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(54) **Process for forming image.**

(57) A process for forming an image by imagewise forming a mobile dye by heating in a substantially waterfree condition and transferring the dye into a dye fixing layer using water. The amount of water used to transfer the dye is within the range from 0.1 time the total weight of all coatings to the difference obtained by subtracting the total weight of all coatings from the weight of water corresponding to the maximum swollen volume of all coatings.

ACTORUM AG

PROCESS FOR FORMING IMAGE

FIELD OF THE INVENTION

The present invention relates to a novel process for forming an image which comprises imagewise forming a mobile dye by heating in a substantially water-free condition and transferring the dye into a dye fixing layer using water. More particularly, the present invention relates to a process for forming an image, wherein a small amount of water is used in transferring an imagewise formed dye into a dye fixing layer.

BACKGROUND OF THE INVENTION

A photographic process utilizing a silver halide has heretofore been widely used due to its excellent photographic characteristics such as sensitivity, control of gradation, etc., as compared with other photographic processes such as an electrophotographic process or a diazo photographic process. In recent years, with respect to image formation process for light-sensitive materials using a silver halide, a technique capable of easily and quickly obtaining an image has been developed by changing the conventional wet development process using, for example, a developing solution, into a dry development process such as a process using heat.

A heat-developable light-sensitive material has been known in the field of this technique. Such heat-developable light-sensitive materials and processes therefor are described in, for example, Shashin Kogaku no Kiso, Corona Co., Ltd., pages 553-555, Eizó Joho, April 1978, page 40, Neblets Handbook of Photography and Reprography, 4th Ed., Van Nostrand Reinhold Co., pages 32-33, U.S. Patents 3,152,904, 3,301,678, 3,392,020 and 3,457,075, British Patents 1,131,108 and 1,167,777, and Research Disclosure, June 1978, pages 9-15 (RD-17029).

Namely processes for obtaining color images by a dry process have been proposed.

With respect to processes for forming color images by the reaction of an oxidation product of a developing agent with a coupler, it has been proposed to use a p-phenylenediamine type reducing agent and a phenolic coupler or an active methylene coupler as described in U.S. Patent 3,531,286, a p-aminophenol type reducing agent as described in U.S. Patent 3,761,270, a sulfonamidophenol type reducing agent as described in Belgian Patent 802,519 and Research Disclosure, pages 31 and 32 (Sept. 1975) and the combination of a sulfonamidophenol type reducing agent and a 4-equivalent coupler as described in U.S. Patent 4,021,240.

These processes, however, are disadvantageous in that turbid color images are formed, because a reduced silver image and a color image are simultaneously formed on the exposed area after heat-development. In order to eliminate these disadvantages, there have been proposed a process which comprises removing a silver image by liquid processing or a process which comprises transferring only the dye to another layer, for example, a sheet having an image receiving layer. However, the latter process is not desirable because it is not easy to transfer only the dye as distinguishable from unreacted substances.

Another process which comprises introducing a nitrogen containing heterocyclic group into a dye, forming a silver salt and releasing a dye by heat-development has been described in Research Disclosure, No. 16966, pages 54 to 58 (May 1978). According to this process, clear images cannot be obtained, because it is difficult to control the release of dyes from nonexposed areas, and thus it is not a conventionally applicable process.

Also, processes for forming a positive-working color image by a silver dye bleach process utilizing heat, with useful dyes and methods for bleaching have been described, for example, in Research Disclosure, No. 14433, pages 30 to 32 (April 1976), ibid., No. 15227, pages 14 and 15 (Dec. 1976) and U.S. Patent 4,235,957.

However, this process requires an additional step and an additional material for accelerating bleaching of dyes, for example, heating with a superposed sheet with an activating agent. Furthermore, it is not desirable because the resulting color images are gradually reduced and bleached by coexisting free silver during long periods of preservation.

Moreover, a process for forming a color image utilizing a leuco dye has been described, for example, in U.S. Patents 3,985,565 and 4,022,617. However, this process is not desirable because it is difficult to stably incorporate the leuco dye in the photographic material and coloration gradually occurs during preservation.

Furthermore, these processes described above generally have disadvantages that a relatively long period of time is required for development and only images having a high level of fog and a low density are obtained.

SUMMARY OF THE INVENTION

The present invention provides a novel process for obtaining a visible image by transferring a dye which is imagewise obtained by heating in a substantially water-free condition into a dye fixing layer and overcomes the problems involved in the conventional materials.

Accordingly, an object of the present invention is to provide a novel process for forming an image by transferring a mobile hydrophilic dye released by heating in a substantially water-free condition into a dye fixing layer using a small amount of water to obtain a dye image.

Another object of the present invention is to provide a novel process for obtaining a smear-free, uniform and sharp color image.

These objects of the present invention are attained by a process for forming an image which comprises

heating a light-sensitive material comprising a support having thereon at least a light-sensitive silver halide, a binder and a dye releasing redox compound which is reductive to the light-sensitive silver halide and capable of releasing a hydrophilic dye upon reaction with the light-sensitive silver halide by heating, after imagewise exposure or simultaneously with imagewise exposure, in a substantially water-free condition to imagewise form a mobile dye; and

transferring the mobile dye into a dye fixing material using water,

wherein when the dye is transferred, the amount of water present in all the coating of the light-sensitive material and dye fixing material is within a range of from 0.1 time the total weight of all the coatings to the difference obtained by subtracting the total weight of all the coatings from the weight of water corresponding to the maximum swollen volume of all the coatings.

DETAILED DESCRIPTION OF THE INVENTION

The maximum amount of water required to transfer the dye imagewise formed by heating to the dye fixing layer depends on the total thickness of the light-sensitive material and dye fixing material when swollen.

One of the features of the present invention is that the mobile dye is transferred with water in an amount not exceeding the total weight of water corresponding to the swollen volume of the light-sensitive material and dye fixing material when those are sufficiently swollen by applying a sufficient amount of water; in other words, the mobile dye is transferred in the condition that the coatings of the light-sensitive material and dye fixing material are not sufficiently swollen. If the amount of water exceeds the above specified upper limit, a smear occurs on the color image transferred and this is undesirable.

The minimum amount of water is sufficient if such can transfer the dye. The preferred minimum amount is 0.1 time the total weight of all the coatings of the light-sensitive material and image-receiving material in a dry state and in this amount, the dye released can be completely transferred.

The coatings are unstable in a swollen state. Depending on conditions, a smear may occur locally. In order to prevent such smearing, the amount of water is preferably 0.8 time or less the difference obtained by subtracting the amount of water corresponding to the total weight of all the coatings from the amount of water corresponding to the maximum swollen volume of all the coatings of the light-sensitive and dye fixing materials.

The amount of water in the present invention is an amount of water present in all the coatings at the time of transferring. The amount of water is controlled within the range as specified above by applying water in a previously determined amount falling within the above specified range or by applying a sufficient amount of water and then adjusting the amount of water within the specified range by squeezing by means of, e.g., rollers, or drying by heating.

Any binder can be used in the preparation of the coatings of the present invention so long as transferring the dye with water can be made. These coatings can contain a light-sensitive silver halide, a dye releasing redox compound, a mordant, a high boiling organic solvent, and the like. Those additives do not change the nature of the present invention described above.

In the case of a coating (4 g/m^2) with gelatin as a binder, assuming that its thickness in a swollen state is 12 μ, the amount of water required for transferring the dye is from 0.4 to 8 ml per square meter of the coating.

The maximum swollen volume can be determined as follows:

A light-sensitive material or dye fixing material having a coating to be measured is immersed in water for transferring and sufficiently swollen. When sufficiently swollen, the thickness of the coating to be measured is determined by measuring the length of the cross-section by means of, e.g., a microscope. By multiplying the thickness as determined above by the surface area of the coating to be measured, the maximum swollen volume can be obtained.

A method of measuring a degree of swelling is described in Photographic Science Engineering, Vol. 16, page 449 (1972).

The degree of swelling of a gelatin coating greatly varies depending on a degree of hardening of the coating. The degree of hardening is usually controlled so that the thickness of the coating in a maximum swollen state is 2 to 6 times the thickness thereof in a dry state.

The photographic light-sensitive material of the present invention may contain inorganic or organic hardeners in hydrophilic colloid layers such as a photographic emulsion layer. Examples of the hardeners which can be used include chromium salts (e.g., chromium alum and chromium acetate), aldehydes (e.g., formaldehyde, glyoxal, and glutaraldehyde), N-methylol compounds (e.g., dimethylol urea and methyloldimethyl hydantoin), dioxane derivatives (e.g., 2,3-dihydroxydioxane), active vinyl compounds (e.g., 1,3,5-triacryloyl-hexahydro-s-triazine and 1,3-vinylsulfonyl-2-propanol), active halogen compounds (e.g., 2,4-dichloro-6-hydroxy-s-triazine), and mucophenoxyhalogeno acids (e.g., mucochloric acid and mucophenoxychloric acid). These compounds can be used alone or in mixtures thereof.

Water may be applied to either of the dye fixing material or the light-sensitive material. Alternatively, water may be applied to both the dye fixing material and the light-sensitive material.

In the process of the present invention, water may be applied in any suitable manner. Water may be jetted through a small hole, or the material can be wet with water using a web roller. In addition, a procedure in which pods containing water are smashed may be used. The present invention is not limited to these methods.

The water as used herein is not limited to "pure water" and includes water in a sense widely and customarily used.

General drinking water, industrial water, etc. can be used in the present invention. With regard to water which is used as the general drinking water, there are the water quality standards for tap water as adopted in U.S.A. and the water quality standards as prescribed by WHO (World Health Organization). Water satisfying these standards, of course, can be used in the process of the present invention. In addition, water ordinarily used in various industries can be used in the process of the present invention. The standards for quality of water used in various industries are described in, for example, Eisei Kogaku Handbook, Asakura Shoten, page 356 (1967). Water satisfying the above standards can be used as the water intended in the present invention, even though it is underground water or river water, or water to which medicines are added for some purpose.

The term "dye image" as used herein includes both multicolor and monocolor dye images. The monocolor dye image includes a multicolor image obtained by mixing at least two dyes.

According to the image formation process for the present invention, a silver image and in the portion corresponding the silver image, a mobile dye and a silver image, are simultaneously given by merely heating after imagewise exposure.

That is, when the light-sensitive material is imagewise exposed and heat developed in a substantially water-free condition, a redox reaction occurs between a light-sensitive silver halide and/or an organic silver salt oxidizing agent and a reducing dye releasing redox compound with the exposed light-sensitive silver halide as a catalyst and a silver image is formed on the exposed area. In this step, the dye releasing redox compound is oxidized by the organic silver salt oxidizing agent to form its oxidation product. As a result, a hydrophilic mobile dye is released and a silver image and mobile dye are formed on the exposed area. The presence of a dye releasing aid at this stage accelerates the above described reaction. By transferring the thus formed mobile dye using a small amount of water into the dye fixing layer, the dye image is formed.

The above explanation has been given in a case where a negative-working emulsion is used. In the case of an autopositive-working emulsion, the same is applicable with the exception that the silver image and mobile dye are formed on the non-exposed area.

The redox reaction between the light-sensitive silver halide and the dye releasing redox compound and the subsequent dye releasing reaction according to the present invention are characterized in that such occur at high temperature and also in a substantially water-free dry condition. The term "high temperature" as used herein means a temperature of 80°C or more. The term "substantially water-free dry condition" means a condition which is in an equilibrium state with respect to water in air but with no addition of water from outside the system. This condition is described in T.H. James edit., <u>The Theory of the Photographic Process</u>, 4th ed., Macmillan, page 374. Showing a sufficient reactivity even in a substantially water-free dry condition can be confirmed by the fact that the reactivity of a test sample does not drop even if it is vacuum dried at

10^{-3} mmHg for one day.

The dye releasing reaction has conventionally been believed to occur due to the attack of a nucleophilic reagent and is usually carried out in a liquid having a pH of at least 10. Therefore, it is unexpected that high reactivity can be obtained at high temperature and also in a substantially water-free dry condition. The dye releasing redox compound of the present invention can undergo a redox reaction with a silver halide without the help of the so-called auxiliary developing agent. This is an unexpected result which could not be anticipated from the knowledge of conventional wet development system which is carried out at temperatures near ordinary temperature.

The above-described reactions proceed smoothly particularly in the presence of an organic silver salt oxidizing agent, producing a high image density. It is therefore particularly preferred in the present invention that such an organic silver salt oxidizing agent is used in combination.

It is unexpected from conventional teachings that in the process of the present invention, a sharp dye image can be formed by applying a small amount of water within the above specified range when transferring the mobile dye imagewise formed into the dye fixing layer.

The dye releasing redox compound which releases a hydrophilic diffusible dye used in the present invention is a compound described in European Patent Application (OPI) No. 76,492 as a dye releasing compound and is represented by the following general formula:

$$R_a\text{-}SO_2\text{-}D$$

wherein R_a represents a reducing group capable of being oxidized by the silver halide; and D represents an image forming dye portion containing a hydrophilic group.

The above-described compound is oxidized corresponding to or reversely corresponding to latent image distributed imagewise in the silver halide and releases imagewise a mobile dye.

The detail definitions of R_a and D, examples of the specific compounds and synthesis examples thereof are described in European Patent Application (OPI) No. 76,492.

As the dye releasing redox compounds used in the present invention, the compounds as described, for example, in U.S. Patent 4,055,428, Japanese Patent Application (OPI) Nos. 12642/81, 16130/81, 16131/81, 650/82 and 4043/82, U.S. Patents 3,928,312 and 4,076,529, U.S. Published Patent Application B 351,673, U.S. Patents

4,135,929 and 4,198,235, Japanese Patent Application (OPI) No. 46730/78, U.S. Patents 4,273,855, 4,149,892, 4,142,891 and 4,258,120, etc., are also effective in addition to the above-described compounds.

Further, the dye releasing redox compounds which release a yellow dye as described, for example, in U.S. Patents 4,013,633, 4,156,609, 4,148,641, 4,165,987, 4,148,643, 4,183,755, 4,246,414, 4,268,625 and 4,245,028, Japanese Patent Application (OPI) Nos. 71072/81, 25737/81, 138744/80, 134849/80, 106727/77, 114930/76, etc., can be effectively used in the present invention.

The dye releasing redox compounds which release a magenta dye as described, for example, in U.S. Patents 3,954,476, 3,932,380, 3,931,144, 3,932,381, 4,268,624 and 4,255,509, Japanese Patent Application (OPI) Nos. 73057/81, 71060/81, 134850/80, 40402/80, 36804/80, 23628/78, 106727/77, 33142/80 and 53329/80, etc., can be effectively used in the present invention.

The dye releasing redox compounds which release a cyan dye as described, for example, in U.S. Patents 3,929,760, 4,013,635, 3,942,987, 4,273,708, 4,148,642, 4,183,754, 4,147,544, 4,165,238, 4,246,414 and 4,268,625, Japanese Patent Application (OPI) Nos. 71061/81, 47823/78, 8827/77 and 143323/78, etc., can be effectively used in the present invention.

Two or more of the dye releasing redox compounds can be used together. In these cases, two or more dye releasing redox compounds may be used together in order to represent the same color or in order to represent black color.

The dye releasing redox compounds are suitably used in a range from 10 mg/m^2 to 15 g/m^2 and preferably in a range from 20 mg/m^2 to 10 g/m^2 in a total.

The dye releasing redox compound used in the present invention can be introduced into a layer of the light-sensitive material by known methods such as a method as described in U.S. Patent 2,322,027. In this case, an organic solvent having a high boiling point or an organic solvent having a low boiling point as described below can be used. For example, the dye releasing redox compound is dispersed in a hydrophilic colloid after dissolved in an organic solvent having a high boiling point, for example, a phthalic acid alkyl ester (for example, dibutyl phthalate, dioctyl phthalate, etc.), a phosphoric acid ester (for example, diphenyl phosphate, triphenyl phosphate, tricresyl phosphate, dioctylbutyl phosphate, etc.), a citric acid ester (for example, tributyl acetylcitrate, etc.), a benzoic acid ester (for example, octyl benzoate, etc.), an alkylamide (for example, diethyl laurylamide, etc.), an aliphatic acid ester (for example, dibutoxyethyl succinate, dioctyl azelate, etc.), a trimesic acid ester (for example, tributyl trimesate, etc.), etc., or an organic solvent having a boiling point of about 30°C to 160°C, for example, a lower alkyl acetate such as ethyl acetate, butyl acetate, etc., ethyl propionate, secondary butyl alcohol, methyl isobutyl ketone, β-ethoxyethyl acetate, methyl cellosolve acetate, cyclohexanone, etc.

The above-described organic solvents having a high boiling point and organic solvents having a low boiling point may be used as a mixture thereof.

Further, it is possible to use a dispersion method using a polymer as described in Japanese Patent Publication No. 39853/76 and Japanese Patent Application (OPI) No. 59943/76. Moreover, various surface active agents can be used when the dye releasing redox compound is dispersed in a hydrophilic colloid. For this purpose, the surface active agents illustrated in other part of the specification can be used.

In the present invention, if necessary, a reducing agent may be used. The reducing agent in this case is the so-called auxiliary developing agent, which is oxidized by the silver halide and/or the organic silver salt oxidizing agent to form its oxidized product having an ability to oxidize the reducing group R_a in the dye releasing redox compound.

Examples of useful auxiliary developing agents include the compounds specifically described in European Patent Application (OPI) No. 76,492.

The silver halide used in the present invention includes silver chloride, silver chlorobromide, silver chloroiodide, silver bromide, silver iodobromide, silver chloroiodobromide and silver iodide, etc.

In the embodiment of the present invention in which the organic silver salt oxidizing agent is not used together with but the silver halide is used alone, particularly preferred silver halide is silver halide partially containing a silver iodide crystal in its grain. That is, the silver halide which shows the X-ray diffraction pattern of pure silver iodide is particularly preferred.

In photographic materials a silver halide containing two or more kinds of halogen atoms can be used. Such a silver halide is present in the form of a completely mixed crystal in a conventional silver halide emulsion. For example, the grain of silver iodobromide shows X-ray diffraction pattern at a position corresponding to the mixed ratio of silver iodide crystal and silver bromide crystal but not at a position corresponding to pure silver iodide crystal and pure silver bromide crystal separately.

Particularly preferred examples of silver halide used in the present invention include silver chloroiodide, silver iodobromide, and silver chloroiodobromide each containing silver iodide crystal in its grain and showing X-ray diffraction pattern of silver iodide crystal.

The process for preparing those silver halides is explained taking the case of silver iodobromide. That is, the silver iodobromide is prepared by first adding silver nitrate solution to potassium bromide solution to form silver bromide particles and then adding potassium iodide to the mixture.

Two or more kinds of silver halides in which a particle size and/or a halogen composition are different from each other may be used in mixture.

An average particle size of the silver halide used in the present invention is preferably from 0.001 μm to 10 μm and more preferably from 0.001 μm to 5 μm.

The silver halide used in the present invention may be used as is. However, it may be chemically sensitized with a chemical sensitizing agent such as compounds or sulfur, selenium or tellurium, etc., or compounds of gold, platinum, palladium, rhodium or iridium, etc., a reducing agent such as tin halide, etc., or a combination thereof. The details thereof are described in T.H. James, The Theory of the Photographic Process, the Fourth Edition, Chapter 5, pages 149 to 169.

In the particularly preferred embodiment of the present invention, an organic silver salt oxidizing agent is used together. The organic silver salt oxidizing agent is a silver salt which forms a silver image by reacting with the above-described image forming substance or a reducing agent coexisting, if necessary, with the image forming substance, when it is heated to a temperature of above 80°C and, preferably, above 100°C in the presence of exposed silver halide. By coexisting the organic silver salt oxidizing agent, the light-sensitive material which provides higher color density can be obtained.

The silver halide used in this case is not always necessarily to have the characteristic in that the silver halide contains pure silver iodide crystal in the case of using the silver halide alone. Any silver halide which is known in the art can be used.

Examples of such organic silver salt oxidizing agents include those described in European Patent Application (OPI) No. 76,492.

A silver salt of an organic compound having a carboxy group can be used. Typical examples thereof include a silver salt of an aliphatic carboxylic acid and a silver salt of an aromatic carboxylic acid.

In addition, a silver salt of a compound containing a mercapto group or a thione group and a derivative thereof can be used.

Further, a silver salt of a compound containing an imino group can be used. Examples of these compounds include a silver salt of benzotriazole and a derivative thereof as described in Japanese Patent Publication Nos. 30270/69 and 18416/70, for example, a silver salt of benzotriazole, a silver salt of alkyl substituted benzotriazole such as a silver salt of methylbenzotriazole, etc., a silver salt of a halogen substituted benzotriazole such as a silver salt of 5-chlorobenzotriazole, etc., a silver salt of carboimidobenzotriazole such as a silver salt of butylcarboimidobenzotriazole, etc., a silver salt of 1,2,4-triazole or 1-H-tetrazole as described in U.S. Patent 4,220,709, a silver salt of carbazole, a silver salt of saccharin, a silver salt of imidazole and an imidazole derivative, and the like.

Moreover, a silver salt as described in Research Disclosure, Vol. 170, No. 17029 (June, 1978) and an organic metal salt such as copper stearate, etc., are the organic metal salt oxidizing agent capable of being used in the present invention.

Methods of preparing these silver halide and organic silver salt oxidizing agents and manners of blending them are described in Research Disclosure, No. 17029, Japanese Patent Application (OPI) Nos. 32928/75 and 42529/76, U.S. Patent 3,700,458, and Japanese Patent Application (OPI) Nos. 13224/74 and 17216/75.

A suitable coating amount of the light-sensitive silver halide and the organic silver salt oxidizing agent employed in the present invention is in a total of from 50 mg/m^2 to 10 g/m^2 calculated as an amount of silver.

The light-sensitive silver halide and the organic silver salt oxidizing agent used in the present invention are prepared in the binder as described below. Further, the dye releasing redox compound is dispersed in the binder described below.

The binder which can be used in the present invention can be employed individually or in a combination thereof. A hydrophilic binder can be used as the binder according to the present invention. The typical hydrophilic binder is a transparent or translucent hydrophilic colloid, examples of which include a natural substance, for example, protein such as gelatin, a gelatin derivative, etc., a cellulose derivative, a polysaccharide such as starch, gum arabic, etc., and a synthetic polymer, for example, a water-soluble polyvinyl compound such as polyvinyl alcohol, polyvinyl pyrrolidone, acrylamide polymer, etc. Another example of the synthetic polymer compound is a dispersed vinyl compound in a latex form which is used for the purpose of increasing dimensional stability of a photographic material.

The silver halide used in the present invention can be spectrally sensitized with methine dyes or other dyes. Suitable dyes which can be employed include cyanine dyes, merocyanine dyes, complex cyanine dyes, complex merocyanine dyes, holopolar cyanine dyes, hemicyanine dyes, styryl dyes, and hemioxonol dyes. Of these dyes, cyanine dyes, merocyanine dyes and complex merocyanine dyes are particularly useful. Any conventionally utilized nucleus for cyanine dyes, such as basic heterocyclic nuclei, can be contained in these dyes. That is, a pyrroline nucleus, an oxazoline nucleus, a thiazoline nucleus, a pyrrole nucleus, an oxazole nucleus, a thiazole nucleus, a selenazole nucleus, an imidazole nucleus, a tetrazole nucleus, a pyridine nucleus, etc., and further, nuclei formed by condensing alicyclic hydrocarbon rings with these nuclei and nuclei formed by condensing aromatic hydrocarbon rings with these nuclei, that is, an indolenine nucleus, a benzindolenine nucleus, an indole nucleus, a benzoxazole nucleus, a naphthoxazole nucleus, a benzothiazole nucleus, a naphthothiazole nucleus, a benzoselenazole nucleus, a benzimidazole nucleus, a quinoline nucleus, etc., are appropriate. The carbon atoms of these nuclei may also be substituted.

As nuclei having a ketomethylene structure, 5- or 6-membered heterocyclic nuclei such as a pyrazolin-5-one nucleus, a thiohydantoin nucleus, a 2-thiooxazolidin-2,4-

dione nucleus, a thiazolidin-2,4-dione nucleus, a rhodanine nucleus, a thiobarbituric acid nucleus, etc., may also be used in mérocyanine dyes and complex merocyanine dyes.

These sensitizing dyes can be employed individually, and can also be employed in combination thereof. A combination of sensitizing dyes is often used, particularly for the purpose of supersensitization. Representative examples thereof are described in U.S. Patents 2,688,545, 2,977,229, 3,397,060, 3,522,052, 3,527,641, 3,617,293, 3,628,964, 3,666,480, 3,672,898, 3,679,428, 3,703,377, 3,769,301, 3,814,609, 3,837,862 and 4,026,707, British Patents 1,344,281 and 1,507,803, Japanese Patent Publication Nos. 4936/68 and 12375/78, Japanese Patent Application (OPI) Nos. 110618/77 and 109925/77, etc.

The sensitizing dyes may be present in the emulsion together with dyes which themselves do not give rise to spectrally sensitizing effects but exhibit a supersensitizing effect or materials which do not substantially absorb visible light but exhibit a supersensitizing effect. For example, aminostilbene compounds substituted with a nitrogen-containing heterocyclic group (e.g., those described in U.S. Patents 2,933,390 and 3,635,721), aromatic organic acid-formaldehyde condensates (e.g., those described in U.S. Patent 3,743,510), cadmium salts, azaindene compounds, etc., can be present. The combinations described in U.S. Patents 3,615,613, 3,615,641, 3,617,295 and 3,635,721 are particularly useful.

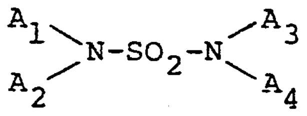
A support used in the light-sensitive material or used as the dye fixing material, if desired, according to the present invention is that which can endure at the processing temperature. As an ordinary support, not only glass, paper, metal or analogues thereto may be used, but also an acetyl cellulose film, a cellulose ester film, a polyvinyl acetal film, a polystyrene film, a polycarbonate film, a polyethylene terephthalate film, and a film related thereto or a plastic material may be used. Further, a paper support laminated with a polymer such as polyethylene, etc., can be used. The polyesters described in U.S. Patents 3,634,089 and 3,725,070 are preferably used.

In the present invention, various kinds of dye releasing activators can be used. The dye releasing activator means a substance which accelerates the oxidation-reduction reaction between the light-sensitive silver halide and/or the organic silver salt oxidizing agent and dye releasing redox compound, or accelerates release of a dye by means of its nucleophilic action to the oxidized dye releasing redox compound in the dye releasing reaction subsequently occurred, and a base and a base precursor can be used. It is particularly advantageous to use these dye releasing activators in order to accelerate the reactions in the present invention.

Examples of preferred bases are amines which include trialkylamines, hydroxylamines, aliphatic polyamines, N-alkyl substituted aromatic amines, N-hydroxyalkyl substituted aromatic amines and bis[p-(dialkylamino)phenyl]methanes. Further, betaine tetramethylammonium iodide and diaminobutane dihydrochloride as described in U.S. Patent 2,410,644, and urea and organic compounds including amino acids such as 6-aminocaproic acid as described in U.S. Patent 3,506,444 are useful. The base precursor is a substance which releases a basic component by heating. Examples of typical base precursors are described in British Patent 998,949. A preferred base precursor is a salt of a carboxylic acid and an organic base, and examples of the suitable carboxylic acids include trichloroacetic acid and trifluoroacetic acid and examples of the suitable bases include guanidine, piperidine, morpholine, p-toluidine and 2-picoline, etc. Guanidine trichloroacetate as described in U.S. Patent 3,220,846 is particularly preferred. Further, aldonic amides as described in Japanese Patent Application (OPI) No. 22625/75 are preferably used because they decompose at a high temperature to form bases.

These dye releasing activators can be used in an amount of a broad range. A useful range is up to 50% by weight based on the amount of a dry layer coated of the light-sensitive material. A range of 0.01% by weight to 40% by weight is more preferred.

It is advantageous to use a compound represented by the general formula described below in the heat-developable color photographic material in order to accelerate development and accelerate release of a dye.



$$\begin{matrix}A_1 \\ A_2\end{matrix} \rangle N-SO_2-N \langle \begin{matrix}A_3 \\ A_4\end{matrix}$$

(A)

wherein A_1, A_2, A_3 and A_4, which may be the same or different, each represents a hydrogen atom or a substituent selected from an alkyl group, a substituted alkyl group, a cycloalkyl group, an aralkyl group, an aryl group, a substituted aryl group and a heterocyclic group; and A_1 and A_2 or A_3 and A_4 may combine with each other to form a ring.

The above-described compound can be used in an amount of broad range. A useful range is up to 20% by weight based on the amount of a dry layer coated of the light-sensitive material. A range of 0.1% by weight to 15% by weight is more preferred.

It is advantageous to use a water releasing compound in the present invention in order to accelerate the dye releasing reaction.

The water releasing compound means a compound which releases water by decomposition during heat development. These compounds are particularly known in the field of printing of fabrics, and $NH_4Fe(SO_4)_2 \cdot 12H_2O$, etc., as described in Japanese Patent Application (OPI) No. 88386/75 are useful.

Further, in the present invention, it is possible to use a compound which activates development and stabilizes the image at the same time. Particularly, it is preferred to use isothiuroniums including 2-hydroxyethylisothiuronium trichloroacetate as described in U.S. Patent 3,301,678, bisisothiuroniums including 1,8-(3,6-dioxaoctane)bis(isothiuronium trifluoroacetate), etc., as described in U.S. Patent 3,669,670, thiol compounds as described in German Patent Application (OLS) No. 2,162,714, thiazolium compounds such as 2-amino-2-thiazolium trichloroacetate, 2-amino-5-bromoethyl-2-thiazolium trichloroacetate, etc., as described in U.S. Patent 4,012,260, compounds having α-sulfonylacetate as an acid part such as bis(2-amino-2-thiazolium)-methylene-bis(sulfonylacetate), 2-amino-2-thiazolium phenylsulfonylacetate, etc., as described in U.S. Patent 4,060,420, and compounds having 2-carboxycarboxamide as an acid part as described in U.S. Patent 4,088,496.

In the present invention, it is possible to use a thermal solvent. The term "thermal solvent" means a non-hydrolyzable organic material which melts at a temperature of heat treatment and melts at a lower temperature of heat treatment when it is present together with other components. Preferred examples of thermal solvents include compounds which can act as a solvent for the developing agent and compounds having a high dielectric constant which accelerate physical development of silver salts. Examples of preferred thermal solvents include those described in European Patent Application (OPI) No. 76,492.

In the present invention, though it is not always necessary to further incorporate substances or dyes for preventing irradiation or halation in the light-sensitive material, because the light-sensitive material is colored by the dye releasing redox compound, it is possible to add filter dyes or light absorbing materials, etc., into the light-sensitive material, as described in Japanese Patent Publication No. 3692/73 and U.S. Patents 3,253,921, 2,527,583 and 2,956,879, etc., in order to further improve sharpness. It is preferred that these dyes have a thermal bleaching property. For example, dyes as described in U.S. Patents 3,769,019, 3,745,009 and 3,615,432 are preferred.

The light-sensitive material used in the present invention may contain, if necessary, various additives known for the heat-developable light-sensitive materials and may have a layer other than the light-sensitive layer, for example, an antistatic layer, an electrically conductive layer, a protective layer, an intermediate layer, an antihalation layer, a strippable layer, etc.

The photographic emulsion layer and other hydrophilic colloid layers in the light-sensitive material of the present invention may contain various surface active agents for various purposes, for example, as coating aids, or for prevention of electrically charging, improvement of lubricating property, emulsification, prevention of adhesion, improvement of photographic properties (for example, acceleration of development, rendering hard tone or sensitization), etc.

For example, it is possible to use nonionic surface active agents such as saponin (steroid saponin), alkylene oxide derivatives (for example, polyethylene glycol, polyethylene glycol/polypropylene glycol condensates, polyethylene glycol alkyl ethers or polyethylene glycol alkylaryl ethers, polyethylene glycol esters, polyethylene glycol sorbitan esters, polyalkylene glycol alkylamines or amides, polyethylene oxide adducts of silicone, etc.), glycidol derivatives (for example, alkenylsuccinic acid polyglycerides, alkylphenol polyglycerides, etc.), polyhydric alcohol aliphatic acid esters or saccharide alkyl esters, etc.; anionic surface active agents containing acid groups such as a carboxy group, a sulfo group, a phospho group, a sulfate group, a phosphate group, etc., such as alkylcarboxylic acid salts, alkylsulfonate salts, alkylbenzenesulfonate salts, alkylnaphthalenesulfonate salts, alkyl sulfuric acid esters, alkylphosphoric acid esters, N-acyl-N-alkyltaurines, sulfosuccinic acid esters, sulfoalkyl polyoxyethylene alkylphenyl ethers, polyoxyethylene alkylphosphoric acid esters, etc.; ampholytic surface active agents such as amino acids, aminoalkylsulfonic acids, aminoalkylsulfuric acid esters or phosphoric acid esters, alkylbetaines, amine oxides, etc.; and cationic surface active agents such as alkylamine salts, aliphatic or aromatic quaternary ammonium salts, heterocyclic quaternary ammonium salts such as pyridinium salts, imidazolium salts, etc., aliphatic or heterocyclic phosphonium salts, aliphatic or heterocyclic sulfonium salts, etc.

Of the above-described surface active agents, polyethylene glycol type nonionic surface active agents having a recurring unit of ethylene oxide in their molecules may be preferably incorporated into the light-sensitive material. It is particularly preferred that the molecule contains 5 or more of the recurring units of ethylene oxide.

The nonionic surface active agents capable of satisfying the above-described conditions are well known as to their structures, properties and methods of synthesis. These nonionic surface active agents are widely used even outside this field. Representative references relating to these agents include: Surfactant Science Series, Vol. 1, Nonionic Surfactants (edited by Martin J. Schick, Marcel Dekker Inc., 1967), and Surface Active Ethylene Oxide Adducts (edited by Schoufeldt N. Pergamon Press, 1969). Among the nonionic surface active agents described in the above-mentioned references, those capable of satisfying the above-described conditions are preferably employed in connection with the present invention.

The nonionic surface active agents can be used individually or as a mixture of two or more of them.

The polyethylene glycol type nonionic surface active agents can be used in an amount of less than 100% by weight, preferably less than 50% by weight, based on a hydrophilic binder.

The light-sensitive material of the present invention may contain a cationic compound containing a pyridinium salt. Examples of the cationic compounds containing a pyridinium group used are described in PSA Journal Section B 36 (1953), U.S. Patents 2,648,604 and 3,671,247, Japanese Patent Publication Nos. 30074/69 and 9503/69, etc.

In the photographic light-sensitive material and the dye fixing material of the present invention, the photographic emulsion layer and other binder layers may contain inorganic or organic hardeners. It is possible to use chromium salts (chromium alum, chromium acetate, etc.), aldehydes (formaldehyde, glyoxal, glutaraldehyde, etc.), N-methylol compounds (dimethylolurea, methylol dimethylhydantoin, etc.), dioxane derivatives (2,3-dihydroxydioxane, etc.), active vinyl compounds (1,3,5-triacryloylhexahydro-s-triazine, 1,3-vinylsulfonyl-2-propanol, etc.), active halogen compounds (2,4-dichloro-6-hydroxy-s-triazine, etc.), mucohalogenic acids (mucochloric acid, mucophenoxychloric acid, etc.), etc., which are used individually or as a combination thereof.

Examples of various additives include those described in Research Disclosure, Vol. 170, No. 17029 (June, 1978), for example, plasticizers, dyes for improving sharpness, antihalation dyes, sensitizing dyes, matting agents, fluorescent whitening agents and fading preventing agents, etc.

If necessary, two or more layers may be coated at the same time by the method as described in U.S. Patent 2,761,791 and British Patent 837,095.

Various means for exposure can be used in the present invention. Latent images are obtained by imagewise exposure by radiant rays including visible rays. Generally, light sources used in this invention include tungsten lamps, mercury lamps, halogen lamps such as iodine lamps, xenon lamps, laser light sources, CRT light sources, fluorescent tubes and light-emitting diodes, etc.

In the present invention, after the heat-developable color photographic material is exposed to light, the resulting latent image can be developed by heating the whole material to a suitably elevated temperature, for example, about 80°C to about 250°C for about 0.5 second to about 300 seconds. A higher temperature or lower temperature can be utilized to prolong or shorten the heating time, if it is within the above-described temperature range. Particularly, a temperature range of about 110°C to about 160°C is useful.

As the heating means, a simple heat plate, iron, heat roller, heat generator utilizing carbon or titanium white, etc., or analogues thereto may be used.

In the present invention, a specific method for forming a color image by heat development comprises transfer of a hydrophilic mobile dye. For this purpose, the heat-developable color photographic material of the present invention is composed of a support having thereon a light-sensitive layer (I) containing at least silver halide, if necessary, an organic silver salt oxidizing agent, a dye releasing redox compound which is also a reducing agent for the organic silver salt oxidizing agent and a binder, and a dye fixing layer (II) capable of receiving the hydrophilic diffusible dye formed in the light-sensitive layer (I).

The above-described light-sensitive layer (I) and the dye fixing layer (II) may be formed on the same support, or they may be formed on different supports, respectively. The dye fixing layer (II) can be stripped off the light-sensitive layer (I). For example, after the heat-developable color photographic material is exposed imagewise to light, it is developed by heating uniformly and thereafter the dye fixing layer (II) or the light-sensitive layer (I) is peeled apart. Also, when a light-sensitive material having the light-sensitive layer coated on a support and a fixing material having the dye fixing layer (II) coated on a support are separately formed, after the light-sensitive material is exposed imagewise to light and uniformly heated, the mobile dye can be transferred on the dye fixing layer (II) by superposing the fixing material on the light-sensitive layer.

Further, there is a method wherein only the light-sensitive layer (I) is exposed imagewise to light and heated uniformly by superposing the dye fixing layer (II) on the light-sensitive layer (I).

The dye fixing layer (II) can contain, for example, a dye mordant in order to fix the dye. In the present invention, various mordants can be used, and polymer mordants are particularly preferred. In addition to the mordants, the dye fixing layer may contain the bases, base precursors and thermal solvents. In particular, it is particularly preferred to incorporate the bases or base precursors into the dye fixing layer (II) in the cases wherein the light-sensitive layer (I) and the dye fixing layer are formed on different supports.

Preferred polymer mordants used in the present invention can be polymers containing secondary and tertiary amino groups, polymers containing nitrogen-containing heterocyclic moieties, polymers having quaternary cation groups thereof, having a molecular weight of from 5,000 to 200,000, and particularly from 10,000 to 50,000.

For example, vinylpyridine polymers and vinylpyridinium cation polymers as disclosed in U.S. Patents 2,548,564, 2,484,430, 3,148,061 and 3,756,814, etc., polymer mordants capable of cross-linking with gelatin as disclosed in U.S. Patents 3,625,694, 3,859,096 and 4,128,538, British Patent 1,277,453, etc., aqueous sol type mordants as disclosed in U.S. Patents 3,958,995, 2,721,852 and 2,798,063, Japanese Patent Application (OPI) Nos. 115228/79, 145529/79 and 126027/79, etc., water-insoluble mordants as disclosed in U.S. Patent 3,898,088, etc., reactive mordants capable of forming cobalent bonds with dyes used as disclosed in U.S. Patent 4,168,976 (Japanese Patent Application (OPI) No. 137333/79), etc., and mordants disclosed in U.S. Patents 3,709,690, 3,788,855, 3,642,482, 3,488,706, 3,557,066, 3,271,147 and 3,271,148, Japanese Patent Application (OPI) Nos. 71332/75, 30328/78, 155528/77, 125/78 and 1024/78, etc., can be illustrated.

In addition, mordants disclosed in U.S. Patents 2,675,316 and 2,882,156 can be used.

The dye fixing layer (II) can have a white reflective layer. For example, a layer of titanium dioxide dispersed in gelatin can be provided on the mordant layer on a transparent support. The layer of titanium dioxide forms a white opaque layer, by which reflection color images of the transferred color images which can be observed through the transparent support is obtained.

Typical dye fixing material used in the present invention is obtained by mixing the polymer containing ammonium salt groups with gelatin and applying the mixture to a transparent support.

The transfer of dyes from the light-sensitive layer to the dye fixing layer can be carried out using a dye transfer assistant. Examples of useful dye transfer assistant include water and an alkaline aqueous solution containing sodium hydroxide, potassium hydroxide and an inorganic alkali metal salt. Further, a solvent having a low boiling point such as methanol, N,N-dimethylformamide, acetone, diisobutyl ketone, etc., and a mixture of such a solvent having a low boiling point with water or an alkaline aqueous solution can be used. The dye transfer assistant can be employed by wetting the image receiving layer with the transfer assistant or by incorporating it in the form of water of crystallization or microcapsules into the material.

EXAMPLE 1

A method of preparing a silver iodobromide is described below.

40 g of gelatin and 26 g of potassium bromide (KBr) were dissolved in 3,000 ml of water and the solution was stirred while maintaining the temperature at 50°C. A solution containing 34 g of silver nitrate dissolved in 200 ml of water was then added to the above prepared solution over 10 minutes. Subsequently, a solution containing 3.3 g of potassium iodide (KI) dissolved in 100 ml of water was added thereto over 2 minutes.

By controlling the pH of the resulting silver iodobromide emulsion, precipitates were formed and excess salts were removed. The pH of the emulsion was adjusted to 6.0 and 400 g of the silver iodobromide emulsion was obtained.

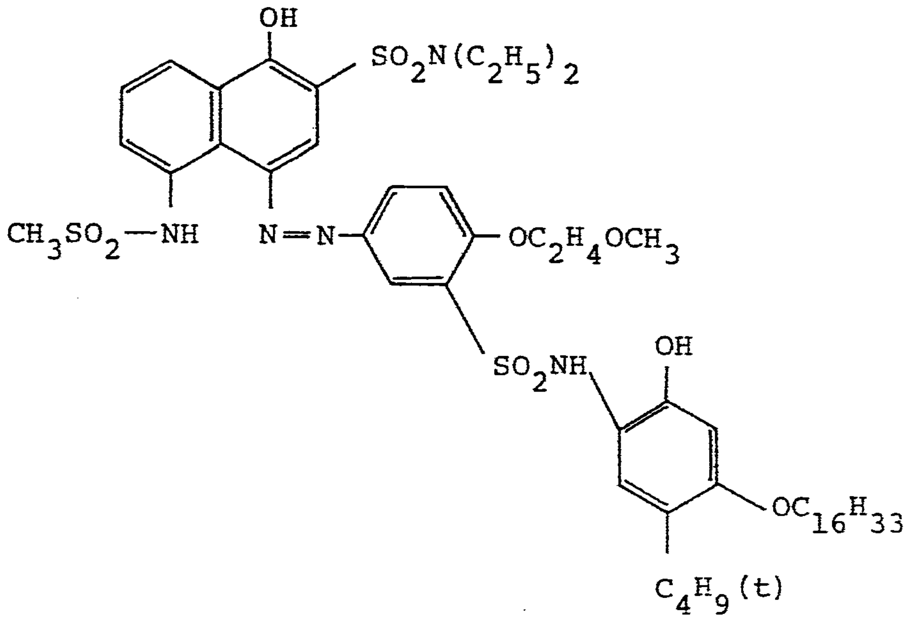
A method of preparing a silver benzotriazole emulsion is described below.

28 g of gelatin and 13.2 g of benzotriazole were dissolved in 3,000 ml of water and the solution was stirred while maintaining the temperature at 40°C. A solution containing 17 g of silver nitrate dissolved in 100 ml of water was then added to the above prepared solution over 2 minutes.

By controlling the pH of the resulting silver benzotriazole emulsion, precipitates were formed and excess salts were removed. The pH of the emulsion was adjusted to 6.0 and 400 g of the silver benzotriazole emulsion was obtained.

The method of preparing a gelatin dispersion of a dye releasing redox compound is described below.

5 g of magenta dye releasing redox compound of the following formula:

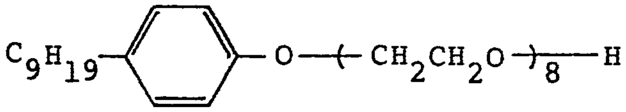


0.5 g of sodium 2-ethyl-hexylsuccinate sulfonate, and 5 g of tri-cresyl phosphate (TCP) were weighed and added to 30 ml of ethyl acetate and dissolved therein by heating at about 60°C to form a uniform solution. This solution was mixed with 100 g of a 10 wt% aqueous solution of lime-treated gelatin and then dispersed at 10,000 rpm for 10 minutes using a homogenizer.

This dispersion is designated a dispersion of the magenta dye releasing redox compound.

A method of preparing light-sensitive coatings A and B is described below.

(a)	Silver iodobromide emulsion prepared above	25 g
(b)	Dispersion of magenta dye-releasing redox compound prepared above	33 g
(c)	5 wt% Aqueous solution of compound having the formula:	10 ml



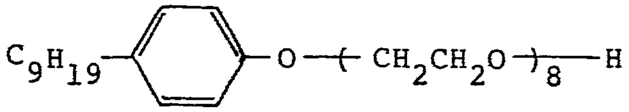
(d)	10 wt% Ethanol solution of guanidine trichloroacetic acid	15 ml
(e)	10 wt% Ethanol solution of dimethylsulfamide	4 ml

The above components (a) to (e) were mixed and dissolved and the resulting mixture solution was coated on a polyethylene terephthalate film in a wet thickness of 30 μm and dried.

(i)	10 wt% Aqueous solution of gelatin	35 g
(ii)	10 wt% Ethanol solution of guanidine trichloroacetic acid	6 ml
(iii)	1 wt% Aqueous solution of sodium 2-ethylhexylsuccinate sulfonate	4 ml
(iv)	Water	55 ml

The above components (i) to (iv) were mixed and coated on the above prepared coating layer as a protective layer in a wet thickness of 25 μm to produce a light-sensitive coating A.

(a)	Silver benzotriazole emulsion	10 g
(b)	Silver iodobromide emulsion	20 g
(c)	Dispersion of the magenta dye releasing redox compound prepared above	33 g
(d)	5 wt% Aqueous solution of compound having the following formula:	10 ml



(e)	10 wt% Ethanol solution of guanidine trichloroacetic acid	17 ml
(f)	10 wt% Aqueous solution of dimethylsulfoamide	4 ml

The above components (a) to (f) were mixed and dissolved and the resulting mixture solution was coated on a polyethylene terephthalate film in a wet thickness of 30 μm and dried. A protective layer having the same formulation for the coating A was provided to produce a light-sensitive coating B.

A method of preparing a dye fixing material is described below.

10 g of poly(methylacrylate-co-N,N-trimethyl-N-vinylbenzylammonium chloride (the ratio of methyl acrylate to vinylbenzylammonium chloride is 1:1) was dissolved in 200 ml of water and the solution was uniformly mixed with 100 g of 10 wt% aqueous solution of lime-treated gelatin. This mixture solution was uniformly coated on a paper support

with a titanium-dispersed polyethylene laminated thereon in a wet thickness of 90 μm. The sample was dried to be used as a dye fixing material having a mordant layer.

The light-sensitive coatings A and B were each imagewise exposed at 2,000 lux for 10 seconds using a tungsten lamp and then uniformly heated for 20 seconds on a heat block which had been heated at 130°C.

Water was applied onto the coating side of the dye fixing material in an amount of 80 ml per square meter of the coating. Each of the light-sensitive coatings A and B which had been heat treated as described above was superposed on the dye fixing material in such a manner that the coating surface of the coating and dye fixing material were in contact with each other.

They were heated for 6 seconds on a heat block which had been heated at 80°C and the dye fixing material was peeled away from the light-sensitive material, whereby a negative-working magenta color image was formed on the dye fixing material.

This magenta image had a remarkable color smear and could not be said to be a sharp image.

The same procedure as above was repeated except that water was applied onto the coating side of the dye fixing material in an amount of 10 ml per square meter of the coating layer. As a result, a sharp and smear-free magenta image could be formed.

The weights of the light-sensitive coatings A and B and the coating of the dye fixing material were about 4 g/m^2, about 4 g/m^2, and about 5 g/m^2, respectively. The maximum swollen thicknesses in the water used were 12 μm, 12 μm and 14 μm, respectively. Hence, the amount of water in the above described system was 0.9 to 17 ml/m^2.

EXAMPLE 2

The light-sensitive coating A prepared in Example 1 was exposed and then heated in the same manner as in Example 1.

Water was applied onto the coating side of the dye fixing material in various amounts: 0.5 ml, 1.2 ml, 2.1 ml, 2.7 ml, 4.4 ml, 7.8 ml, 9.3 ml, 15 ml and 20.5 ml, each being per square meter of the coating.

The light-sensitive material and the dye fixing material were superposed in such a manner that their coating surfaces were in contact with each other and then heated for 6 seconds on a heat block which had been heated at 80°C. The dye fixing material was peeled away from the light-sensitive material. The maximum density of the magenta color image formed on the dye fixing material was measured with a Macbeth reflection densitometer (RD-519). The results were as follows:

Amount of Water	Maximum Density
0.5	0.20
1.2	1.00
2.1	1.20
2.7	1.55
4.4	1.82
7.8	1.95
9.3	1.90
15	2.05
20.5	1.88

When water was applied in an amount of 20.5 ml/m^2, smears were recognized on the image.

The maximum swollen thickness of the light-sensitive coating A and the dye fixing material of Example 1 was 26 µm, and the total amount of all the coatings was 9 g/m^2. Therefore, the range as specified in the present invention of this material is 0.9 ml/m^2 to 17 ml/m^2. It can be seen from the above results that within this range the dye can be sufficiently transferred satisfactorily without causing any smears.

EXAMPLE 3

The procedure of Example 2 was repeated except for using water in an amount of 2.72 ml and varying the heating time on the heat block. As a result, the maximum density of

1.85 could be obtained by heating for 12 seconds.

Also, the procedure of Example 2 was repeated except for using water in an amount of 1.2 ml. As a result, the maximum density of 1.60 could be obtained by heating for 30 seconds.

It can be seen from the above results that when the amount of water applied is small, a sharp and smear-free image can be formed by increasing the heating time for transferring the dye.

EXAMPLE 4

In the light-sensitive coating A of Example 1, only the weight of the protective layer was changed to 14 g/m^2 (the maximum swollen thickness was 40 μm).

Using this light-sensitive coating, the procedure of Example 2 was repeated. When water was applied in an amount of 2.1 ml or more, a sufficiently high color density could be obtained. Even when the amount of water applied was 20.5 ml, a sharp and smear-free image could be obtained.

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

WHAT IS CLAIMED IS:

1. A process for forming an image which comprises

heating a light-sensitive material comprising a support having thereon at least a light-sensitive silver halide, a binder, and a dye releasing redox compound which is reductive to the light-sensitive silver halide and is capable of releasing a hydrophilic dye upon reaction with the light-sensitive silver halide by heating, after imagewise exposure or simultaneously with imagewise exposure, in a substantially water-free condition to imagewise form a mobile dye; and

transferring the mobile dye into a dye fixing material using water, wherein in transferring the mobile dye, the amount of water present in all the coatings of the light-sensitive material and the dye fixing material is within a range of from 0.1 time the total weight of all the coatings to the difference obtained by subtracting the total weight of all the coatings from the weight of water corresponding to the maximum swollen volume of all the coatings.

2. A process as claimed in Claim 1, wherein the amount of water is 0.8 time or less the difference.

3. A process as claimed in Claim 1, wherein the water is supplied to at least one of the dye fixing material and the light-sensitive material.

4. A process as claimed in Claim 1, wherein the heating is conducted at a temperatuer of 80°C or more.

5. A process as claimed in Claim 1, wherein the substantially water-free condition is a condition which is in an equilibrium state with respect to water in air and with no addition of water from outside.