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(54) **Silver halide color photographic light-sensitive material.**

(57) A silver halide photographic light-sensitive material which has improved image quality and storage stability is disclosed. The material comprises a support having thereon at least two silver halide light-sensitive emulsion layers, said layers having sensitivity in the same spectral region but different speed from each other, and at least one non-light-sensitive hydrophilic colloid layer, wherein said hydrophilic colloid layer is interposed between a higher speed layer and a lower speed layer of said silver halide light-sensitive emulsion layers, and said higher speed layer has a dry layer thickness of 1.6μ or less and said hydrophilic colloid layer has a dry thickness of 0.2 to 1.5μ.

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Description

SILVER HALIDE COLOR PHOTOGRAPHIC LIGHT-SENSITIVE MATERIAL

FIELD OF THE INVENTION

The present invention relates to a silver halide color photographic light-sensitive material, and, particularly, to a silver halide color photographic light-sensitive material that can achieve an improved image quality and an improved storage stability.

BACKGROUND OF THE INVENTION

With recent spread of miniature cameras (for example, a disc camera and a half-size camera), there is a strong demand for achieving a high image quality of silver halide photographic light-sensitive materials. In particular, the above demand is increasingly strong in silver halide color photographic materials that have become very popular.

The silver halide color photographic materials are formed with plural color-sensitive layers, and on the other hand an effort is made for reducing the thickness of photographic component layers of the light-sensitive materials according to the demand for achieving the high image quality.

With progress of reducing the thickness, an oxidized product of a developing agent may react in a higher speed layer adjacent to a lower speed layer with a coupler present in said higher speed layer and thereafter an excessive oxidized product may diffuse even in the lower speed layer to react with a coupler present in said lower speed layer, causing a lowering of the graininess in the lower speed layer because of the so-called diffusion projection effect, and bringing about a situation in which the image irregularity is caused.

To solve such problems, it is practiced to provide a non-light-sensitive intermediate layer between the above respective layers to prevent the deterioration of the graininess.

However, according to a finding made by the present inventors, there is a fear of the deterioration of sharpness if only the intermediate layer is provided, and thus it does not necessarily follow that the above problems can be settled only by providing the intermediate layer.

SUMMARY OF THE INVENTION

An object of the present invention is to provide a silver halide color photographic light-sensitive material that can have a good graininess and sharpness of an image and have an excellent image quality and development processing stability.

The above object can be achieved by a silver halide color photographic light-sensitive material comprising a support having thereon at least two silver halide emulsion layers having the same color sensitivity and different speed from each other and at least one non-light-sensitive hydrophilic colloid layer, wherein said hydrophilic colloid layer is interposed between a higher speed layer and a lower speed layer of said silver halide emulsion layers, and said higher speed layer has a dry layer thickness of 1.6 μ or less and said hydrophilic colloid layer has a dry layer thickness of 0.2 to 1.5 μ.

DETAILED DESCRIPTION OF THE INVENTION

In the present invention, the non-light-sensitive hydrophilic colloid layer is interposed between a higher speed layer and a lower speed layer of the silver halide emulsion layers.

For the formation of the hydrophilic colloid layer, there is used gelatin, protein, or synthetic hydrophilic polymers, and particularly may preferably be used gelatin. The hydrophilic colloid layer is non-light-sensitive, and being non-light-sensitive is meant to be not sensitive to the exposure when an image is exposed to light or sensitive to light in a negligible degree.

Thus, the presence of such an intermediate layer can effectively prevent the diffusion projection effect originating from the higher speed layer. In order to more enhance the effect of preventing the same, it is possible for said layer to contain a substance capable of reacting an oxidized product diffusing from the higher speed layer into said layer.

In the present invention, the intermediate layer may be interposed between the higher speed layer and the lower speed layer, but may preferably be in direct contact with the higher speed layer and the lower speed layer. Also, the above silver halide emulsion layers are comprised of at least two layers having the same color sensitivity and different speed from each other. The "same color sensitivity" herein used refers to a property of absorbing the light of the wavelength of substantially the same wavelength region. Such color sensitive layers may be of a single color or multi-colors, but preferably of multi-colors. Particularly preferred is a full color type, and in the case of the full color type they are formed with the provision of a blue-sensitive emulsion layer, a green-sensitive emulsion layer and a red-sensitive emulsion layer.

The above color sensitive layers generally include a blue-sensitive layer, a green-sensitive layer and a red-sensitive layer, and when used for the full color type, all of these color sensitive layers are laminated to from the layers. The present invention can be particularly effective in the green-sensitive layer.

Then, the above green-sensitive layer contains a magenta coupler, and as the magenta coupler, there can be preferably used known 5-pyrazolone type couplers, pyrazolobenzimidazole type couplers, pyrazolotriazole type couplers and open chain acylacetonitrile type couplers.

The red-sensitive layer contains a cyan coupler, and as the cyan coupler, there can be preferably used naphthol type couplers and phenol type couplers.

The blue-sensitive layer contains a yellow coupler, and as the yellow coupler, there can be preferably used, for example, acylacetanilide type couplers. Among these, preferred are benzoylacetanilide type and pivaloylacetanilide type compounds.

It is also possible to further promote the reduction of the thickness of the color sensitive layers by newly providing a highly color-forming coupler. As the highly color-forming coupler, various types have been reported, including, for example, the polymer couplers described in Japanese Patent Publication Open to Public Inspection (hereinafter referred to as Japanese Patent O.P.I. Publication) 36249/1984, the pyrazolotriazole type magenta couplers and benzoyl type yellow couplers described Japanese Patent O.P.I. Publication No.246748/1986. Accordingly, it is preferable to use the highly color-forming couplers as a means for reducing the thickness.

The silver halide emulsion layers are further provided with the higher speed layer and the lower speed layer. The terms "higher speed" and "lower speed" herein used are based on a strictly relative evaluation. Thus, in an instance of a three-layer structure in which the layers are formed with high, intermediate and low speed layers, the intermediate speed layer corresponds to the lower speed layer mentioned in the present invention when, for example, the hydrophilic colloid layer is interposed between the respective high speed layer and intermediate speed layer, or the intermediate speed layer corresponds to the higher speed layer mentioned in the present invention when it is interposed between the intermediate speed layer and the low speed layer.

In the present invention, the emulsion layers preferably used, for example, the layers of the three layer structure comprising the high speed layer, the intermediate layer and the low speed layer, may preferably have a difference in the sensitivity between the respectively adjacent layers, generally of 0.1 to 1.0 in log E (E represents an exposure amount). The gradation of an image can be controlled by providing at least two of such layers having the same color sensitivity and different speed from each other.

The control of sensitivity is carried out generally by changing the grain size of silver halide grains contained in the silver halide emulsion layers, and the control of sensitivity in the present invention can be carried out by use of the means like this or any other various means.

In the present invention, the higher sensitivity layer have a dry layer thickness of 1.6 μ or less, preferably 0.5 to 1.5 μ.

The layer thickness of more than 1.6 μ may cause a strong tendency of the scattering of transmitted light in said higher speed layer, resulting in the deterioration of sharpness. On the contrary, an overly thin layer thickness may result in no practical usefulness in relation to silver halide emulsions, additives, etc., giving a lower limit to be necessarily set up. Here, the dry layer thickness refers to a layer thickness measured under the conditions in which the humidity has been controlled to 55 % at 23° C. The layer thickness in the plurality of layers can be determined by taking an enlarged photograph with use of a scanning electron microscope and measuring the layer thickness of the respective layers.

The lower limit of the total of the dry layer thickness of all the hudrophilic colloid layers in the above photographic component layers has the limit as mentioned above depending on the volume held by silver halide emulsion layers, oily agents such as couplers, additives, and binders such as gelatin, and the layer thickness on the emulsion layer side may preferably be in the range of 5 μ to 8 μ, more preferably 10 μ to 16 μ. Also, the thickness from an outermost surface of the emulsion layer side to the bottom end of an emulsion layer nearest to the support may preferably be 14 μ or less, and the thickness therefrom to the bottom end of an emulsion layer having the color sensitivity different from the first-mentioned emulsion layer and near to the support in the next place of the first-mentioned emulsion layer may preferably be 10 μ or less.

A silver halide emulsion forming the above color-sensitive layer will be described below. In the silver halide emulsion, there can be used any of those used as silver halides in usual silver halide emulsion layers, such as silver bromide, silver iodobromide, silver iodochloride, silver chlorobromide and silver chloride, but preferred is an emulsion in which silver iodobromide is used. Silver halide grains used in the silver halide emulsion may be any of those obtained by an acidic method, a neutral method and an ammoniacal method. The grains may be allowed to grow at one time, or grow after seed grains have been formed. The manner to prepare the seed grains and the manner to grow them may be same or different. The silver halide emulsion containing such grains may be obtained by simultaneously mixing halide ions and silver ions, or by preparing an aqueous solution in which either one of them is present and then mixing in it the other of them. Alternatively, taking into account the critical growth rate of silver halide crystals, it may be formed by successively simultaneously adding halide ions and silver ions while controlling pH and pAg in a mixing vessel. According to this method, it is possible to obtain silver halide grains that are regular in crystal form and substantially uniform in grain size. it is also possible to change the silver halide formulation in the grains after growth, with use of a conversion method. With regard to the silver halide emulsion thus obtained, there may be used those having any grain size distribution. Thus, there may be used a emulsion having a broad grain size distribution (called a polydispersed emulsion), or may be used an emulsion having a narrow grain size distribution (called a monodispersed emulsion), which may be used alone or as a mix of several kinds. Alternatively, a mixture of the polydispersed emulsion and monodispersed emulsion may also be used. Preferably used is the monodispersed emulsion.

Here, the "monodispersed" in the monodispersed emulsion is meant to be an emulsion having the variation coefficient in the grain size distribution of the silver halide grains to be contained in the silver halide emulsion, of 22 % or less, preferably 15 % or less. The variation coefficient refers to a coefficient showing the width of

grain size distribution, and can be defined by the formulas shown below.

$$\text{Variation coefficient} = \frac{\text{Standard deviation of grain size distribution}}{\text{Average grain size}} \times 100$$

$$\text{Standard deviation of grain size distribution} = \sqrt{\frac{\Sigma\,(\bar{r} - r_i)^2 n_i}{\Sigma\, n_i}}$$

$$\text{Average grain size} = \frac{\Sigma\, n_i r_i}{\Sigma\, n_i} = \bar{r}$$

Here, ri represents grain size of the respective grains, and ni, the number thereof. The average grain size $\bar{r}$ indicates the length of a side in the case of a cubic silver halide grain, and, in the case of a spherical one, an average value of the length of a side when converted into a cube. When the grain size of the respective grains is ri and the number thereof is ni, the above formulas can be applied.

The above grain size can be measured according to all sort of methods generally used for the above purpose in the present technical field. A typical method is described in "Method for Analysis of Grain Size", A.S.T.S. Symposium on Light Microscopy, 1955, pp.94-122, or Chapter II in "The Theory of The Photographic Process", coauthored by Meas and James, Third Edition, published by Macmillan Publishing Co., Inc. (1966).

In working the present invention, it can be effective for improving the graininess if the "compound that captures an oxidized product of a color developing agent but does not contribute to the image formation" described in Japanese Patent O.P.I. Publication No. 205343/1987 is contained in at least the higher speed layer. Particularly effective as the above compound is a gallic acid derivative. It may be added preferably in an amount of 4.0×10^{-5} mol to 3.5×10^{-2} mol, more preferably 4.0×10^{-4} mol to 1.7×10^{-2} mol, per mol of silver halide contained in an emulsion layer. To add the compound in an emulsion, it may be added by dissolving a given amount of the above compound in an organic solvent such as methanol, or may be added by dispersing it together with couplers or singularly.

In the present invention, the above hydrophilic colloid layer may have a dry layer thickness of 0.2 to 1.5 μ, more preferably 0.3 to 1.0 μ. In the absence of this layer, there can be obtained no action of preventing the diffusion projection effect owing to the higher speed layer, causing a lowering of the graininess of the lower speed layer. The layer thickness less than 0.2μ may result similarly. On the contrary, the layer thickness more than 1.5 μ may cause a lowering of the permeability of the developing agent to deteriorate the developing performance, and also cause a lcwering of the transmittance of light to deteriorate the graininess.

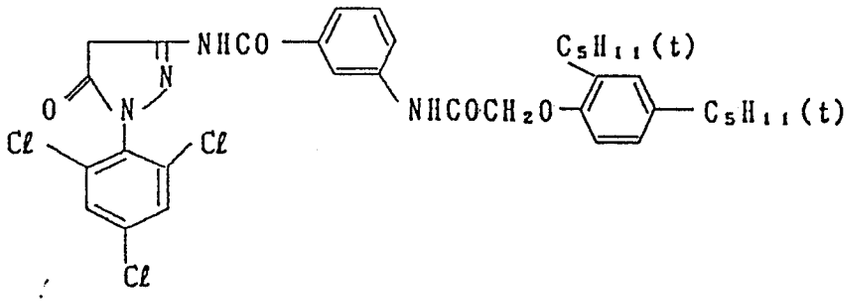
In working the present invention, a diffusible DIR compound can be preferably used. The diffusible DIR compound refers to a compound whose diffusibility of a development inhibitor or of a compound capable of releasing a developing inhibitor, which is capable of being split off from the DIR compound through the reaction with an oxidized product of a color developing agent, is 0.40 or more in terms of the diffusibility according to the evaluation method described below.

The diffusibility can be evaluated according to the following method:

Samples (I) and (II) are prepared.

Sample (I): A sample having a green-sensitive silver halide emulsion layer.

A gelatinous coating solution containing silver iodobromide having been spectrally sensitized to have green sensitivity (silver iodide: 6 mol %; average grain size: 0.48 μm) and 0.07 mol, per mol of silver, of the coupler shown below is coated to have a coated silver amount of 1.1 g/m^2 and a gelatin amount of 3.0 g/m^2, and a gelatinous coating solution containing silver iodobromide having been not chemically sensitized nor spectrally sensitized (silver iodide: 2 mol %; average grain size: 0.08 μm) is coated thereon as a protective layer to have a coated silver amount of 0.1 g/m^2 and a gelatin amount of 0.8 g/m^2.



Sample (II): A sample in which the silver iodobromide has been removed from the protective layer.

In the respective layers, a hardening agent and a surface active agent are contained in addition to the above.

Samples (I) and (II) are exposed to white light with use of a wedge, followed by processing according to the processing method shown below. As developing solutions, used are a solution to which a development inhibitor of every kind is added in the amount sufficient for suppressing the sensitivity of sample (II) to 60 % ($-\Delta$ log E = 0.22 in terms of a logarithmic indication), and a solution to which no development inhibitor is added.

Processing steps (38°C):

Color developing	2 min. 40 sec.
Bleaching	6 min. 30 sec.
Washing	3 min. 15 sec.
Fixing	6 min. 30 sec.
Washing	3 min. 15 sec.
Stabilizing	1 min. 30 sec.
Drying	

Processing solutions used in the respective processing steps have the formulation as follows.

[Color developing solution]

4-Amino-3-methyl-N-ethyl-N-(β-hydroxyethyl)-aniline sulfate	4.75 g
Anhydrous sodium sulfite	4.25 g
Hydroxylamine 1/2 sulfate	2.0 g
Anhydrous potassium carbonate	37.5 g
Sodium bromide	1.3 g
Trisodium nitrilotriacetate (monohydrate)	2.5 g
Potassium hydroxide	1.0 g

Made up to 1 liter by adding water.

[Bleaching solution]

Ferric ammonium ethylenediaminetetraacetate	100.0 g
Diammonium ethylenediaminetetraacetate	10.0 g
Ammonium bromide	150.0 g
Glacial acetic acid	10.0 g

Made up to 1 liter by adding water, and adjusted to pH = 6.0 with use of ammonia water.

[Fixing solution]

Ammonium thiosulfate	175.0 g
Anhydrous sodium sulfite	8.5 g
Sodium metasulfite	2.3 g

Made up to 1 liter by adding water, and adjusted to pH = 6.0 with use of acetic acid.

[Stabilizing solution]

Formalin (a 37 % aqueous solution)	1.5 ml
Konidax (produced by Konishiroku Photo Industry Co., Ltd.)	7.5 ml

Made up to 1 liter by adding water.

Assuming as S_0 the sensitivity of sample (I) when no development inhibitor is added, as S_0' the sensitivity of sample (II), as S_I the sensitivity of sample (I) when the development inhibitor is added, and as S_{II} the sensitivity of sample (II), the following can be represented:

Desensitization degree of sample (I): $\Delta S = S_0 - S_I$

Desensitization degree of sample (II): $\Delta S_0 = S_0' - S_{II}$

Diffusibility = $\Delta S/\Delta S_0$

provided that all the sensitivities are assumed as logarithms of reciprocals (-log E) of the exposure amount at the density point of fog density + 0.3.

The dispersibility determined according to the above method in respect of several kinds of development inhibitors is exemplified in the following table.

Table

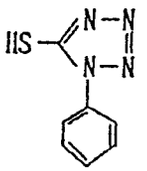
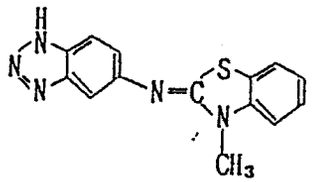
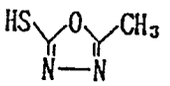
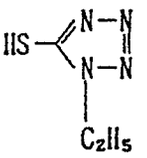
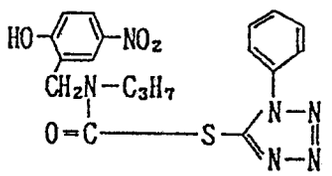
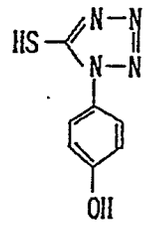
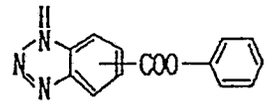
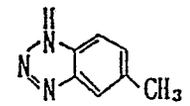
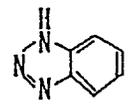
Structure	Amount (mol/ℓ)	Desensitization degree ΔS_0	Desensitization degree ΔS	Diffusibility $\Delta S/\Delta S_0$
	1.3×10^{-5}	0.22	0.05	0.23
	1.3×10^{-5}	0.23	0.08	0.34
	2.5×10^{-5}	0.22	0.10	0.45
	3.0×10^{-5}	0.21	0.10	0.48
	1.4×10^{-5}	0.23	0.11	0.48

Table (Cont'd)

Structure	Amount (mol/ℓ)	Desensitization degree ΔS_0	Desensitization degree ΔS	Diffusibility $\Delta S/\Delta S_0$
	2.5×10^{-5}	0.22	0.13	0.59
	3.5×10^{-5}	0.23	0.15	0.65
	4.3×10^{-5}	0.22	0.16	0.73
	1.7×10^{-4}	0.21	0.20	0.95

As the diffusible DIR compounds used in the present invention, any types of the compound can be used without regard to their chemical structure so long as the diffusibility of the released group is in the above range.

Typical structural formula thereof is shown below.

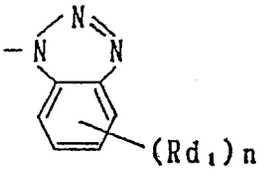
General Formula (D-1)

$A-(Y)_m$

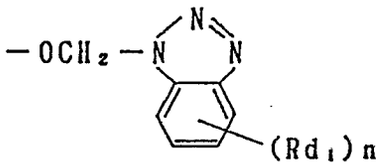
Symbol A represents a coupler residual group; m represents 1 or 2; Y represents a group capable of bonding to the coupling position of the coupler residual group A and capable of being split off through the reaction with an oxidized product of a color developing agent, and is a development inhibiting group or a group capable of releasing a development inhibitor, having 0.40 or more of diffusibility.

In General Formula (D-1), Y is typically represented by any of General Formulas (D-2) to (D-19) shown below.

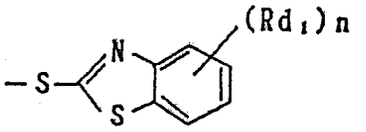
Formula (D-2)



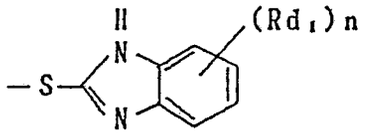
Formula (D-3)



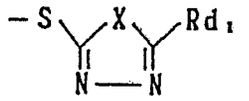
Formula (D-4)



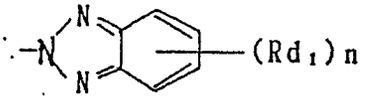
Formula (D-5)



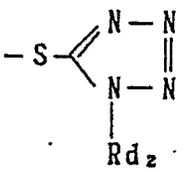
Formula (D-6)



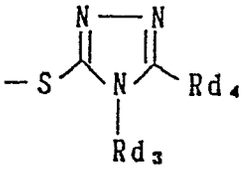
Formula (D-7)



Formula (D-8)



Formula (D-9)



In Formulas (D-2) to (D-7), Rd_1 represents a hydrogen atom, a halogen atom, or an alkyl, alkoxy, acylamino, alkoxycarbonyl, thiazolylideneamino, aryloxycarbonyl, acyloxy, carbamoyl, N-alkylcarbamoyl, N,N-dialkylcarbamoyl, nitro, amino, N-arylcarbamoyloxy, sulfamoyl, N-alkylcarbamoyloxy, hydroxy, alkoxycarbonylamino, alkylthio, arylthio, aryl, heterocyclic, cyano, alkylsulfonyl or aryloxycarbonylamino group; and n represents 0, 1 or 2, and Rd_1 may be the same or different when n is 2. The sum of the number of the carbon atoms contained in n of Rd_{15} is 0 to 10. Also, the number of the carbon atoms contained in Rd_1 in Formula (D-6) is 0 to 15.

In Formula (D-6), X represents an oxygen atom or a sulfur atom.

In Formula (D-8), Rd_2 represents an alkyl group, aryl group or heterocyclic group.

In Formula (D-9), Rd_3 represents a hydrogen atom, or an alkyl, cycloalkyl, aryl or heterocyclic group, Rd_4 represents a hydrogen atom, a halogen atom, or an alkyl, cycloalkyl, aryl, acylamino, alkoxycarbonylamino, aryloxycarbonylamino, alkanesulfonamide, cyano, heterocyclic, alkylthio or amino group.

When Rd_1, Rd_2, Rd_3 or Rd_4 represents an alkyl group, this alkyl group may include those having a substituent and may be any of straight chain or branched alkyls.

When Rd_1, Rd_2, Rd_3 or Rd_4 represents an aryl group, the aryl group may include those having a substituent.

When Rd_1, Rd_2, Rd_3 or Rd_4 represents heterocyclic group, this heterocyclic group may include those having a substituent, and preferably include a single ring or condensed ring of 5 or 6 members containing as a hetero atom at least one selected from a nitrogen atom, an oxygen atom and a sulfur atom, for example, a pyridyl, quinolyl, furyl, benzothiazolyl, oxazolyl, imidazolyl, thiazolyl, triazolyl, benzotriazolyl, imide or oxazine group.

In Formulas (D-6) and (D-8), the number of the carbon atoms contained in Rd_2 is 0 to 15.

In the above Formula (D-9), the sum of the number of the carbon atoms contained in Rd_3 and Rd_4 is 0 to 15.

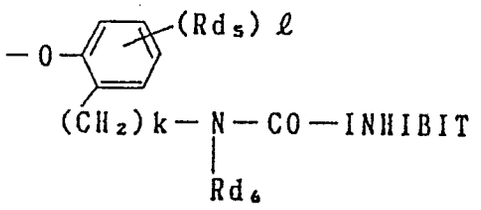
Formula (D-10)

-TIME-INHIBIT

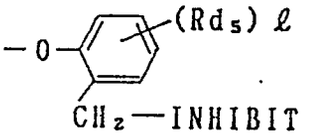
In the formula, -TIME- represents a group capable of entering into combination with the coupling position of A and undergoing cleavage through the reaction with an oxidized product of a color developing agent, and is a group capable of releasing an -INHIBIT group with appropriate control, after cleavage from a coupler. The -INHIBIT group is a group formed into a development inhibiter by the above releasing (e.g. the group represented by any of the above Formulas (D-2) to (D-9)).

In Formula (D-10), the -TIME-INHIBIT- group is typically represented by any of Formulas (D-11) to (D-19) shown below.

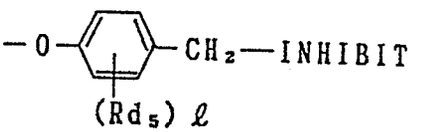
Formula (D-11)



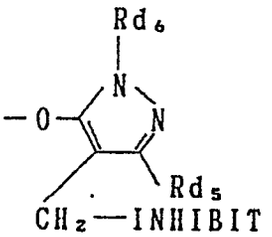
Formula (D-12)



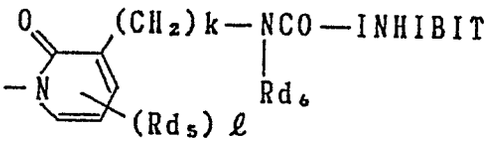
Formula (D-13)



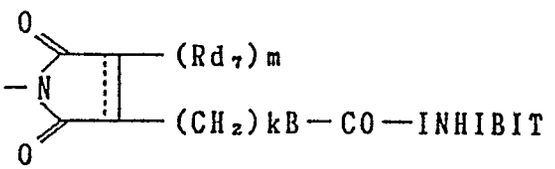
Formula (D-14)



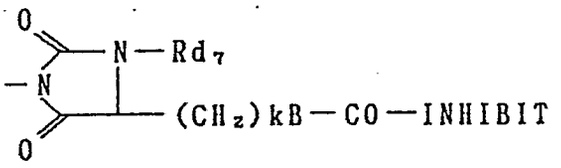
Formula (D-15)



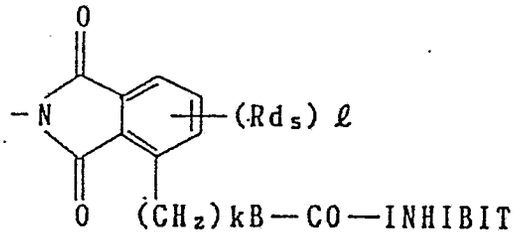
Formula (D-16)



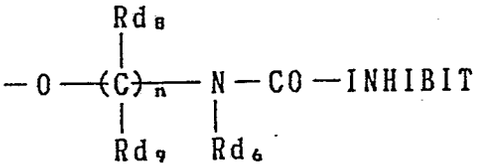
Formula (D-17)



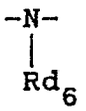
Formula (D-18)



Formula (D-19)



In Formulas (D-11) to (D-15) and (D-18), Rd_5 represents a hydrogen atom, or an alkyl, cycloalkyl, alkenyl, aralkyl, alkoxy, alkoxycarbonyl, anilino, acylamino, ureido, cyano, nitro, sulfonamide, sulfamoyl, carbamoyl, aryl, carboxy, sulfo, hydroxyl or alkanesulfonyl group. In Formulas (D-11) to (D-13), (D-15) and (D-18), Rd_5 may each other combine to form a condensed ring. In Formulas (D-11), (D-14), (D-15) and (D-19), Rd_6 represents an alkyl, alkenyl, aralkyl, cycloalkyl, heterocyclic or aryl group. In Formulas (D- 16) and (D-17), Rd_7 represents a hydrogen atom, or an alkyl, alkenyl, aralkyl, cycloalkyl, heterocyclic or aryl group. Rd_8 and Rd_9 in Formula (D-19) each represent a hydrogen atom or an alkyl group (preferably an alkyl group having 1 to 4 carbon atoms); k in Formulas (D-11) and (D-15) to (D-18) represents an integer of 1 or 2; in Formulas (D-11) to (D-13), (D-15) and (D-18) represents an integer of 1 to 4; m in Formula (D-16) represents an integer of 1 or 2, and the respective Rd_7 may be the same or different when m is 2; n in Formula (D-19) represents an integer of 2 to 4, and Rd_8 and Rd_9 in the number of n may each be the same or different; B in Formulas (D-16) to (D-18) represents an oxygen atom or



(Rd_6 has the same meaning as defined above); ..... in Formula (D-16) indicates that the bond may be either a single bond or a double bond, wherein m is 2 when it is a single bond, and m is 1 when it is a double bond; and INHIBIT has the same meaning as the general formulas defined for Formulas (D-2) to (D-9) except for the carbon atom number.

In the INHIBIT group, the sum of the number of carbon atoms contained in Rd_1 in Formulas (D-2) to (D-7) is 0 to 32, that of the number of the carbon atoms contained in Rd_2 in Formula (D-8) is 1 to 32, and that of the number of the carbon atoms contained in Rd_3 and Rd_4 in Formula (D-9) is 0 to 32.

When Rd_5, Rd_6 and Rd_7 each represent an alkyl group, an aryl group or a cycloalkyl group, they may include those having a substituent.

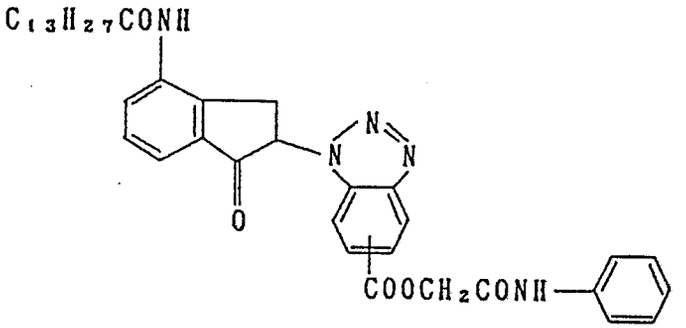
Among the diffusible DIR compound, preferred are those wherein Y is represented by Formula (D-2), (D-3) or (D-10). Among (D-10), preferred are those wherein INHIBIT is represented by Formulas (D-2), (D-6) (particularly when X in Formula (D-6) is an oxygen atom) or (D-8) (particularly when Rd_2 in Formula (D-8) is a hydroxyl aryl group or an alkyl group having 1 to 3 carbon atoms).

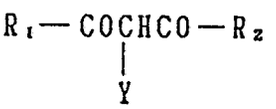
The coupler component represented by A in Formula (D-1) may include a yellow dye image-forming coupler residual group, a magenta dye image-forming coupler residual group, a cyan dye image-forming coupler residual group, and non-coloring coupler residual group.

Examples of the diffusible DIR compound used in working the present invention include the compounds as shown below, but by no means limited to these.

Exemplary compounds:

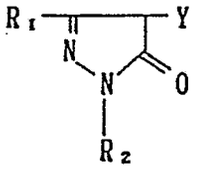
D'-1



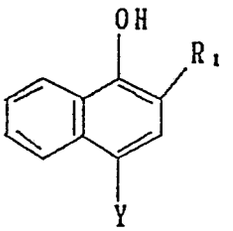


Compound No.	R_1	R_2	Y
D'-2	(1)	(1)	(30)
D'-3	(2)	(3)	(30)
D'-4	(2)	(4)	(30)
D'-5	(5)	(6)	(31)
D'-6	(2)	(4)	(32)
D'-7	(2)	(3)	(32)
D'-8	(7)	(8)	(33)

The groups are specified by the number in the parentheses. The groups corresponding to the respective numbers are listed at the end of this exemplary compound, hereinafter having the same meaning in specifying the exemplary compounds.

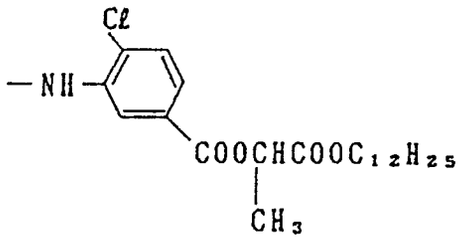


Compound No.	R_1	R_2	Y
D′ − 9	(9)	(10)	(30)
D′ −10	(11)	(10)	(30)
D′ −11	(12)	(7)	(34)
D′ −12	(12)	(13)	(35)
D′ −13	(9)	(14)	(36)
D′ −14	(15)	(16)	(37)



Compound No.	R_1	Y
D′ −15	(17)	(38)
D′ −16	(17)	(39)
D′ −17	(18)	(40)
D′ −18	(20)	(41)
D′ −19	(18)	(42)
D′ −20	(18)	(43)
D′ −21	(18)	(44)
D′ −22	(18)	(45)
D′ −23	(19)	(46)
D′ −24	(21)	(47)
D′ −25	(21)	(48)
D′ −26	(22)	(49)
D′ −27	(22)	(50)
D′ −28	(22)	(51)
D′ −29	(23)	(52)
D′ −30	(18)	(53)
D′ −31	(18)	(54)
D′ −32	(23)	(49)

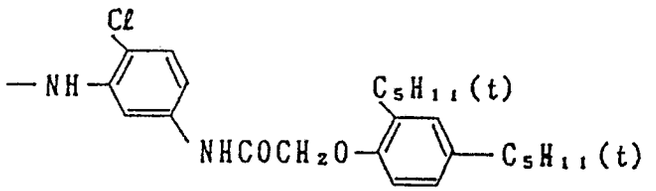
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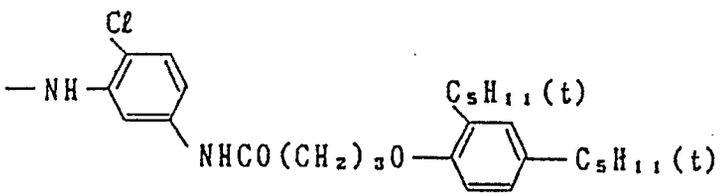
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$-C(CH_3)_3$,

3



4



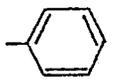
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$-OCH_3$,

6

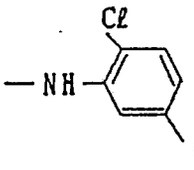


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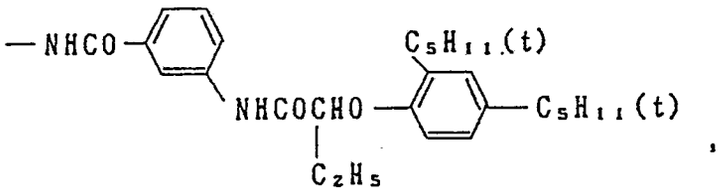
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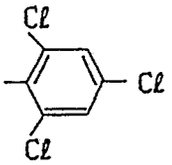
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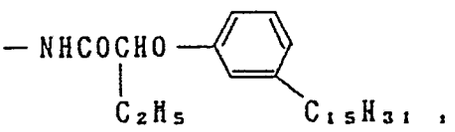
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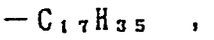
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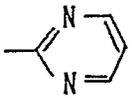
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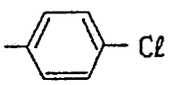
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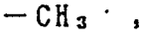
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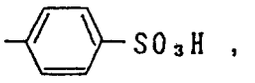


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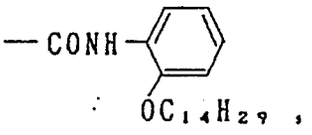
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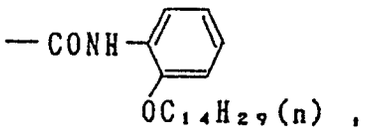


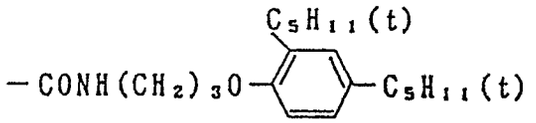
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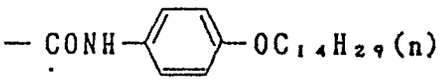
16 

17 $-CONHC_{18}H_{37}$,

18 

19 

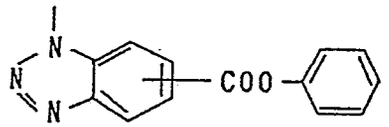
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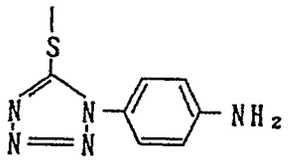
22 $-CONHCH_2CH_2COOH$

23 $-CONHCH_2CH_2COOCH_3$

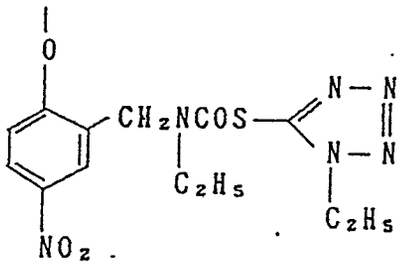
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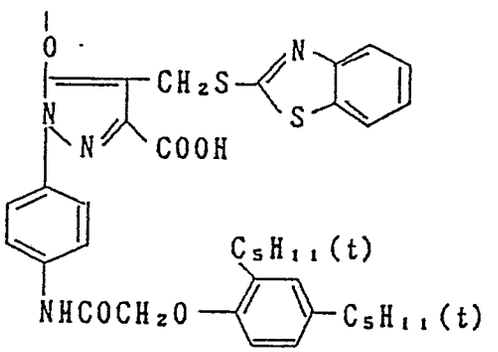
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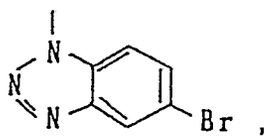
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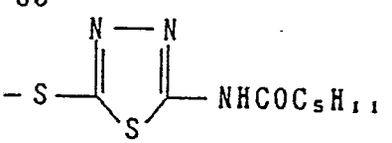
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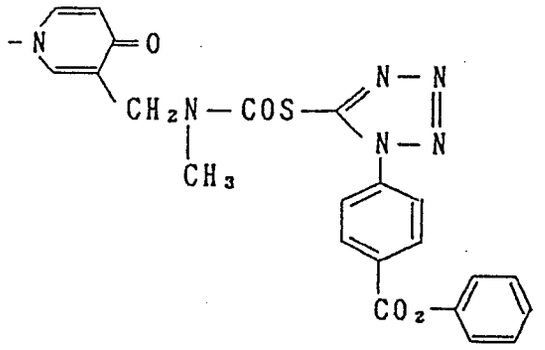
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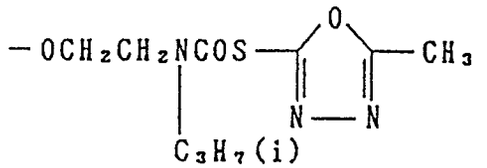
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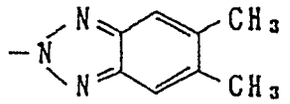
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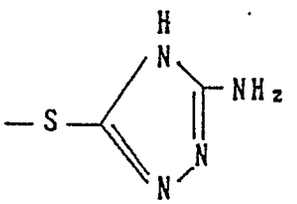
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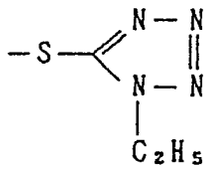
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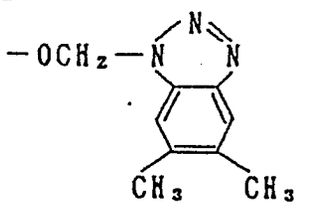
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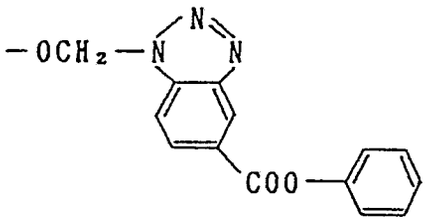
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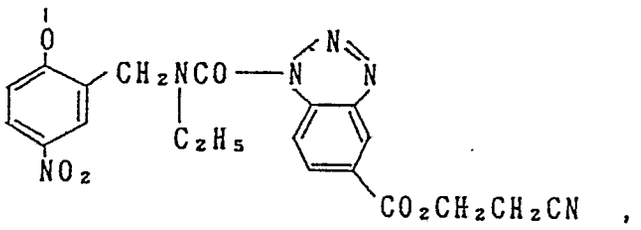
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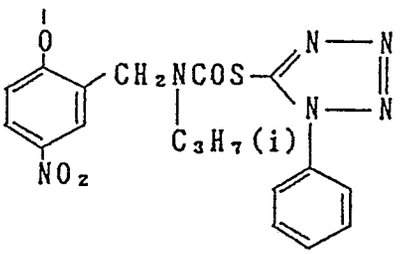
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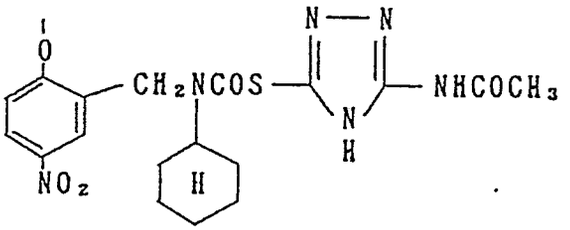
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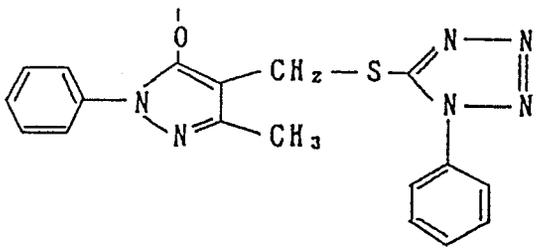
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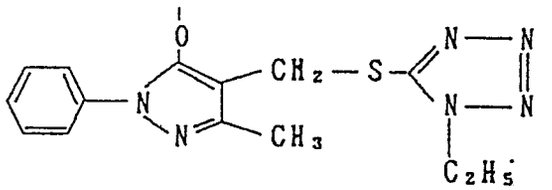
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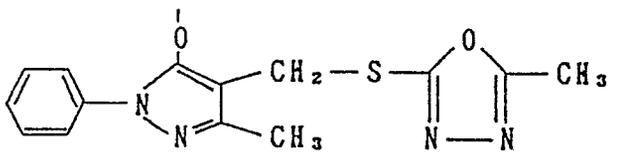
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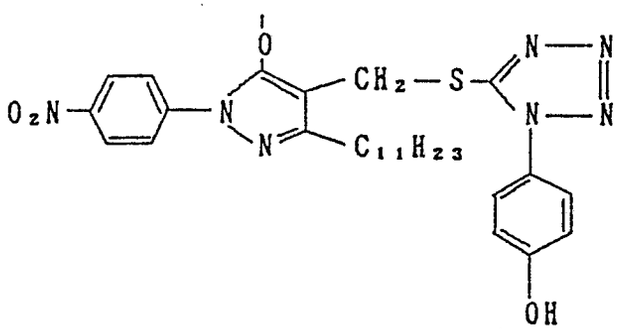
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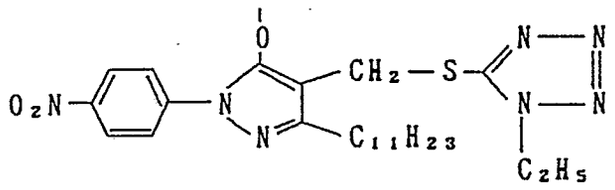
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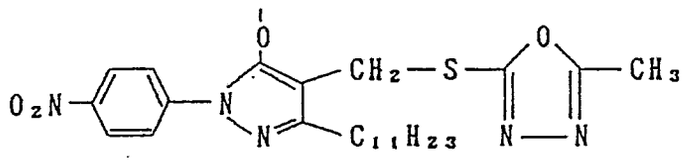
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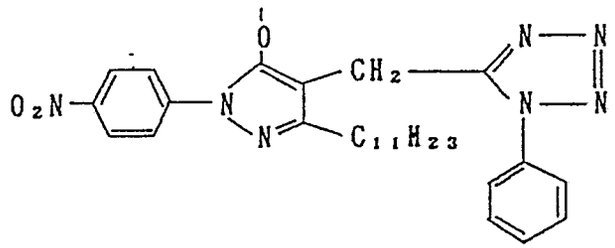
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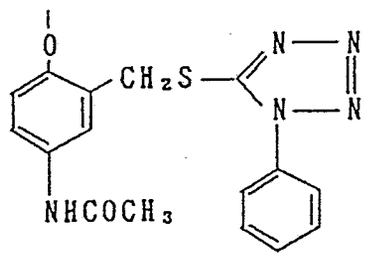
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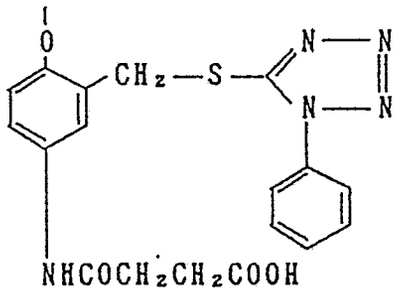
52



53



54



Including these, specific examples of the diffusible DIR compound that can be used in the present invention are described in U.S. Patents No. 4,234,678, No. 3,227,554, No. 3,617,291, No. 3,958,993, No. 4,149,886 and No. 3,933,500, Japanese Patent O.P.I. Publications No. 56837/1982 and No. 13239/1976, U.S. Patents No. 2,072,363 and No. 2,070,266, Research Disclosure No. 21228 of December, 1981, etc.

Various additives can be added in the photographic component layers formed by the above emulsions and so forth, and all kinds of photographic additives such as a wetting agent, a film property improver and a coating auxiliary can be also added depending on the purpose. As other photographic additives, there can be further used a plasticizer, a surface active agent, an ultraviolet absorbent, a pH adjuster, an antioxidant, an antistatic agent, a thickening agent, a graininess improver, a dye, a mordant, a brightening agent, a development speed regulator, a matting agent, and so forth.

Also, in order to prevent the color fading owing to active light with short wavelength, of a dye image, it is useful to use the ultraviolet absorbent, for example, thiazolidone, benzotriazole, acrylonitrile or benzophenone compounds.

In the silver halide emulsion layers used in the above light-sensitive layers, a suitable gelatin derivative can be used as a protective colloid or binding agent (binder) depending on the purpose, in additon to gelatin, and other hydrophilic binding agent (binder) can be also contained depending on the purpose. They can be added in the photographic component layers such as an emulsion layer, an intermediate layer, a protective layer, a filter layer and a subbing layer in the above light-sensitive photographic material, and the above hydrophilic binder may further contain a suitable plasticizer or wetting agent depending on the purpose.

The photographic component layers of the above light-sensitive material can also be hardened with use of a suitable hardening agent.

The silver halide color photographic light-sensitive material according to the present invention is particularly

suited for a negative light-sensitive photographic material.

EXAMPLES

Specific examples of the present invention will be described below, but working embodiments of the present invention are by no means limited to these.

In all examples shown below, the amount for the addition in the silver halide photographic light-sensitive material indicates an amount per 1 m^2, unless particularly mentioned. Silver halide and colloidal silver are indicated in terms of silver.

On a triacetyl cellulose film support, layers having the composition as shown below were formed in sequence from the support side to produce multi-color photographic elements, samples Nos. 1 to 18

Sample No. 1 (Comparative example)

First layer:

Anti-alation layer (HC-1) (1.1 μm).

A gelatin layer containing black colloidal silver.

Second layer:

Intermediate layer (I.L.) (0.8 μm)

A gelatin layer containing an emulsification dispersion of 2,5-di-t-octyl hydroquinone.

Third layer:

Low speed red-sensitive silver halide emulsion layer (RL-1) (2.2 μm)

A monodispersed emulsion (emulsion I) comprising AgBrI containing 6 mol % of AgI and having an average grain size ($\bar{r}$) of 0.40 μm .....

1.8 g/m^2 in coated silver amount

Sensitizing dye I .....

5.0×10^{-4} mol per mol of silver

Sensitizing dye II .....

0.8×10^{-4} mol per mol of silver

Cyan coupler (C-1) ..... 0.05 mol per mol of silver

Colored cyan coupler (CC-1) .....

0.005 mol per mol of silver

DIR compound (A) ..... 0.0015 mol per mol of silver

DIR compound (D'-25) ... 0.002 mol per mol of silver

Fourth layer:

High speed red-sensitive silver halide emulsion layer (RH-1) (1.8 μm)

A monodispersed emulsion (emulsion II) comprising AgBrI containing 6 mol % of AgI and having an average grain size ($\bar{r}$) of 0.8 μm .....

1.3 g/m^2 in coated silver amount

Sensitizing dye I .....

2.5×10^{-4} mol per mol of silver

Sensitizing dye II .....

1.0×10^{-4} mol per mol of silver

Cyan coupler (C-2) ..... 0.07 mol per mol of silver

Cyan coupler (C-3) ..... 0.027 mol per mol of silver

Colored cyan coupler (CC-1) .....

0.0015 mol per mol of silver

Fifth layer:

Intermediate layer (I.L.) (0.8 μm)

A gelatin layer same as the second layer.

Sixth layer:

Low speed green-sensitive silver halide emulsion layer (GL-1) (3.2 μm)

Emulsion I ..... 1.5 g/m^2 in coated silver amount

Sensitizing dye III .....

1.5×10^{-4} mol per mol of silver

Sensitizing dye IV .....

1.2×10^{-4} mol per mol of silver

Magenta coupler (M-1) ...0.062 mol per mol of silver

Colored magenta coupler (CM-1) .....

0.004 mol per mol of silver

DIR compound (D'-32) .....

0.003 mol per mol of silver

Seventh layer:

An intermediate layer (DCL).

A gelatin layer containing an emulsification dispersion of 2,5-di-t-octyl hydroquinone and having a dry layer thickness as shown in Table 1 below.

Eighth layer:

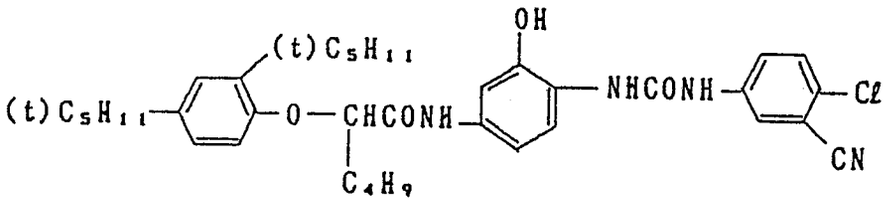
High speed green-sensitive silver halide emulsion layer (GH-1) having a dry layer thickness as shown in Table 1 below.
Emulsion II ..... 1.5 g/m^2 in coated silver amount
Sensitizing dye III .....
1.2 x 10^{-4} mol per mol of silver
Sensitizing dye IV .....
0.8 x 10^{-4} mol per mol of silver
Magenta coupler (M-1) ...0.015 mol per mol of silver
Colored magenta coupler (CM-1) .....
0.002 mol per mol of silver
Ninth layer:
Yellow filter layer (YC-1)
A gelatin layer containing yellow colloidal silver and an emulsification dispersion of 2,5-di-t-octyl hydroquinone.
Tenth layer:
Low speed blue-sensitive silver halide emulsion layer (BL-1)
A monodispersed emulsion (emulsion III) comprising AgBrI containing 4 mol % of AgI and having an average grain size of 0.48 μm .....
0.9 g/m^2 in coated silver amount
Sensitizing dye V .....
1.3 x 10^{-4} mol per mol of silver
Yellow coupler (Y-1) ... 0.18 mol per mol of silver
DIR compound (D'-25) .....
0.003 mol per mol of silver
Eleventh layer:
High speed blue-sensitive silver halide emulsion layer (BH-1)
A monodispersed emulsion (emulsion IV) comprising AgBrI containinging 7 mol % of AgI and having an average grain size of 0.8 μm .....
Emulsion IV ..... 0.5 g/m^2 in coated silver amount
Sensitizing dye V .....
1.0 x 10^{-4} mol per mol of silver
Yellow coupler (Y-1) ...0.08 mol per mol of silver
Twelfth layer:
First protective layer (Pro-1)
A gelatin layer containing:
silver iodobromide (AgI: 1 mol %, average grain size: 0.07 μm) .....
0.5 g/m^2 in coated silver amount; and
ultraviolet absorbents UV-1 and UV-2.
Thirteenth layer:
Second protective layer (Pro-2)
A gelatin layer containing polymethyl methacrylate particles (diameter: 1.5 μm) and formalin scavenger (HS-1).

In the respective layers, gelatin-hardening agents (H-1) and (H-2) and a surface active agent were added in addition to the above composition.

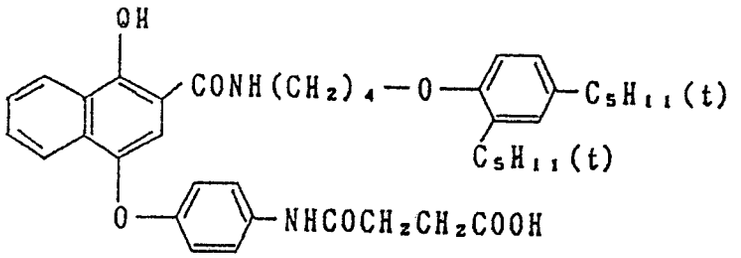
The compounds contained in the respective layers of sample 1 are as follows:

Sensitizing dye I: Anhydro-5,5'-dichloro-9-ethyl-3,3'-di-(3-sulfopropyl)thiacarbocyanine hydroxide
Sensitizing dye II: Anhydro-9-ethyl-3,3'-di-(3-sulfopropyl)-4,5,4',5'-dipenzothiacarbocyanine hydroxide
Sensitizing dye III: Anhydro-5,5'-diphenyl-9-ethyl-3,3'-di-(3-sulfopropyl)oxacarbocyanine hydroxide
Sensitizing dye IV: Anhydro-9-ethyl-3,3'-di-(3-sulfopropyl)-5,6,5',6'-dibenzoxacarbocyamine hydroxide
Sensitizing dye V: Anhydro-3,3'-di-(3-sulfopropyl)-4,5-benzo-5'-methoxythiacyanine hydroxide

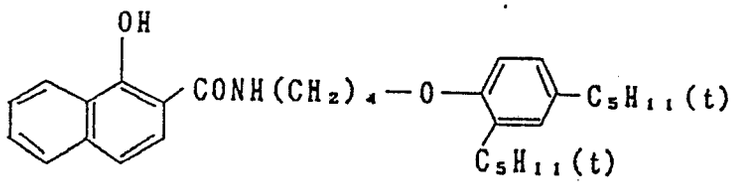
C − 1


(t)C5H11
OH
(t)C5H11
O—CHCONH
NHCONH
Cℓ
CN
C4H9

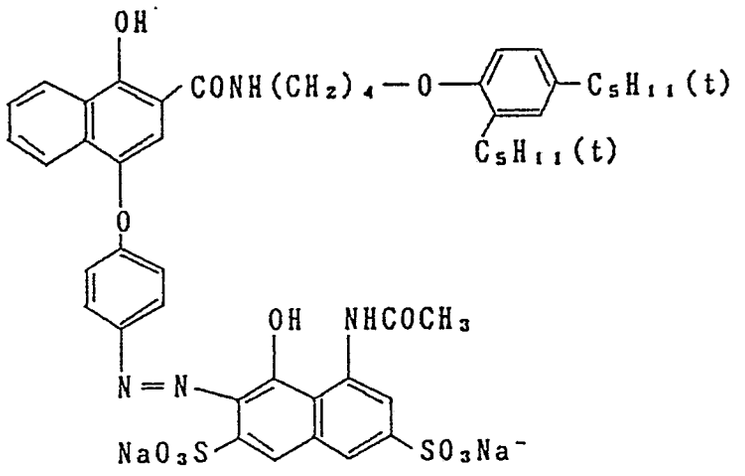
C − 2


OH
CONH(CH2)4—O
C5H11(t)
C5H11(t)
O
NHCOCH2CH2COOH

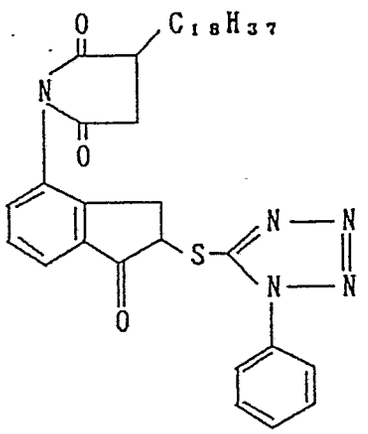
C − 3


OH
CONH(CH2)4—O
C5H11(t)
C5H11(t)

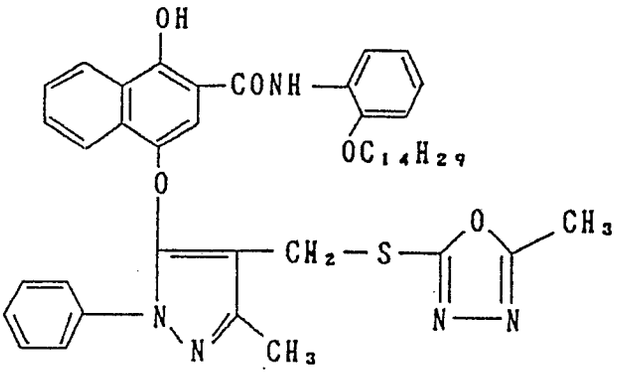
CC－1



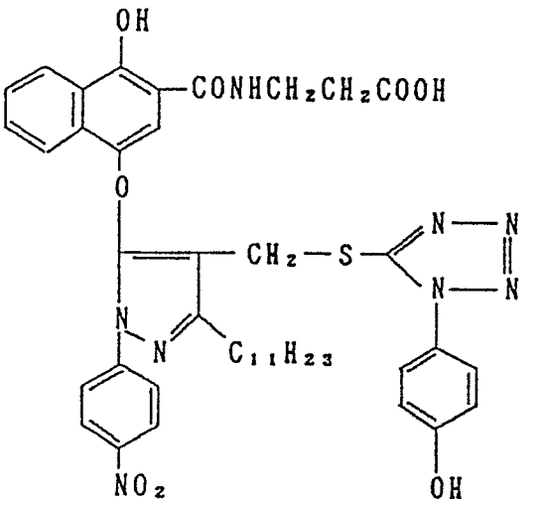
A



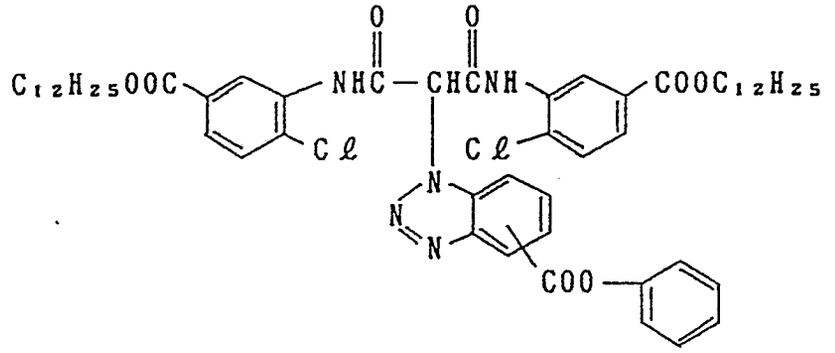
D′ −25



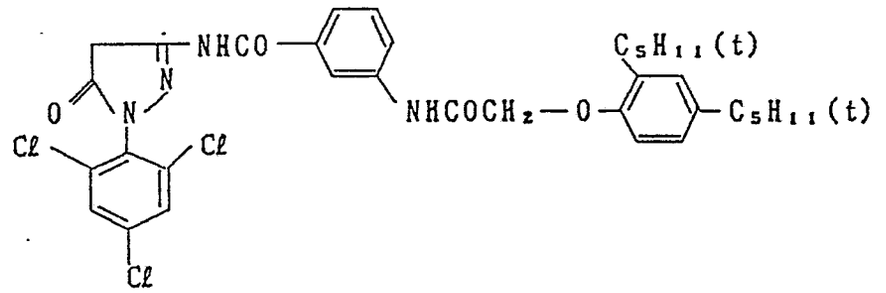
D′ −32



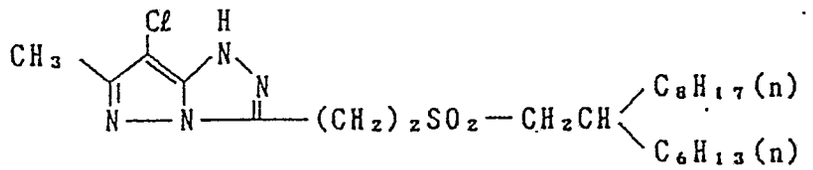
D ' − 4



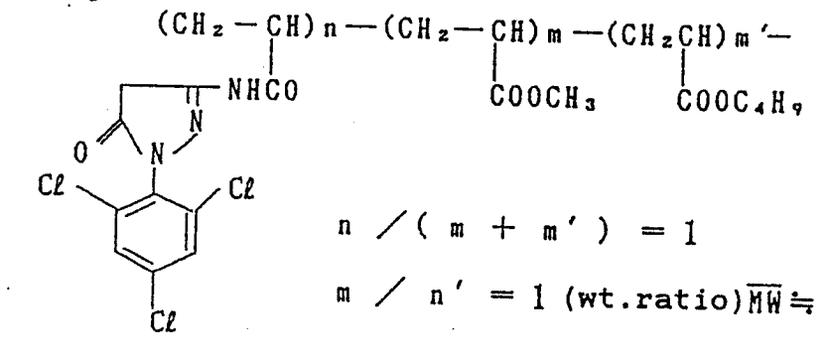
M − 1



M − 2



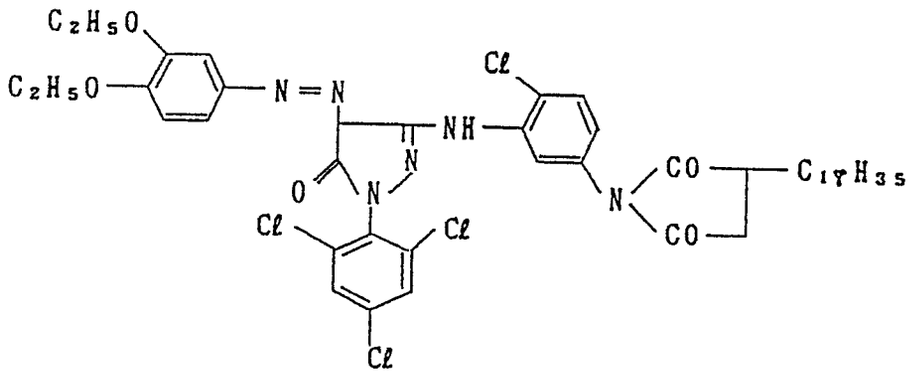
M − 3



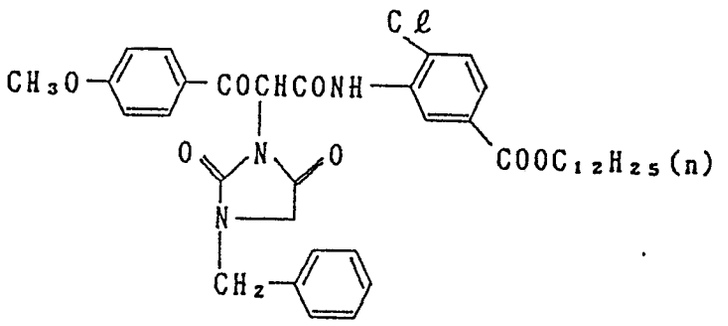
n / (m + m') = 1

m / n' = 1 (wt.ratio) $\overline{MW}$ ≒ 3000

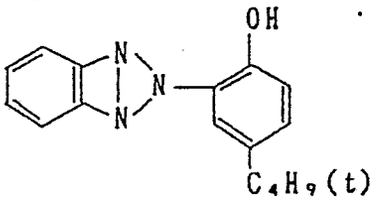
C M − 1



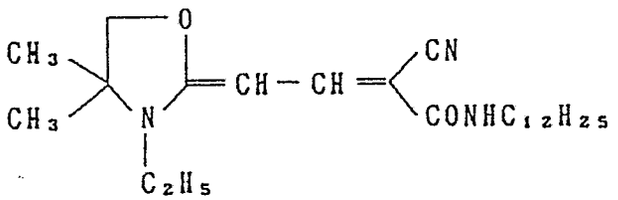
Y − 1



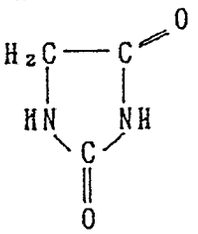
UV－1



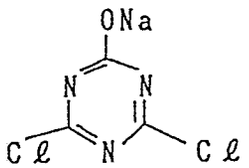
UV－2



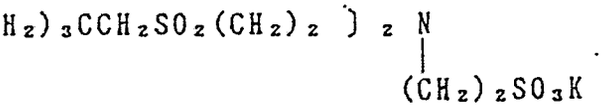
HS－1



H－1



H－2



The respective samples Nos. 1 to 18 were exposed to light with use of white light, followed by development processing according to the following:

Processing steps (38°C):

Color developing	3 min. 15 sec.
Bleaching	6 min. 30 sec.
Washing	3 min. 15 sec.
Fixing	6 min. 30 sec.
Washing	3 min. 15 sec.
Stabilizing	1 min. 30 sec.
Drying	

Processing solutions used in the respective processing steps had the formulation as follows.

[Color developing solution]

4-Amino-3-methyl-N-ethyl-N-(β-hydroxyethyl)-aniline sulfate	4.75 g
Anhydrous sodium sulfite	4.25 g
Hydroxylamine 1/2 sulfate	2.0 g
Anhydrous potassium carbonate	37.5 g
Sodium bromide	1.3 g

Trisodium nitrilotriacetate (monohydrate) 2.5 g
Potassium hydroxide 1.0 g
Made up to 1 liter by adding water.

[Bleaching solution]
Ferric ammonium ethylenediaminetetraacetate 100.0 g
Diammonium ethylenediaminetetraacetate 10.0 g
Ammonium bromide 150.0 g
Glacial acetic acid 10.0 g
Made up to 1 liter by adding water, and adjusted to pH = 6.0 with use of ammonia water.

[Fixing solution]
Ammonium thiosulfate 175.0 g
Anhydrous sodium sulfite 8.5 g
Sodium metasulfite 2.3 g
Made up to 1 liter by adding water, and adjusted to pH = 6.0 with use of acetic acid.

[Stabilizing solution]
Formalin (a 37 % aqueous solution) 1.5 ml
Konidax (produced by Konishiroku Photo Industry Co., Ltd.) 7.5 ml
Made up to 1 liter by adding water.

After the above processing was carried out, the sharpness and graininess of the images obtained in the above respective samples were measured to obtain the results as shown in Table 2.

For the measurement, exposure was made with use of white light, and the sharpness (MTF) and granularity (RMS) of the above green-sensitive layer was measured with use of green light.

Obtaining MTF (modulation transfer function) of a dye image, the effect of improving the sharpness of samples Nos. 1 to 18 was indicated in terms of a relative value (assuming as 100 the value for sample No. 1) of the MTF at 10 lines/mm.

The RMS value was indicated in terms of a 1,000 time value of the standard deviation of the variation in density values caused when the density at minimum density + 1.0 was scanned with a microdensitometer having an open scanning area of 250 μm^2.

Table 1

Sample No.	High speed layer thickness(μ)	DCL layer thickness (μ)	Graininess (RMS relative value)	Sharpness (MTF relative value)	Outside or inside the present invention
1	1.2	–	100	100	Outside
2	0.8	0.3	79	99	Inside
3	0.8	0.8	68	99	Inside
4	0.8	1.5	81	98	Inside
5	0.8	2.0	84	85	Outside
6	1.2	0.3	81	99	Inside
7	1.2	0.8	70	98	Inside
8	1.2	1.5	82	97	Inside
9	1.2	2.0	84	83	Outside
10	1.5	0.3	85	98	Inside
11	1.5	0.8	74	97	Inside
12	1.5	1.5	85	95	Inside
13	1.5	2.0	88	80	Outside
14	2.0	0.8	80	80	Outside
15	2.0	2.0	93	78	Outside
*16	1.2	–	104	100	Outside
*17	0.8	0.8	75	99	Inside
*18	1.2	0.8	73	98	Inside

In the above Table, the samples other than those marked with "*" are those in which 8.5×10^{-4} mol of gallic acid ester per mol of silver was added by dispersing it simultaneously with the couplers.

The above Table 1 tells the following:

(1) Comparing sample No. 14 in which the dry layer thickness of the high speed layer is greater than the range of the present invention and an intermediate layer (DCL layer in the table) was interposed, with sample No. 3 according to the present invention and provided with an intermediate layer of the same type, the sharpenss, though improved a little more than sample No. 1 having no intermediate layer, is poorer than that of sample No. 3. Also, sample No. 14 is greatly poorer than sample No. 3 in the sharpness. The reason why the sharpness is poor is presumably that there is a great influence by the scattering of light in the thicker high speed layer. This tendency is observed also in samples Nos. 7 and 11.

(2) Comparing comparative sample No. 1 with samples Nos. 6 to 8 produced in the same manner as sample No. 1 except for the interposition of the intermediate layer, it is understood that all of samples Nos. 6 to 8 show better graininess than that of sample No. 1 and sufficiently secure the sharpness.

(3) Next, when the dry layer thickness of the intermediate layer is varied from 0.3 μ to 1.5 μ while making constant the dry layer thickness of the high speed layer within the range of the present invention, the graininess at 0.8 μ is found to be best.

(4) When the thickness of the intermediate layer becomes greater than the range of the present invention, it is understood that as will be clear also from samples Nos. 5, 9 and 13, the sharpness is poorer than sample No. 1 although the graininess is improved. Accordingly, it is meant that the layer thickness must be in the range of the present invention in order to keep the sharpness.

(5) Comparing sample No. 3 with sample No. 17 for instances in which the gallic acid ester is added or not in the emulsion layer, the graininess is seen to be further improved when the gallic acid ester is added.

Example 2

Next, tests were carried out on the storage stability and developing performance of the silver halide color photographic light-sensitive materials. For carrying out them, used were samples shown in Table 2 below among those used in Example 1.

The storage stability was evaluated by the sensitivity (S) and gamma value (γ) of the images obtained by the above respective samples, after unexposed light-sensitive materials were stored for 3 days in an environment of a temperature of 50°C and a relative humidity of 80 %.

On the other hand, the developing performance was evaluated by the sensitivity (S) and gamma value (γ) of the above respective samples, obtained when processed with use of respective developing solutions, using color developing solutions whose pH was set to 9.90 and 10.20.

Table 2

Sample No.	High speed layer thickness (μ)	DCL layer thickness (μ)	After 3 days storage, 50°C, 80 % RH		Developing solution pH change				*(A)
					pH=9.90		pH=10.20		
			S		S		S		
1	1.2	-	83	91	86	83	120	117	X
6	1.2	0.3	91	94	91	93	107	110	Y
7	1.2	0.8	94	97	94	96	104	107	Y
8	1.2	1.5	94	95	93	95	104	105	Y
9	1.2	2.0	86	91	88	85	115	115	X
11	1.5	0.8	91	96	92	93	105	109	Y
12	1.5	1.5	92	94	92	93	106	106	Y
13	1.5	2.0	85	90	87	85	117	116	X
14	2.0	0.8	84	93	87	85	119	115	X
15	2.0	2.0	82	86	83	83	122	120	X
18	1.2	0.8	94	97	92	93	107	109	Y

*(A): Inside or outside the present invention

X : Outside

Y : Inside

The results shown in the above Table 2 tell the following:

(1) Comparing sample No. 14 in which the high speed layer has a dry layer thickness greater than the range of the present invention with samples Nos. 7 and 11 each provided with an intermediate layer same as in sample No. 14 having the intermediate layer interposed (DCL layer in the table), sample No. 14 having a greater layer thickness shows the sensitivity (S) and gamma value (γ) both greater in the amount of change than samples Nos. 7 and 11, assuming as 100 the sensitivity (S) and gamma value (γ) observed before storage. Thus sample No. 14 is seen to have poorer storage stability, particularly showing great deterioration in the sensitivity (S).

On the other hand, reviewing the developing performance assuming the sensitivity (S) and gamma value (γ) as 100 when pH = 10.05 was regarded as a standard, both samples No. 7 and 11 show not so great variation quantity as the comparative sample No. 14, though suffering the variation by the influence of pH, and thus they are provided to be stable.

(2) Reviewing the influence by the intermediate layer, comparing comparative sample No. 1 having no intermediate layer interposed and samples Nos. 6 to 8 of the present invention, having the intermediate layer interposed and comprising the high speed layer having the same layer thickness as said sample No. 1, there is shown the same tendency as in the above (1), and the above sample No. 1 shows particularly great variation quantity in the sensitivity (S).

On the other hand, also in respect of the developing performance, the above sample No. 1 shows great variation, but samples Nos. 6 to 8 show a small variation quantity and proved to be stable.

Reviewing also the influence by the change in the layer thickness of the intermediate layer in respect of samples Nos. 6 to 8 of the present invention, both the storage stability and developing performance do not change so much depending on the change in the layer thickness, and they are seen to be stable in

both of these.

It is further recognized, as will be clear from comparative samples Nos. 9 and 13, that if the layer thickness of the intermediate layer is greater than the range of the present invention, they show poor storage stability and tend to be affected by the pH value of the developing solution, proving to be unstable, even if the layer thickness of the high speed layer is in the range of the present invention.

(3) Next, reviewing samples Nos. 7, 11 and 14 in which the layer thickness of the intermediate layer was made constant (0.8 μ) and the layer thickness of the high speed layer was varied, it is seen that the samples Nos. 7 and 11 of the present invention are good in both the storage stability and processing performance, but comparative sample No. 14 is poorer in the both than samples Nos. 7 and 11. Accordingly, it is understood that the layer thickness of the high speed layer must be 1.5 μ or less.

(4) In sample No. 15 in which the layer thickness of both the high speed layer and intermediate layer was made to be greater than the range of the present invention, the storage stability and processing performance are seen to have been worsened.

As described in the foregoing, it is possible according to the present invention to obtain a silver halide color photographic light-sensitive material having good graininess and sharpness of an image and also having good storage stability and development processing stability.

Claims

1. A silver halide photographic light-sensitive material comprising a support having thereon at least two silver halide light-sensitive emulsion layers, said layer having sensitivity in the same spectral region but different speed from each other, and at least one non-light-sensitive hydrophilic colloid layer, wherein said hydrophilic colloid layer is interposed between higher speed layer and a lower speed layer of said silver halide light-sensitive emulsion layers, and said higher speed layer has a dry layer thickness of 1.6μ or less and said hydrophilic colloid layer has a dry thickness of 0.2 to 1.5μ.

2. The silver halide photographic light-sensitive material of claim 1, wherein said hydrophilic colloid layer is directly in contact with said higher speed layer and said lower speed layer.

3. The silver halide photographic light-sensitive material of claim 1, wherein said silver halide light-sensitive emulsion layer is blue-sensitive, green-sensitive or red-sensitive.

4. The silver halide photographic light-sensitive material of claim 1, wherein said silver halide light-sensitive emulsion layer is green-sensitive.

5. The silver halide photographic light-sensitive material of claim 4, wherein said silver halide light-sensitive emulsion layer contains a dye-forming coupler.

6. The silver halide photographic light-sensitive material of claim 4, wherein said silver halide light-sensitive emulsion layer contains a magenta dye-forming coupler.

7. The silver halide photographic light-sensitive material of claim 1, wherein difference in speed in terms of logE, in which E stands for amount of exposure, between said said higher speed layer and said lower speed layer is 0.1 to 1.0.

8. The silver halide photographic light-sensitive material of claim 1, wherein higher speed layer has a dry layer thickness of 0.5 to 1.5μ.

9. The silver halide photographic light-sensitive material of claim 1, wherein the dry thickness of said hydrophilic colloid layer is 0.2 to 1.5μ.

10. The silver halide photographic light-sensitive material of claim 8, wherein the dry thickness of said hydrophilic colloid layer is 0.2 to 1.5μ.

11. The silver halide photographic light-sensitive material of claim 9, wherein the dry thickness of said hydrophilic colloid layer is 0.3 to 1.0μ.

12. The silver halide photographic light-sensitive material of claim 10, wherein the dry thickness of said hydrophilic colloid layer is 0.3 to 1.0μ.

13. The silver halide photographic light-sensitive material of claim 1, wherein said material contains a compound which is capable of reacting with the oxidation prodduct of a color developing agent.

14. The silver halide photographic light-sensitive material of claim 4, wherein said higher speed silver halide emulsion layer contains a compound which is capable of reacting with the oxidation product of a color developing agent.

15. The silver halide photographic light-sensitive material of claim 4, wherein said hydrophilic colloid layer contains a compound which is capable of reacting with the oxidation product of a color developing agent.

16. The silver halide photographic light-sensitive material of claim 14, wherein said hydrophilic colloid layer contains a compound which is capable of reacting with the oxidation product of a color developing agent.

17. The silver halide photographic light-sensitive material of claim 1, wherein said material contains a non-diffusible development inhibitor releasing compound.