

June 3, 1969

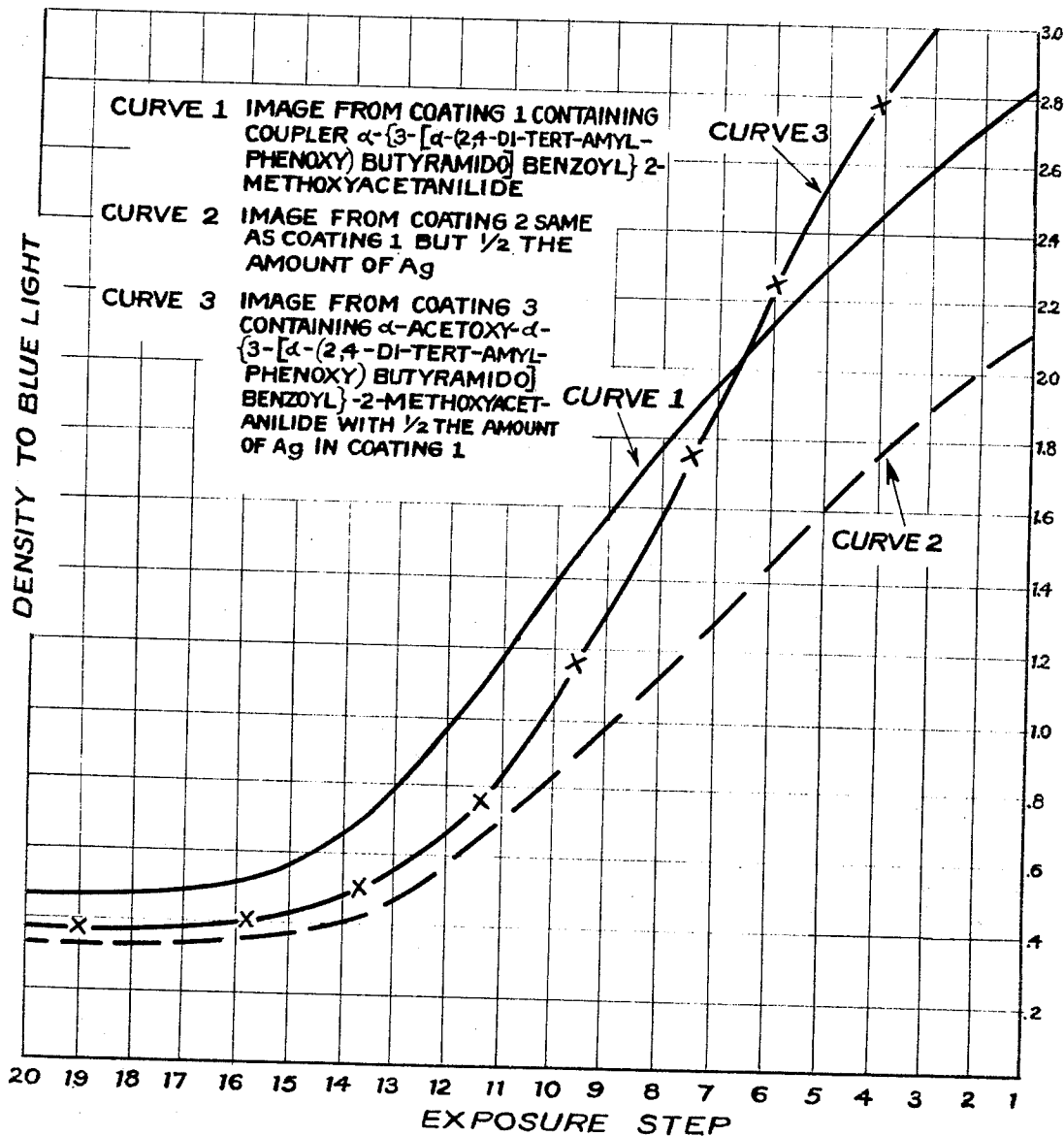
A. LORIA
SILVER HALIDE EMULSION CONTAINING TWO-EQUIVALENT YELLOW
DYE-FORMING COUPLER

3,447,928

Filed July 26, 1965

Sheet / of 2

Fig 1



Anthony Loria

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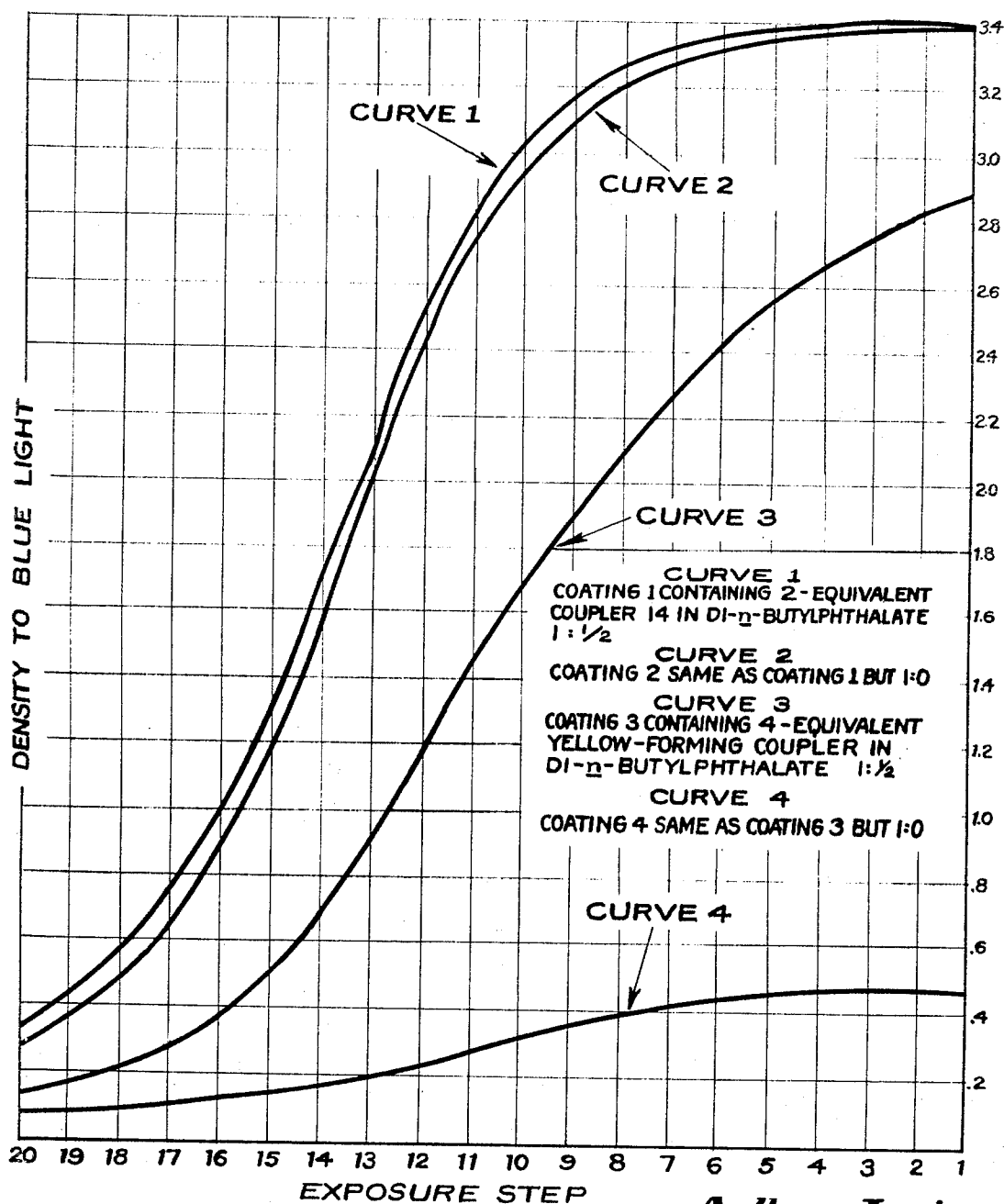
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Sheet 2 of 2

Fig. 2



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3,447,928

SILVER HALIDE EMULSION CONTAINING TWO-EQUIVALENT YELLOW DYE-FORMING COUPLER

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Continuation-in-part of application Ser. No. 246,931, Dec. 26, 1962. This application July 26, 1965, Ser. No. 477,353

Int. Cl. G03c 7/32

U.S. Cl. 96-100

15 Claims

ABSTRACT OF THE DISCLOSURE

Uncolored open-chain reactive methylene two-equivalent yellow dye-forming couplers having one of the hydrogens of the active methylene group replaced with an acyloxy substituent have good coupling reactivity with oxidized primary aromatic amine color developing agents to form good yellow dye images and are advantageously incorporated in light-sensitive photographic silver halide emulsion layers to form yellow dye images.

This is a continuation-in-part of Loria U.S. Ser. No. 246,931, filed Dec. 26, 1962, now abandoned.

This invention relates to photography and particularly to compounds which form dyes upon coupling with oxidized color developing agent and to photographic elements containing these compounds.

The formation of colored photographic images by the coupling of oxidized aromatic primary amino developing agents with color forming or coupling compounds is well known. In these processes the subtractive process of color formation is ordinarily used and the image dyes are intended to be cyan, magenta, and yellow, the colors that are complementary to the primary colors. Usually phenol or naphthol couplers are used to form the cyan dye image, pyrazolone couplers are used to form the magenta dye image and couplers containing a methylene group having one or two carbonyl groups attached to it are used to form the yellow dye image.

In these color developing processes the color forming coupler may be either in the developer solution or incorporated in the light-sensitive photographic emulsion layer so that during development it is available in the emulsion layer to react with the color developing agent that is oxidized by silver image development. Diffusible type couplers are used in color developer solutions. Fischer type couplers and nondiffusing couplers are incorporated in photographic emulsion layers. When the dye image formed is to be used in situ, couplers are selected which form nondiffusing dyes. The dye image used for image transfer processes should be diffusible but capable of being mordanted or fixed in the receiving sheet. For this purpose a coupler is selected which will produce this type of dye.

Conventional color-forming couplers are four-equivalent, that is, they require the development of four mols of exposed silver halide in order to supply one mol of oxidized color developing agent that is free to couple and form one mol of dye. Two-equivalent couplers require the development of only two mols of exposed silver halide to bring about the formation of one mol of dye. Two-equivalent couplers are very desirable for color photography, since only one-half the usual amount of silver halide is needed and the light-sensitive coatings can thus be made thinner. Certain of the available two-equivalent couplers tend to produce more stain than is desired, and

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others have not had the desired coupling reactivity. New classes of two-equivalent couplers are needed.

It is, therefore, an object of my invention to provide a new class of colorless two-equivalent dye-forming couplers which have a high degree of reactivity.

Another object is to provide a new class of dye-forming two-equivalent couplers which not only have a high coupling reactivity but which do not produce stains caused by nonimage forming coupling that is a problem with some of the prior art two-equivalent couplers.

Another object of my invention is to provide valuable acyloxy substituted two-equivalent couplers for forming dye images that have good spectral absorption characteristics, and good stability to prolonged exposure to light, heat, and high humidity.

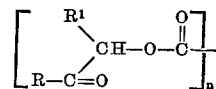
Another object is to provide acyloxy substituted two-equivalent couplers which have good coupling reactivity and which include the diffusible type coupler, the Fischer type and the nondiffusing type couplers which are readily incorporated in light-sensitive hydrophilic colloid-silver halide emulsion layers in a wide range of coupler to solvent ratios.

Still another object is to provide photographic hydrophilic colloid-silver halide emulsion layers containing my acyloxy substituted two-equivalent couplers.

These and other objects will appear from the following specification and claims.

These and other objects are accomplished by the synthesis and use of my novel couplers. The couplers of my invention are open-chain reactive methylene two-equivalent couplers wherein one of the hydrogens of the active methylene group is replaced with an acyloxy substituent. The acyloxy group includes, for example, unsubstituted and substituted alkylacyloxy, arylacyloxy, and heterocyclicacyloxy. These groups may be either mono- or divalent, i.e., they can contain either one or two coupler moieties. The effectiveness of my couplers as two-equivalent couplers is not dependent on the specific composition of the coupler moiety, and it will be understood that this moiety may be varied widely to meet such requirements as spectral absorptivity, reactivity, solubility, and diffusibility, as may be imposed by the photographic system in which the couplers are to be used. Typical examples of different classes of couplers which are included in my invention are alpha-acylacetamides, 2-(alpha-cyanoacetyl)coumarones, and alpha-acylacetonitriles, etc. My two-equivalent couplers are derived to advantage from any of the known four-equivalent open chain reactive methylene couplers.

Accordingly, the couplers of my invention may be further represented by the following general formula:



wherein n represents an integer of from 1 to 2; R represents an alkyl radical (either a straight or branched chain alkyl radical of from 1 to 30 carbon atoms, a mono- or bicyclic alkyl radical such as cyclohexyl, terphenyl, such as, a norbornyl radical, etc., said alkyl radicals being unsubstituted or substituted alkyl radicals, containing substituent groups such as halogen, nitro, hydroxyl, carboxyl, amino, substituted amino, aryl, carboxyl esters (e.g., carboethoxy, carbophenoxy, etc.), amido (e.g., acetamido, butyramido, ethylsulfonamido, N-propylbenzamido, etc.), carbamyl (e.g., N-methylcarbamyl, N-octadecylcarbamyl, N-methyl-N-phenylcarbamyl, etc.), sulfamyl (e.g., N-propylsulfamyl, N-tolylsulfamyl, etc.), alkoxy (e.g., ethoxy, octadecoxy, etc.), aryloxy (e.g., phenoxy, tolyloxy, etc.), sulfo, sulfoesters (e.g., methylsulfonfyl octadecylsulfonfyl, ethoxysulfonfyl, dodecoxysulfonfyl, phenyl-

sulfonyl, tolylsulfonyl, phenoxy sulfonyl, etc.), an aryl radical such as phenyl or naphthyl, said aryl radicals being unsubstituted or substituted with groups such as defined above for the substituted alkyl groups, and a heterocyclic radical containing a heterocyclic ring with 5 to 6 atoms in the ring, typical radicals including furanyl, benzofuranyl, benzothiazolyl, naphthothiazolyl, thiazolyl, benzoxazolyl, oxazolyl, imidazolyl, quinolyl, pyridyl, etc., said heterocyclic groups being unsubstituted or substituted with groups as defined for the substituted alkyl groups above; R_1 represents the cyano group, a carbamyl group (e.g., an unsubstituted carbamyl), an alkyl carbamyl group in which the alkyl group is either straight or branched chain having from 1 to 30 carbon atoms, a mono- or bicyclic group such as cyclohexyl, terphenyl (e.g., a norbornyl group, etc.), a dialkylcarbamyl group in which a combination of any two of the above-mentioned alkyl groups are substituted on the nitrogen atom of the carbamyl group, an aryl-carbamyl group in which the aryl group is a group such as phenyl, naphthyl, etc., an aralkylcarbamyl group in which a combination of any two of the above alkyl and aryl groups are substituted on the nitrogen atom of the carbamyl group (e.g., an N-methyl-N-phenylcarbamyl group, an N-butyl-N-tolylcarbamyl group, etc.), a heterocyclic carbamyl group in which the heterocyclic group is a group having a heterocyclic ring with 5 to 6 atoms in the ring, typical radicals including a thiazolyl group, a benzothiazolyl group, a naphthothiazolyl group, an oxazolyl group, a pyridyl group, a quinolyl group, etc., an alkyl heterocyclic carbamyl group in which a combination of any two of the above alkyl groups and heterocyclic groups, respectively, are substituted on the nitrogen atom of the carbamyl group (e.g., an N-(2-benzothiazolyl)-N-methylcarbamyl group, etc.), an aryl heterocyclic carbamyl group in which a combination of any two of the above aryl groups and heterocyclic groups, respectively, are substituted on the nitrogen atom of the carbamyl group (e.g., an N-phenyl-N-(2-thiazolyl)carbamyl group, etc.), in which the alkyl, aryl, and heterocyclic groups on the carbonyl are unsubstituted or substituted with groups such as defined for substituted alkyl in R above; R^2 , when n is the integer 1, represents an alkyl, aryl, or heterocyclic group as defined in R above, and, when n is the integer 2, R^2 represents alkylene (e.g., ethylene, hexylene, etc.), arylene (e.g., 1,2-phenylene, 1,3-phenylene, 1,4-phenylene, 1,4-naphthylene, ethylenebis(1,4-phenylene), sulfonylbis(1,4-phenylene), etc.), a divalent heterocyclic radical

derived from any of the heterocyclic groups defined above for R (e.g., a 3,5-pyridylene, a 4,8-quinolyne, a 4,5-thiazolylene, a 5,6-benzothiazolylene, a 6,7-naphthothia-

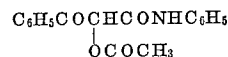
zolylene, a 4,5-oxazolylene, a 4,5-imidazolylene, etc.), the said alkyl, aryl and heterocyclic groups being unsubstituted or substituted with any of the groups such as defined for substituted alkyl in R above.

My two-equivalent couplers are characterized by having an acyloxy group on the coupling position of the coupler which gives them good coupling activity and other valuable properties. Nonimage-forming but stain-producing reactions characteristic of certain prior art two-equivalent couplers are not exhibited by my couplers. Some of my nondiffusible couplers have good coupling reactivity when incorporated in emulsion layers with no high-boiling coupler solvents, while the others are dispersed to advantage in high-boiling solvent solutions in a wide range of coupler to solvent ratios.

The acyloxy group provides a means of attaching other substituents to my couplers such as, for example, preformed dyes useful in color correction, dye transfer processes, and the like. Additional ballasting groups can also be conveniently introduced via the acyloxy group. Such groups, i.e., preformed dyes, ballasting groups, etc., are then eliminated during the coupling reaction of my couplers with oxidized color developer which forms the image dyes.

The following typical couplers will serve to illustrate but not limit my invention.

(1) α -Acetoxy- α -benzoylacetanilide



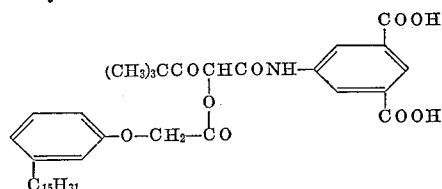
(2) α -Acetoxy- α -benzoyl-2-methoxyacetanilide

(3) α -Acetoxy- α -benzoyl-3,5-dicarboxymethoxyacetanilide

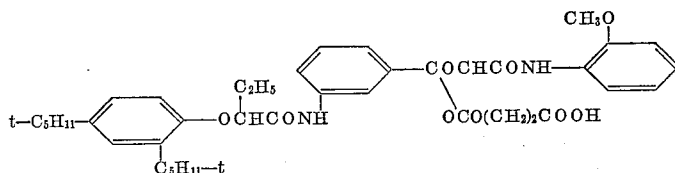
(4) α -Acetoxy- α -benzoyl-3,5-dicarboxyacetanilide

(5) α -Benzoyl- α -stearoyloxy-3,5-dicarboxyacetanilide

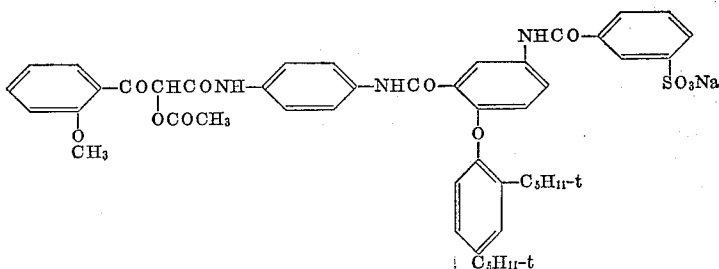
(6) α -Pivalyl- α -[α -(3-pentadecylphenoxy)acetoxy]-3,5-dicarboxyacetanilide



(7) α -(β -Carboxypropionyloxy)- α -{3-[α -(2,4-di-tert-amylphenoxy)butyramido]benzoyl}-2-methoxyacetanilide

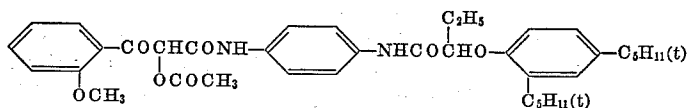


(8) α -Acetoxy- α -(2-methoxybenzoyl)-4-[2-(2,4-di-tert-amylphenoxy-5-(3-sulfobenzamido)benzabido]acetanilide sodium salt

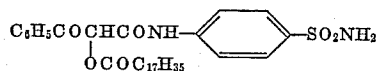


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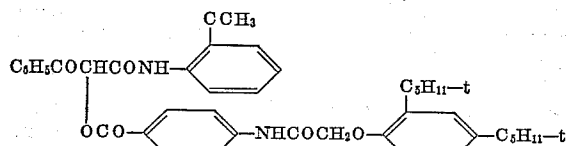
- (9) α -Acetoxy- α -(2-methoxybenzoyl)-4-[α -(2,4-di-tert-amylphenoxy)butyramido]acetanilide



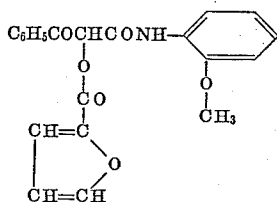
- (10) α -Benzoyl- α -stearoyloxy-2-methoxyacetanilide
 (11) α -Acetoxy- α -{3-[α -(2,4-di-tert-amylphenoxy)butyramido]benzoyl}-2-methoxyacetanilide
 (12) α -Acetoxy- α -{3-[γ -(2,4-di-tert-amylphenoxy)butyramido]benzoyl}-2-methoxyacetanilide
 (13) α -Pivalyloxy- α -{3-[γ -(2,4-di-tert-amylphenoxy)butyramido]benzoyl}-2-methoxyacetanilide
 (14) α -Benzoyl- α -[α -(2,4-di-n-amylphenoxy)acetoxy]-2-methoxyacetanilide
 (15) α -Benzoyl- α -stearoyloxy-4-sulfamylacetanilide



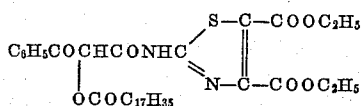
- (16) α -Pivalyl- α -stearoyloxy-4-sulfamylacetanilide
 (17) α -Benzoyl- α -stearoyloxy-2-chloroacetanilide
 (18) α -Benzoyl- α -[α -(2,4-di-tert-amylphenoxy)acetoxy]-2-methoxyacetanilide
 (19) α -Benzoyl- α -[α -(3-pentadecylphenoxy)acetoxy]-2-methoxyacetanilide
 (20) α -Benzoyl- α -stearoyloxy-3-chloroacetanilide
 (21) α -Benzoyl- α -[α -(2,4-di-tert-amylphenoxy)butyryloxy]-2-methoxyacetanilide
 (22) α -Benzoyl- α -{4-[α -(2,4-di-tert-amylphenoxy)acetamido]benzoyloxy}-2-methoxyacetanilide



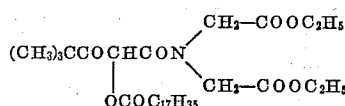
- (23) α -[α -(2,4-di-n-amylphenoxy)acetoxy]- α -pivalyl-2-chloroacetanilide
 (24) α -Benzoyl- α -(2-furoyloxy)-2-methoxyacetanilide



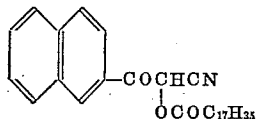
- (25) α -{3-[α -(2,4-di-tert-amylphenoxy)butyramido]benzoyl}- α -(2-furoyloxy)-2-methoxyacetanilide
 (26) α -Benzoyl- α -stearoyloxy-N-(4,5-di-carbethoxy-2-thiazolyl)acetamide



- (27) α -Pivalyl- α -stearoyloxy-N-(5-carbethoxy-2-thiazolyl)acetamide
 (28) α -Pivalyl- α -[α -(3-pentadecylphenoxy)acetoxy]-N-(2-pyridyl)acetamide
 (29) α -Pivalyl- α -stearoyloxy-N,N-bis(carbethoxymethyl)acetamide

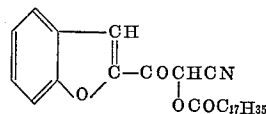


- (30) α -(β -Naphthoyl)- α -stearoyloxyacetoneitrile

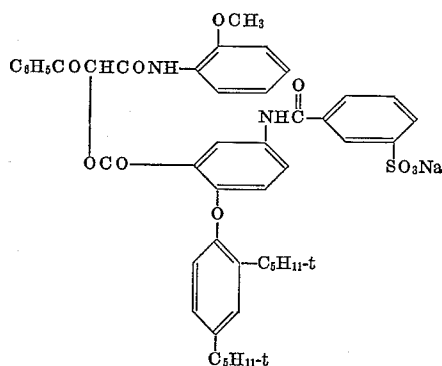


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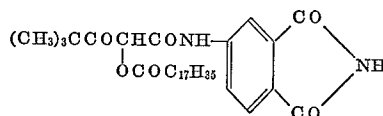
- (31) α -(β -Naphthoyl)- α -acetoxyacetoneitrile
 (32) 2-(α -cyano- α -stearoyloxyacetyl)coumarone



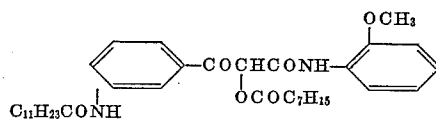
- (33) α -Benzoyl- α -[2-(2,4-di-tert-amylphenoxy)-5-(3-sulfobenzamido)benzoyloxy]-2-methoxyacetanilide sodium salt



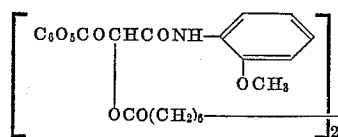
- (34) α -Pivalyl- α -stearoyloxy-3,5-dicarboximidoacetanilide



- (35) α -Pivalyl- α -stearoyloxy-3-chloroacetanilide
 (36) α -Pivalyl- α -stearoyloxy-4-(N,N-dimethylsulfamyl)acetanilide
 (37) α -Pivalyl- α -stearoyloxy-4-methylsulfonamidoacetanilide
 (38) α -Benzoyl- α -stearoyloxy-5-chloro-2-methoxyacetanilide
 (39) α -Benzoyl- α -stearoyloxyaceto-o-toluidide
 (40) α -Benzoyloxy- α -pivalyl-2-chloro-5-[γ -(2,4-di-tert-amylphenoxy)butyramido]acetanilide
 (41) Alpha-acetoxy-alpha-pivalyl-2-chloro-5-[γ -2,4-di-t-amylphenoxy]butyramido]acetanilide
 (42) Alpha-(3-dodecanamidobenzoyl)-alpha-octanoyloxy-2-methoxy-acetanilide



- (43) Alpha-alpha'-tetradecanedioxybis(alpha-benzoyl-2-methoxy-acetanilide)



The diffusible couplers of my invention, such as Couplers 1, 2 and 3, are used to advantage in color developer solutions used to color develop light-sensitive elements used for color photography which do not contain the color-forming coupler. Any of the well known primary

aromatic amino color-forming silver halide developing agents such as the phenylenediamines, e.g.,

diethyl-p-phenylenediamine hydrochloride,
monomethyl-p-phenylenediamine hydrochloride,
dimethyl-p-phenylenediamine hydrochloride,
2-amino-5-diethylaminotoluene hydrochloride,
2-amino-5-(N-ethyl-N-laurylamino)toluene,
N-ethyl-N-(beta-methanesulfonamidoethyl)-3-methyl-4-aminoaniline sulfate,
N-ethyl-N-(beta-methanesulfonamidoethyl)-4-aminoaminoaniline,
4-[N-ethyl-N-(beta-hydroxyethyl)amino]aniline, etc.,

the p-aminophenols and their substitution products where the amino group is unsubstituted may be used in the alkaline developer solution with my couplers. Various other materials may be included in the developer solutions depending upon the particular requirements, for example, an alkali metal sulfite, carbonate, bisulfite, bromide, iodide, etc., and the thickening agents used in viscous developer compositions such as carboxymethyl cellulose, carboxyethyl cellulose, etc. The following is a typical developer solution given to illustrate but not limit the invention.

2-amino-5-diethylaminotoluene HCl	-----g--	2.0
Sodium sulfite (anhydrous)	-----g--	2.0
Sodium carbonate (anhydrous)	-----g--	20.0
Potassium bromide	-----g--	1.0
Coupler	-----g--	2.0
Water, to	-----ml--	1000.0

The other coupler examples used to illustrate my invention are nondiffusing and are used to advantage in photographic emulsion layers. Couplers such as 7 and 8 illustrate those that are incorporated as Fischer type couplers. The other nondiffusing couplers are incorporated in emulsion layers by methods such as are described by Mannes et al. U.S. Patent 2,304,939, issued Dec. 15, 1942, Jelley et al. U.S. Patent 2,322,027, issued June 15, 1943, etc., in which high-boiling organic solvents are used to dissolve the coupler, and by methods described in Vittum et al. U.S. Patent 2,801,170, and Fierke et al. U.S. Patent 2,801,171, both issued July 30, 1957, and Julian U.S. Patent 2,949,360, issued Aug. 16, 1960 in which low-boiling or water-soluble organic solvents are used with or in place of the high-boiling solvent. Not only can emulsion layers containing my couplers be made thinner because they require only one-half the silver halide required by conventional couplers (i.e., four-equivalent couplers) but some of my couplers are sufficiently reactive that they do not require any high-boiling coupler solvent that is usually required by couplers. Thin image-forming layers are very desirable because they cause less light scattering and produce sharper images.

My nondiffusing couplers 5 and 6 form diffusible dye images upon color development and are used to advantage either in image transfer elements or in emulsion layers that contain my coupler as a nonimage-forming competing coupler along with an image-forming coupler. Couplers 15 and 16 have a sulfamyl group (on a noncoupling position) which ionizes and forms a diffusible dye upon color development at a pH above 11.

The other nondiffusing couplers used to illustrate my invention form nondiffusing dyes and are used to advantage in any photographic element where incorporated image-forming couplers are desired. Couplers No. 15 and 16 when developed with a color developer with a pH lower than 11 will produce nondiffusing dye images.

Couplers 30, 31 and 32 form magenta image dyes on coupling with oxidized color developer.

My couplers are used in the color development of photographic hydrophilic colloid-silver halide emulsion layers of the developing-out type either in the color developer solution or in the emulsion layer. The emulsions may contain silver chloride, silver bromide, silver iodide, silver

chlorobromide, silver bromide, silver chlorobromide, etc., as the light-sensitive material.

Hydrophilic colloids used to advantage include gelatin, colloidal albumin, a cellulose derivative, or a synthetic resin, for instance, a polyvinyl compound. Some colloids which may be used are polyvinyl alcohol or a hydrolyzed polyvinyl acetate as described in U.S. Patent 2,286,215, of Lowe; a far hydrolyzed cellulose ester, such as cellulose acetate hydrolyzed to an acetyl content of 19-26% as described in U.S. Patent 2,327,808 of Lowe and Clark, a water-soluble ethanolamine cellulose acetate as described in U.S. Patent 2,322,085 of Yutzy; a polyacrylamide having a combined acrylamide content of 30-60% and a specific viscosity of 0.25-1.5 on an imidized polyacrylamide of like acrylamide content and viscosity as described in U.S. Patent 2,541,474 of Lowe, Minsk and Kenyon; zein as described in U.S. Patent 2,563,791 of Lowe; a vinyl alcohol polymer containing urethane carboxylic acid groups of the type described in U.S. Patent 2,768,154 of Unruh and Smith, or containing cyano-acetyl groups, such as the vinyl alcohol-vinyl cyano-acetate copolymer as described in U.S. Patent 2,808,331 of Unruh, Smith and Priest; or a polymeric material which results from polymerizing a protein or a saturated acrylated protein with a monomer having a vinyl group as described in U.S. Patent 2,852,382 of Illingsworth, Dann and Gates.

The emulsions used in the photographic element of my invention can be chemically sensitized by any of the accepted procedures. The emulsions can be digested with naturally active gelatin, or sulfur compounds can be added, such as those described in Sheppard U.S. Patent 1,574,944; Sheppard and Punnett U.S. Patent 1,623,499; and Sheppard and Brigham U.S. Patent 2,410,689.

The emulsions can also be treated with salts of the noble metals, such as ruthenium, rhodium, palladium, iridium and platinum. Representative compounds are ammonium chloropalladate, potassium chloroplatinate, and sodium chloropalladate, which are used for sensitizing in amounts below that which produces any substantial fog inhibition, as described in Smith and Trivelli U.S. Patent 2,448,060 and as antifoggants in higher amounts, as described in Trivelli and Smith U.S. Patents 2,566,245 and 2,566,263.

The emulsions can also be chemically sensitized with gold salts as described in Waller, Collins and Dodd U.S. Patent 2,399,083 or stabilized with gold salts as described in Damschroder U.S. Patent 2,597,856 and Yutzy and Leermakers U.S. Patent 2,597,915. Suitable compounds are potassium chloroaurate, potassium aurithiocyanate, potassium chloroaurate, auric trichloride and 2-aurosulfobenzothiazole methochloride.

The emulsions can also be chemically sensitized with reducing agents, such as stannous salts (Carroll U.S. Patent 2,487,850), polyamines, such as diethylene triamine (Lowe and Jones U.S. Patent 2,518,698), polyamines, such as spermine (Lowe and Allen U.S. Patent 2,521,925), or bis(beta-aminoethyl)sulfide and its water-soluble salts (Lowe and Jones U.S. Patent 2,521,926).

The emulsions can also be optically sensitized with cyanine and merocyanine dyes, such as those described in Brooker U.S. Patents 1,846,301, 1,846,302 and 1,942,854; White U.S. Patent 1,990,507; Brooker and White U.S. Patents 2,112,140, 2,165,338, 2,493,747 and 2,739,964; Brooker and Keyes U.S. Patent 2,493,748; Sprague U.S. Patents 2,503,776 and 2,419,001; Heseltine and Brooker U.S. Patent 2,666,761; Heseltine U.S. Patent 2,734,900; Van Lare U.S. Patent 2,739,149; and Kodak Limited British 450,958.

The emulsions may also contain speed-increasing compounds of the quaternary ammonium type of Carroll U.S. Patent 2,271,623; Carroll and Allen U.S. Patent 2,288,226; and Carroll and Spence U.S. Patent 2,334,864; and the polyethylene glycol type of Carroll and Beach U.S. Patent 2,708,162.

The above-described emulsions can be coated on a wide variety of photographic emulsion supports. Typical supports include cellulose nitrate film, cellulose acetate film, polyvinyl acetal film, polystyrene film, polyethylene terephthalate film, polyethylene film, polypropylene film, and related films of resinous materials, as well as paper, glass and others.

The couplers of my invention may also be used to advantage in image-forming layers, either alone or with image-forming compounds other than silver halide such as ZnO, ZnS, CdS, CdSe, NiS, etc., either with or without binders such as gelatin, polyvinyl alcohol, etc.

Usually my emulsions are coated on photographic supports in the form of multilayer color photographic elements wherein at least three differently sensitized emulsion layers are coated over one another on the support. Usually the support is coated in succession with a red-sensitive layer, a green sensitive layer and a blue-sensitive layer either with or without a Carey Lea filter layer between the blue-sensitive and green-sensitive layers. The three differently color sensitized layers may be arranged in any other order over one another that is desirable;

10 ml. of a gelatino-silver halide emulsion with stirring at 40° C. The resultant emulsion was coated at 0.009 inch wet thickness.

An image exposed strip of this coating was processed by contacting it for 7 minutes at 80° F. with a mordant containing receiving sheet which had been presoaked in the following developer solution:

Developer formula

Sodium carbonate monohydrate	g--	22.000
Ascorbic acid	g--	0.385
Potassium bromide	g--	0.825
Sodium sulfite	g--	2.000
4 - [N - Ethyl - N - (Beta - hydroxyethyl)-amino]	g--	11.000
Water, to pH 11.0	ml--	1100.000

The following photographic data were obtained from the processed strips, i.e., from the mordant receiving sheets containing the transferred dye images.

Mordant in Receiving Sheet	D _{max}	λ _{max}	Light Fade to Xenon Arc		Heat Fade: 140°F., 70% R.H.		Specular Density
			30 hrs.	70 hrs.	1 wk.	2 wks.	
Dimethyl-β-hydroxyethyl-α-(octadecyl-amido)propylammonium dihydrogen phosphate	.86	437	.15	.38	.13	.30	.86
Octadecyltributylammonium thiocyanate + 1,4-dihexadecyl-1,4-diazobicyclo[2,2,2]octane dithiocyanate in 2,4-ditert-amylphenol	.80	434	.11	.32	0	0	.81

however, the Carey Lea filter layer obviously would not be put over the blue-sensitive layer. Preferably, these light-sensitive layers are arranged on the same side of the support.

Elements made for image transfer processing may use a separate reception sheet which is contacted with the light-sensitive layer during its development or the reception layer may be an integral part of the light-sensitive element. Any of the support materials mentioned previously may be used for a separate reception sheet. The reception layer comprises a hydrophilic colloid layer containing a cationic mordant, e.g., the polymers of amino guanidine derivatives of vinyl methyl ketone such as described in Minsk U.S. Patent 2,882,156, granted Apr. 14, 1959. Other mordants include the 2-vinyl pyridine polymer metho-p-toluene sulfonate and similar compounds described in Sprague et al. U.S. Patent 2,484,430, granted Oct. 11, 1949, and cetyl trimethyl ammonium bromide, etc. Particularly effective mordanting compositions are described in copending applications of Knechel et al. U.S. Ser. No. 211,095, filed July 19, 1962, now U.S. Patent 3,271,148 and Bush U.S. Ser. No. 211,094, filed July 19, 1962, now U.S. Patent 3,271,147. Additional variations of the image transfer elements and processes in which couplers of my invention (such as 5, 6, 15, 16 and 37) can be used to advantage, are described in copending application of Whitmore and Mader U.S. Ser. No. 222,105, filed Sept. 7, 1962, now U.S. Patent 3,227,550.

My invention is still further illustrated by the following typical examples.

EXAMPLE 1

To a solution of 0.32 g. of Coupler No. 6 in 10 ml. of water, 6 drops of 10% sodium hydroxide, 13 ml. of 10% bone gelatin and 1 g. of 5% formalin was added

EXAMPLE 2

Single layer gelatino-silver bromide emulsions containing yellow-forming coupler and the coupler solvent, di-n-butylphthalate, were made for my couplers No. 10, No. 11 and No. 12 and control couplers α-{3-[α-(2,4-di-tert-amylphenoxy)butyramido]benzoyl}-2-methoxyacetanilide (control for coupler No. 10 and coupler No. 11) and α-{3-[γ-(2,4-di-tert-amylphenoxy)butyramido]benzoyl}-2-methoxyacetanilide (control for coupler No. 12). The amounts of gelatin, silver bromide, coupler and coupler solvent used in the coatings are listed in Table I. Strips of the two coatings were given 1/25-second exposures on a 1B intensity scale sensitometer and processed through the following process.

Time:	Process steps
30''	Water dip.
10'	Development.
5'	Stop bath.
5'	Ferricyanide bleach.
5'	Wash.
5'	Fixing bath.
10'	Wash.
30''	Photo-Flo (wetting agent solution) dry.

The following developer solutions were used in this process:

Developer 1

Sodium sulfite	g--	2
2-amino-5-diethylaminotoluene HCl	g--	2
Sodium carbonate	g--	20
Potassium bromide	g--	2
Water, to pH 10.86	l--	1

11

Developer 2

Benzyl alcohol	ml.	4.00
Sodium sulfite	g.	2.00
N - ethyl - N - (beta - methanesulfonamidoethyl) 4-aminoaniline sulfate	g.	5.00
Sodium carbonate	g.	50.00
Sodium bromide	g.	0.86
Sodium hydroxide (20% solution)	ml.	4.00
Sodium hexametaphosphate	g.	0.50
Water, to pH 10.75	l.	1.00

The resulting yellow dye images were observed with a spectrophotometer to determine the $\lambda_{\max}$ values, and examined with a densitometer to obtain $D_{\max}$ values and the density data needed to calculate the γ and speed values. These data are summarized in Table I.

TABLE I

Coating Number	Gel, mg./ft. ²	AgBr, mg./ft. ²	Coupler, mg./ft. ²	Coupler Solvent, mg./ft. ²	Developer Formula No.	Photographic Data		
						$\lambda_{\max}$	γ	Speed
1	386	146	90—Control for 11	45	1	453	2.08	2.37
2	386	146	90—No. 11	45	1	451	2.82	2.46
3	386	146	90—Control for 11	45	2	447	1.74	2.26
4	386	146	90—No. 11	45	2	446	2.90	2.33
5	500	200	90—Control for 12	45	1	452	1.88	2.33
6	500	200	90—No. 12	45	1	453	3.06	2.33
7	580	45	90—Control for 10	45	1	452	.59	-----
8	580	45	90—No. 10	45	1	442	1.08	-----

The results obtained from coatings 1 through 6 show that in each instance the coating of my invention (i.e., 2, 4 and 6) with a two-equivalent coupler gave higher dye yields as evidenced by higher γ and $D_{\max}$ values than were obtained with the corresponding control coatings No. 1, 3 and 5, respectively, with the corresponding four-equivalent couplers even though the amount of silver halide used in these emulsion coatings was not limited.

The results from coatings 7 and 8 further show the advantage of coatings containing my two-equivalent coupler No. 10 compared to a four-equivalent coupler.

EXAMPLE 3

Two multilayer coatings were prepared which were identical except that in the second one, i.e., coating No. 2, the silver halide level was reduced to one-half that contained in coating No. 1. These coatings each contained the four-equivalent coupler α -{3-[α -(2,4-di-tert-amylphenoxy)butyramido]benzoyl}-2-methoxyacetanilide as a control coupler. A third coating, No. 3, was prepared which was identical to coating No. 2 excepting that an equimolar amount of coupler No. 11 was used in place of the control coupler in the blue-sensitive layer. Each multilayer coating contained along with the yellow-forming layer, a light-sensitive layer with a magenta-forming coupler and one with a cyan-forming coupler.

Strips of these coatings were each given a blue light step tablet exposure in a sensitometer, and then processed as follows.

Processing solution:	Time in minutes
Color developer	11.5
Acid stop bath	6.0
Formalin hardener	4.0
Water wash	4.0
Ferricyanide bleach	6.5
Water wash	4.0
Hypo fix	4.5
Water wash	8.0

The processing solutions had essentially the compositions set out below:

Color developer	
Benzyl alcohol	ml. 5.00
Sodium hexametaphosphate	g. 2.50
Sodium sulfite	g. 2.50
Sodium bromide	g. 1.40
Potassium bromide	mg. 0.50
Sodium hydroxide	g. 13.10
Borax	g. 39.60

12

4 - amino - N - ethyl - 3 - methyl - N - (β - methylsulfonamidoethyl) aniline	g.	50.00
Water, to make	ml.	1000.00

Acid stop bath

Acetic acid (glacial)	ml.	8.6
Water, to make	ml.	1000.0

Formalin hardener

Formalin (37% by weight formaldehyde in water)	ml.	20.00
Sodium bisulfite	g.	5.00
Borax	g.	3.82
Sodium hydroxide	g.	4.50
Water, to make	ml.	1000.00

Ferricyanide bleach

Potassium ferricyanide	g.	18.0
Sodium bromide	g.	12.8
Borax	g.	7.3
Boric acid	g.	15.0
Water, to make	ml.	1000.0

Hypo fix

Sodium sulfite	g.	5.90
Sodium thiosulfate	g.	320.00
Sodium bisulfite	g.	1.47
Water, to make	ml.	1000.00

The yellow dye density to blue light in each step on the processed strips was measured with a densitometer and these density data were plotted against the step number of the step tablet. Consecutive steps represented 0.15 log exposure increments. A comparison of the yellow dye density curves 1 and 2 for the step tablet images obtained in coating No. 1 and No. 2, respectively, shows that the dye yield is proportional to the silver level in the coating. A comparison of the yellow dye density, curve 3, for the step tablet image obtained in coating No. 3 containing coupler No. 11 at the low silver level showed a high dye yield that was comparable to the yield obtained with coating No. 1 but with one-half the amount of silver that was used in coating No. 1.

The following example will illustrate the high reactivity of my couplers even in nonsolvent dispersions.

EXAMPLE 4

Two single layer coatings were made of a hydrophilic colloid-silver halide emulsion containing coupler 14. In coating No. 1 the coupler was dispersed as a solution in the high boiling coupler solvent, di-n-butylphthalate, at a coupler to solvent ratio of 1:½. In coating No. 2 this ratio was 1:0. These coatings contained 228 mg. of silver (as silver halide) and 90 mg. of coupler per square foot. Similar coatings No. 3 and No. 4 were made containing equimolar amounts of the coupler α -pivalyl-2-chloro-5-[γ -(2,4-di-tert-amylphenoxy)butyramido]acetanilide and silver at coupler to solvent ratios of 1:½ and 1:0, respectively.

Strips from each of these coatings were given identical exposures and processed as described in Example 3. The yellow dye density (to blue light) of each step on the processed strips was measured with a densitometer and plotted against the exposure step number.

The curves resulting from the plots of density vs. exposure showed that the strips containing coupler 14 gave

excellent dye image reproductions of the original step tablet image with very little difference between the curves 1 and 2 for this coupler at coupler to solvent ratios of 1:½ and 1:0, respectively. The curve 3 for the strip containing the dispersion of coupler α -pivalyl-2-chloro-5-[γ -(2,4-di-tert-amylphenoxy)butyramido]acetanilide in the solvent at the ratio of 1:½ had a substantially lower dye density at each step than the strip containing coupler No. 14. The curve 4 for the strip containing the nonsolvent dispersion of the coupler used in coating No. 3 showed that there was very little dye produced and that no useful image was produced of the original step tablet.

Similarly other couplers of my invention are used to advantage in hydrophilic colloid-silver halide emulsions when dispersed in coupler solvent solution or dispersed without coupler solvent and exhibit high reactivities.

EXAMPLE 5

Single layer coatings were made containing coupler No. 16. Coating No. 1 contained 400 mg. of gelatin, 146 mg. silver bromide, 63 mg. of coupler 16, and 31 mg. of di-n-butylphthalate per square foot of coating. Coating No. 2 had the same composition except that 63 mg. of diethylauramide per square foot of coating was used in place of the di-n-butylphthalate.

35 mm. strips of coating No. 1 were exposed and processed to produce a nondiffusible image dye as described in Example 2 using developer 1 for one strip and developer 2 for another strip. Measurements made of λ_{max} , D_{max} , the amount of "light fade," that is, density loss caused by 30 hours' exposure to a Xenon arc, the amount of "heat fade," that is, the density loss caused by holding the processed film strip in an oven at 140° F. and 70% R.H. for one week, are tabulated as follows:

Coating	Developer	λ_{max}	Light Fade	Heat Fade	D_{max}
1.-----	1	450	0.13	0.12	2.52
1.-----	2	445	0.09	0.13	2.47

35 mm. strips of coating No. 2 were exposed and processed as described in Example 1 but using a mordanted receiving sheet presoaked in a developer solution having the composition:

Sodium hydroxide -----g--	7.0
Ascorbic acid -----g--	0.4
Potassium bromide -----g--	2.0
Sodium sulfite -----g--	2.0
4-N-ethyl-N-(β -hydroxyethylamino)aniline --g--	2.0
Water, to -----ml--	1100.0

The dye transfer images formed in the mordanted receiving sheets were measured for D_{max} , λ_{max} , percent light fade after 30 and 100 hours' exposure to a Xenon arc, and percent heat fade after 1 week and after 2 weeks' storage in an oven at 140° F., 75% R.H. These values are summarized as follows:

Receiving Sheet Mordant and Support	D_{max} in Density Units	λ_{max} in m μ	Percent Light Fade		Percent Heat Fade	
			30 hrs.	100 hrs.	1 wk.	2 wks.
N-cetyl-N-ethylmorpholinium ethosulfate + methyltrilaurylammonium bromide coacervate on a suitably subbed cellulose acetate sheet-----	1.47	436	6	19	18	33
Above mordant coacervate on white pigmented cellulose acetate support-----	2.82	435	6	18	16	24
Poly(4-vinylpyridine) on white pigmented cellulose acetate support-----	1.61	440	10	22	6	18

Couplers, such as No. 16 that have a sulfamyl group on a noncoupling position can be used to produce either a diffusible or a nondiffusible image dye. By using a high pH developer solution, this group ionizes (to the anion $-\text{SO}_2\text{NH}^-$) and thus causes the dye to diffuse and to be mordanted on a cationic type of mordant. If the pH of the developer solution is kept low enough, e.g., below 11, the sulfamyl group does not ionize; consequently, the dye which is formed does not diffuse.

The following example will illustrate the use of still other representative couplers of my invention.

EXAMPLE 6

Single layer coatings were made and coated to contain 400 mg. gelatin, 45 mg. of silver bromide, 90 mg. of coupler, and either 45 mg. or 0 mg. of di-n-butylphthalate per square foot of coating. These were exposed and developed as described in Example 2 using developer solution No. 1. The λ_{max} value in m μ and the D_{max} value were measured and recorded in the following Table II.

TABLE II

Coating Number	Coupler Number	Di-n-butylphthalate, mg./ft. ²	λ_{max} , m μ	D_{max}
1	17	45	449	2.68
2	17	0	451	2.60
3	18	45	445	2.05
4	18	0	447	1.64
5	19	45	443	2.39
6	19	0	445	2.53
7	20	45	450	2.52
8	20	0	455	2.57
9	21	45	444	1.78
10	21	0	447	1.34
11	22	45	448	2.24
12	22	0	450	1.10

Similarly, the other couplers of my invention having Formula I are used to advantage in color photography as illustrated previously with representative couplers.

In general, my couplers of Formula I are prepared from the parent coupler having the formula:



wherein R and R' are as described previously by first replacing one of the hydrogen atoms on the active methylene group, with a halogen atom, (e.g., fluorine, bromine, chlorine, etc.) for example, by reacting the parent coupler with a chlorinating agent, such as, thionyl chloride, sulfonyl chloride, etc., as described by McCrossen, Vittum and Weissberger in U.S. Patent 2,728,658, issued Dec. 27, 1955, by reacting the sodium enolate of the parent coupler with perchloryl fluoride as described in Loria, Salminen and Weissberger U.S. Ser. No. 51,549, filed Aug. 24, 1960, by reacting the parent coupler with a brominating agent, such as, N-bromosuccinimide in acetic acid, etc., then by reacting the α -halo derivative of the coupler with the appropriate acylating agent, such as an organic acid or alkali metal salt of the organic acid having the formula:



wherein R_2 is a defined previously and M represents an alkali metal, such as, sodium, potassium, lithium, etc., in the presence of a suitable solvent, such as, acetic acid, glacial acetic acid, acetonitrile, dioxane, tetrahydrofuran, diethylcellosolve, methylcellosolve, dimethylformamide, etc. It is preferable that the reactants be anhydrous. An amine, such as, triethylamine is used to advantage as a condensing agent in this reaction when the acylating agent is an organic acid. The reaction is usually conducted at an elevated temperature, e.g., the reflux temperature of the reaction mixture. The following preparation of cou-

pler 1 will illustrate the synthesis of my yellow-forming couplers 1 and 41.

Coupler 1

A mixture of 8 g. of α -benzoyl- α -chloroacetanilide (prepared by the method given in McCrossen et al. U.S. Patent 2,728,658, issued Dec. 27, 1955), 5 g. of anhydrous sodium acetate, and 150 ml. of acetic acid was refluxed for 18 hours. Sodium chloride began to separate after about 3 hours of heating. Water was added to the hot solution to just dissolve the sodium chloride and

bring incipient crystallization. The solution was stirred while cooling to room temperature. The white crystalline product which formed was collected, washed on the funnel with cold water, and air dried. This solid was recrystallized twice from absolute alcohol to yield a product which melted at 139°–139.5° C.

Couplers 2 through 41 are prepared by similar reactions using the coupler intermediate, acylating agent, condensing agent (where needed) and solvent reaction medium listed in Table III. The melting points found for my couplers are given in Table III.

TABLE III

Compound No.	Coupler Intermediate	Acylating Agent	Condensing Agent	Solvent (Reaction Medium)	M.P. of Product (° C.)
1	α -Benzoyl- α -chloroacetanilide	Sodium acetate		Acetic acid	139–139.5
2	α -Benzoyl- α -fluoro- α -methoxyacetanilide (Loria et al., U.S. Serial No. 51,549, filed August 24, 1960).	do.		do.	128–129
3	α -Benzoyl- α -bromo-3,5-dicarboxymethoxyacetanilide.	do.		do.	169–171
4	α -Benzoyl- α -bromo-3,5-dicarboxyacetanilide.	do.		do.	108–110
5	do.	Sodium stearate		Dimethylformamide	>200
6	α -Bromo- α -pivalyl-3,5-dicarboxyacetanilide.	m-Pentadecylphenoxy acetic acid sodium salt (Bush et al., U.S. 2,908,573, issued October 13, 1959).		Ethyl alcohol	90
7	α -Chloro- α -[3-(α -(2,4-di-tert-amylphenoxy)butyramido)benzoyl]-2-methoxyacetanilide (McCrossen et al., U.S. Patent 2,875,057, issued February 24, 1959).	Succinic acid monosodium salt.		Dioxane	>180
8	α -Chloro- α -(α -methoxybenzoyl)-4-[2-(2,4-di-tert-amylphenoxy)-5-(3-sulfobenzamido)benzamido]acetanilide sodium salt.	Sodium acetate		Acetic acid	*>170
9	α -Chloro- α -(2-methoxybenzoyl)-4-[α -(2,4-di-tert-amylphenoxy)butyramido]acetanilide (Yule U.S. 2,407,211, issued Sept. 3, 1946 and Loria et al., U.S. 2,728,659, issued Dec. 27, 1955).	do.		do.	91–92
10	α -Benzoyl- α -chloro-2-methoxyacetanilide.	Stearic acid	Triethylamine	Acetonitrile	66–68
11	As in Compound 7 above.	Acetic acid	do.	do.	>200
12	α -Chloro- α -[3-(γ -(2,4-di-tert-amylphenoxy)butyramido)benzoyl]-2-methoxyacetanilide.	do.	do.	do.	98–99
13	As in Compound 12 above.	Pivalic acid	do.	do.	107–108
14	As in Compound 2 above.	α -(2,4-di-n-amylphenoxy)acetic acid sodium salt.		Dimethylformamide	89–91
15	α -Benzoyl- α -chloro-4-sulfamylacetanilide.	Stearic acid	Triethylamine	do.	116–118
16	α -Chloro- α -pivalyl-4-sulfamylacetanilide.	do.	do.	do.	108–110
17	α -Benzoyl- α -fluoro-2-chloroacetanilide (Loria U.S. Serial No. 51,549).	Sodium stearate		do.	62–63
18	As in Compound 2 above.	α -(2,4-di-tert-amylphenoxy)acetic acid (McCrossen et al., U.S. 2,875,057, issued Feb. 24, 1959 and McCrossen et al., U.S. 2,728,658, issued Dec. 27, 1955).		Acetonitrile	106–108
19	As in Compound 2 above.	As in Compound 6 above.		Ethyl alcohol	82–87
20	α -Benzoyl- α -fluoro-3-chloroacetanilide.	Sodium stearate		Dimethylformamide	75–77
21	As in Compound 2 above.	α -(2,4-di-tert-amylphenoxy) butyric acid sodium salt.		do.	123–125
22	As in Compound 2 above.	4-[α -(2,4-di-tert-amylphenoxy)acetamido] benzoic acid.	Triethylamine	Acetonitrile	161–162
23	α -Chloro- α -pivalyl-2-chloro acetanilide.	α -(2,4-di-n-amylphenoxy) acetic acid	do.	Dimethylformamide	52–54
24	As in Compound 2 above.	2-furoic acid.	do.	do.	
25	As in Compound 7 above.	do.	do.	Acetonitrile	
26	α -Benzoyl- α -chloro-N-(4,5-dicarbethoxy-2-thiazolyl)acetamide.	Sodium stearate		Dimethylformamide	
27	α -Chloro- α -pivalyl-N-(5-carbethoxy-2-thiazolyl)acetamide.	do.		do.	
28	α -Bromo- α -pivalyl-N-(2-pyridinyl)-acetamide.	As in Compound 6 above.		Ethyl alcohol	
29	α -Bromo- α -pivalyl-N,N-bis(carbethoxymethyl)acetamide.	Sodium stearate		Dimethylformamide	
30	α -Chloro- α -(β -naphthoyl)acetanitrile.	do.		Dioxane	
31	As in Compound 30 above.	Sodium acetate		Acetic acid	
32	α -Chloro- α -cyanoacetyl coumarone.	Sodium stearate		Dimethylformamide	
33	As in Compound 2 above.	5-(3-chlorosulfonylbenzamido)-2-(2,4-di-tert-amylphenoxy)benzoic acid (with subsequent acid hydrolysis of chlorosulfonyl group to sulfonic acid).		Ethyl alcohol	>180
34	α -Chloro- α -pivalyl-3,5-dicarboximidoacetanilide.	Stearic acid	Triethylamine	Dimethylformamide	118–120
35	α -Chloro- α -pivalyl-3-chloroacetanilide.	do.	do.	do.	66–67
36	α -Chloro- α -pivalyl-4-(N,N-dimethylsulfamyl)acetanilide.	do.	do.	do.	100–101
37	α -Chloro- α -pivalyl-4-(methylsulfonamido)acetanilide.	do.	do.	do.	74–76
38	α -Benzoyl- α -chloro-3-chloro-2-methoxyacetanilide.	do.	do.	do.	72–74
39	α -Benzoyl- α -chloroaceto-o-toluidide.	do.	do.	do.	81–82
40	α -Chloro- α -pivalyl-2-chloro-5-[α -(2,4-di-tert-amylphenoxy)butyramido]acetanilide.	Benzoic acid	do.	do.	140–142
41	As in Compound 40 above.	Sodium acetate		Acetic acid	77–79
42	α -Chloro- α -(3-decanamidobenzoyl)-2-methoxyacetanilide.	Octanoic acid	Triethylamine	Acetonitrile	67–69
43	As in Compound 10 above.	Tetradecanedioic acid	do.	do.	*95

*Decomposition.

My invention is still further illustrated by the accompanying drawings FIG. 1 and FIG. 2.

FIG. 1 shows the curves 1, 2 and 3 representing yellow dye image density to blue light vs. exposure obtained for coatings 1, 2, and 3, respectively, described in Example 3.

FIG. 2 shows the curves 1, 2, 3 and 4 representing the yellow dye image density to blue light vs. exposure obtained for coatings 1, 2, 3 and 4, respectively, as described in Example 4.

The two-equivalent image-forming couplers of my invention are distinguished from other two-equivalent couplers by having an acyloxy group substituted on the coupling position of the coupler molecule. My couplers are characterized by not producing stain, a problem with certain prior art two-equivalent couplers, and by having very good coupling reactivity. My nondiffusing couplers are dispersed readily in emulsion layers as a coupler solvent solution having a wide range of coupler to solvent ratios.

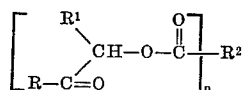
My couplers are not only valuable for the reasons cited but because photographic emulsion layers color developed with them require only $\frac{1}{2}$ the amount of silver halide required by four-equivalent couplers. Of particular value are certain of my nondiffusing couplers which have high coupling reactivity when dispersed in photographic emulsion layers without any high boiling solvent. These couplers are coated to advantage in particularly thin layers that produce good sharp images.

The invention has been described in detail with particular reference to preferred embodiments thereof but it will be understood that variations and modifications can be effected within the spirit and scope of the invention as described hereinabove and as defined in the appended claims.

I claim:

1. A yellow dye image-forming layer containing light-sensitive silver halide emulsion and an uncolored two-equivalent coupler which is capable of forming a yellow dye on coupling with the oxidation product of a primary aromatic amino color developing agent, said coupler containing at least one open-chain reactive methine group joined directly to two activating groups, at least one of said activating groups being a carbonyl group, said active methine group also joined directly to an acyloxy group, which acyloxy group is eliminated on the coupling of said coupler with said oxidation product.

2. A yellow dye image-forming layer containing light-sensitive silver halide emulsion and an uncolored two-equivalent yellow dye-forming coupler of the formula:



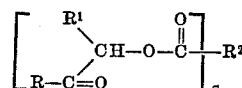
wherein n represents an integer of from 1 to 2; R represents a group selected from the class consisting of an alkyl radical, an aryl radical, and a heterocyclic radical containing a 5 to 6 membered heterocyclic ring; R^1 represents a group selected from the class consisting of a carbamyl radical and the cyano radical; R^2 represents, when n is the integer 1, a group selected from the class consisting of an alkyl radical, an aryl radical, and a heterocyclic radical containing a 5 to 6 membered heterocyclic ring, and, when n is 2, a group selected from the class consisting of an alkylene radical, an arylene radical, and a divalent heterocyclic radical.

3. A yellow dye image-forming layer of claim 2 in which the silver halide is in a gelatin silver halide emulsion.

4. A multilayer, multicolor photographic element containing silver halide and incorporated image dye-forming couplers which are capable of forming dyes on coupling with the oxidation product of a primary aromatic amino

color developing agent, said element containing for the formation of yellow image dye a coupler containing at least one open-chain reactive methine group joined directly to two activating groups, at least one of said activating groups being a carbonyl group, said active methine group also joined directly to an acyloxy group, which acyloxy group is eliminated on the coupling of said coupler with said oxidation product.

5. A multilayer, multicolor photographic element containing silver halide and incorporated image-dye-forming couplers which are capable of forming dyes on coupling with the oxidation product of a primary aromatic amino color developing agent, said element containing for the formation of yellow image dye a coupler of the formula:



wherein n represents an integer of from 1 to 2; R represents a group selected from the class consisting of an alkyl radical, an aryl radical, and a heterocyclic radical containing a 5 to 6 membered heterocyclic ring; R^1 represents a group selected from the class consisting of a carbamyl radical and a cyano radical; R^2 represents, when n is 1, a group selected from the class consisting of an alkyl radical, an aryl radical, and a heterocyclic radical containing a 5 to 6 membered heterocyclic ring, and when n is 2, a group selected from the class consisting of an alkylene radical, an arylene radical, and a divalent heterocyclic radical.

6. An image-forming layer of claim 2 in which the R group of the uncolored two-equivalent coupler formula is an alkyl radical, said layer containing a light-sensitive hydrophilic colloid-silver halide emulsion.

7. An image-forming layer of claim 2 in which the R group of the uncolored two-equivalent coupler formula is a pivalyl radical, said layer containing a light-sensitive hydrophilic colloid-silver halide emulsion.

8. An image-forming layer of claim 2 in which the R group of the uncolored two-equivalent coupler formula is an aryl radical, said layer containing a light-sensitive hydrophilic colloid-silver halide emulsion.

9. An image-forming layer of claim 2 in which the R group of the uncolored two-equivalent coupler formula is a heterocyclic radical, said layer containing a light-sensitive hydrophilic colloid-silver halide emulsion.

10. A light-sensitive hydrophilic colloid-silver halide emulsion layer containing as the sole dye image-former the two-equivalent coupler α -acetoxy- α -{3-[γ -(2,4-di-t-amylphenoxy)butyramido]benzoyl}-2-methoxyacetanilide.

11. A light-sensitive hydrophilic colloid-silver halide emulsion layer containing as the sole dye image-former the two-equivalent coupler α -benzoyl- α -[α -(2,4-di-n-amylphenoxy)acetoxy]-2-methoxyacetanilide.

12. A light-sensitive hydrophilic colloid-silver halide emulsion layer containing as the sole dye image-former the two-equivalent coupler α -pivalyl- α -stearoyloxy-4-sulfamylacetanilide.

13. A light-sensitive hydrophilic colloid-silver halide emulsion layer containing as the sole dye image-former the two-equivalent coupler α -pivalyl- α -[α -(3-pentadecylphenoxy)acetoxy]-3,5-dicarboxyacetanilide.

14. A light-sensitive hydrophilic colloid-silver halide emulsion layer containing as the sole dye image-former the two-equivalent coupler α -acetoxy- α -{3-[α -(2,4-di-t-amylphenoxy)butyramido]benzoyl}-2-methoxyacetanilide.

15. A light-sensitive hydrophilic colloid-silver halide emulsion layer containing as the sole dye image-former

the two-equivalent coupler alpha-(3-dodecanamidoben-
zoyl) alpha-octanoyloxy-2-methoxyacetanilide.

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