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3,320,063

PHOTOGRAPHIC DEVELOPING AGENTS AND USES THEREOF

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11 Claims. (Cl. 96—29)

This invention relates to photography and more particularly to processes and compositions for the development of photosensitive silver halide elements. In one aspect, this invention relates to transfer processes which involve the development of a latent image in an exposed silver halide emulsion and the transfer of components from said emulsion to another layer to effect the formation of a print in said other layer, and to products useful in the performance of said transfer processes.

It has been proposed to carry out transfer processes by applying an aqueous solution of a silver halide developing agent to a silver halide stratum and an image-receptive stratum which are in superposed relation. The developing agent reduces exposed silver halide to silver, said developing agent becoming oxidized in the process. Thereafter, the remaining unoxidized developing agent is transferred in an imagewise distribution to the image-receptive stratum to form a photographic image.

It is one object of the present invention to provide novel compounds characterized by their ability to develop photographic latent images with a high degree of discrimination, and processes for preparing such compounds.

A further object is to provide novel processes for the development of silver halide emulsions, in which the developing agent is capable of developing a photographic latent image and imparting a reversed or positive image of said developed image to a superposed image-receiving element.

A further object is to provide novel processes suitable for use in preparing monochromatic and multichromatic photographic images.

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

The invention accordingly comprises the process involving the several steps and the relation and order of one or more of such steps with respect to each of the others, and the products and compositions 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.

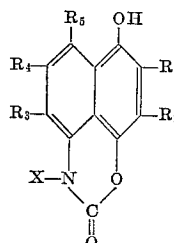
For a fuller understanding of the nature and objects of the invention, reference should be had to the following detailed description.

It has now been discovered that certain compounds, herein referred to as leuco compounds or as leuco developing agents, are effective as discriminating silver halide developing agents, and in addition to developing the exposed silver halide emulsion, are effective to impart a positive image of the developed silver image to an image-receiving layer, as will be further described herein. These leuco developing agents are referred to herein as keto-peri-naphthoxazines. Specifically, these compounds may be designated as 2-keto-7-hydroxy-1,3-peri-naphthox-

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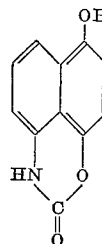
azines, and may be represented by the formula:

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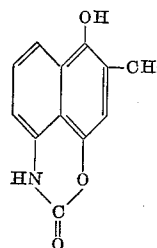


wherein R_1 and R_2 are hydrogen or lower alkyl, R_3 , R_4 , and R_5 are each members from the group consisting of hydrogen, hydroxyl, halogen, lower alkyl, substituted lower alkyl, aryl, and substituted aryl, and X is hydrogen or lower alkyl.

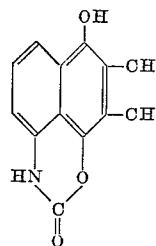
As specific examples of the leuco compounds within the scope of this invention, mention may be made of the following:



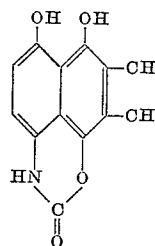
2-keto-7-hydroxy-1,3-peri-naphthoxazine



2-keto-7-hydroxy-8-methyl-1,3-peri-naphthoxazine



2-keto-7-hydroxy-8,9-dimethyl-1,3-peri-naphthoxazine



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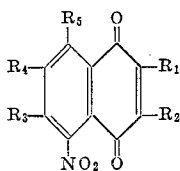
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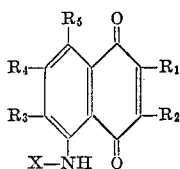
2-keto-7-dihydroxy-8,9-dimethyl-1,3-peri-naphthoxazine
 2-keto-7-hydroxy-8-ethyl-1,3-peri-naphthoxazine
 2-keto-7-hydroxy-8,9-diethyl-1,3-peri-naphthoxazine
 2-keto-7-hydroxy-6,8,9-trimethyl-1,3-peri-naphthoxazine
 2-keto-7-hydroxy-6-methyl-8,9-diethyl-1,3-peri-naphthoxazine
 2-keto-7-hydroxy-4,6,8,9-tetramethyl-1,3-peri-naphthoxazine
 2-keto-7-hydroxy-4,6-diethyl-8,9-dimethyl-1,3-peri-naphthoxazine
 2-keto-7-hydroxy-8,9-dichloroethyl-1,3-peri-naphthoxazine
 2-keto-7-hydroxy-4,6-dichloroethyl-8,9-dimethyl-1,3-peri-naphthoxazine
 2-keto-6,7-dihydroxy-8,9-dichloroethyl-1,8-peri-naphthoxazine

The leuco compounds contemplated herein may be prepared by the following general procedure:

Initially, nitro naphthoquinones having the general formula:

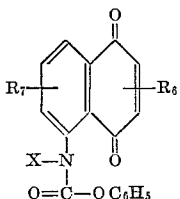


wherein R_1 and R_2 are hydrogen or lower alkyl, R_3 , R_4 , and R_5 are each members from the group consisting of hydrogen, halogen, hydroxyl, lower alkyl, substituted lower alkyl, aryl, and substituted aryl, are synthesized, either from the corresponding naphthoquinone by nitration as described in U.S. Patent 2,687,939, to Mario F. Sartori, issued August 31, 1954, or by other known methods, such as those described in U.S.S.R. Patent 108,812, abstracted in Chemical Abstracts, vol. 52, p. 11942i, and in U. U. Vorozhtsov et al., Zhur, Vsesoyuz Khim. Obshchestva im. D. I. Mendeleev, vol. 5, pp. 474-5. Thereafter, the compounds produced thereby are reduced to the corresponding amino compounds. Satisfactory techniques for this step are also described in U.S. Patent 2,687,939. Additionally, methods are described therein for the introduction of selected substituents into the naphthalene ring. The resultant compounds are amino naphthoquinones having the general formula:



wherein R_1 and R_2 are hydrogen or lower alkyl, R_3 , R_4 , and R_5 are each members from the group consisting of hydrogen, halogen, hydroxyl, lower alkyl, substituted lower alkyl, aryl, and substituted aryl, and X is hydrogen or lower alkyl; they provide the starting materials for the novel synthesis described below. It will be understood that any commercially available substituted or unsubstituted naphthoquinone can be treated as described above and reduced to ultimately produce a keto-*peri*-naphthoxazine which is similarly substituted.

The amino naphthoquinones are treated with phenyl chlorocarbonate in a non-aqueous solvent to form phenyl-5-(1,4-naphthoquinonyl)-carbamates having the general formula:



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wherein R_6 represents R_1 and R_2 in Formula 1, R_7 represents R_3 , R_4 , and R_5 in Formula 1, and X is hydrogen or lower alkyl. Thereafter, the resulting phenyl-5-(1,4-naphthoquinonyl)-carbamates may be treated with a reducing agent in an aqueous alkaline solution to form the desired 2-keto-7-hydroxy-1,3-*peri*-naphthoxazines. Suitable reducing agents are those which will reduce the quinone moiety to hydroquinone, such as alkali metal dithionites, alkali metal borohydrides, and zinc in alkaline solution. The solution should have a pH of at least about 9 and for this purpose, a weak base, such as sodium carbonate or ammonia should be employed. Moreover, it may be advantageous to deaerate the solution throughout the course of the reaction in the interest of inhibiting oxidation of the products.

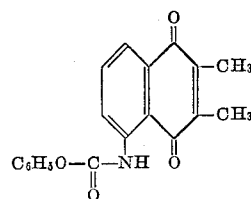
In both of the above steps, i.e., the carbamate formation and subsequent reduction, the relative proportions of reactants are not critical; however, it is advantageous to have present an excess of reactants, i.e., phenyl chlorocarbonate and reducing agent, in order to insure good yields of the desired materials.

The following are examples of the procedure for making compounds as described herein, and are presented by way of illustration only:

EXAMPLE 1

2-keto-7-hydroxy-8,9-dimethyl-1,3-*peri*-naphthoxazine

(A) Preparation of phenyl-5-(2,3-dimethyl-1,4-naphthoquinonyl)-carbamate. 4.5 grams (0.22 mole) of 5-amino-2,3-dimethyl-1,4-naphthoquinone were dissolved in 500 ml. of dry toluene containing 2.4 grams (0.03 mole) of dry pyridine. To the above solution, 7.0 grams (0.044 mole) of phenyl chlorocarbonate were gradually added over a period of 1/2 hour. Thereafter, the solution was refluxed for two hours. Following concentration of the solution and the addition of hexane, 5 grams of a yellow precipitate were recovered, said yellow precipitate melting at 190-195° C. The material was recrystallized from toluene, and the product obtained therefrom melted at 199-200° C. and was found upon analysis to be phenyl-5-(2,3-dimethyl-1,4-naphthoquinonyl)-carbamate, having the formula



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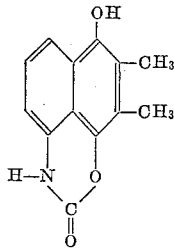
Calc. for $C_{19}H_{15}O_4N$: C, 71.03; H, 4.71; N, 4.36. Found: C, 70.72; H, 4.60; N, 4.30.

(B) Preparation of 2-keto-7-hydroxy-8,9-dimethyl-1,3-*peri*-naphthoxazine. 12 grams of phenyl-5-(2,3-dimethyl-1,4-naphthoquinonyl)-carbamate prepared by the method of Step A above were powdered and added to a deaerated mixture of 350 ml. of ethanol and 250 ml. of water. 24 grams of sodium carbonate were added, care being taken to keep the reaction medium flushed with nitrogen. After dissolution of the sodium carbonate, 12 grams of sodium dithionite was added and the reaction refluxed under nitrogen for about twenty minutes. The reaction was then made acidic with excess acetic acid, concentrated to one-half its volume, and poured into about 2 liters of an ice-water mixture. The solid was filtered off, washed with water, and dried. After crystallization from ethanol using the Soxhlet extraction, the product melted at 260-265° C. The compound was recrystallized from ethyl acetate-ethanol, M.P. 265° C. dec.

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Upon analysis, the product was found to be 2-keto-7-hydroxy-8,9-dimethyl-1,3-peri-naphthoxazine, having the following configuration:



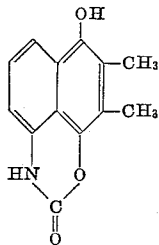
Calc. for $C_{15}H_{11}O_3N$: N, 6.1. Found: N, 6.0; 6.1.

The following is an example of the use of the novel compounds described herein as silver halide developing agents in accordance with this invention, and is given as illustration only:

EXAMPLE 2

An image-receiving element was prepared by coating a subcoated baryta paper with a partial butyl ester of poly(ethylene/maleic anhydride), followed by a layer of polyvinyl alcohol and a layer of a mixture of polyvinyl alcohol and poly-4-vinylpyridine. A detailed description of the process for preparing such image-receiving elements may be found in the copending U.S. patent application of Edwin H. Land, Serial No. 234,864, filed November 1, 1962.

A photosensitive element for use with the above image-receiving element was prepared by coating a silver-iodobromide emulsion (of the type used in Type 42 Polaroid Land Picture Rolls) with an acetone solution comprising 2% cellulose acetate hydrogen phthalate and 2.1% of a keto hydroxy-peri-naphthoxazine having the formula:



as prepared in Example 1 above, and drying the coating.

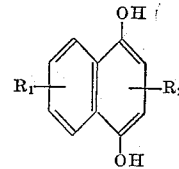
The photosensitive element was exposed to a test object which was designed to completely expose a portion of the element and to leave the remaining portion totally unexposed. An aqueous liquid processing composition comprising:

Water	-----cc	100
Potassium hydroxide	-----grams	11.2
Hydroxyethyl cellulose (high viscosity) [commercially available from Hercules Powder Co., Wilmington 99, Delaware, under the trade name Natrasol 250]	-----grams	3.7
Potassium thiosulfate	-----do	0.5
Zinc nitrate	-----do	0.5
Benzotriazole	-----do	3.5
Lithium nitrate	-----do	0.5
N-benzyl- α -picolinium bromide	-----do	2.0

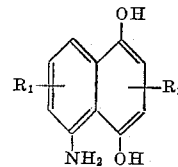
was then spread in a layer approximately 0.0026" thick between the photo-sensitive element and the image-receiving element as said elements were brought into superposed relationship. After an imbibition period of approximately one minute, the image-receiving element was separated. Thereafter, the maximum densities in the exposed and unexposed portions of the developed negative were determined by means of an IR Densichron. The density measurement for the exposed portion was 0.58; for the unexposed portion, 0.18, thereby indicating the utility of the keto-hydroxy-peri-naphthoxazine as a discriminating developing agent.

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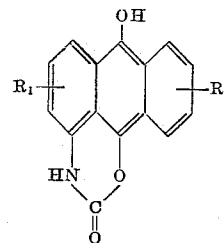
The success with which the keto-hydroxy-peri-naphthoxazines described herein can be used as developing agents is especially surprising and unexpected in view of the fact that similar analogous compounds, such as naphthoquinones,



wherein R_1 and R_2 represent a variety of substituents such as those described above, 5-amino-1,4-naphthoquinones,



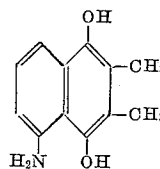
and keto hydroxy-peri-anthroxazines,



are markedly less effective, as illustrated by the following examples:

EXAMPLE 3

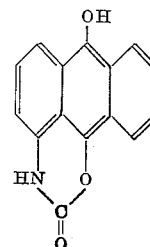
The process described in Example 2 was followed, except that an aminonaphthoquinone having the formula:



was substituted for the keto-hydroxy-peri-naphthoxazine. The developed negative showed a D_{max} of 0.25 and a D_{min} of 0.16.

EXAMPLE 4

The process described in Example 2 was followed, except that a keto-hydroxy-peri-anthroxazine having the formula:



was substituted for the keto-hydroxy-peri-naphthoxazine. The final developed negative was similar to that obtained in Example 3, in that there was relatively little difference in density between the exposed and unexposed portions.

The leuco developing agents described herein are substantially colorless or only faintly colored, but upon oxidation, assume various colors depending upon the nature of the groups substituted thereon. Specifically, referring

Formula 1, the substituents R₃, R₄, and R₅ can be selected to determine the final color of the oxidized developing agent, R₅ having the most pronounced effect. Accordingly, it is possible to predetermine a given desired color in the final image by appropriate selection of substituents.

The color-forming ability of the developing agents described herein renders them suitable for the formation of colored images in photographic products and processes of the type described, for example, in U.S. Letters Patent Nos. 2,647,049, 2,698,244, 2,909,430, and 2,968,554. In these patents, examples of diffusion transfer reversal processes are disclosed wherein a photographic material, such as a photosensitive element comprising an exposed silver halide emulsion layer containing a latent image, is processed to impart to an image-carrying or image-receiving element a reversed or positive dye image of said latent image by permeating into said emulsion layer a suitable liquid processing composition and bringing said photosensitive element into superposed relationship with an appropriate image-receiving element.

In using the keto-peri-naphthoxazines in the above procedures, a photosensitive element comprising a silver halide emulsion is exposed and a liquid processing composition applied thereto, as by immersing, coating, spraying, blowing, etc., in the dark. In a preferred embodiment, the photosensitive element contains a layer of one of the above leuco compounds, and the liquid processing composition is applied to the exposed photosensitive element in a uniform layer as this element is brought into superposed relationship with an image-receiving element. The liquid processing composition permeates the emulsion and provides a solution of leuco compounds substantially uniformly distributed therein. As the latent image is developed, the leuco compound is oxidized and immobilized in exposed areas. This immobilization is apparently primarily due to a change in the solubility characteristics of the leuco compound upon oxidation, and especially as regards its solubility in alkaline solution. The unoxidized leuco compound retains its solubility in the liquid processing composition, thereby providing an imagewise distribution of unoxidized leuco compound, at least a part of which is transferred, by imbibition, to a superposed image-receiving element. The latter element receives a depthwise diffusion, from the emulsion, of unoxidized leuco compound without appreciably disturbing its imagewise distribution, thereby giving a reversed, or positive, colored image of the developed negative image. The image-receiving element may contain agents adapted to oxidize, mordant, or otherwise fix the diffused, unoxidized leuco compound. Imbibition periods of approximately one minute have been found to give good results, but this contact period may be adjusted where necessary to compensate for variation in temperature or other conditions. The desired positive image is revealed by stripping the image-receiving element from the photosensitive element at the end of the imbibition period.

The leuco compounds disclosed herein may be disposed in the liquid processing composition in the image-receiving element or in the photosensitive element, for example, in, on, or behind the silver halide emulsion layer. When employed in either element the leuco compound becomes solubilized by the liquid processing composition as it permeates the element. In a preferred embodiment, a coating or layer of the leuco compound is incorporated in the photosensitive element, preferably behind the silver halide layer, i.e., on the side of the emulsion adapted to be located most distant from the photographed subject when the photosensitive element is exposed. Placing the leuco compound behind the emulsion layer has the advantage of providing increased contrast in the positive image, and also minimizes any light-filtering action by the leuco compound. If the leuco compound is colorless or otherwise non-absorptive of light complementary in color to the sensitivity of the emulsion, it may be incorporated

in the silver halide emulsion without adverse light-filtering effects.

Where the leuco compound is employed in a layer within the photosensitive element, it may be applied by using coating solutions containing about 0.5 to 8%, by weight, of the leuco compound. Similar concentrations may be used if the leuco compound is utilized as a component of the liquid processing composition. A particularly useful method of incorporating the leuco compound is to dissolve it in a water-immiscible solvent, disperse the resulting solution in gelatin, and apply the resulting dispersion to provide the layer of leuco compound.

The liquid processing composition referred to above comprises at least an aqueous solution of an alkaline material, such as diethylamine, sodium hydroxide, potassium hydroxide, or sodium carbonate. If the liquid processing composition is to be applied to the emulsion by being spread thereon, preferably in a relatively thin, uniform layer, it may also include a viscosity-increasing compound constituting film-forming material of the type which, when spread over a water-absorbent base, will form a relatively firm and relatively stable film. A preferred film-forming material is a high molecular weight polymer such as a polymeric water-soluble ether which is inert to an alkaline solution, as, for example, a hydroxyethyl cellulose or sodium carboxymethyl cellulose. Other film-forming material or thickening agents may be employed whose ability to increase viscosity is substantially unaffected when left in solution for a long period of time.

The unoxidized leuco compound which diffuses to the image-receiving element is oxidized to provide the desired positive dye image. This oxidation may be effected by merely exposing the stripped image-receiving element to the air. In a preferred embodiment, the image-receiving element has incorporated therein an oxidizing agent for the purpose of speeding up dye formation and providing full image density immediately upon stripping. The incorporation of an oxidizing agent in the image-receiving element for the purpose of oxidizing unreacted developing agent permeated therein is described in U.S. Patent No. 2,559,643. As examples of suitable oxidizing agents, mention may be made of peroxy compounds such as sodium or potassium perborate, certain dichromates such as ammonium dichromate, and compounds having polyvalent metallic elements in higher valent form such as copper, iron, and cerium, wherein the metallic element is in a higher valent form. Examples of these latter-mentioned agents are cupric salts, such as cupric sulfate. Another suitable oxidizing agent is benzoyl peroxide. Other useful oxidizing agents are quinones, and particularly 2,3-dicyano-1,4-benzoquinone.

The following non-limiting example illustrates the use of the keto-peri-naphthoxazines described herein as leuco dyes in the formation of colored images.

EXAMPLE 5

An image-receiving element was prepared by coating a 2:1 mixture, by weight, of polyvinyl alcohol and poly-4-vinylpyridine on a cellulose acetate-subcoated baryta paper.

A photosensitive element for use with the above image-receiving element similar to that in Example 2, and containing the same keto-peri-naphthoxazine, was prepared.

The photosensitive element was exposed to a test object which was designed to completely expose a portion of the element and to leave the remaining portion totally unexposed. An aqueous liquid processing composition comprising:

	Grams
Sodium hydroxide -----	5.17
Hydroxyethyl cellulose (high viscosity) [commercially available from Hercules Powder Co., Wilmington 99, Delaware, under the trade name Natrasol 250] -----	4.03

	Grams
Sodium thiosulfate	1.15
Benzotriazole	2.3
N-benzyl- α -picolinium bromide	2.3

was then spread in a layer approximately 0.0026" thick between the photosensitive element and the image-receiving element as said elements were brought into superposed relationship. After an imbibition period of approximately one minute, the image-receiving element was separated and examined. An image was observed which was salmon pink in color, the density of which increased after exposure to air.

It will be apparent that by appropriate selection of the image-receiving element from among suitable known opaque and transparent materials, it is possible to obtain either a colored positive reflection print or a colored positive transparency. Likewise, the inventive concepts herein set forth are adaptable for multicolor work by the use of special photographic materials, for example, screen type negatives or film materials of the type containing two or more photosensitized elements associated with a suitable number of image-receiving elements and adapted to be treated with one or more liquid processing compositions, appropriate developing agents suitable to impart the desired subtractive colors being incorporated in the photosensitive elements or in the liquid processing composition. Examples of such photographic materials are disclosed in the previously mentioned patents.

In the preceding portions of the specification, the expression "color" has been frequently used. This expression is intended to include the use of a plurality of colors to obtain black.

It should be noted that the leuco developing agents of this invention provide the desired colored image upon oxidation and do not depend upon or require coupling reactions to produce the desired color. They thus provide a complete departure from photographic color processes in which the color is produced by a coupling reaction involving the oxidized developing agent.

It will be noted that a conventional silver halide developing agent may be incorporated with the leuco compounds described herein. This conventional developing agent is preferably used in a quantity sufficient to accelerate development of the latent image. Use of such a developing agent has been found helpful in improving the tonal range or steps in the positive image. Some of the unreacted leuco compound may be oxidized by an energy transfer reaction with oxidized auxiliary developing agent. Among the developing agents suitable for this purpose are p-N-benzylamino-phenol, ascorbic acid, p-anilino-phenol, 1-amino-2-naphthol, diethylhydroxylamine, and Metol (p-N-methyl-amino-phenol).

In employing the leuco compounds of this invention in film units suitable for use in self-developing cameras, such as the Polaroid Land camera, sold by Polaroid Corporation, Cambridge, Massachusetts, the film unit may comprise a photosensitive element, an image-receiving element adapted to be superposed on said photosensitive element, and a rupturable container holding a processing composition and adapted to release its contents for spreading over predetermined areas of said elements after rupture. Suitable structures of this type are described in the herein-mentioned U.S. patents. The leuco compound may be located in the processing composition or in a layer of the photosensitive element.

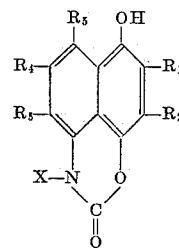
The developing agents of this invention may be used also in conventional photographic processes, such as tray or tank development of conventional photosensitive films, plates, or papers to obtain monochromatic or toned prints or negatives. By way of example, a developer composition suitable for such use may comprise an aqueous solution of approximately 1-2% of the leuco compound, 1% sodium hydroxide, 2% sodium sulfite, and 0.5% potassium bromide. After development is completed, any unreacted leuco compound is washed out of the photosensitive element, preferably with an alkaline washing me-

diu or other medium in which the oxidized leuco compound is soluble. The expression "toned" is used to designate photographic images wherein the silver is retained with the resultant dye, whereas "monochromatic" is intended to designate dye images free of silver.

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

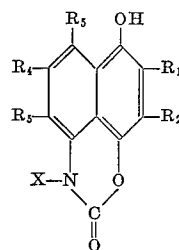
What is claimed is:

1. A process of developing an exposed photosensitive silver halide emulsion which comprises treating said emulsion with an aqueous alkaline solution of a leuco compound having the formula:



wherein R₁ and R₂ are each members selected from the group consisting of hydrogen and lower alkyl, R₃, R₄, and R₅ are each members selected from the group consisting of hydrogen, hydroxyl, halogen, lower alkyl, substituted lower alkyl, aryl, and substituted aryl, and X is a member selected from the group consisting of hydrogen and lower alkyl.

2. In a process of forming photographic images in color, the steps which comprise developing an exposed silver halide emulsion with an aqueous alkaline solution of a leuco compound having the formula:



wherein R₁ and R₂ are each members selected from the group consisting of hydrogen and lower alkyl, R₃, R₄, and R₅ are each members selected from the group consisting of hydrogen, hydroxyl, halogen, lower alkyl, substituted lower alkyl, aryl, and substituted aryl, and X is a member selected from the group consisting of hydrogen and lower alkyl, to thereby provide in said emulsion a predetermined distribution of unoxidized leuco compound, transferring at least part of said distribution of unoxidized leuco compound by imbibition from said emulsion to an image-receiving layer in superposed relationship with said emulsion, and oxidizing the diffused, transferred, unoxidized leuco compound to impart to said image-receiving layer a reversed, positive dye image of the developed image.

3. The process as defined in claim 2 wherein said leuco compound is disposed, prior to exposure, in a photosensitive element comprising said exposed silver halide emulsion, and the solution containing said leuco compound is formed by permeating the photosensitive element with an aqueous alkaline liquid capable of solubilizing said leuco compound.

4. The process as defined in claim 2 wherein said liquid is introduced by being spread in a substantially uniform layer between said photosensitive element and said image-receiving layer as said photosensitive element and said image-receiving layer are brought into superposed relationship.

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5. The process as defined in claim 4 wherein the difused, transferred, unoxidized leuco compound is oxidized by exposure to air.

6. The process as defined in claim 4 wherein said difused, transferred, unoxidized leuco compound is oxidized by an oxidizing agent incorporated in the image-receiving layer.

7. The process as defined in claim 5 wherein said liquid contains a thickener for increasing viscosity and for facilitating the spreading thereof between said photosensitive element and said image-receiving layer.

8. The process as defined in claim 4 wherein said aqueous alkaline solution contains a silver halide developing agent in addition to said leuco compound.

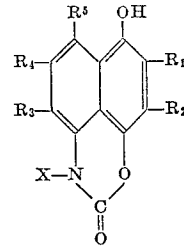
9. The process as defined in claim 4 wherein said leuco compound is 2-keto-7-hydroxy-1,3-peri-naphthoxazine.

10. The process as defined in claim 4 wherein said leuco compound is 2-keto-7-hydroxy-8,9-dimethyl-1,3-peri-naphthoxazine.

11. The photographic product comprising a support, a silver halide emulsion in a layer carried on said support, and a keto-hydroxy-peri-naphthoxazine in a layer on the

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same side of said support as said silver halide emulsion, said keto-hydroxy-peri-naphthoxazine having the formula:



wherein R_1 and R_2 are each members from the group consisting of hydrogen and lower alkyl, R_3 , R_4 , and R_5 are each members from the group consisting of hydrogen, hydroxyl, halogen, lower alkyl, substituted lower alkyl, aryl, and substituted aryl, and X is a member from the group consisting of hydrogen and lower alkyl.

No references cited.

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