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3,585,028

DIFFUSION TRANSFER COLOR PRODUCTS AND PROCESSES SIMULTANEOUSLY
UTILIZING EXPOSED AND UNEXPOSED SILVER HALIDES
Filed May 9, 1969

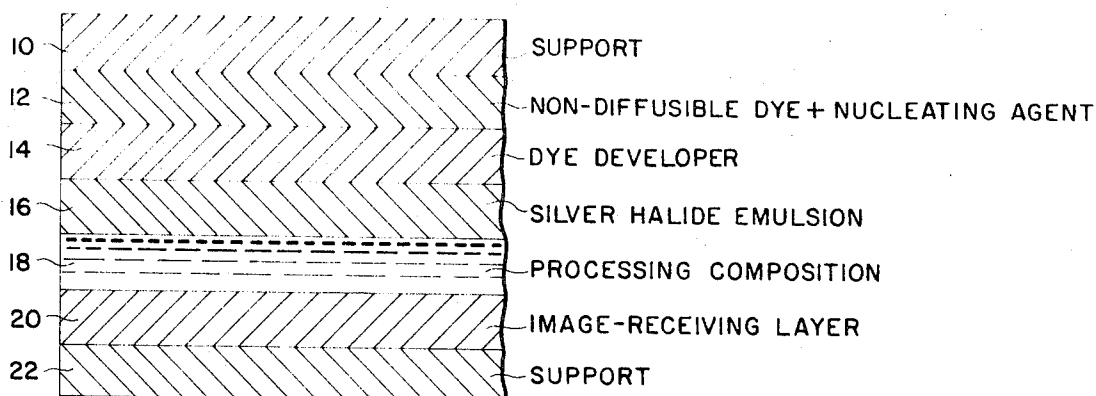


FIG. 1

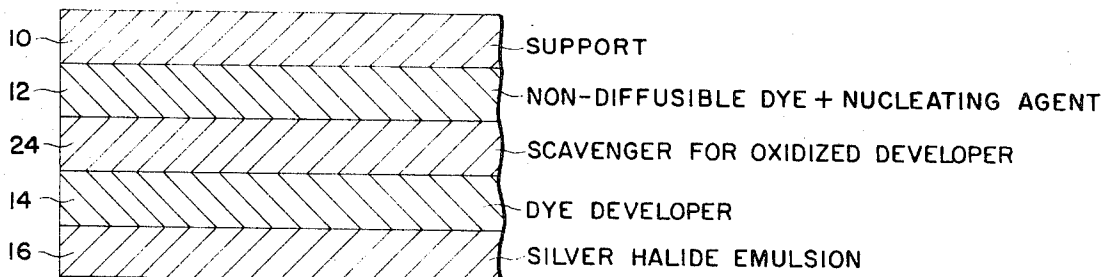


FIG. 2

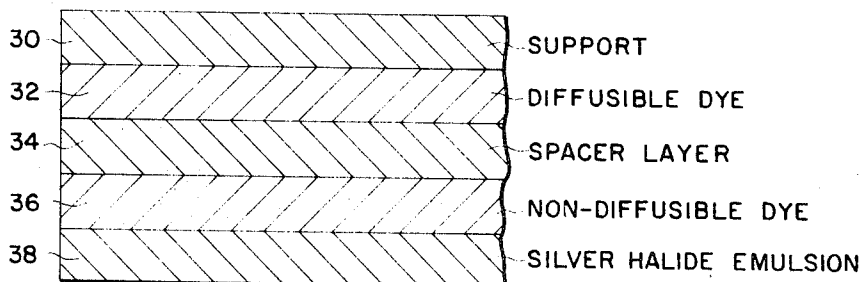


FIG. 3

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3,585,028

DIFFUSION TRANSFER COLOR PRODUCTS AND PROCESSES SIMULTANEOUSLY UTILIZING EXPOSED AND UNEXPOSED SILVER HALIDES

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Int. Cl. G03c 7/00, 5/54

U.S. Cl. 96—3

25 Claims

ABSTRACT OF THE DISCLOSURE

Photographic systems for preparing color transfer images employing a photosensitive element having at least one light-sensitive silver halide emulsion having associated therewith a first color-providing material which is normally diffusible in an aqueous alkaline processing medium and a second color-providing material which is not, wherein the photosensitive element is exposed and thereafter developed, as a function of development the first-named material is selectively immobilized to provide a first imagewise distribution of mobile and diffusible color-providing material and said second-named material is selectively rendered diffusible to provide a second imagewise distribution of mobile and diffusible color-providing material in areas corresponding to said first imagewise distribution, and said two imagewise distributions of mobile and diffusible color-providing material are then transferred to a dyeable stratum to impart thereto a color transfer image. The photographic systems are also characterized as employing exposed silver halide to control transfer of one color-providing material; and unexposed silver halide to control transfer of the other.

BACKGROUND OF THE INVENTION

U.S. Pat. No. 2,983,606 issued to Howard G. Rogers discloses processes for forming color transfer images utilizing dye developers (dyes which are also silver halide developing agents). In such systems, a photosensitive element containing at least one light-sensitive silver halide emulsion and associated dye developer is exposed to form a developable image; the thus exposed element is then contacted with an aqueous alkaline processing composition to develop the image and, as a function of development, to form an imagewise distribution of mobile and diffusible dye developer; and this imagewise distribution is then transferred, at least in part, to a superposed dyeable stratum to impart thereto a color transfer image. Multicolor images may be obtained by having two or more emulsion layers and associated dye developers. Accurate color reproductions may be obtained by providing a blue-sensitive, a green-sensitive and a red-sensitive silver halide emulsion layer, the emulsion layers having associated therewith a yellow, a magenta and a cyan dye developer, respectively. Multilayer photosensitive elements of this nature are described with more particularity in U.S. Pat. No. 3,345,163 issued to Edwin H. Land and Howard G. Rogers.

The copending application of Stanley M. Bloom, Ser. No. 655,338 filed July 24, 1967, describes and claims novel compounds containing a color-providing moiety and an "anchoring" or immobilizing moiety. These compounds are immobile and non-diffusible in an aqueous alkaline medium, but are capable of providing, upon oxidation, an oxidation product which may auto-react intramolecularly to form a heterocyclic ring and, as a function of this ring formation, to split off a mobile and diffusible color-providing material. This reaction may also be defined as a ring-closure of the oxidation product resulting

in a separation or "freeing" of the mobile color-providing moiety from the "anchoring moiety."

The copending application of Stanley M. Bloom and Howard G. Rogers, Ser. No. 655,440 filed July 24, 1967 now U.S. Pat. 3,443,940, discloses various photographic systems employing compounds such as those described and claimed in the aforementioned application Ser. No. 655,338 to obtain color transfer images. In one of the disclosed systems, a photosensitive element containing at least one light-sensitive silver halide emulsion and associated immobile compound such as described above is exposed to provide a developable image; the thus exposed element is contacted with an aqueous alkaline processing composition including a silver halide developing agent and a silver halide solvent to develop the image and to form an imagewise distribution of a soluble silver complex in terms of undeveloped areas of the emulsion according to principles well known in silver diffusion transfer; the resulting soluble silver complex is then transferred to contact the color-providing material, whereby the color-providing material reduces the silver complex and is in turn oxidized to provide an oxidation product which then autoreacts intramolecularly to effect ring-closure and to split off the mobile and diffusible color-providing moiety to provide in turn an imagewise distribution of mobile and diffusible color-providing material in terms of unexposed areas of the emulsion; and this imagewise distribution is then transferred, at least in part, by imbibition, to a superposed dyeable stratum to impart thereto a positive color transfer image.

The copending application of Stanley M. Bloom and Robert K. Stephens, Ser. No. 655,436, filed July 24, 1967 now U.S. Patent 3,443,939, disclosed systems employing the aforementioned non-diffusible compounds to obtain negative color transfer images. According to this copending patent application a photosensitive element containing at least one light-sensitive silver halide emulsion and associated compound of the foregoing description is exposed and then developed with an aqueous alkaline processing composition including a silver halide developing agent which upon development provides an oxidation product capable of being reduced by a redox reaction with the aforementioned compound, the developer further being sufficiently mobile in its oxidized state so as to be capable of migrating to the layer containing the color-providing compound; oxidizing the developing agent as a function of development to provide an imagewise distribution of oxidized developing agent in terms of exposed and undeveloped areas of the emulsion; transferring this imagewise distribution at least in part, to contact the color-providing compound, whereby a redox reaction takes place to reduce the developing agent and to oxidize the color-providing compound to form an oxidation product which then ring-closes to split off the mobile color-providing moiety in turn to form an imagewise distribution of mobile and diffusible color-providing compound in terms of exposed areas of the emulsion; and transferring this latter imagewise distribution, at least in part, by imbibition to a superposed dyeable stratum to impart thereto a negative color transfer image.

The copending application of Howard G. Rogers, Ser. No. 655,502 filed July 24, 1967 now U.S. Patent 3,443,941, disclosed various systems for forming color transfer images utilizing silver ions to control color transfer. One of the disclosed systems utilizes silver ions to immobilize a color-providing material which is normally mobile and diffusible in an aqueous alkaline medium. In such a system, a photosensitive element containing at least one light-sensitive silver halide emulsion and associated color-providing material is exposed to form a developable image and then developed by applying a processing composition including a silver halide solvent. In known manner, an imagewise dis-

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tribution of soluble silver complex is formed in terms of unexposed areas of the emulsion. This imagewise distribution is transferred to the color-providing material where it immobilizes the color-providing material in terms of unexposed areas of the emulsion, thereby providing an imagewise distribution of mobile and diffusible color-providing material in terms of exposed areas. This latter imagewise distribution is then transferred, at least in part, by imbibition, to a superposed strata to impart thereto a negative color transfer image.

The present invention utilizes the principles of the inventions described and claimed in the aforementioned patent and copending applications in a novel manner to form color transfer images.

SUMMARY OF THE INVENTION

According to the present invention, diffusion transfer systems for preparing color images are provided employing photosensitive elements comprising at least one light-sensitive silver halide emulsion having associated therewith a color-providing material normally diffusible in an aqueous alkaline medium and a color-providing material normally non-diffusible in an aqueous alkaline medium. Reaction mechanisms are employed to form, as a function of development, an imagewise distribution of color-providing ingredients containing each of these color-providing materials, which is then transferred to a superposed stratum to form a color transfer image.

In one embodiment of this invention, a dye developer is employed as the normally diffusible color-providing material and a compound containing a color-providing moiety and an anchoring moiety, e.g., a compound as described in the aforementioned application Ser. No. 655,338, is employed as the non-diffusible color-providing material.

In this embodiment of the invention, as a function of development, dye developer is oxidized and immobilized for transfer in terms of exposed areas of the light-sensitive emulsion to provide an imagewise distribution of unoxidized and diffusible dye developer in terms of unexposed areas; and at substantially the same time a soluble silver complex formed in terms of unexposed areas is employed to release the diffusible dye moiety from the anchoring moiety of the normally non-diffusible compound, thereby forming a second imagewise distribution of color-providing material in terms of unexposed areas of the emulsion. Both of these imagewise distributions are then transferred to a superposed stratum to impart thereto a positive color transfer image.

In a second embodiment of the invention, a compound of the type described above in connection with the aforementioned U.S. Patent 3,443,941 is employed as the normally diffusible color-providing material and one of the aforementioned non-diffusible compounds containing a diffusible color-providing moiety and an anchoring moiety is employed as the non-diffusible compound. In this embodiment, an imagewise distribution of the former is obtained according to the procedures described in the aforementioned U.S. Patent 3,443,941; and an imagewise distribution of the latter is obtained by the redox reaction described in the aforementioned U.S. Patent 3,443,939. These two imagewise distributions are then transferred to a superposed stratum, by imbibition, to form a negative color transfer image.

The present invention may be said to be predicated upon the use of "negative silver," i.e., exposed silver in the photosensitive element, to control transfer of one color-providing material, and "positive silver," i.e., unexposed silver in the form of a soluble silver complex, to control transfer of another color-providing material to form a color transfer image in terms of both; as distinguished from prior color transfer processes which utilize either "negative" or "positive" silver, but not both, to form color transfer images. The ability to employ both "negative" and "positive" silver in two non-competing reaction mechanisms to control dye transfer provides certain significant advantages,

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some of which should be apparent and others which will be discussed with particularity hereinafter.

DESCRIPTION OF DRAWING

FIG. 1 is a partially schematic, enlarged, fragmentary, sectional view illustrating one previously exposed photographic product of this invention during processing thereof;

FIG. 2 is a partially schematic, enlarged, fragmentary, sectional view of another photographic product of this invention; and

FIG. 3 is a similar view of yet another photographic product of this invention.

As was indicated previously, this invention relates to photography and more particularly to novel products and processes for preparing color transfer images.

A primary object of this invention therefore is to provide novel products and processes for preparing monochromatic or multicolor transfer images.

Another object is to provide novel color diffusion transfer systems wherein two different reaction mechanisms are employed substantially simultaneously to provide, as a function of development, two imagewise distributions of color-providing material available for transfer to a superposed stratum to impart thereto a color transfer image.

Still another object is to provide novel color diffusion transfer systems employing at least one color-providing material normally diffusible in an aqueous alkaline processing medium and at least one color-providing material normally non-diffusible in an aqueous alkaline processing medium.

A still further object is to provide novel products and processes of the foregoing description.

Yet another object is to provide novel color transfer systems wherein both exposed and unexposed silver halide are employed to control transfer of color-providing materials.

Other objects of the invention will in part be obvious and will in part appear hereinafter.

The invention accordingly comprises the several steps and the relation and order of one or more of such steps with respect to each of the others, and the product possessing the features, properties and the relation of elements which are exemplified in the following detailed disclosure, and the scope of the application of which will be indicated in the claims.

For a fuller understanding of the nature and objects of the invention, reference should be had to the following detailed description taken in connection with the accompanying drawing.

Color transfer processes are known which use unexposed silver halide to control the transfer of color-providing material to a superposed stratum to form a color transfer image. Color transfer processes are also known which use exposed silver halide to control the transfer of color-providing material to a superposed stratum. The photographic systems described and claimed in the aforementioned U.S. Pats. 3,443,941 and 3,443,939 are examples of the former; and the systems described and claimed in the aforementioned U.S. Pat. No. 2,983,606 are examples of the latter.

The essence of the present invention is providing a system which combines the two so that the color transfer image is formed in terms of at least two color-providing materials, one being a color-providing material whose transfer is controlled by unexposed silver halide; the other being a color-providing material whose transfer is controlled by exposed silver halide. These color-providing materials may be complete dyes or they may be couplers or dye intermediates, i.e., materials which form complete dyes at some time during or after transfer. Because of the ready availability of suitable complete dyes, these materials are preferred to dye intermediates which require a subsequent action, e.g., a coupling reaction, to provide the desired color image. In any event the two color-providing materials may possess substantially the same spec-

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tral absorption characteristics, for example, they both may be yellow, magenta or cyan dyes, or they may be of entirely different colors. In the former case, the resulting color transfer image would be of substantially the same color as either of the color-providing materials alone; whereas in the latter case, the color of the resulting image would be entirely different, being a product of the sum total of the two. Thus, for example, if the two color-providing materials are complementary in color, and substantially equivalent amounts in terms of covering power or density are transferred, the resulting image would be essentially neutral in color, i.e., black or a shade of black.

Common to all of the systems contemplated by this invention is the use of a photosensitive element having at least one light-sensitive silver halide emulsion having associated therewith a normally diffusible color-providing material and a normally non-diffusible color-providing material; and an aqueous alkaline processing composition including a silver halide solvent.

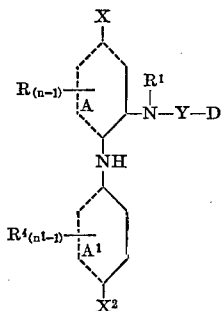
As used herein and in the appended claims, the term "diffusible" refers to the property of the color-providing material of being mobile and transferable in the aqueous alkaline processing medium from the layer in which it is initially contained to a superposed stratum during processing. The term "non-diffusible" therefore refers to the converse property.

In one aspect of this invention, use is made of a dye developer, i.e., a dye which is also a silver halide developing agent, as the normally diffusible color-providing material; and a compound containing a diffusible color-providing moiety and an "anchoring" moiety rendering the compound non-diffusible as the non-diffusible color-providing material.

Dye developers are per se well known in the art and suitable dye developers of various structures and colors are disclosed, for example, in U.S. Pat. No. 2,983,606, in various patents cited therein, and in many other patents, as well. In general, a dye developer is a compound containing a dye moiety and a silver halide developing function linked to the dye moiety through a covalent bond or a divalent organic linking group. As is well known, the developing function generally is a benzene nucleus containing amino and/or hydroxy groups in either the ortho or para positions. Dihydroxyphenyl substituents such as hydroquinonyl and substituted derivatives thereof are generally preferred. The dye moiety may comprise any chromophoric system contributing the desired chemical and physical characteristics. Included are azo, anthraquinone, azomethine, phthalocyanine dyes, etc. The dye developer may, if desired, be in the form of a metal complex, e.g., a chromium-complexed dye developer.

As was mentioned previously, useful compounds containing a diffusible color-providing moiety and an "anchoring" moiety rendering the compound non-diffusible include those described and claimed in copending application Ser. No. 655,338 and which may be represented by the formula:

(A)



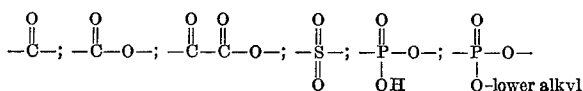
wherein:

A and A¹ each represent the atoms necessary to complete a benzene or naphthalene ring, which ring may be further substituted;

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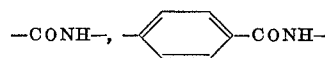
D is a color-providing moiety, e.g., a complete dye such as a monoazo, disazo or anthraquinone dye which may, if desired, be metallized in known manner;

Y is any substituent which completes or forms an amide with, and reduces the basic character of the amino group in the 3-position, such as the residue of an acid, linking the color-providing moiety D to the 3-nitrogen atom, and which is capable of being eliminated during the ring formation to be described with more particularity herein-after, e.g.,



etc.;

R and R⁴ each represent an "anchoring" or immobilizing substituent rendering the compound non-diffusible, e.g., higher alkyl such as decyl, dodecyl, stearyl, oleyl, etc. linked directly to the aromatic nucleus or linked indirectly thereto through an appropriate linking group, e.g.,



etc., an aromatic ring, e.g., of the benzene or naphthalene series, or a heterocyclic ring, which rings may be either bonded to a single carbon atom of the aromatic nucleus or fused thereto, i.e., bonded to a pair of adjacent carbon atoms; or R and/or R⁴ may be a plurality of short chain radicals which together provide the anchoring moiety, each of said short chain radicals being linked directly or indirectly to a different carbon atom of the aromatic nucleus formed by the A and/or A¹ moieties;

X is hydrogen, hydroxy, amino, e.g., a primary, secondary or tertiary amino substituent of the formula:



wherein each R³ may be hydrogen, a hydrocarbon radical, e.g., alkyl, such as methyl, ethyl, butyl, dodecyl, etc., aryl such as phenyl or naphthyl attached through a carbon atom thereof to the nitrogen atom, a cyclic alkyl such as cyclopentyl or cyclohexyl, i.e., where both R³'s are alkylene comprising together with the nitrogen atom a heterocyclic ring, a substituted alkyl, such as hydroxyethyl, methoxyethoxyethyl, polyglycoloxyethyl, carboxymethyl, benzyl, phenylethyl, sulfo-phenylethyl, acetyl-amino-phenylethyl, succinylamino-phenylethyl, furanemethyl, etc.; or a substituted aryl such as methylphenyl, ethylphenyl, etc., or the anchoring substituent R;

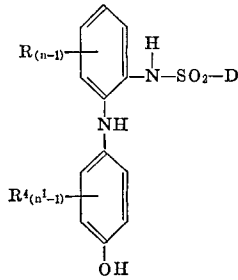
X² is hydrogen, hydroxy, an amino group such as may be contained by said X moiety, as previously described, or the anchoring substituent R⁴, provided that one of said X and X² moieties must be hydroxy or amino;

R¹ is hydrogen, an alkyl such as methyl, ethyl, propyl, butyl, hexyl, octyl, dodecyl, cyclohexyl, etc.; or a substituted alkyl such as 2-hydroxyethyl, 3-dihydroxypropyl, carboxymethyl, carboxyethyl, carboxybutyl, carboxydecyl, hydroxyethyl-ether, polyglycoloxyethyl, furanemethyl, benzyl, phenylethyl, carboxyphenylethyl, sulfo-phenylethyl, acylaminophenylethyl, etc.; and

n and n¹ each is a positive integer from 1 to 2, provided that when R¹ or R³ alone or together comprise one of those heretofore named substituents rendering the compound non-diffusible, either or both of n and n¹ may be 1 but when R¹ and R³ alone or together do not provide such an anchoring moiety at least one of n and n¹ must be 2.

Preferred are those compounds within the scope of the above formula which are of the following formula:

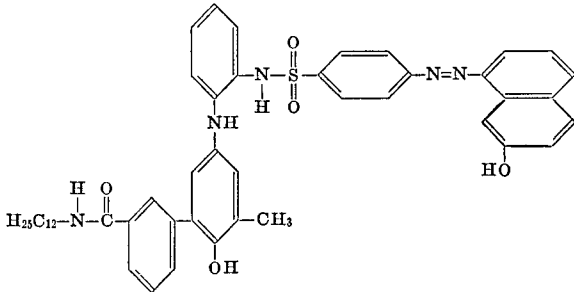
(B)



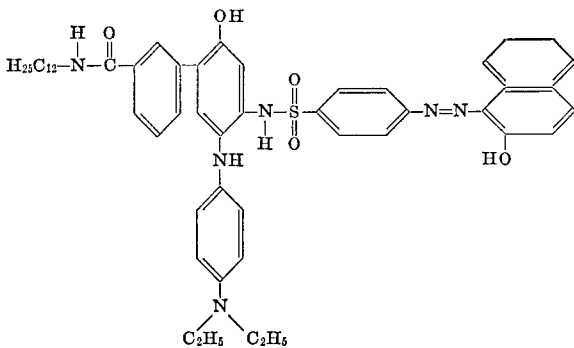
wherein the anchoring moiety R or R⁴ comprises a long chain amide, e.g., of at least 13 carbon atoms; and nuclear substituted derivatives thereof, e.g., where any of the nuclear carbon atoms not specifically substituted may contain a carboxy, alkyl, alkoxy, amino, chloro, hydroxy or amide substituent.

As examples of such compounds, mention may be made of the following:

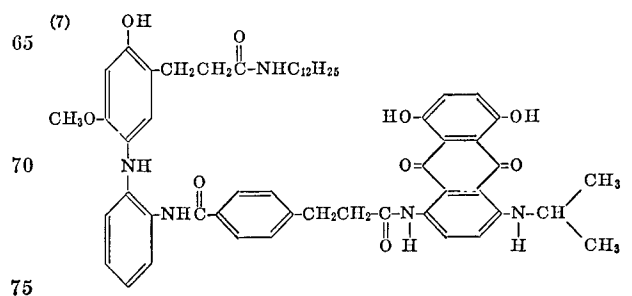
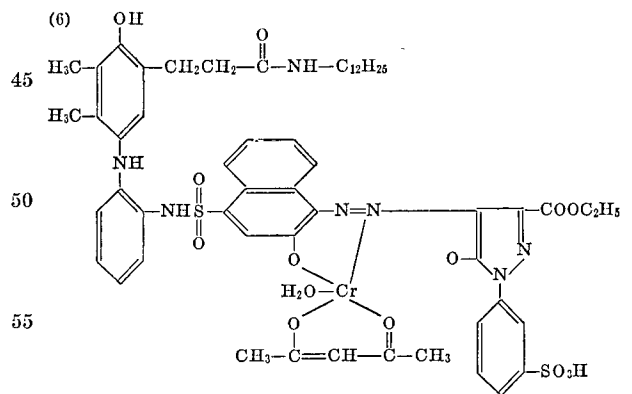
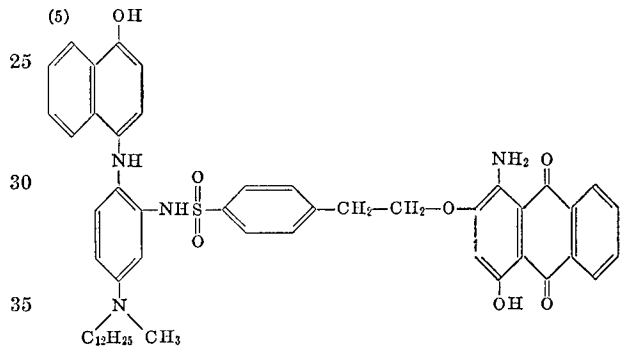
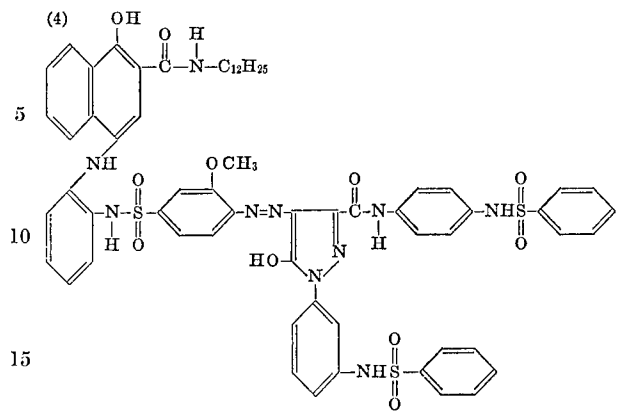
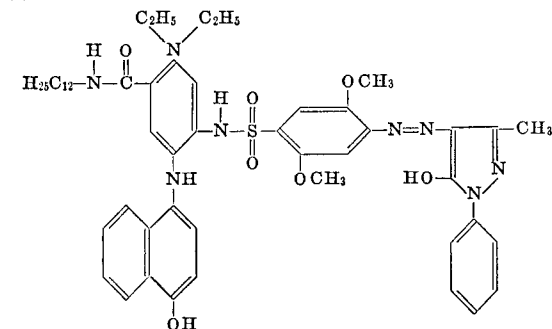
(1)



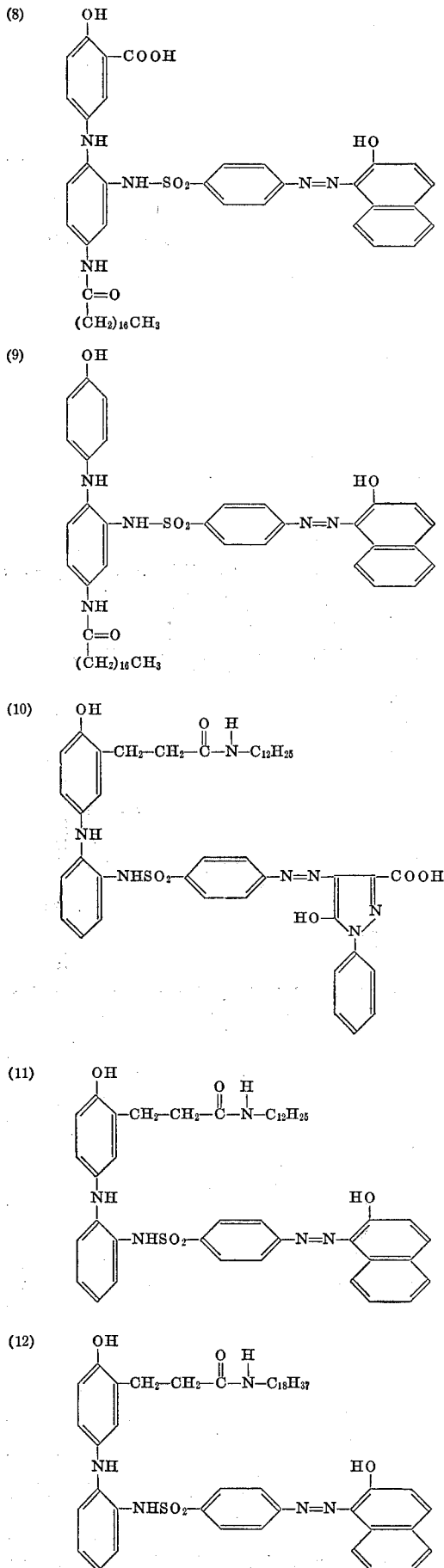
(2)



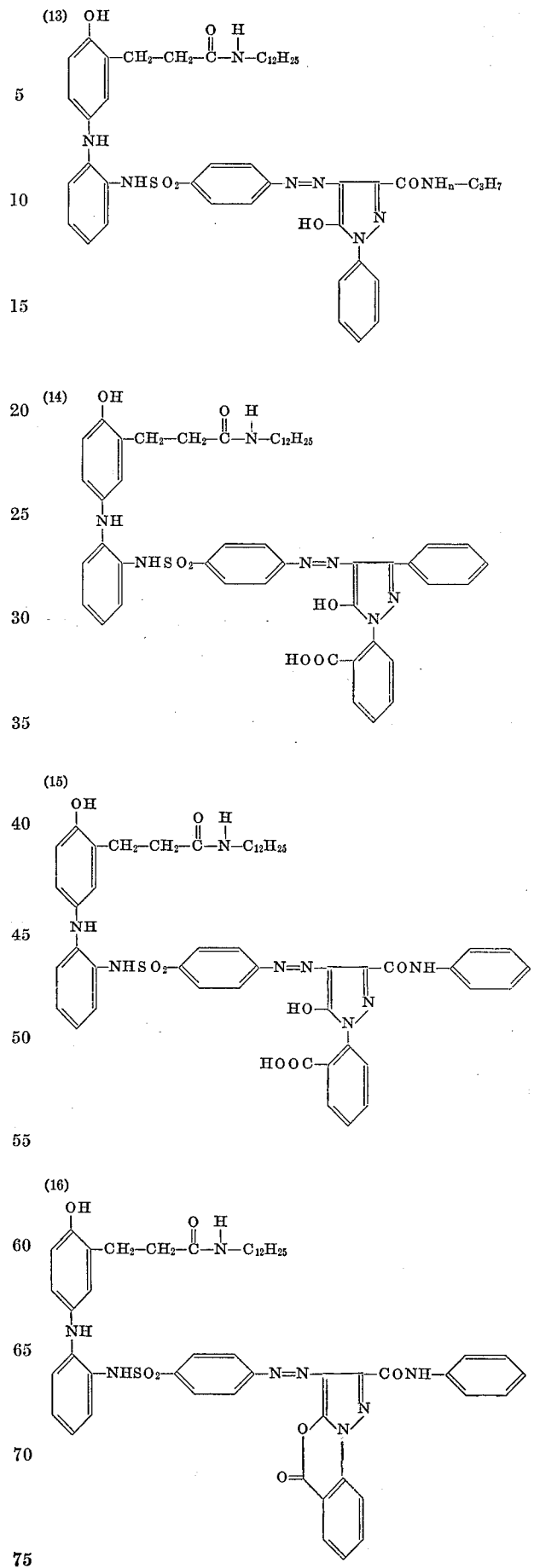
(3)



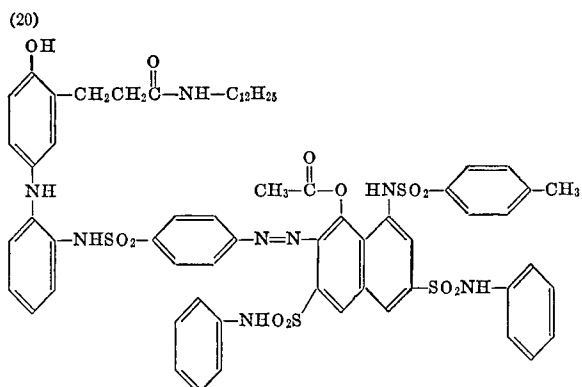
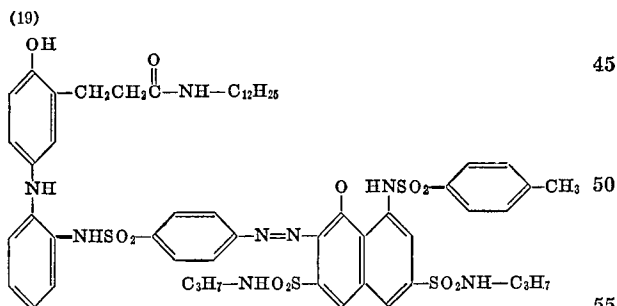
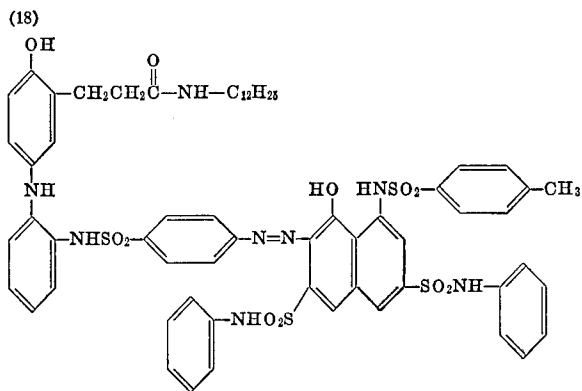
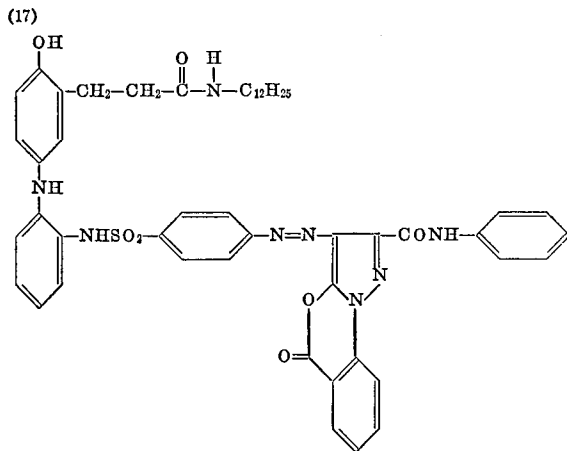
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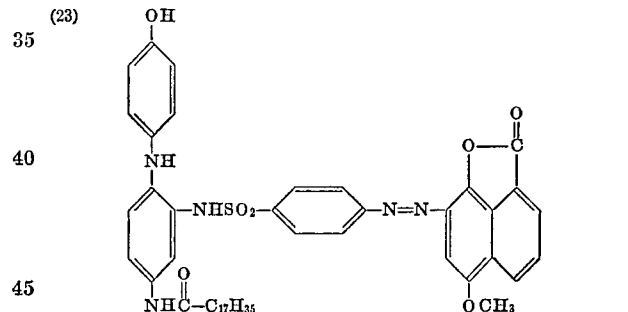
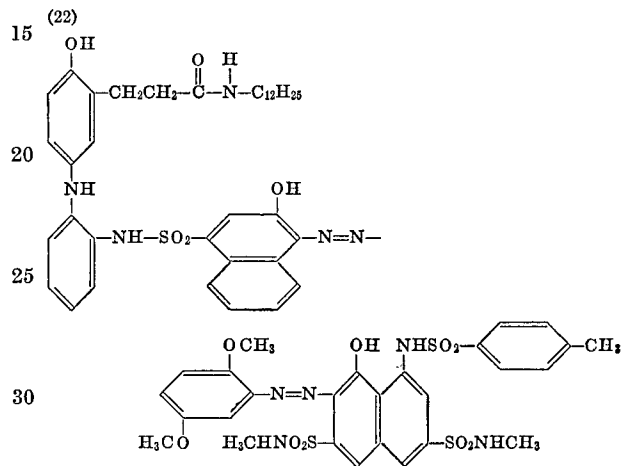
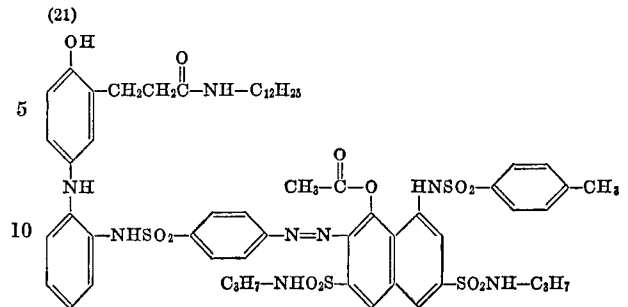
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In the embodiment of the invention using the combination of a dye developer and a non-diffusible color-providing material of the foregoing description, a photosensitive element including at least one light-sensitive silver halide emulsion is first exposed to form a developable image. This image is then developed by contacting it with an aqueous alkaline processing composition including a dye developer and a silver halide solvent to develop the image. In known manner, exposed silver halide controls the diffusibility of the dye developer by being reduced and as a function of this development causing the dye developer to in turn be oxidized to provide an oxidation product which is less mobile and diffusible than the unoxidized dye developer present in terms of unexposed and undeveloped areas of the emulsion. The dye developer is preferably contained initially in a layer associated with the silver halide emulsion, the dye developer solution being obtained by contacting the photosensitive element with the aqueous alkaline medium. However, in monochromatic processes, the dye developer may be contained initially in the processing composition in the manner known in the art.

In either event, at substantially the same time as the exposed silver halide is developed to provide an image-wise distribution of diffusible dye developer in terms of unexposed areas of the emulsion, an image-wise distribution of soluble silver complex is formed in terms of unexposed and undeveloped silver halide, according

to principles well known in silver diffusion transfer photography. This imagewise distribution of soluble silver complex migrates or is transferred in the processing composition to a layer associated with the silver halide emulsion, containing the non-diffusible color-providing material where the two react to reduce the silver complex and as a function thereof to oxidize the color-providing material. In the manner described with particularity in the aforementioned copending application Ser. No. 655,440, an oxidation product of this color-providing material is formed which then ring-closes intramolecularly and splits off the diffusible color-providing moiety, thereby providing an imagewise distribution of this diffusible color-providing moiety in terms of unexposed areas of the emulsion. It will thus be seen that the foregoing reaction mechanisms provide two image-wise diffusible distributions which are virtually identical in terms of the point-to-point degree of exposure of the silver halide emulsion. These two imagewise distributions are then transferred, by imbibition, to a superposed layer to impart thereto a color image in terms of the two.

This aspect of the invention is illustrated in FIGS. 1 and 2. FIG. 1 shows an exposed photosensitive element of this aspect of the invention during development. The photo-sensitive element comprises a suitable opaque or transparent support 10 containing a layer 12 of non-diffusible dye and a nucleating agent; layer 14 of the dye developer; and silver halide emulsion layer 16. At some time during or after exposure, processing composition 18 is spread between the thus exposed photosensitive element and a superposed image-receiving element comprising an image-receiving layer or dyeable stratum 20 contained on a suitable support 22. After a suitable processing time, e.g., 60 seconds, the respective elements are then separated to reveal a positive color image on the image-receiving element.

Supports 10 and 22 may comprise any of the materials heretofore employed for such purposes, e.g., a cellulose ester such as cellulose acetate, polyvinyl acetal, polystyrene, polyethylene terephthalate, polyethylene, paper, glass, etc.

Layer 12 comprises the non-diffusible color-providing material and silver precipitating or physical development nuclei contained in a suitable alkali-permeable matrix, e.g., gelatin. Such nuclei, which are well known in the art, include noble metals such as silver or gold; colloidal metal sulfides, selenides, and tellurides; metal proteinates such as silver proteinates; and such organic sulfur-containing compounds as thiourea, xanthates, etc. While the precipitating nuclei are shown to be contained in the same layer, they may be contained in a contiguous layer, as discussed in the aforementioned U.S. Patent 3,443,940.

Dye developer layer 14 may, of course, be prepared by any of the methods heretofore known and described in numerous patents, e.g., by coating from a cellulose acetate hydrogen phthalate, gel or solid dispersions, etc.

Emulsion layer 16 comprises any of the conventional silver halide emulsions, e.g., silver chloride, silver bromide, silver bromoiodide, silver chlorobromide or silver chlorobromoiodide. It may thus be a high speed or a low speed emulsion. The emulsion layer may also contain the various additives frequently employed in such layers, e.g., optical sensitizers, antifoggants, hardeners, plasticizers, coating aids, speed-increasing materials, etc. The dispersing agent or substrate for the silver halide may be gelatin or some other material.

Receiving layer 20 comprises a dyeable stratum generally including a mordant. A preferred image-receiving element is one comprising a layer of a 2:1 mixture by weight of polyvinyl alcohol and poly-4-vinyl pyridine; a layer of polyvinyl alcohol, and a layer of a half-butyl ester of poly-(ethylene/maleic anhydride) coated on a baryta paper support, as disclosed in U.S. Pat. No. 3,362,819 issued to Edwin H. Land.

Processing composition 18 includes at least an aqueous solution of an alkaline material such as sodium or potassium hydroxide, and a silver halide solvent such as sodium or potassium thiosulfate. It may also contain additional reagents performing specific desired functions, e.g., viscous film-forming reagents such as hydroxyethyl cellulose, sodium carboxymethyl cellulose, etc., antifoggants and the like, as well as accelerating or auxiliary silver halide developing agents such as p-methylphenylhydroquinone. Any of the aforementioned components of the processing composition may, if desired, be present initially in the film unit, in which case the processing composition providing the desired ingredients is formed by contacting the film unit with the aqueous medium therefor. As was mentioned previously, in monochromatic systems, the dye developer may also be contained initially in the processing composition. In any event, the processing composition may, if desired, be confined in a frangible container or "pod" such as described, for example, in U.S. 2,543,181 and 2,634,886, issued to Edwin H. Land.

As was discussed in the aforementioned U.S. Pats. 3,443,940 and 3,443,939, in systems employing non-diffusible compounds of the foregoing description two competing reactions are possible: (1) the reaction between the soluble silver complex and the color-providing material; and (2) a redox reaction between the color-providing material and oxidized silver halide developing agent, i.e., developing agent oxidized as a function of development of exposed silver halide. It is possible for both reactions to occur at the same time so that a uniform transfer of the released diffusible color-providing moiety occurs in both exposed and unexposed areas. In this aspect of the invention it will, therefore, be apparent that any redox reaction between the non-diffusible compound and oxidized developer should be avoided. Since the oxidized dye developer is not appreciably mobile, in many instances selection of appropriate dye developers and imbibition times will suffice. In other instances, it may be desirable to employ appropriate controls for optimum efficiency. It may be entirely satisfactory to employ a spacer layer of gelatin, cellulose acetate hydrogen phthalate or some other alkali and dye permeable material between layers 12 and 14 to restrain migration of oxidized dye developer and/or accelerating developing agent (if any). Another system described in U.S. Pat. 3,443,940 for obviating the redox reaction is to include in the photosensitive element a material which will intercept any oxidized developer and render it innocuous before it can migrate to the color-providing material in layer 26, e.g., by reducing the oxidized developing agent. Such materials may be defined as being scavengers for oxidized developer. Suitable scavengers of this nature are described in U.S. Pat. 3,443,940 and copending applications of Stanley M. Bloom et al., Ser. Nos. 655,324 and 655,309, both filed July 24, 1967 now U.S. Pats. 3,482,971 and 3,459,548 respectively.

FIG. 2 illustrates the use of a scavenger of this description, the scavenger being shown to be contained in a layer 24 between the dye developer layer and the layer containing the non-diffusible color-providing material.

In the aforementioned embodiment of this invention for preparing positive images, reference has been made to the presence of silver-precipitating nuclei in the same layer or in a layer adjacent to the non-diffusible dye. These nuclei are employed to render the soluble silver complex reducible and the color-providing material oxidized. It will be appreciated, however, that if the color-providing material is a non-discriminating silver halide developer, silver precipitating nuclei need not be employed. It should likewise be apparent that the invention is not restricted to the specific color-providing materials disclosed above and described and claimed in copending application Ser. No. 655,338.

Another embodiment of this invention employs non-diffusible color-providing materials such as mentioned

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above and normally diffusible color-providing materials reactable with silver ions to be rendered non-diffusible to form negative color transfer images. In this embodiment, as distinguished from the prior embodiment, exposed silver halide is employed to control transfer of the non-diffusible color-providing material and unexposed silver halide is employed to control transfer of the diffusible color-providing material.

Again, the non-diffusible color-providing material may be one such as is described and claimed in Ser. No. 655,338.

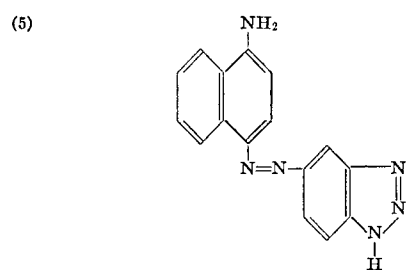
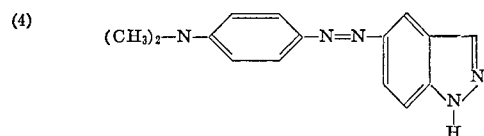
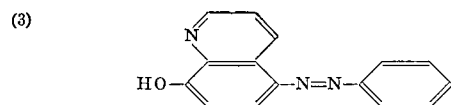
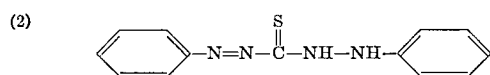
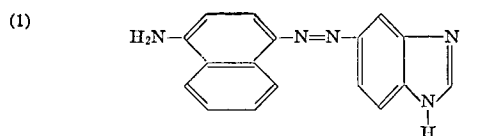
As examples of diffusible materials which can be rendered non-diffusible by reaction with silver ions, e.g., a soluble silver complex formed in terms of unexposed areas of the emulsion, mention may be made of those compounds disclosed in U.S. Pat. 3,443,941 which are complete dyes containing a moiety known in the photographic art as an organic antifogging agent. This moiety reacts with silver ions, for example, by forming addition complexes, to provide a relatively stable, non-diffusible silver-containing reaction product. As examples of such antifoggant moieties, mention may be made of benzimidazoles, thiocarbazonates, benzotriazoles, mercaptothiazoles, hydroxyquinolines, azoethionates, azopyridines, bis-pyrazoles, tetrazolindenes, etc.

Such compounds may be defined as dyes which are diffusible in an aqueous alkaline medium and which may be represented by the following formula:

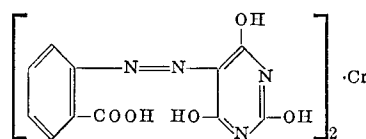
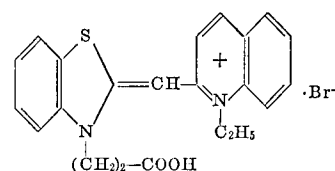
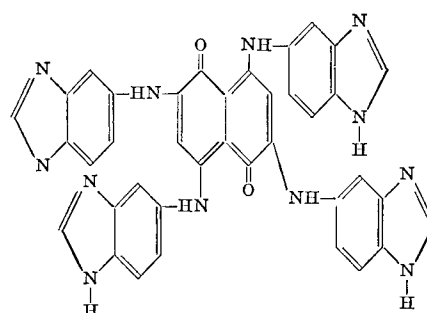
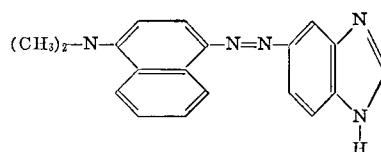
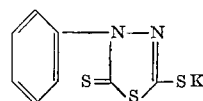
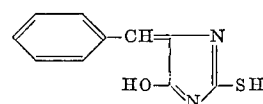
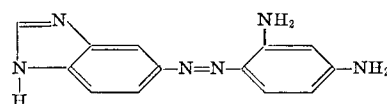
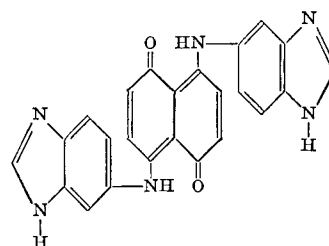
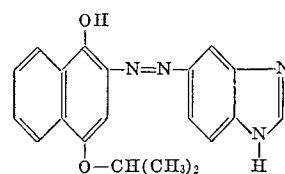


wherein A is a substituent reactable with silver ions to provide a reaction product which is non-diffusible in an aqueous alkaline medium, e.g., a monovalent radical of an organic antifogging agent, and D is the remainder of the dye molecule.

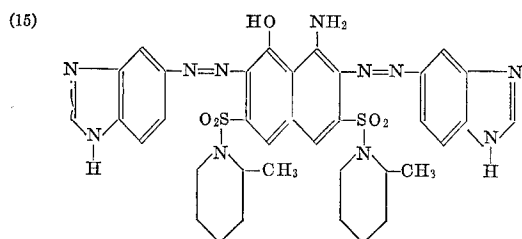
As examples of compounds of this description, mention may be made of the following:



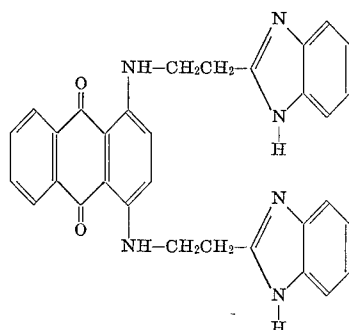
16



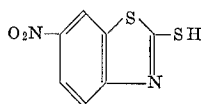
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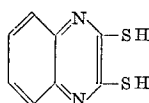
(17)



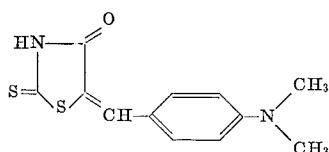
(18)



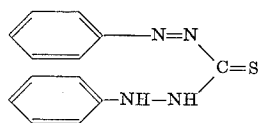
(19)



21)



(22)



In this embodiment of the invention, exposed silver halide causes the non-diffusible color-providing material to be oxidized and in turn to ring-close and release the diffusible color-providing moiety to form an imagewise distribution of the same in terms of exposed areas of the emulsion; while the soluble silver complex formed in terms of unexposed and undeveloped areas migrates or diffuses to the diffusible color-providing moiety to render it non-diffusible, thereby forming a second imagewise distribution of this latter diffusible material in areas where it is unreacted with the soluble silver complex. As in the first-named embodiment, the two imagewise distributions are then transferred, by imbibition, to a superposed dyeable stratum to impart thereto a color transfer image. In the latter instance, however, it will be seen that a negative color transfer image is obtained.

The control of the non-diffusible material by exposed silver halide, i.e., the reaction by which the material is oxidized to provide an oxidation product which autoreacts intramolecularly to ring-close and release the diffusible color-providing moiety, is in accordance with the pro-

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cedures described in the aforementioned U.S. Pat. No. 3,443,939.

This may be accomplished by a redox reaction with oxidized developing agent obtained by development of the exposed silver halide emulsion with a processing composition containing a silver halide developing agent, the oxidation product of which is mobile and reducible. In such a system, exposed silver halide is reduced to silver and as a function of development the developing agent is in turn oxidized to provide an imagewise distribution of oxidized developing agent in terms of exposed areas of the silver halide emulsion. This imagewise distribution of oxidized developing agent possesses sufficient mobility to migrate and to contact the color-providing material whereby a redox reaction occurs in which the developing agent is reduced and the color-providing material is in turn oxidized in an imagewise pattern corresponding to exposed areas of the emulsion to effect ring-closure and subsequent release of the mobile color-providing moiety in terms of exposed areas of the emulsion. In the preparation of monochromatic color transfer images by this procedure, the non-diffusible color-providing material may be present initially in the photosensitive element or in the processing composition.

As examples of silver halide developing agents contemplated by the above description, mention may be made of dihydroxybenzene developers such as the hydroquinones, aminophenol developers such as metol, diamino-benzene developers such as 2-amino-5-diethylamino-toluene, etc.

As is disclosed in U.S. Pat. 3,443,939, the non-diffusible color-providing material may also be oxidized in terms of exposed areas of the associated emulsion by a system employing an aqueous alkaline processing composition containing no additional silver halide developing agent. In this instance, the color-providing material, which is itself a silver halide developing agent, is the only silver halide developing agent employed. In this system, the non-diffusible color-providing material-developing agent associated with the emulsion is contacted with an aqueous alkaline medium to bring it into contact with the exposed emulsion so that exposed silver halide is developed and the material is in turn oxidized. Although the non-diffusible color-providing material is substantially immobile, i.e., of very low mobility in the processing fluid, since it is of close proximity to the silver halide emulsion, upon a suitable imbibition time, e.g., two minutes, it will migrate to and develop exposed silver halide. It will be noted, however, that because of its relative immobility, longer imbibition times are needed than in the previously disclosed redox reaction with oxidized developing agent.

The aspect of this invention relating to the formation of negative color transfer images is illustrated in FIG. 3.

As shown therein, support 30 carries a layer 32 of diffusible dye reactable with silver ions to form a non-diffusible reaction product; a layer 36 of non-diffusible dye capable, upon development of exposed silver halide, of releasing a diffusible color-providing moiety for transfer; and a silver halide emulsion layer 38. A spacer layer 34 is preferably provided between layers 32 and 36.

Support 30 may be of the type described above in connection with FIGS. 1 and 2, as may emulsion layer 38 be similar to the emulsion layer of FIGS. 1 and 2.

Layer 32 comprises a diffusible color-providing material, e.g., an antifoggant dye of the type mentioned above and described in U.S. Pat. 3,443,941, contained in a suitable alkali-permeable matrix of gelatin or the like.

Spacer layer 34 may be of gelatin, cellulose acetate hydrogen phthalate, etc.

Layer 36 comprises a non-diffusible color-providing moiety such as those mentioned above and described and claimed in Ser. No. 655,338, contained in an alkali-permeable matrix of the type mentioned. It will be noted that in this embodiment of the invention, unlike that previously described, layer 36 does not contain any silver-precipitat-

ing nuclei. As discussed previously, if it did, the soluble silver complex formed as a function of development would be reduced and the non-diffusible color-providing material oxidized to release the color-providing moiety, thereby permitting both competing reactions to occur so that a uniform distribution of material in layer 36 is released for transfer.

To form a negative color transfer image with the photosensitive element of FIG. 3, the element is exposed and then developed by applying between the thus exposed element and a superposed dyeable stratum an aqueous alkaline processing composition containing at least a silver halide solvent. In the preferred embodiment, the redox reaction with oxidized developing agent is employed and the processing composition accordingly contains a silver halide developing agent of the foregoing description which can provide an oxidation product capable of undergoing a redox reaction with the non-diffusible color-providing material in layer 36. As in the previous embodiment, any of the ingredients of the processing composition may be contained initially in the film unit, in which event the desired composition is obtained by applying the aqueous medium therefor.

Upon applying the processing composition to the exposed element, exposed silver halide is developed and as a function thereof an imagewise distribution of diffusible dye moiety is released in terms of exposed areas from the material in layer 36 in accordance with the reaction mechanism previously discussed. At substantially the same time, an imagewise distribution of a soluble silver complex is formed in terms of unexposed areas of the emulsion. This distribution of soluble silver complex is transferred through layers 36 and 34 to the diffusible material in layer 32 where, as described above, the diffusible material is rendered non-diffusible by reaction with the diffusing silver complex, thereby leaving remaining in layer 32 an imagewise distribution of diffusible material in areas unreacted with unexposed silver halide. Both of these imagewise distributions of color-providing material are then transferred, at least in part, by imbibition to a superposed dyeable stratum to impart thereto a negative color transfer image.

It will be appreciated that the elements illustrated in FIGS. 1 through 3 are capable of various modifications in structure and in components and materials employed without departing from the scope of this invention. For example, reference has been made in FIG. 1 to a separate element containing the image-receiving layer. In lieu thereof, this layer and the other layers may be contained on a common support. In one useful system a layer of a suitable opacifying material may be provided between the emulsion layer and the receiving layer. This layer of opacifying material effectively masks the negative image formed in the emulsion layer and provides the desired background for viewing the color transfer image formed in or on the receiving layer so that a composite print is thereby formed which is viewable without separation of the receiving layer as a positive color reflection print. As examples of useful opacifying materials, mention may be made of pigments, particularly white pigments such as titanium dioxide, barium sulfate, magnesium oxide, etc.

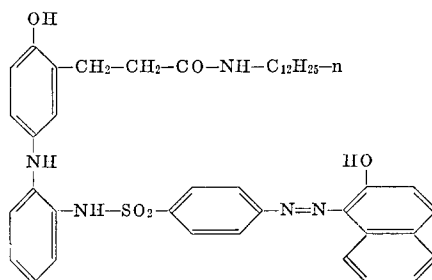
While for purposes of illustration, the photosensitive elements have been shown to contain a single silver halide emulsion and associated color-providing materials, a plurality of such emulsions and associated materials may be employed. Accordingly, both monochromatic and multicolor transfer images are contemplated by this invention.

The following examples illustrate the practice of this invention.

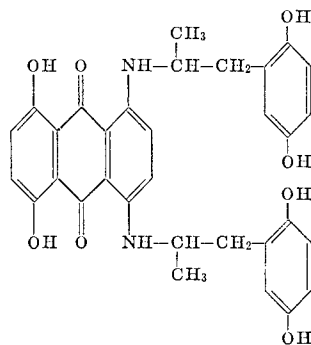
Example 1

On a cellulose triacetate support was coated a layer containing, per square foot of surface area, 15 mgms.

of a colloidal silver precipitating agent and 200 mgms. of a non-diffusible reddish dye of the formula:



in a gelatin matrix. Over this was applied a spacer layer containing 100 mgms./ft.² of gelatin; a layer containing 125 mgms./ft.² of a cyan dye developer of the formula:



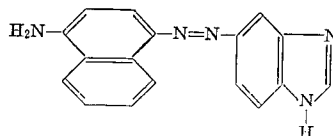
followed by a silver halide emulsion layer containing about 100 mgms. per square foot of silver; and an outer layer containing 30 mgms. per square foot of gelatin. The resulting photosensitive element was photoexposed for about one second and then processed by spreading between the thus exposed element and a superposed image-receiving element of the type described in the aforementioned U.S. Pat. No. 3,362,819, at a gap of about .0036 inch, a processing composition comprising the following proportions of ingredients:

Water—100.0 cc.
Hydroxyethyl cellulose—3.9 g.
Sodium hydroxide—5.0 g.
Sodium thiosulfate—8.0 g.

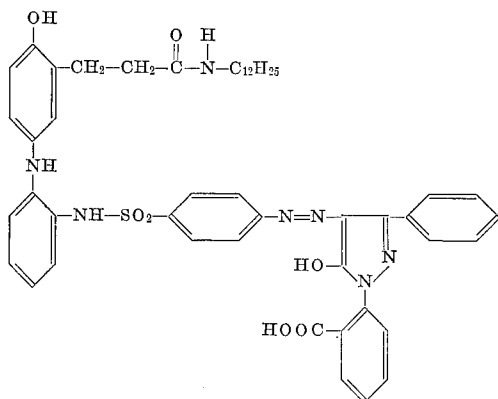
After an imbibition period of about 90 seconds, the respective elements were separated to reveal a neutral tone positive color transfer image.

Example 2

To prepare a negative color transfer image, one may employ a photosensitive element such as shown in FIG. 3, using as the diffusible color-providing material a yellow dye of the formula:



and as the non-diffusible color-providing material, a yellow dye of the formula:



Exposure and development may be accomplished as in Example 1, employing a similar developing composition including about 1.6 g. of a suitable developing agent such as metol, to obtain a yellow negative transfer image.

From the foregoing description and illustrative examples, it will be seen that the present invention provides a system wherein two color-providing materials are transferred for each silver halide emulsion layer contained in the photosensitive elements, the color of the resulting image being a function of the spectral absorption characteristics of the mixture of dyes so transferred, in accordance with principles well known, for example, in the dye art. If two complementary dyes are transferred, e.g., yellow-blue; magenta-green; or cyan-red, a neutral tone image in the black or blue-black family will be obtained. If the two materials providing substantially the same color are transferred, the resulting image will be likewise of substantially the same color. These facts in turn form the basis for certain significant advantages provided by this invention.

Regardless of the colors of the materials employed, the present invention permits of a more efficient use of silver. In prior processes, image formation is based upon a reaction involving either exposed or unexposed silver halide, so that a given amount of this silver halide must be present to obtain a color image of a given density. In the present invention, use is made of both unexposed and exposed silver halide so that it is theoretically possible to get the same dye density with half as much silver, or, stated another way, twice as much dye transferred with the same silver. In other words, the present invention makes it possible to employ emulsion layers containing less silver, thereby reducing the cost of the product or, conversely, to obtain extremely dense color images with an emulsion containing more or less standard amounts of silver. In addition, since both exposed and unexposed silver halide are utilized to control transfer, it is possible to obtain appreciably higher film speeds.

Where images of a given color are desired, particularly in multicolor systems for providing reproductions of the highest fidelity and accuracy of color rendition, the present invention provides a novel system for color correction. By way of illustration, in multicolor transfer systems, use is customarily made of a blue-sensitive emulsion having a yellow dye associated therewith; a green-sensitive emulsion having a magenta dye associated therewith; and a red-sensitive emulsion having a cyan dye associated therewith. (Such a system employing dye developers has been previously mentioned.) In these color systems, each of the requisite dyes must ideally combine several features including the requisite color, transferability and stability. It is sometimes difficult to obtain a single dye possessing all these features to such an extent in the system employed that a multicolor image of the desired high standards is obtained. It is sometimes the case that a dye that is otherwise acceptable is not quite of

the right color. The present invention provides a color correction system wherein a second dye may be employed in association with the emulsion layer, both dyes possessing essentially the same spectral absorption characteristics, but one being slightly different to correct the color deficiency of the other, so that the sum total of dye transferred provides just the right color. To accomplish this, substantially equal amounts of each dye may be transferred, or the amounts may be unequal. For instance, it is contemplated that one of the two dyes may be present initially in relatively small amounts where only a slight amount of this dye need be transferred to provide the requisite color.

On the other hand, by providing two complementary dyes, a system is provided for obtaining dense neutral dye or black-and-white images. Either positive or negative images of this type can be prepared, according to the system employed. It will, therefore, be apparent that this embodiment of the invention is not just of interest in black-and-white photography per se, but also in special applications such as document copying and X-ray. The above-noted efficient use of silver makes the invention particularly useful in such fields.

Moreover, in certain uses such as X-ray and document copying, a color image other than black-and-white is sometimes desirable. It will be seen that the present invention also permits one to obtain dense monochromes of any color desired.

It will, therefore, be seen that the present invention provides a color system of use in a variety of different photographic applications.

While reference has been made in the foregoing description and illustrative examples to contain color-providing materials which may be employed in the practice of this invention, it will be appreciated that the invention is not limited thereto. By way of illustration, it is contemplated that one may employ in the practice of this invention the materials disclosed, for example, in U.S. Pats. Nos. 3,227,550, 3,227,551, 3,227,552, and 3,243,294.

Since certain changes may be made in the above product and process without departing from the scope of the invention herein involved, it is intended that all matter contained in the above description or shown in the accompanying drawing shall be interpreted as illustrative and not in a limiting sense.

What is claimed is:

1. A photographic product comprising a photosensitive element having at least one light sensitive silver halide layer having associated therewith a first color providing material which is normally diffusible in an aqueous alkaline medium and a second color providing material which is normally non-diffusible in said medium, said diffusible material being reactable in the presence of an aqueous alkaline medium with one or the other of the exposed or non-exposed image patterns formed by exposing the product to a pattern of activating radiation and upon reaction with said pattern providing a non-diffusible reaction product; said non-diffusible color providing material being reactable in the presence of said aqueous alkaline medium with the other remaining silver halide image pattern to provide an imagewise distribution of diffusible dyes whereby said color providing materials form two imagewise distributions of diffusible dyes which are substantially identical in terms of the point to point degree of exposure of said silver halide.

2. A product as defined in claim 1 wherein said diffusible color-providing material is a dye developer.

3. A product as defined in claim 1 wherein said diffusible color-providing material is a dye of the formula:

D-A

wherein A is a substituent reactable with silver ions to provide a reaction product which is non-diffusible in an aqueous alkaline medium; and D is the remainder of the dye molecule.

4. A product as defined in claim 1 wherein said non-diffusible color-providing material is a compound containing a diffusible color-providing moiety and an anchoring moiety rendering said compound non-diffusible, said compound upon oxidation forming an oxidation product which can autoreact intramolecularly to effect ring-closure and to eliminate said diffusible color-providing moiety.

5. A photographic product comprising a photosensitive element having at least one light-sensitive silver halide layer having associated therewith a first color-providing material which is diffusible in an aqueous alkaline medium and a second color-providing material which is non-diffusible in said medium, said first material being capable, in the presence of said medium, of reacting with exposed silver halide in said layer to form a non-diffusible product, said second material being capable of reacting with unexposed silver halide to form a diffusible product.

6. A photographic product comprising a photosensitive element having at least one light-sensitive silver halide layer having associated therewith a first color-providing material which is diffusible in an aqueous alkaline medium and a second color-providing material which is non-diffusible in said medium, said first material being reactable with unexposed silver halide to form a non-diffusible product, said second material being capable of forming a diffusible product, as a function of developing exposed silver halide in said layer by contacting said exposed silver halide with an aqueous alkaline processing medium.

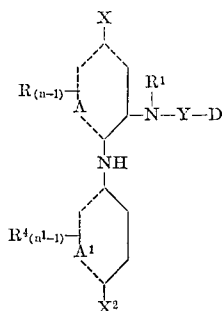
7. A photographic product comprising a photosensitive element including at least one light-sensitive silver halide layer having associated therewith a dye developer diffusible in an aqueous alkaline medium and a compound containing a diffusible color-providing moiety and an anchoring moiety rendering said compound non-diffusible, said compound being capable of reacting with silver ions to release said diffusible color-providing moiety, said compound and said dye developer being contained in separate layers in said element.

8. A product as defined in claim 7 wherein said layer containing said non-diffusible compound also includes silver-precipitating nuclei.

9. A product as defined in claim 7 wherein silver-precipitating nuclei are present in a layer contiguous with said layer of non-diffusible compound.

10. A product as defined in claim 7 wherein said compound upon oxidation forms an oxidation product which can autoreact intramolecularly to effect ring-closure and to eliminate said diffusible color-providing moiety.

11. A product as defined in claim 10 wherein said compound is of the formula:



wherein:

each of A and A¹ represents the atoms necessary to complete an aromatic ring;

D is a color-providing moiety;

Y is a substituent which completes an amide with and reduces the basic character of the 3-amino substituent bonded thereto, said Y substituent further being a divalent radical linking said D moiety to said 3-nitrogen atom;

X and X² each is hydrogen, hydroxy, amino or the substituent R, provided that at least one of X and X² must be hydroxy or amino;

R and R⁴ each represents an anchoring moiety rendering said compound non-diffusible;

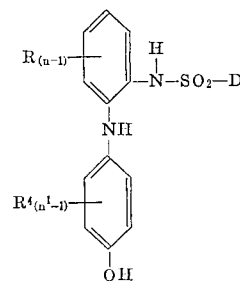
R¹ is hydrogen, alkyl or substituted alkyl; and

n and n¹ each is 1 or 2, provided that when R¹ is an alkyl radical or X or X² is a secondary or tertiary amino comprising an anchoring moiety rendering said compound non-diffusible or when R¹, X and X² together contribute an anchoring moiety, n and n¹ may be 1, but when said substituents alone or together do not contribute an anchoring moiety at least one of n and n¹ must be 2.

12. A product as defined in claim 7 wherein said dye developer and said compound are of a similar color.

13. A product as defined in claim 7 wherein said dye developer and said compound are of complementary colors.

14. A photographic product comprising a support carrying on one side thereof a light-sensitive silver halide emulsion having associated therewith a layer of diffusible dye developer and a layer containing silver precipitating nuclei and a non-diffusible dye of the formula:



wherein:

each of R and R⁴ comprises an amide of at least 13 carbon atoms, said amide being bonded directly to a nuclear carbon atom of the shown benzene moiety or linked thereto through a phenylene or alkylene substituent;

each of n and n¹ is 1 or 2, provided that at least one of said n and n¹ is 2; and

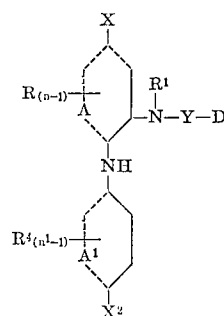
D is a monoazo, disazo or anthraquinone dye moiety.

15. A product as defined in claim 14 including a scavenger for oxidized silver halide developing agent disposed in a layer between said layer containing said emulsion and said layer containing said non-diffusible dye.

16. A photographic product comprising a support carrying on one side thereof a light-sensitive silver halide emulsion having associated therewith a dye diffusible in an aqueous alkaline medium and a dye which is not diffusible in said medium, said dyes being disposed in different layers in said element, said diffusible dye being of the formula:



wherein A is a substituent reactable with silver ions to provide a reaction product which is non-diffusible in an aqueous alkaline medium; and D is the remainder of the dye molecule; said non-diffusible dye being of the formula:



wherein:

each of A and A¹ represents the atoms necessary to

complete an aromatic ring;
 D is a color-providing moiety;
 Y is a substituent which completes an amide with and reduces the basic character of the 3-amino substituent bonded thereto, said Y substituent further being a divalent radical linking said D moiety to said 3-nitrogen atom;
 X and X² each is hydrogen, hydroxy, amino or the substituent R, provided that at least one of X and X² must be hydroxy or amino;
 R and R⁴ each represents an anchoring moiety rendering said compound non-diffusile;
 R¹ is hydrogen, alkyl or substituted alkyl; and
 n and n¹ each is 1 or 2, provided that when R¹ is an alkyl radical or X or X² is a secondary or tertiary amino comprising an anchoring moiety rendering said compound non-diffusile or when R¹, X and X² together contribute an anchoring moiety, n and n¹ may be 1, but when said substituents alone or together do not contribute an anchoring moiety at least one of n and n¹ must be 2.

17. A process for forming color transfer images comprising the steps of exposing a photosensitive element including at least one light-sensitive silver halide layer having associated therewith a first color-providing material which is normally diffusile in an aqueous alkaline processing medium and a second color-providing material which is normally non-diffusile in said medium, to form a developable image;

contacting the exposed photosensitive element with an aqueous alkaline processing medium to develop said image and, as a function of development, selectively immobilizing said first-named material to provide a first imagewise distribution of mobile and diffusile color-providing material and selectively rendering said second-named material diffusile to provide a second imagewise distribution of mobile and diffusile color-providing material in areas corresponding to said first imagewise distribution;

and transferring said two images, at least in part, by imbibition, to a superposed stratum to impart thereto a color transfer image.

18. A process as defined in claim 17 wherein said two imagewise distributions are formed in terms of unexposed areas of said silver halide layer and said transfer image is a positive color image.

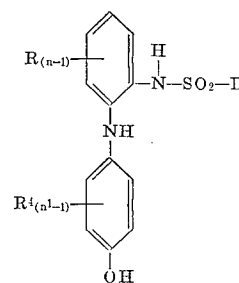
19. A process as defined in claim 17 wherein said color-providing materials are of complementary colors.

20. In a process wherein a photosensitive element including at least one light-sensitive silver halide layer is exposed to provide exposed silver halide areas containing a developable image and unexposed silver halide areas and said element is contacted with a processing composition to develop said exposed silver halide areas and to form an imagewise distribution of image-forming constituents which are transferred to a stratum to impart thereto a transfer image; the improvement which comprises the steps of employing exposed silver halide in said layer to control transfer of a first color-providing material and unexposed silver halide to control transfer of a second color-providing material; as a function thereof forming two imagewise distributions of diffusile color-providing material for each said silver halide layer in said element, said two imagewise distributions being substantially identical in terms of the point-to-point degree of exposure of said layer; and transferring said two imagewise distributions to a superposed stratum to impart thereto a color transfer image.

21. A process for forming images in color comprising exposing a photosensitive element including at least one light-sensitive silver halide layer to form a developable image; contacting said exposed layer with an aqueous alkaline medium including a silver halide solvent and a dye developer; selectively oxidizing said dye developer as a function of development to provide an imagewise dis-

tribution of unreacted and unoxidized dye developer in terms of unexposed areas, and at substantially the same time forming an imagewise distribution of soluble silver complex in terms of unexposed silver halide; transferring said soluble silver complex to a layer containing a compound having a color-providing moiety diffusile in said aqueous alkaline medium and an anchoring moiety rendering said compound non-diffusile in said medium; reacting said compound with said complex to release said diffusile color-providing moiety from said anchoring moiety, thereby providing an imagewise distribution of said released diffusile moiety in terms of unexposed areas of said silver halide layer; and transferring both of said imagewise distribution of unoxidized dye developer and said imagewise distribution of diffusile color-providing moiety, by imbibition, to a superposed stratum to impart thereto a color transfer image.

22. A process as defined in claim 21 wherein said compound is of the formula:



wherein:

each of R and R⁴ comprises an amide of at least 13 carbon atoms, said amide being bonded directly to a nuclear carbon atom of the shown benzene moiety or linked thereto through a phenylene or alkylene substituent;

each of n and n¹ is 1 or 2, provided that at least one of said n and n¹ is 2; and

D is a monoazo, disazo or anthraquinone dye moiety.

23. A process for forming negative color transfer images comprising the steps of exposing a photosensitive element including at least one light-sensitive silver halide emulsion to form a developable image; developing said image and as a function of development selectively oxidizing in terms of exposed areas of said emulsion a non-diffusile compound containing a diffusile dye moiety and an anchoring moiety rendering said compound non-diffusile, said compound upon oxidation forming an oxidation product which autoreacts intramolecularly to effect ring-closure and to eliminate said diffusile dye, thereby forming a first imagewise distribution of diffusile dye in terms of exposed areas of said emulsion; forming from unexposed silver halide in said layer an imagewise distribution of a soluble silver complex; transferring this imagewise distribution of soluble silver complex into an adjacent layer containing a diffusile dye reactable with silver ions to form a non-diffusile reaction product; reacting said complex with said diffusile dye to form a non-diffusile reaction product, thereby leaving in said adjacent layer a second imagewise distribution of dye in terms of exposed areas of said emulsion; and transferring both of said imagewise diffusile dye distributions to a superposed stratum to impart thereto a negative color transfer image.

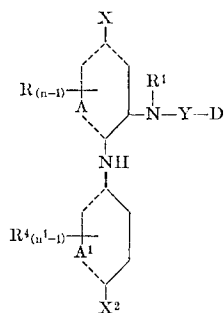
24. A process as defined in claim 23 wherein said exposed element is developed by contacting it with an aqueous alkaline processing composition including a silver halide solvent and a silver halide developing agent, and said non-diffusile compound is oxidized by a redox reaction with oxidized developing agent obtained as a function of developing exposed silver halide.

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25. A process as defined in claim 24 wherein said diffusible dye is of the formula:



wherein A is a substituent reactable with silver ions to provide a reaction product which is non-diffusible in an aqueous alkaline medium and D is the remainder of the dye molecule; and said non-diffusible dye is of the formula:



wherein:

each of A and A¹ represents the atoms necessary to complete an aromatic ring;

D is a color-providing moiety;

Y is a substituent which completes an amide with and reduces the basic character of the 3-amino substituent bonded thereto, said Y substituent further being

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a divalent radical linking said D moiety to said 3-nitrogen atom;

X and X² each is hydrogen, hydroxy, amino or the substituent R, provided that at least one of X and X² must be hydroxy or amino;

R and R⁴ each represents an anchoring moiety rendering said compound non-diffusible;

R¹ is hydrogen, alkyl or substituted alkyl; and

n and n¹ each is 1 or 2, provided that when R¹ is an alkyl radical or X or X² is a secondary or tertiary amino comprising an anchoring moiety rendering said compound non-diffusible or when R¹, X and X² together contribute an anchoring moiety, n and n¹ may be 1, but when said substituents alone or together do not contribute an anchoring moiety at least one of n and n¹ must be 2.

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