sup>5</sup>)<sub>2</sub>; or  
R<sup>2</sup>+R<sup>3</sup>=the ring members required to complete an isocyclic or heterocyclic ketomethylene ring;  
R<sup>4</sup>=hydrogen, alkyl having up to 4 carbon atoms, hydroxyl, alkoxy having up to 4 carbon atoms or aryl;  
R<sup>5</sup>=a saturated or an unsaturated aliphatic group or aryl;  
x=oxygen or sulfur;  
n=0, 1 or 2;  
Q=the members required to complete a 5-membered or 6-membered heterocyclic ring.

2. The combination according to claim 1, characterized in that Q is a benzothiazole ring.  
3. The combination according to claim 1, characterized in that R<sup>2</sup> and R<sup>3</sup> form a thiobarbituric acid ring.  
4. The combination according to claim 1, characterized in that the sensitizer is an [N-alkylbenzothiazole]-dimethine-[1,3-dialkylthiobarbituric acid] merocyanine.  
5. The combinations according to claim 1, characterized in that the sensitizer is:



6. The combination according to claim 1, characterized in that the sensitizer is:

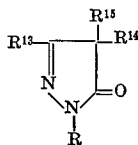


7. The combination according to claim 1, characterized in that the image producing compound is a 1-hydroxy-4-alkoxynaphthalene.  
8. The combination according to claim 7, characterized in that the image producing compound is 1-hydroxy-4-methoxynaphthalene.  
9. The combination according to claim 1, characterized in that the image producing compound is 1,2,3,4-tetrahydro-8-hydroxyquinoline.  
10. The combination according to claim 1, characterized in that the image producing compound is a 3-pyrzolidone.

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11. The combination according to claim 10, characterized in that the image producing compound is 1-phenyl-4-methyl-3-pyrazolidone.

12. The combination according to claim 1, characterized in that the image producing compound is a pyrazolin-5-one compound of the following formula:



wherein

R<sup>12</sup>=(1) hydrogen, (2) a saturated or olefinically unsaturated aliphatic group, (3) aryl, (4) a heterocyclic group or (5) cycloalkyl;

R<sup>13</sup>=(1) hydrogen, (2) a saturated or olefinically unsaturated aliphatic group, (3) aryl, (4) a heterocyclic group, (5) cycloalkyl, (6) hydroxyl, (7) amino or (8) an alkoxy-carbonyl group;

R<sup>14</sup>=(1) hydrogen, (2) a saturated or olefinically unsaturated aliphatic group, (3) aryl, (4) amino or (5) alkoxy; or R<sup>13</sup> and R<sup>14</sup> together represent the members required to complete a carbocyclic or heterocyclic ring; and

R<sup>15</sup>=hydrogen or a 4-aminophenylamino group.

13. The combination according to claim 1, characterized in that the image receiving layer contains a heavy metal compound which is not sensitive to light under the conditions of the process.

14. The combination according to claim 13, characterized in that the heavy metal compound is a silver compound which has little or no sensitivity to light under the conditions of the process.

15. The combination according to claim 14, characterized in that the silver compound is a salt of a long chain aliphatic carboxylic acid having 8 to 24 carbon atoms.

16. The combination according to claim 14, characterized in that the silver compound is a salt of an aliphatic carboxylic acid substituted with a thioether group.

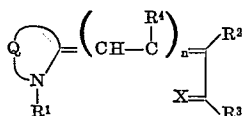
17. The combination according to claim 13, characterized in that the image receiving layer is used contains a toner.

18. The combination according to claim 13, characterized in that the image receiving layer contains a sterically hindered phenol that accelerates the reaction in the image-receiving layer.

19. The combination according to claim 13, characterized in that the image receiving layer contains a white pigment.

20. The combination according to claim 13, characterized in that the image receiving layer contains a metal ion image intensifier.

21. In a light-sensitive photographic layer containing a sensitizer sensitizing the layer to light and an image-producing compound which can be transferred to an image-receiving layer at temperatures of between 80 and 200° C., but which is converted by exposure into non-transferrable reaction product in the presence of the sensitizer, the improvement wherein the sensitizer has the formula:



wherein

R<sup>1</sup>=(1) a saturated or unsaturated aliphatic group, (2) cycloalkyl or (3) aryl;

R<sup>2</sup>=CN, CO—R<sup>5</sup>, CO—N(R<sup>5</sup>)<sub>2</sub> or COOR<sup>5</sup>;

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R<sup>3</sup>=R<sup>5</sup>, O—R<sup>5</sup> or N(R<sup>5</sup>)<sub>2</sub> or

R<sup>2</sup>+R<sup>3</sup>=the ring members required to complete an isocyclic or heterocyclic ketomethylene ring;

R<sup>4</sup>=hydrogen, alkyl having up to 4 carbon atoms, hydroxyl, alkoxy having up to 4 carbon atoms or aryl;

R<sup>5</sup>=a saturated or unsaturated aliphatic group or aryl;

X=oxygen or sulphur;

n=0, 1 or 2;

Q=the members required to complete a 5-membered or

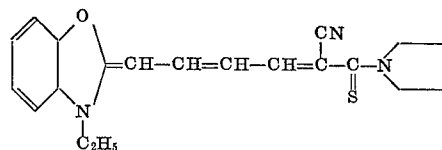
6-membered heterocyclic ring.

22. The combination according to claim 21, characterized in that Q is a benzothiazole ring.

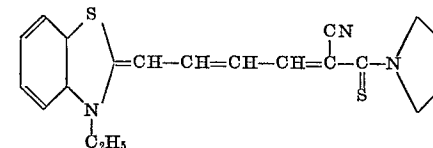
23. The combination according to claim 21, characterized in that R<sup>2</sup> and R<sup>3</sup> form a thiobarbituric acid ring.

24. The combination according to claim 21, characterized in that the sensitizer is a [N-alkylbenzothiazole]-dimethine-[1,3-dialkyl-thiobarbituric acid] merocyanine.

25. The combination according to claim 21, characterized in that the sensitizer is the following sensitizer.



26. The combination according to claim 21, characterized in that the sensitizer is



27. The combination according to claim 21, characterized in that the image-producing compound is a 1-hydroxy-4-alkoxynaphthalene.

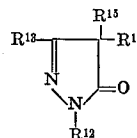
28. The combination according to claim 27, characterized in that the image-producing compounds is 1-hydroxy-4-methoxynaphthalene.

29. The combination according to claim 23, characterized in that the image-producing compound is 1,2,3,4-tetrahydro-8-hydroxyquinoline.

30. The combination according to claim 24, characterized in that the image-producing compound is a 3-pyrazolidone.

31. The combination according to claim 30, characterized in that the image-producing compound 1-phenyl-4-methyl-3-pyrazolidone as image producing compound.

32. The combination according to claim 21, characterized in that the image producing compound is a pyrazolin-5-one compound of the formula:



wherein

R<sup>12</sup>=(1) hydrogen, (2) a saturated or olefinically unsaturated aliphatic group, (3) aryl, (4) a heterocyclic group or (5) cycloalkyl;

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R<sup>13</sup>=(1) hydrogen, (2) a saturated or olefinically unsaturated aliphatic group, (3) aryl, (4) a heterocyclic group, (5) cycloalkyl, (6) hydroxyl, (7) amino or (8) an alkoxy-carbonyl group;

R<sup>14</sup>=(1) hydrogen, (2) a saturated or olefinically unsaturated aliphatic group, (3) aryl, (4) amino or (5) alkoxy; or R<sup>13</sup> and R<sup>14</sup> together represent the ring members required for completing a carbocyclic or heterocyclic ring; and

R<sup>15</sup>=hydrogen or a 4-aminophenylamino group.

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NORMAN G. TORCHIN, Primary Examiner

J. L. GOODROW, Assistant Examiner

U.S. Cl. X.R.

117-36.9; 96-141, 76 R, 3