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DIKETOPYRIMIDINE COUPLER

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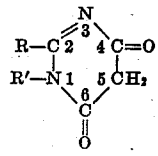
8 Claims. (Cl. 95—88)

This invention relates to color-forming development and particularly to novel coupling components for producing colored images.

It is known that colored photographic images may be formed by using a developer which forms a colored compound on development. The colored compound thus formed is deposited adjacent to the silver grains of the silver image during the development. It is also known that a colored image may be formed by adding to certain developer solutions or by incorporating in the gelatino-silver-halide emulsion before or after exposure a compound which couples during development with the oxidation product of the developing agent and forms a coloring substance which is likewise deposited adjacent to the silver grains of the silver image during development. Such a compound, which is employed in conjunction with a developing agent for the silver and which couples with the oxidation product thereof during development, is referred to herein as a coupling component or coupler.

The present invention concerns new or improved color forming developers comprising a primary aromatic amino compound serving as the developing agent, and a coupler as hereinafter defined and also includes a new or improved color development process which consists in developing a reducible silver salt image in a photographic element with the aid of a primary aromatic amino compound in the presence of a coupler as hereinafter defined. It also includes photographic sensitive elements having such a coupler incorporated in one or more emulsion layers.

I have found that certain diketopyrimidine compounds having the following probable structure are suitable for use as color formers:



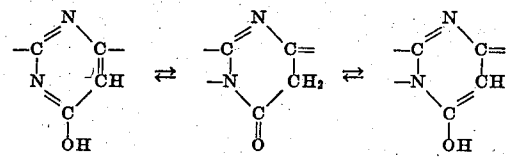
in which R is an alkyl, aryl, alkaryl, or heterocyclic group, R′ is hydrogen, an alkyl, aryl or alkaryl group or R and R′ together constitute the atoms necessary to complete a pyridine ring, that is, a six-membered ring containing one of the nitrogen atoms of the pyrimidine ring.

The substituents R and R′ may be varied either by their molecular weight or by the introduction of substituents to produce different dyes ranging from blue to magenta. The substituent groups R and R′ may be relatively low in molecular weight thus making the coupler suitable for use in the developing solution or the substituent groups may be of such size as to permit the use of these compounds in the emulsion layers of multi-layer coatings.

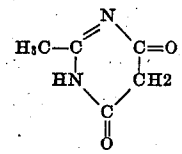
In the formula above, the substituent R may be an alkyl group such as methyl, ethyl, propyl, butyl, or amyl, an aryl group such as phenyl or an alkaryl group such as tolyl. The substituent R′ may be hydrogen or any of the alkyl, aryl or alkaryl groups previously mentioned.

These compounds are believed to be reactive in the color-forming reaction because of the methylene group in the 5-position having carbonyl groups on each side of it in the 4- and 6-positions. The CH_2 group present between two negative centers is a well known type of active group in the coupling process.

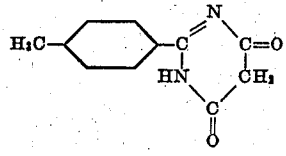
The pyrimidine compounds which I propose to use may have two tautomeric structures and I intend to include within the scope of my invention either structure since in many cases it is impossible to determine which structure exists in the compound as used in color photography. The pyrimidine compounds may exist in the following tautomeric forms:



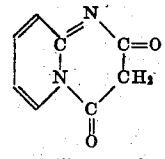
Specific compounds which may be used according to my invention are the following:



2-methyl-4,6-diketotetrahydropyrimidine



2-p-tolyl-4,6-diketotetrahydropyrimidine



1,2-divinylene-4,6-diketotetrahydropyrimidine

The coupling compounds used according to my invention may be prepared as follows:

Alkyl and aryl 4,6-diketotetrahydropyrimidines may be prepared by condensing substituted malonic esters with amidines, by a method similar to that described by Dox and Yoder in J. A. C. S., vol. 44 (1922), page 361.

1,2-divinylene -4,6- diketotetrahydropyrimidine

is prepared as described in Ber., vol. 57, (1924), page 1170.

The couplers used according to my invention may contain other substituents which do not affect the coupling properties of the compound but which alter properties such as solubility or color of the resulting dye. Where I refer in the general formula to aryl and alkyl groups, it is to be understood that these groups may be further substituted provided that the other requirements with respect to coupling are met.

The aromatic amino developing agents used with the coupler compounds of my invention include the mono-, di- and tri-aminoaryl compounds and their derivatives formed by substitution in the amino group as well as in the ring such as alkylphenylenediamines and alkyltoluylenediamines. These compounds are usually used in the salt form, such as the hydrochloride or the sulphate, which are more stable than the amines themselves. Suitable compounds are diethyl - p - phenylenediamine hydrochloride, monoethyl - p - phenylenediaminehydrochloride, dimethyl - p - phenylenediamine - hydrochloride and dimethyl-p-phenylenediamine sulphate. Developing agents containing heterocyclic systems as described in U. S. Patent 2,196,739 are also suitable. The p-aminophenols and their substitution products may also be used where the amino group is unsubstituted. All of these compounds have an unsubstituted amino group which enables the oxidation products of the developer to couple with the color-forming compounds to form a dye image.

The following example illustrates a developing formula which may be used with the compounds of my invention:

A. p-aminodiethylaniline sulfate	grams	2
Sodium sulfite (anhydrous)	do	5
Sodium carbonate (anhydrous)	do	20
Potassium bromide	do	2
Water to	liter	1
B. 2-p-tolyl-4,6-diketotetrahydroprimidine	grams	1–5
Sodium hydroxide (2% solution)	cc	50

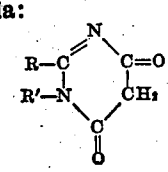
B is added to A

My development process may be employed for the production of colored photographic images in layers of gelatin or other carrier such as collodion, organic esters of cellulose or synthetic resins. The carrier may be supported by a transparent medium such as glass, a cellulose ester, or a non-transparent reflecting medium such as paper of an opaque cellulose ester. The emulsion may be coated as a single layer on the support or as superposed layers on one or both sides of the support. The superposed layers may be differentially sensitized to form natural color photographic images in the well known manner.

The examples and compounds set forth in the present specification are illustrative only and it is to be understood that my invention is limited only by the scope of the appended claims.

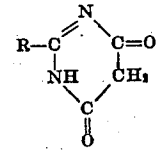
I claim:

1. A color-forming photographic developer, comprising a primary aromatic amino developing agent and a coupler compound having the probable formula:



where R and R' represent substituents in which R is selected from the class consisting of alkyl, aryl, and alkaryl groups, R' is selected from the class consisting of hydrogen, alkyl, aryl and alkaryl groups, or R and R' together represent the atoms necessary to complete a pyridine ring.

2. A color-forming photographic developer, comprising a primary aromatic amino developing agent and a coupler compound having the following probable formula:

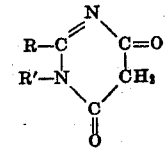


to which R is selected from the class consisting of alkyl, aryl, and alkaryl groups.

3. A color-forming photographic developer, comprising a primary aromatic amino developing agent and 2-p-tolyl-4,6-diketotetrahydropyrimidine.

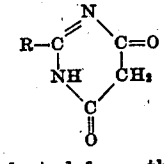
4. A color-forming photographic developer, comprising a primary aromatic amino developing agent and 1,2-divinylene-4,6-diketotetrahydropyrimidine.

5. The method of producing a colored photographic image in a gelatino-silver halide emulsion layer, which comprises exposing said layer and developing it with a primary aromatic amino developing agent in the presence of a coupler compound having the probable formula:



where R and R' represent substituents in which R is selected from the class consisting of alkyl, aryl, and alkaryl groups, R' is selected from the class consisting of hydrogen, alkyl, aryl and alkaryl groups, or R and R' together represent the atoms necessary to complete a pyridine ring.

6. The method of producing a colored photographic image in a gelatino-silver halide emulsion layer, which comprises exposing said layer and developing it with a primary aromatic amino developing agent in the presence of a coupler compound having the probable formula:



in which R is selected from the class consisting of alkyl, aryl, and alkaryl groups.

7. The method of producing a colored photographic image in a gelatino-silver halide emulsion layer, which comprises exposing said layer and developing it with a primary aromatic amine developing agent in the presence of 2-p-tolyl-4,6-diketotetrahydropyrimidine.

8. The method of producing a colored photographic image in a gelatino-silver halide emulsion layer, which comprises exposing said layer and developing it with a primary aromatic amino developing agent in the presence of 1,2-divinylene-4,6-diketotetrahydropyrimidine.

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