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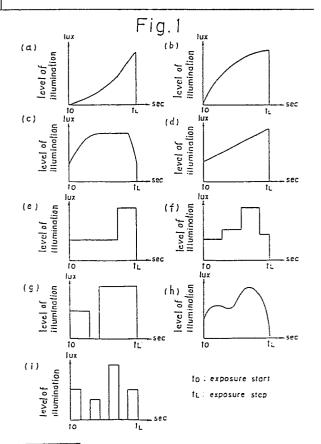
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- (1) Applicant: KONISHIROKU PHOTO INDUSTRY CO. LTD. No. 26-2, Nishishinjuku 1-chome Shinjuku-ku Tokyo 160(JP)
- (72) Inventor: Tosaka, Yasuo 6-44, Ohnodai 1-chome Sagamihara-shi Kanagawa-ken 229(JP)
- 72 Inventor: Ogi, Keiji 698-20, Kamiya-cho Hachioji-shi Tokyo 192(JP)
- (74) Representative: Türk, Dietmar, Dr. rer. nat. et al, Türk, Gille + Hrabal Patentanwälte Bruckner Strasse 20 D-4000 Düsseldorf 13(DE)

#### (54) Method of forming a direct-positive image.

67 In a method of forming a direct-positive image on a silver halide photographic material by first giving an imagewise exposure, and then applying an overall exposure either prior to or during subsequent development, said photographic material having on a support two or more silver halide emulsion layers which have different wavelength ranges of sensitivity and each of which contains internal latent-image forming silver halide grains that are not pre-fogged on the surface, the improvement is disclosed wherein the intensity of said overall exposure is changed at least once during said overall exposure and each of the relative magnitudes of the photographic effects exerted by said overall exposure on the respective silver halide emulsion layers is no greater than 20.



# METHOD OF FORMING A DIRECT-POSITIVE IMAGE

# BACKGROUND OF THE INVENTION

The present invention relates to a method of forming a direct-positive image and, more particularly, to a method of producing a direct-positive color image on an internal latent-image forming silver halide photographic material by the sequence of imagewise exposure and surface development as accompanied by an overall exposure.

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It is well known that a direct-positive photographic image can be formed on a silver halide photographic material without requiring any intervening processing step or forming any negative photographic image. In consideration of practical utility, this can be achieved by one of the following two basic methods. In one type, a prefogged silver halide emulsion is exposed, the fog centers (or latent image) in exposed areas are destroyed by the reversal effect of the solarization region or by the Herschel effect, then the emulsion is developed to obtain a positive image. In the other type, a non-prefogged internal latent-image forming silver halide photographic emulsion is imagewise exposed, then subjected to surface development after and/or during a fogging treatment so as to obtain a positive image.

The internal latent-image forming silver halide photographic emulsion (hereinafter sometimes referred to as the



internal-image silver halide photographic emulsion) means a silver halide photographic emulsion that has sensitivity centers chiefly within silver halide grains and which forms a latent image preferentially within the grains by exposure.

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Compared with the first type of method, the second type of method of forming a direct positive image generally has high sensitivity and is adaptive to applications where high sensitivity is required. The method of the present invention relates to this second type of method of forming a direct positive image.

Various techniques have heretofore been proposed in the technical field of forming a direct positive image with a non-prefogged internal-image silver halide photographic emulsion; principal examples are described in U.S. Patent Nos. 2,592,250, 2,466,957, 2,497,875, 2,588,982, 3,761,266, 3,761,276, 3,796,577, and British Patent No. 1,151,363. These known methods are capable of providing direct-positive

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sensitivity.

Details of the mechanism behind the formation of a direct-positive image have not been explained with complete clarity but one may understand to some extent the process of positive-image formation in terms of "desensitization by the internal latent image" as discussed in Mees and James, "The Theory of the Photographic Process", third edition,

working photographic materials having a comparatively high



P. 161. A plausible explanation is as follows: an "internal latent image" forms within silver halide grains by the first imagewise exposure; the surface sensitizing effect of this "internal latent image" causes fog centers to generate selectively on the surface of the unexposed silver halide grains; then, the surface fog centers are processed by ordinary surface development to form a photographic image in unexposed areas.

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Selective formation of fog centers is conventionally achieved by photo-fogging (i.e., fogging by light ) which depends on applying exposure to the entire surface of the light-sensitive layer or by chemical fogging which uses a reagent such as a foggant. The second method (chemical fogging) requires hostile conditions in that the effect of the foggant is only attained at high phs ( $\geq$ 12); this increases the chance of deterioration of the foggant by aerial oxidation, which leads to a very low fogging effect.

The photo-fogging method has the advantage of convenience for practical use since it does not require such hostile conditions as are necessary for effecting the chemical fogging method. However, even this approach has several technical problems that have to be solved before it can be applied to a broad range of photographic fields to satisfy a variety of purposes. Stated more specifically, the photo-fogging method is based on the formation of fog



centers as a result of photodecomposition of silver halide, so the intensity or amount of exposure which is appropriate for this method is highly dependent on the type and characteristics of the silver halide employed. Japanese Patent Publication No. 12709/1970 describes a proposal for implementing the photo-fogging method by applying a uniform overall exposure with light of low intensity. According to this reference, a satisfactory direct-positive image having a high maximum density and a low minimum density can be obtained by applying an overall exposure of low intensity.

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Applying the internal image direct-positive emulsion to silver halide color photographic material is very useful for practical purposes. The method commonly employed for forming a positive color image comprises: subjecting the silver halide color photographic material to imagewise exposure, subjecting the exposed photographic material to black-and-white development with a black-and-white developer, giving overall exposure or performing overall fogging with a foggant, and subsequently performing color development to This color reversal processing obtain a color reversal image. has the disadvantage that it requires many steps and is quite complicated. On the other hand, a positive color lightsensitive material using an internal-image direct-positive emulsion has the advantage of simple processing since only one development is necessary to produce a positive image.



The present inventors applied an internal-image directpositive emulsion to a color light-sensitive material and made various efforts to obtain an image by the photo-fogging method. As a result, the inventors found that when a uniform overall exposure was given with light of low intensity as shown in Japanese Patent Publication No. 12709/1970, image characteristics that were satisfactory in all respects could not be attained for the images formed in a plurality of layers. According to Unexamined Published Japanese Patent Application No. 137350/1981, photo-fogging exposure is performed under a fluorescent lamp having good color rendering properties. However, the present inventors found that this method had the disadvantage that although good characteristics were obtained with a certain sample of internal-image directpositive color light-sensitive material, only an unsatisfactory positive color image could be obtained with another sample. This finding may be stated more specifically as follows: in order to produce a satisfactory positive color image with an internal-image direct-positive color light-sensitive material by development involving the photo-fogging method, the material must be exposed to light of a limited and comparatively low intensity; if illumination lower than this reference is employed, an adequately high maximum density cannot be attained and, if a higher illumination is used, not only is the maximum density decreased but also the

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minimum density is appreciably increased, with the result that the quality of the positive image in the highlight is significantly impaired. In addition, the intensity of exposure within a certain range that ensures the formation of a satisfactory positive image sometimes differs for each of the silver halide emulsion layers in the positive lightsensitive material having dissimilar wavelength regions of sensitivity, and no satisfactory positive image is attainable In consideration of this possibility, the in such a case. invention described in Unexamined Published Japanese Patent Application No. 137350/1981 employs a light source composed of a fluorescent lamp having good color rendering properties. However, the present inventors found that it was difficult to obtain a desired positive color image by this method when there was a change in the characteristics, with respect to photo-fogging exposure, of the internal-image direct-positive color light-sensitive material used.

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A method for obtaining a satisfactory positive image by giving an overall exposure to a positive color light-sensitive material with an increasing level of illumination has been proposed in Japanese Patent Publication No. 6936/1983. It turned out however that this method was not always capable of producing a completely satisfactory positive color image.

25 A method for obtaining a satisfactory positive image

by applying an overall exposure with the distribution of the energy for overall exposure being varied has been proposed in Unexamined Published Utility Model Application No. 145049/1981; according to the disclosed method, successive overall exposures are given using a plurality of light sources having different energy distributions. However, even this method sometimes fails to produce a completely satisfactory positive color image.

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#### SUMMARY OF THE INVENTION

The principal object, therefore, of the present invention is to provide a method of forming a satisfactory direct-positive image by processing an internal-image direct-positive light-sensitive material employing the photofogging method.

This object of the present invention can be attained by a method of the type wherein a direct-positive image is formed on a silver halide photographic material by first giving an imagewise exposure, then applying an overall exposure either prior to or during subsequent development, said photographic material having on a support two or more silver halide emulsion layers that have different wavelength regions of sensitivity and each of which contains internalimage silver halide grains that are not pre-fogged on the surface, said method being characterized in that the intensity of said overall exposure is changed at least once



during said overall exposure and that each of the relative magnitudes of the photographic effects exerted by said overall exposure on the respective silver halide emulsion layers is no greater than 20.

#### BRIEF DESCRIPTION OF THE DRAWINGS

Figs. 1(a) to (i) show exposure patterns of various shapes that illustrate the changes in the level of illumination provided by overall exposure according to various embodiments of the method of the present invention.

## DETAILED DESCRIPTION OF THE INVENTION

According to the positive-image forming method of the present invention, an imagewise exposed silver halide photographic material is fogged by an overall light exposure either prior to or during subsequent development. One characteristic feature of the present invention lies in 15 the method that is employed to apply an overall exposure to cause fogging by light. According to the present invention, the intensity of irradiation applied in the overall exposure is changed at least once during said exposure. The term "intensity of overall exposure" as used herein means the 20 irradiance and/or energy distribution of the overall exposure. The change in the intensity of overall exposure means a change in the level of illumination, or a change in the wavelength-associated energy distribution of exposure, or changes in both factors. 25



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The method of changing the intensity of overall exposure at least once during said exposure in accordance with the present invention is hereunder described in detail. If the level of illumination of overall exposure is to be changed, it suffices that the level of illumination is changed at least once during overall exposure; alternatively the level of illumination may be changed continuously or stepwise. When the level of illumination of overall exposure is changed, an optimal pattern of exposure level of illumination may slightly differ with the object of use of a silver halide photographic material or its performance. The level-of-illumination pattern may be such that the level of illumination increases monotorically as shown in Figs. 1(a) to (d) or that it increases stepwise as depicted in Figs. 1(e) and (f); the level-of-illumination pattern may change in a complex manner from the start of exposure up to the point of a maximum level of illumination as shown in Figs. (g) to (i). These patterns may be attained by selecting specific means with reference being made to the disclosure in Japanese Patent Publication No. 6936/1983; for example, varying level of illumination may be attained by changing the voltage or current being imposed on a fixed type of light source during the overall exposure; the same effect may be attained with a filter such as a neutral density filter; alternatively, the level of illumination at the

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surface of the light-sensitive material may be changed during the overall exposure by changing the distance between the light source and the light-sensitive material or by changing the angle at which the incident light falls upon the material.

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The intensity of overall exposure may be changed in terms of the energy distribution. According to the present invention, the energy distribution of the irradiation applied in the overall exposure is changed at least once during said exposure; stated more specifically, the energy distribution of overall exposure is allowed to change at least once either discontinuously or continuously within the duration of said overall exposure. The change in the energy distribution of overall exposure is usually accompanied by a change in the level of illumination at the surface of the light-sensitive material but the energy distribution of overall exposure may be changed with the level of illumination at the surface of the light-sensitive material being held constant. Therefore, the method of the present invention wherein the energy distribution of overall exposure is changed includes the two cases, one involving a change in the level of illumination at the surface of the light-sensitive material and other involving no such change in the level of illumination.

In accordance with the present invention, the energy distribution of overall exposure can be changed by various

means: in one method, different types of light sources are used to change the energy distribution during the course of overall exposure; in another method, the distribution of energy from a fixed light source is changed by using an appropriate filter such as a color correcting filter or an interference filter; alternatively, the same result may be attained by changing the optical density of a solution disposed between a fixed light source and the light-sensitive material. Specific methods for performing an overall exposure, with the energy distribution of irradiation being changed at least once during the overall exposure, are described below:

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- (1) Perform an overall exposure with a tungsten lamp for a period of, say, 30 seconds, with the exposure for the first 10 seconds being applied through a magenta color correcting filter (green density: 0.5);
- (2) Perform an overall exposure with a tungsten lamp of 2800 K for a period of, say, 10 seconds, followed by a non-exposure interval of, say, 5 seconds and an overall exposure with a xenon lamp of 5200 K for a period of, say, 10 seconds, with the level of illumination of each overall exposure being 0.2 lux;
- (3) Perform an overall exposure with a white fluorescent lamp for a period of, say, 40 seconds, with tungsten light through an interference filter (640 nm) being

also applied for 20 seconds after the lapse of 10 seconds from the start of exposure;

equipped with blue, green and red gelatin filters, with blue light being applied for a period of 1 minute, green light for 1 minute after the lapse of 20 seconds from the start of exposure with blue light, and red light for 30 seconds after the lapse of 40 seconds from the start of exposure with blue light;

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10 (5) Perform an overall exposure for a period of, say, 10 seconds with a white fluorescent lamp through a solution whose optical density at 450 nm is 1.4 when the length of an optical path in the solution is 10 cm and 0.7 when said length is 5 cm, with the length of the optical path in the solution being gradually decreased from, for example, 10 cm at the start of exposure to 5 cm at the end of exposure.

In methods (1) and (3) to (5), the level of illumination at the surface of the light-sensitive material changes as the energy distribution of the overall exposure changes, but this is not so in method (2) where only the energy distribution of the overall exposure changes.

While several specific examples of the method for performing an overall exposure with the energy distribution of irradiation being changed at least once during the overall

exposure are described above, it should be understood that various modifications may be made without departing from the scope of the present invention. It should also be remembered that in the method of the present invention the overall exposure may be performed with either the level of illumination or energy distribution or both being changed at least once during the overall exposure.

The fogging by light which is performed in the present invention with the intensity of overall exposure being changed during said exposure is especially intended for achieving a level-of-illumination adjustment by artificial means. It should be emphasized that the advantages of the present invention will not be attained if a gradually increasing amount of exposure is continuously applied from a single light source by simply taking advantage of the inherent flashing characteristics of the light source. The method of the present invention may be performed with advantage by using two or more light sources in combination.

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The concept of "the magnitude of photographic effect" as used herein means the magnitude of an effect that a certain type of overall exposure can cause photographically on a certain silver halide emulsion layer and the relative value of this effect can be determined for each silver halide emulsion layer. The magnitude of photographic effect depends on both the energy distribution of overall exposure



and the distribution of the spectral sensitivity of an individual silver halide emulsion layer.

The specific method for determining the magnitude of the photographic effect of an overall exposure is described hereinafter.

First suppose that an internal-image silver halide photographic material that is intended to be processed by the present invention and which has not been subjected to imagewise exposure is given an overall exposure either prior to or during subsequent development; if the density of an image that is formed in a silver halide emulsion layer having a certain wavelength range of sensitivity is 0.2 higher than the density of an image that is obtained by following entirely the same procedures except for the absence of any overall exposure, then the reciprocal of the amount of exposure required to provide that high density is taken as the magnitude of the photographic effect of said overall exposure on that particular silver halide emulsion layer. Similarly, the magnitude of the photographic effect of a silver halide emulsion layer having a different wavelength range of sensitivity may be determined. The two determined values are used to calculate the ratio of the magnitudes of photographic effects.

In carrying out the above procedures for determining the magnitude of the photographic effect of an overall

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exposure, it is necessary to conduct a series of tests with the amount of the overall exposure being varied. The amount of an overall exposure may be changed by using a neutral density filter (hereunder referred to as an ND filter) such as Wratten Gelatin Filter available from Eastman Kodak Company. The ND filter may be inserted at any position of the optical path between the light source and the light-sensitive material so long as it is used in such a manner that it attains uniform attenuation of the quantity of light throughout the duration of overall exposure.

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The ratio of the magnitudes of photographic effects attained in the present invention may be determined by changing the relative amounts of an overall exposure with the aid of an ND filter. The procedures for determining the of the photographic effect of an overall magnitude exposure are identical to the processing steps employed for positive-image formation except that an ND filter is used to change the amount of said overall exposure. More specifically, the duration of the overall exposure given in a test for determining the magnitude of the photographic effect of that exposure is the same as that of the overall exposure applied for positive-image formation; if an overall exposure is to be given during development, the time interval between the start of development and the start of overall exposure is adjusted to be the same for



the two methods.

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If the amount of the overall exposure applied is small, a very low image density results, and as the amount of exposure is increased, the density of the image obtained becomes higher. This dependency of image density on the amount of exposure usually differs for each of the silver halide emulsion layers used.

The present inventors have found that a satisfactory positive image is obtained if an overall exposure is applied either prior to or during the development step after imagewise exposure, with the intensity of irradiation being changed at least once during said overall exposure, and if each of the relative magnitudes of the photographic effects exerted by said overall exposure on a plurality of silver halide emulsion layers is no greater than 20, preferably no greater than 10.

In summary, the conditions of the overall exposure which is applied in the method of the present invention for positive-image formation in such a manner that the intensity of irradiation is changed at least once during said exposure can be determined by the following procedures: a series of experiments are conducted to process samples of the same silver halide photographic material with the intensity of an overall exposure being changed at least once during said exposure; the relative magnitudes of the

photographic effects exerted by said overall exposure on the silver halide emulsion layers in each sample are determined; and the conditions of overall exposure that provide photographic effects the relative magnitudes of which are not greater than 20 are checked.

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by the method of the present invention is one having two or more internal-image silver halide emulsion layers having different wavelength ranges of sensitivity. According to one preferable embodiment of the present invention, the silver halide photographic material comprises a blue-sensitive silver halide emulsion layer capable of forming a yellow image, a green-sensitive silver halide emulsion layer capable of forming a cyan image. The method of the present invention is hereunder described in further detail with reference to the case where the silver halide color photographic material has such a multi-layered structure.

First suppose that a varying amount of overall exposure is applied in a series of experiments for determining the relative magnitudes of the photographic effects exerted by said overall exposure; also suppose that the blue, green and red densities which are respectively measured with blue light corresponding to the yellow image obtained, green

light corresponding to the magenta image, and red light corresponding to the cyan image are 0.2 higher than the blue, green and red densities of the images that are obtained by entirely the same method except that no overall exposure is applied. If the amounts of exposure that have provided those high densities are signified by Eb, Eg and Er, the magnitudes of the photographic effects exerted by the overall exposure in accordance with the present invention are expressed by 1/Eb, 1/Eg and 1/Er, respectively.

In accordance with the present invention,

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$$0.05 \leq \frac{Eb}{Eg} \leq 20 \tag{1}$$

$$0.05 \leq \frac{Eq}{Er} \leq 20 \tag{2}$$

$$0.05 \le \frac{Er}{Eb} \le 20$$
 (3).

The object of the present invention can be attained if an overall exposure is applied either prior to or during development subsequent to imagewise exposure in such a manner that the relative magnitudes of the photographic effects attained by that overall exposure satisfy all of the relations (1) to (3).

The effectiveness of the overall exposure that satisfies
the relations (1) to (3) is in no way limited to a light-

sensitive material comprising a blue-sensitive yellow-image forming layer, a green-sensitive magenta-image forming layer, and a red-sensitive cyan-image forming layer.

In the present invention, the density of a particular image is measured with light having a wavelength in the neighborhood of the maximum absorption of that image.

Stated more specifically, the light is monochromatic light having a maximum absorption within 20 nm from the wavelength for the maximum absorption of that particular image.

The overall exposure applied in the present invention may be implemented with any type of light source that is capable of an adjustment such that each of the relative magnitudes of the photographic effects exerted by said overall exposure on the emulsion layers in the silver halide photographic material used is no greater than 20. Illustrative light sources that may be used include a tungsten lamp, a fluorescent lamp, a halogen lamp, a xenon lamp, a mercury lamp and daylight, which may be employed in appropriate combinations.

The relative magnitudes of the photographic effects exerted by the overall exposure on the silver halide emulsions may be so adjusted as to satisfy the requirement for <20 by changing them in accordance with any one of the commonly employed techniques, such as by changing the energy distribution of the light source used, or by using filters

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such as a color correcting filter or a color temperature converting filter.

The overall exposure applied in the present invention may also be implemented with a plurality of light sources; in a preferable embodiment, separate light sources may be employed to provide blue, green and red light.

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The silver halide emulsion layers in the light-sensitive material to be processed by the method of the present invention has different wavelength regions of sensitivity; this means that the distributions of spectral sensitivity of the individual layers are not completely identical to each other and the possibility of partial overlapping of the wavelength ranges of sensitivity of these layers is by no means precluded.

Any two silver halide emulsion layers having different wavelength ranges of sensitivity are capable of image formation by the method of the present invention and it is preferable to select silver halide emulsion layers which are such that the images they produce create a minimum overlapping of the wavelength ranges of absorption.

In accordance with the present invention, the overall exposure may be performed prior to development and this means that after imagewise exposure, the light-sensitive material is given said overall exposure either in the bath that is used for the processing that precedes the development step or after completion of such processing.



If necessary, the processing bath may contain an additive such as a reducing material, an alkali agent, a restrainer or a desensitizer.

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If the overall exposure is performed during development, it is preferably completed in the early stage of development for the purpose of shortening the duration of the development time. In this case, it is advantageous to start the exposure after the developer has penetrated through the emulsion layers to a reasonable extent.

The surface developer used in the present invention for development purposes means one which is substantially free of any solvent for silver halides. Developing agents that may be incorporated in the surface developer are conventional silver halide developing agents such as polyhydroxybenzenes (e.g., hydroquinone), aminophenols, 3-pyrazolidones, ascorbic acid and derivatives thereof, reductones, phenylenediamines, and mixtures thereof. Specific examples of such developing agents include: hydroquinone, aminophenol, N-methylaminophenol, 1-phenyl-3-pyrazolidone, 1-phenyl-4,4-dimethyl-3pyrazolidone, 1-phenyl-4-methyl-4-hydroxymethyl-3-pyrazolidone, ascorbic acid, N,N-diethyl-p-phenylenediamine, diethylaminoo-toluidine, 4-amino-3-methyl-N-ethyl-N-(β-methanesulfonamidoethyl) aniline, 4-amino-3-methyl-N-ethyl-N- $(\beta$ -hydroxyethyl)aniline, 4-amino-3-methyl-N,N-diethyl-p-phenylenediamine, and 4-amino-3-methyl-N-ethyl-N-β-methoxyethyl-p-phenylene-



diamine. These developing agents may be incorporated in emulsions such that they will act on silver halides when the light-sensitive material is submerged in an aqueous solution of high pH.

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The surface developer may further contain an additive such as an antifoggant or a development restrainer.

These additives may be optionally incorporated in one or more of the constituent layers of the silver halide photographic material. Generally useful antifoggants include heterocyclic thionines and aromatic or aliphatic mercaptocompounds such as benzotriazoles, benzimidazoles, benzothiazoles benzoxazoles and 1-phenyl-5-mercaptotetrazole. The developer may also contain an appropriate development accelerator such as a polyalkylene oxide derivative or a quaternary ammonium salt compound.

The method of the present invention for forming a direct-positive image may be applied not only to general color photographic materials but also to those photographic materials which are designed to be processed by color image transfer, color diffusion transfer or by absorption transfer, as shown in U.S. Patent Nos. 87,817, 3,185,567 and 2,983,606 to Rogers, U.S. Patent No. 3,253,915 to Weyerts et al., U.S. Patent No. 3,227,550 to Whitemore et al., U.S. Patent No. 3,227,552 to Whitemore, and U.S. Patent Nos. 3,415,644, 3,415,645 and

3,415,646 to Land.

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The internal latent-image forming silver halide emulsion used in the present invention is one having silver halide grains which form a latent image predominantly in their interior so that the greater part of the sensitivity centers are present within the grains. The silver halide grains may have any of the silver halide compositions such as silver bromide, silver chloroide, silver chlorobromide, silver iodobromide and silver chloroiodide.

It is preferable that the internal-image silver halide grains used in the present invention are not chemically sensitized on their surfaces or are sensitized only slightly.

The surfaces of the silver halide grains used in the present invention are not prefogged. This means that when an unexposed test piece wherein the emulsion used in the present invention is coated on a transparent support to form a layer having a silver deposit to 35 mg/dm<sup>2</sup> is developed at 20°C for 10 minutes in a surface developer (A) having the formulation indicated below, the density obtained will not exceed 0.6, and will preferably not exceed 0.4: Surface developer (A):

	Methol	2.5 g
	L-ascorbic acid	10.0 g
	NaBO <sub>2</sub> ·4H <sub>2</sub> O	35.0 g
25	KBr	1.0 g
	Water	to make 1,000 ml

The silver halide emulsion used in the present invention is such that the test piece prepared as above will provide an adequate density when it is developed with an internal developer (B) having the formulation shown below after being exposed:

Internal developer (B):

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	Methol			2.0	g
	Sodium sulfite (anhydrous)			90.0	g
	Hydroquinone			8.0	g
10	Sodium carbonate (monohydrat	:e)		52.5	g
	KBr			5.0	g
	KI			0.5	g
	Water	to	make	1,000	m1

Stated more specifically, the silver halide emulsion used in the present invention is such that when part of the above-described test piece is developed with the internal developer (B) at 20°C for 10 minutes after being exposed on a light intensity scale over a predetermined time period of up to about one second, it shows a maximum density at least 5 times, preferably at least 10 times, the density attained by developing another part of the same test piece with the surface developer (A) at 20°C for 10 minutes after being exposed under the same conditions.

Specific examples of the silver halide emulsion that may be used in the present invention include: the conversion

type silver halide emulsion described in U.S. Patent No. 2,592,250; the core/shell emulsion which contains internally chemically sensitized nuclei or which is doped with polyvalent metallic ions, as shown in U.S. Patent Nos. 3,761,266 and 3,761,276; the multi-layered silver halide emulsion described in Unexamined Published Japanese Patent Application Nos. 8524/1975, 38525/1975 and 2408/1978; and emulsions of the types described in Unexamined Published Japanese Patent Application Nos. 156614/1977 and 127549/1980.

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The silver halide emulsion used in the present invention may be spectrally sensitized with any of the commonly employed sensitizing dyes. Combinations of sensitizing dyes that are employed for the purpose of supersensitization of internalimage silver halide emulsions or negative-working silver halide emulsions are also useful for the silver halide emulsion used in the present invention. For the selection of appropriate sensitizing dyes, reference may be made to Research Disclosure Nos. 15162 and 17643.

In order to minimize the surface sensitivity and to provide a lower minimum density and more stable characteristics, the silver halide emulsion used in the present invention may incorporate a commonly employed stabilizer selected from among the compounds having an azaindene ring (a typical example is 4-hydroxy-6-methyl-1,3,3a,7-tetrazaindene) and the mercapto-containing heterocyclic compounds (typically,



1-phenyl-5-mercaptotetrazole).

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The silver halide emulsion used in the present invention may also contain an antifoggant or a stabilizer, which may be selected from among triazole, azaindene and benzothiazolium compounds.

The silver halide emulsion used in the present invention may further contain a variety of photographic addenda which include: a wetting agent such as a dihydroxyalkane; an agent that provides improved film properties and which is advantageously selected from the water-dispersible, fine particulate large molecular-weight substances that are obtained by emulsion polymerization, such as a copolymer of an alkyl acrylate or methacrylate and acrylic or methacrylic acid, a styrene/maleic acid copolymer, and a styrene/maleic anhydride/half alkyl ester copolymer; and a coating agent such as saponin or polyethylene glycol lauryl ether. Other photographic addenda that may be optionally employed are gelatin plasticizers, surfactants, UV absorbers, pH modifiers, antioxidants, antistats, thickeners, granularity improving agents, dyes, mordants, brighteners, development speed control agents, and matting agents.

The thus prepared silver halide emulsion is coated onto a support, with a subbing layer, an anti-halation layer, a filter layer and any other appropriate layers being optionally interleaved to provide an internal-image silver

halide photographic material.

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The silver halide photographic material to be processed by the present invention may incorporate cyan, magenta and yellow dye forming couplers in at least three internal-image silver halide emulsion layers, with one coupler being present in each of the associated layers.

An appropriate yellow-dye image forming coupler is a benzoylacetanilide coupler, a pivaloylacetanilide coupler, or a two-equivalent yellow-dye image forming coupler wherein the carbon atom at the coupling site is replaced by a split-off group, or a substituent which is capable of being eliminated upon coupling reaction. An appropriate magenta-dye image forming coupler is a 5-pyrazolone coupler, a pyrazolotriazole coupler, a pyrazolinobenzimidazole coupler, an indazolone coupler, or a two-equivalent magenta-dye image forming coupler having a split-off group. An appropriate cyan-dye image forming coupler is a phenolic coupler, a naphtholic coupler, a pyrazoloquinazolone coupler, or a two-equivalent cyan-dye image forming coupler having a split-off group.

In order to prevent the dye images from becoming faded by exposure to actinic radiation of short wavelength, it is advisable to use a UV absorber such as thiazolidone, benzotriazole, acrylonitrile or benzophenone compound. It is particularly useful to employ Tinuvin PS 320, 326, 327 and 328 (all being from Ciba Geigy AG.) either individually or

in combination.

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Any support may be used with the silver halide photographic material in the present invention; typical examples include optionally subbed polymer films (e.g., polyethylene terephthalate, polycarbonate, polystyrene, polypropylene and cellulose acetate), glass plates, baryta paper, and polyethylenelaminated paper.

Constituent layers such as emulsion layers, intermediate layers, filter layers, backing layer and protective layer in the silver halide photographic material to be processed by the present invention typically use gelatin as a hydrophilic binder. A suitable gelatin derivative may also be used depending on a specific object and illustrative gelatin derivatives include acylated gelatin, guanidylated gelatin, carbamylated gelatin, cyanoethanolated gelatin and esterified Other hydrophilic binders in common use may also gelatin. be incorporated depending on the specific object. hydrophilic binders may contain plasticizers, lubricants or any other appropriate additives.

20 The constituent layers of the silver halide photographic material may be hardened with any appropriate film hardener which is selected from, for example, chromium salts, zirconium salts, aldehyde compounds such as formaldehyde and mucohalogenic acids, halotriazine compounds, polyepoxy compounds, ethyleneimines, vinylsulfone compounds and acryloyl compounds.

The silver halide photographic material to be processed by the present invention may have a number of constituent layers formed on a support, such as emulsion layers, filter layers, intermediate layers, a protective layer, a subbing layer, a backing layer, and an anti-halation layer.

The following examples are provided for the purpose of further illustrating the present invention but should in no sense be taken as limiting the possible embodiments of the present invention.

 $\underline{\text{Example 1}}$ 

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A core/shell emulsion was prepared in the following manner: to an aqueous solution of gelatin that was controlled at 50°C, equimolar amounts of aqueous solutions of silver nitrate and potassium bromide were added simultaneously by the controlled double-jet method over a period of 40 minutes so as to obtain an emulsion comprising 0.35 µm cubic silver bromide grains; to the so obtained core emulsion, 2.0 mg of sodium thiosulfate and 3.0 mg of potassium chloroaurate, each being based on one mole of silver, were added and chemical ripening was performed at 55°C for 120 minutes; to the so treated emulsion, aqueous solutions of silver nitrate and potassium bromide were added simultaneously to obtain a silver bromide emulsion comprising 0.5 µm cubic grains. To the so prepared core/shell emulsion, 2.0 mg of sodium thiosulfate and 2.0 mg of potassium chloroaurate,



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each being based on one mole of silver, were added and chemical ripening was performed at 55°C for 90 minutes. The thus obtained emulsion was referred to as Emulsion A.

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The Emulsion A was divided into three portions; the first portion was spectrally sensitized with dye (I) having the formula shown below, so as to form a green-sensitive emulsion; the second portion was spectrally sensitized with dyes (II) and (III) (see below) to form a red-sensitive emulsion; and the third portion was used as a blue-sensitive emulsion without being subjected to any spectral sensitization.

Dye [ ] ]

$$\begin{array}{c} C_2H_5 \\ O \\ CH = C - CH \\ O \\ CH_2)_3 SO_3 \\ O \\ CH_2)_3 SO_3N_{\Delta} \\ \end{array}$$

Dye [ II ]

$$Cl \xrightarrow{S} CH = C - CH \xrightarrow{S} Cl$$

$$(CH_2)_3 SO_3 \xrightarrow{\Theta} (CH_2)_3 SO_3 NA$$

Dye [ II ]

$$\begin{array}{c|c}
C_{2}H_{5} \\
C_{1}H_{5} \\
C_{2}H_{5} \\
C_{3}H_{5} \\
C_{4}H_{5} \\
C_{5}H_{5} \\
C_{7}H_{5} \\
C_{8}H_{5} \\
C_{8}H_{5$$

A resin-coated paper support was coated with the following layers in the order written.

- (1) Red-sensitive emulsion layer
  This layer contained the previously obtained red-sensitive emulsion (silver deposit, 5 mg/dm²) and an oil-protected cyan coupler, or 2,4-dichloro-3-methyl-6-[α-(2,4-di-tert-amylphenoxy)butylamido]phenol (0.45 moles per mole of silver halide);
- (2) Intermediate layer

  This layer contained oil-protected 2,5-di-tertoctylhydroquinone.
- This layer contained the previously obtained greensensitive emulsion (silver deposit, 5 mg/dm²) and
  an oil-protected magenta coupler, or 1-(2,4,6trichlorophenyl)-3-(2-chloro-5-octadecylsuccinimidoanilino)-5-pyrazolone (0.25 moles per mole of silver
  halide);
- (4) Yellow filter layer
  This layer contained yellow colloidal silver and oil-protected 2,5-di-tert-octylhydroquinone;
  - (5) Blue-sensitive emulsion layer This layer contained the previously obtained blue-sensitive emulsion (silver deposit, 6 mg/dm $^2$ ) and an oil-protected yellow coupler, or  $\alpha$ -[4-(1-benzyl-2-

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phenyl-3,5-dioxo-1,2,4-triazolidinyl)]- $\alpha$ -pivalyl-2-chloro-5-[ $\gamma$ -(2,4-di-tert-amylphenoxy)butylamido]-acetanilide (0.45 moles per mole of silver halide); and

5 (6) Protective layer

This was a gelatin layer.

The web was dried to obtain Sample 1.

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To an aqueous solution of gelatin that was controlled at 50°C, equimolar amounts of aqueous solutions of silver nitrate and potassium bromide were added simultaneously by the controlled double-jet method over a period of 90 minutes so as to obtain a silver bromide emulsion comprising 0.9- $\mu m$ To the so obtained core emulsion, 1.0 mg of cubic grains. sodium thiosulfate and 2.0 mg of potassium chloroaurate, each being based on one mole of silver, were added and chemical ripening was effected at 55°C for 180 minutes. To the so treated emulsion, aqueous solutions of silver nitrate and potassium bromide were added simultaneously to obtain a silver bromide emulsion comprising 1.0- $\mu m$  cubic grains. To the so prepared core/shell emulsion, 1.0 mg of sodium thiosulfate and 1.5 mg of potassium chloroaurate, each being based on one mole of silver, were added and chemical ripening was effected at 60°C for 120 minutes. The thus obtained emulsion was referred to as Emulsion B.

This emulsion B was spectrally sensitized with dye (IV) having the formula shown below, so as to form a blue-sensitive



emulsion:

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Dye (IV)

Sample 2 was prepared by repeating the procedures for the preparation of Sample 1 except that the above-obtained emulsion was used as a blue-sensitive emulsion.

Each of the samples 1 and 2 was exposed through a sensitometric optical wedge with a sensitometer (this exposure is hereinafter referred to as wedge exposure). The exposed samples were further given an overall exposure for 20 minutes with xenon light in accordance with the following exposure schedules:

- (1) Exposed for 10 seconds at 0.25 lux and, immediately thereafter, exposed for 10 seconds at 10 lux;
- (2) Exposed at in (1) except that a color correcting yellow filter (blue density D<sub>B</sub>: 0.3) was employed, with the level of illumination being measured in the absence of any yellow filter;
  - (3) Exposed as in (2) except that  $D_{\overline{B}}$  was 0.6;
  - (4) Exposed as in (2) except that  $D_{\rm B}$  was 0.9;
- 25 (5) Exposed as in (2) except that  $D_B$  was 1.2;



- (6) Exposed as in (2) except that  $D_{\overline{B}}$  was 1.5;
- (7) Exposed as in (2) except that  $D_B$  was 1.8;
- (8) Exposed as in (2) except that  $D_{\mathrm{B}}$  was 2.1; and
- (9) Exposed as in (2) except that  $D_{\rm B}$  was 2.4.

Each sample was developed at 20°C for 5 minutes with a developer having the formulation indicated below. The overall exposure specified above was started 20 seconds after the samples were submerged in the developer.

## Developer formulation

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10	4-Amino-3-methyl-N-ethyl-N-( $\beta$ -methanesulfonamidoethyl)aniline s	ılfate		5 g
	Sodium sulfite (anhydrous)			2 g
	Sodium carbonate (monohydrate)		1	5 g
	Potassium bromide			1 g
	Benzyl alcohol		1	0 ml
15	Water	to mak	se 1,00	0 ml.

The developed samples were bleached, fixed, washed with water and dried by routine procedures. The maximum density (Dmax) and minimum density (Dmin) of the positive image formed in each of the samples were measured, and the results are shown in Tables 1 and 2, wherein the overall rating of the image is indicated by o (good) and x (poor).

Table 1 (Sample 1)

		<del></del> 1		<del></del> -			<del></del> 1		·		
•	Image quality							X ( yellow had	low Dmax )	X ( do. )	× ( do. )
		0	0	0	0	0	0	<u>×</u>		×	<u> </u>
r L	Omin	0.03	0.09	0.08	0.09	0.00	0.10	0.10		0.09	0.09
cyan	D max	2.14	2.07	2.15	2.15	2.14	2.17	2.13	-	2.16	2.11
magenta	Omin	0.15	0.14	0.15	0.15	0.17	0.14	0.16		0.17	0.16
mage	D max	2.15	2.14	2.19	2.09	2.21	2.13	2.17		2.20	2.19
low	Omin	0.10	0.17	0.18	0.17	0.17	0.17	0.19		0.17	0.17
vellow	Dmax	2.13	2.15	2.17	2.14	2.05	2.09	1.65		1.32	1.20
	Overall Amaye -	0	0	(P)	<b>(</b>	9	@	0		@	6

Table 2 (Sample 2)

	Image quality	X ( yellow had	high Dmin )	X ( dc. )	0	0	0	0	0	0	0
cyan	Dmin	0.09		0.09	0.10	0.08	0.08	0.09	0.09	0.09	0.10
су	Dınax	2. 18		2.24	2.21	2.19	2.20	2.17	2.23	2.24	2.19
magenta	Dmin	0.15		0.14	0.15	0.16	0.15	0.17	0.16	0.15	0.16
ma	D max	2.21		2.19	2.24	2.24	2.15	2.08	2.14	2.20	2.23
yellow	Dmin	0.46		0.31	0.19	0.17	0.18	0.17	0.18	0.19	0.18
yel	Dwax	1,85		1.97	2.17	2.09	2.13	2.24	2.15	2.18	2.17
Owerall Tmade	sure	<b>(</b>		0	(3)	<b>(</b>	9	9	0	(9)	<b>6</b>

The relative magnitudes of the photographic effects of the overall exposures (1) to (9) were determined by the following procedures.

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Some of the test pieces of samples 1 and 2 were developed, bleached and fixed as shown above, except that neither wedge exposure nor overall exposure was applied. The image densities obtained with sample 1 were 0.12 (yellow), 0.11 (magenta) and 0.07 (cyan).

Some of the test pieces of sample 1 were not subjected to wedge exposure but were given an overall exposure according to schedule (1), with an ND filter capable of stepwise change in density from 0.2 to 3.0 at intervals of 0.2 being disposed between the light source and each test piece. The procedures of development, bleaching, fixing and washing were the same as those employed in the processing of the samples that had been subjected to wedge exposure. The image densities obtained with the test pieces were measured and the density of the ND filter that was required to provide a density 0.2 higher than the density of each of the yellow, magenta and cyan images on the test pieces that were not given any overall exposure was also measured. The results were 2.35 (yellow), 1.96 (magenta) and 1.30 (cyan). Therefore, the relative magnitudes of the photographic effects of overall exposure (1) were calculated as follows:



Eb/Eg = 
$$10^{-2.35}/10^{-1.96}$$
  
Eg/Er =  $10^{-1.96}/10^{-1.30}$   
Er/Eb =  $10^{-1.30}/10^{-2.35}$ 

Overall exposure (1) was within the scope of the present invention since it satisfied all of the following relations:

$$0.05 \le Eb/Eg \le 20$$
  
 $0.05 < Eg/Er \le 20$ 

$$0.05 \le Er/Eb \le 20$$
.

Similarly, the relative magnitudes of the photographic effects exerted by each of the overall exposures (2) to (9) on the test pieces of sample 1 and by each of the overall exposures (1) to (9) on sample 2 were determined.

The results are shown in Tables 3 and 4.

Table 3 (Sample 1)

Overall exposure	Eb/Eg	Eg /Er	Er/Eb	Remarks
1	0.41	0.22	11.2	0
2	0.85	0.29	4.1	Ö
3	1.54	0.30	2.2	0
<b>4</b>	3.2	0.27	0.86	0
(5)	5.9	0.24	0.71	. 0
6	13.2	0.31	0.24	0
0	29	0.27	0.128	х
8	45	0.24	0.0118	х
9	59	0.51	0.033	Х

 $\ensuremath{\textsc{0}}$  : within the scope of the invention

 ${\tt X}$  : outside the scope of the invention

Table 4 ( Sample 2 )

Overall				
exposure	Eb/Eg	Eg /Er	Er/Eb	Remarks
1	0.063	0.23	69	х
2	0.143	0.18	39	Х
3	0.22	0.18	17	0
4	0.55	0.22	8.3	.0
<b>⑤</b>	0.85	0.20	5.9	0
(6)	1. 79	0. 194	2.9	0
Ō	3.5	0.27	1.06	0
8	7.6	0.23	0.57	0
9	12.4	0.22	0.37	0

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As the above data shows, positive images having high maximum densities and low minimum densities can be formed by applying an overall exposure under the conditions that satisfy the requirement specified by the present invention. The image characteristics obtained are satisfactory for each of the dye images formed in a multi-layered silver halide photographic material. The range of the conditions for overall exposure that ensure the production of a satisfactory photographic image differs between sample 1 and sample 2; the method of the present invention enables a satisfactory image to be obtained irrespective of the type of light-sensitive material but this is very difficult to attain if methods that are outside the scope of the present invention are used.

15 Example 2

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exposure. It was then given an overall exposure for 10 seconds with a white fluorescent lamp being used as a light source under the following condition: a first exposure was given at 0.3 lux and, immediately thereafter, a second exposure was given for 8 seconds at 5 lux. In another run, the sample was given an overall exposure with the same fluorescent lamp under the same condition except that a color correcting magenta filter (green density = 1.0) was used, with the level of illumination being measured in the

absence of any magenta filter.

The test pieces were developed, bleached, fixed and washed with water as in Example 1. The maximum and minimum densities of the positive image formed in each test piece were measured, and the results are shown in Table 5.

The relative magnitudes of the photographic effects exerted by applying an overall exposure in the two different manners are shown in Table 6.

Table 5

10 Image Yellow Magenta Cyan Image quality Overall Dmax Dmin Dmax Dmin Dmax Dmin exposure White fluorescent 1.99 0.17 1.79 0.30 2.14 0.09 X lamp (magenta had high Dmin) White fluorescent lamp + magenta 2.04 0.18 2.21 0.15 2.20 0.09 0 15 filter

Table 6

Overall exposure Eb/Eg Eg/Er Er/Eb Remarks White fluorescent lamp 42 0.14 0.17 Outside the scope of the invention White fluorescent lamp Within the scope of 10.5 0.43 0.22 + magenta filter the invention

The above data shows that a satisfactory positive image having a high maximum density and a low minimum density can be obtained by applying an overall exposure in accordance with the present invention.

Example 3

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A core/shell emulsion was prepared in the following to an aqueous solution of gelatin that was controlled at 50°C, equimolar amounts of aqueous solutions of silver nitrate and potassium bromide were added simultaneously by the controlled double-jet method over a period of 40 minutes so as to obtain an emulsion comprising 0.35- $\mu m$  octahedral silver bromide grains; to the so obtained core emulsion, 2.0 mg of sodium thiosulfate and 3.0 mg of potassium chloroaurate, each being based on one mole of silver, were added and chemical ripening was performed at 60°C for 120 minutes; to the so treated emulsion, aqueous solutions of silver nitrate and potassium bromide were added simultaneously to obtain a silver bromide emulsion comprising 0.5-µm octahedral To the so prepared core/shell emulsion, 2.0 mg of grains. sodium thiosulfate and 2.0 mg of potassium chloroaurate, each being based on one mole of silver, were added and chemical ripening was conducted at 55°C for 120 minutes. The thus obtained emulsion was referred to as Emulsion C.

The emulsion C was divided into three portions; the first portion was spectrally sensitized with dye (I) (same

as employed in Example 1) to form a green-sensitive emulsion; the second portion was spectrally sensitized with dyes (II) and (III) (same as used in Example 1) to form a red-sensitive emulsion; and the third portion was used as a blue-sensitive emulsion without being subjected to any spectral sensitization.

A resin-coated paper support was coated with the following layers in the order written.

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- (1) Red-sensitive emulsion layer
  This layer contained the previously obtained red-sensitive
  emulsion (silver deposit, 5 mg/dm²) and an oil-protected
  cyan coupler, or 2,4-dichloro-3-methyl-6-[α-(2,4-di-tert-amylphenoxy)butylamido]phenol (0.45 moles per mole of silver halide);
- (2) Intermediate layer
  This layer contained oil-protected 2,5-di-tert-octyl-hydroquinone.
- This layer contained the previously obtained greensensitive emulsion (silver deposit, 5 mg/dm²) and an

  oil-protected magenta coupler, or 1-(2,4,6-trichlorophenyl)3-(2-chloro-5-octadecylsuccinimidoanilino)-5-pyrazolone

  (0.25 moles per mole of silver halide);
  - (4) Yellow filter layer

    This layer contained yellow colloidal silver and oilprotected 2,5-di-tert-octylhydroquinone;



- Blue-sensitive emulsion layer (5) This layer contained the previously obtained bluesensitive emulsion (silver deposit, 6 mg/dm<sup>2</sup>) and an oil-protected yellow coupler, or  $\alpha-[4-(1-benzyl-2$ phenyl-3,5-dioxo-1,2,4-triazolidinyl)]- $\alpha$ -pivalyl-2chloro-5-[\gamma-(2,4-di-tert-amylphenoxy)butylamido]acetanilide (0.45 moles per mole of silver halide); and
  - Protective layer (6) This was a gelatin layer.
- The web was dried to obtain a light-sensitive material. 10 It was then subjected to wedge exposure. The exposed sample was further given an overall exposure under three tungsten lamps which were respectively equipped with a blue filter (Wratten Gelatin Filter No. 47B of Eastman Kodak Company), a green filter (Wratten Filter No. 61), and a red filter 15 (Wratten Filter No. 29). The overall exposure was started by turning on the three tungsten lamps simultaneously. Exposure to green and red light was continued for 3 seconds, but the duration of exposure to blue light was varied as (1) 0.5 seconds, (2) 1 second, (3) 2 seconds, 20 5 seconds, (5) 10 seconds, (6) 20 seconds, (4)

  - 40 seconds, (8) 80 seconds, (9) 160 seconds, and (7)
  - 280 seconds. (10)

The test pieces of the sample were developed at 20°C for 5 minutes with a developer having the formulation 25



indicated below. The overall exposure specified above was started 20 seconds after they were submerged in the developer. Developer formulation

4-Amino-3-methyl-N-ethyl-N-( $\beta$ -methanesulfonamidoethyl) aniline	sul	Eate	5	g
Sodium sulfite (anhydrous)			2	g
Sodium carbonate (monohydrate)			15	g
Potassium bromide			1	g
Benzyl alcohol			10	ml
Water	to	make	1,000	ml.

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The developed test pieces were bleached, fixed, washed with water and dried by routine procedures. The maximum density (Dmax) and minimum density (Dmin) of the positive image formed in each of the test pieces were measured, and the results are shown in Table 7, wherein the overall rating the image is indicated by o (good) and x (poor).

Table 7

	Image quality	X (.yellow had	low Dmax · )	đo. )	do. )							
	Imag	× ×	JQ.	×	×	0	0	0	0	0	0	0
a	Omin	60.0		0.10	009	0.09	0.03	0.11	0.09	0.09	0.08	0.03
cyan	Опах	2.14		2.09	2.17	2.21	2.20	2. 18	2.26	. 2. 19	2.33	2.25
nta	Omin	0.15		. 0.14	0.15	0.15	0.15	0.16	0.15	0.15	0.17	0.15
magenta	O max	2.10		2.09	2, 15	2.20	2.21	2.24	2, 19	2.23	2.19	2.27
ΦO	Omin	0.17		. 0.17	. 71.0	0.17	0.18	0.18	0.18	0.19	0.19	0.18
vellow	O max	1.04		1.39	1.62	2.04	2.04	2.18	2.22	2.25	2.23	2.29
1	/	to blu	light for 0.0 sec	(2) 1 sec	(3) 2 sec	(d) 5 sec	(5) 10 sec	(B) 20 sec	Ø 40 sec	(B) 80 sec	(a) 160 sec	(D . 280 sec



The relative magnitudes of the photographic effects of the overall exposures (1) to (10) were determined by the following procedures.

Some of the test pieces were developed, bleached, fixed and washed with water as shown above except that neither wedge exposure nor overall exposure was applied. The image densities obtained were 0.12 (yellow), 0.10 (magenta) and 0.07 (cyan).

Other test pieces were not subjected to wedge exposure but were given an overall exposure according to schedule (1), with an ND filter capable of stepwise change in density from 0.2 to 3.0 at intervals of 0.2 being disposed between the light sources and each test piece. The procedures of development, bleaching, fixing and washing were the same as those employed in the processing of the samples that had been subjected to wedge exposure. The image densities obtained with the test pieces were measured and the density of the ND filter that was required to provide a density 0.2 higher than the density of each of the yellow, magenta and cyan images on the test pieces that were not given any overall exposure was also measured. The results were: 0.85 (yellow), 2.70 (magenta) and 2.40 (cyan). Therefore, the relative magnitudes of the photographic effects of overall exposure (1) were calculated as follows:



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$$Eb/Eg = 10^{-0.85}/10^{-2.7}$$

$$Eg/Er = 10^{-2.7}/10^{-2.4}$$

$$Er/Eb = 10^{-2.4}/10^{-0.85}$$

Overall exposure (1) was outside the scope of the present invention since Eb/Eg > 20 and Er/Eb < 0.050.

Similarly, the relative magnitudes of the photographic effects exerted by each of the overall exposures (2) to (10) on the test pieces were determined. The results are shown in Table 8.

Table 8

Overall exposure	Eb/Eg	Eg/Er	Er/Eb	Remarks
1	70.8	0.50	0.028	x
2	68	0.45	0.033	Х
3	33	0.43	0.070	Х
<b>4</b>	13.6	0.40	0.184	0
(5)	6.4	0.43	0.36	0
6	3.0	0.45	0.74	0
7	1.7	0.50	1.18	0
8	1.25	0.43	1.86	0
9	0.52	0.45	4.3	0
0	0.32	0.50	6.3	0





As the above data shows, a positive image having a high maximum density and a low minimum density for each of the ays images can be obtained by performing an overall exposure under the conditions that satisfy the requirement specified by the present invention.

## Example 4

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A core/shell emulsion was prepared in the following manner: to an aqueous solution of gelatin that was controlled at 50°C, equimolar amounts of aqueous solutions of silver nitrate and potassium bromide were added simultaneously by the controlled double-jet method over a period of 70 minutes so as to obtain a silver bromide emulsion comprising 0.8- $\mu m$ cubic grains; to the so obtained core emulsion, 1.0 mg of sodium thiosulfate and 2.0 mg of potassium chloroaurate, each being based on one mole of silver, were added and chemical ripening was conducted at 60°C for 170 minutes; to the so treated emulsion, aqueous solutions of silver nitrate and potassium bromide were added simultaneously to obtain a silver bromide emulsion comprising 1.0-µm cubic grains. To the so prepared core/shell emulsion, 1.0 mg of sodium thiosulfate and 1.5 mg of potassium chloroaurate, each being based on one mole of silver, were added and chemical ripening was conducted at 60°C for 120 minutes. The thus obtained emulsion was referred to as Emulsion D. This emulsion D was spectrally sensitized with dye (IV)



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(same as used in Example 1), so as to form a blue-sensitive emulsion.

A coated light-sensitive material was prepared by repeating the procedures of Example 3 except that the above-obtained emulsion was used as a blue-sensitive emulsion.

The test pieces of the coated sample were subjected to wedge exposure as in Example 3 and subsequently given an overall exposure according to schedules (1) to (10). They were developed, bleached, fixed and washed as in Example 3 and the maximum and minimum densities of the positive images formed were measured. The results are shown in Table 9 together with the overall rating the image quality.

Some of the test pieces were processed as in Example 3 to determine the relative magnitudes of the photographic effects exerted by applying an overall exposure in accordance with schedules (1) to (10). The results are shown in Table 10.



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Table 9

	low Dmin		mage Опах	magenta nax Dmin	Cyan		Image quality
2.01 0.17	0.1	_	2.13	0.15	2.20	0.09	0
2.04 0.17	0.1	_	2.09	0.15	2.21	0.09	0
2.15 0.18	0.1	8	2.20	0.16	2.09	0.09	0
2.21 0.17	0.1	1	2.24	0.15	2.15	0.08	0
2.24 0.17	0.1	1	2. 19	0.17	2.14	0.09	0
2.29 0.19	0.1	6	2.21	0.17	2.16	0.10	0
2.07 0.27	0.2	7	2.17	0.16	2.22	0.09	X (yellow had
- <del></del>							high Dmin )
2.02 0.	0.	0.34	2.25	0.17	2.17	0.09	× ( do. )
1.95 0.41	0.	41	2.20	0.15	2.23	0.11	× ( do. )
2.05 0	·-	0.38	2.21	0.18	2.25	0.09	X ( do. )

Table 10

Overall exposure	Eb/Eg	Eg /Er	Er/Eb	Remarks
①	6.8	0.44	0.33	0
2	3.1	0.47	0.69	0
3	1.1	0.40	2.27	0
4	0.59	0.50	3.4	0
<b>(5)</b>	0.24	0.47	1.3	0
6	0.17	0.44	14.7	0
7	0.088	0.46	24.7	. <b>X</b>
8	0.040	0.49	51	Х
9	0.015	0.42	158	Х
0	0.015	0.40	167	Х

As the above data shows, a positive image having a high maximum density and a low minimum density for each dye image can be obtained by applying an overall exposure under the conditions that satisfy the requirement specified by the present invention. Different light-sensitive materials were used in Examples 3 and 4, so that the range of the conditions for overall exposure that ensured the production of a satisfactory positive image differed between the two samples. With this fact taken into consideration, the foregoing data also shows that the method of the present invention enables a satisfactory image to be obtained irrespective of the type of light-sensitive material whereas this is a very difficult result to attain if methods that are outside the scope of the present invention are used.

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Example 5

A light-sensitive material was prepared as in Example 4 and this sample was subjected to wedge exposure with a sensitometer. It was then given an overall exposure for 20 seconds with a white fluorescent lamp being used as a light source. In another run, the same sample was given an overall exposure for 20 seconds with the same fluorescent lamp, with the light being passed through a magenta filter (green density = 1.0) for 15 seconds after the lapse of 5 seconds from the start of exposure.

The test pieces were developed, bleached, fixed and



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washed with water as in Example 3. The maximum and minimum densities of the positive image formed in each test piece were measured, and the results are shown in Table 11. The relative magnitudes of the photographic effects exerted by applying an overall exposure in the two different manners are shown in Table 12.

Table 11

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I	[mage	Yel	low	Mage	nta	Су	an	Image	
Overall exposure		Dmax	Dmin	Dmax	Dmin	Dmax	Dmin	quality	
White fluore	escent	1.95	0.17	1.85	0.35	2.17	0.10	X (magenta had high Dmin)	
White fluore lamp + mager filter		2.00	0.19	2.20	0.15	2.25	0.09	0	

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Table 12

Overall exposure	Eb/Eg	Eg/Er	Er/Eb	Remarks
White fluorescent lamp	35	0.069	0.41	Outside the scope of the invention
White fluorescent lamp + magenta filter	8.3	0.22	0.55	Within the scope of the invention

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The above data shows that a positive image having a high maximum density and a low minimum density can be obtained by applying an overall exposure in accordance with the present invention.



It should be noted here that in Examples 3 to 5 both the energy distribution and level of illumination were allowed to change during the overall exposure.

# Example 6

A sample of light-sensitive material was prepared as in Example 4 and subjected to wedge exposure with a sensitometer. The sample was subsequently processed as in Example 3 except that the condition of overall exposure was changed to the following: 20 seconds after it was submerged in the developer, the sample was given an overall exposure for 5 seconds under a while fluorescent lamp at 1 lux, then given an overall exposure for 10 seconds under a tugsten lamp at 1 lux.

The maximum and minimum densities of the positive image obtained were measured as in Example 3, and the results are shown in Table 13 together with the overall rating of the image quality. Some of the test pieces were reserved for determining the relative magnitudes of the photographic effects exerted by the overall exposure as in Example 3, and the results are shown in Table 14.

Table 13

Image	Yel	.low	Mage	nta	Су	an	Image
Overall exposure	Dmax	Dmin	Dmax	Dmin	Dmax	Dmin	quality
White fluorescent lamp + tungsten lamp	2.02	0.19	2.17	0.17	2.21	0.09	0

Table 14

Overall exposure	Eb/Eg	Eg/Er	Er/Eb	Remarks
White fluorescent lamp + tungsten lamp	2.8	2.4	0.15	Within the scope of the invention

The data in Tables 13 and 14 show that a satisfactory positive image having a high maximum density and a low minimum density can be obtained by applying an overall exposure in accordance with the present invention even if only the energy distribution of radiation is changed and its level of illumination maintained constant throughout the duration of the overall exposure.

## WHAT IS CLAIMED IS:

- 1. In a method of forming a direct-positive image on a silver halide photographic material by first giving an imagewise exposure, and then applying an overall exposure either prior to or during subsequent development, said photographic material having on a support two or more silver halide emulsion layers which have different wavelength ranges of sensitivity and each of which contains internal latent-image forming silver halide grains that are not pre-fogged on the surface, the improvement wherein the intensity of said overall exposure is changed at least once during said overall exposure and each of the relative magnitudes of the photographic effects exerted by said overall exposure on the respective silver halide emulsion layers is no greater than 20.
- 2. A method according to Claim 1 wherein said intensity of the overall exposure is the level of illumination of radiation.
- 3. A method according to Claim 1 wherein said intensity of the overall exposure is the energy distribution of radiation.
- 4. A method according to Claim 3 wherein both the energy distribution and the level of illumination of radiation are changed at least once during said overall exposure.

- 5. A method according to any one of the preceding Claims wherein each of the silver halide emulsion layers having different wavelength ranges of sensitivity is optically sensitized with a sensitizing dye.
- 6. A method according to any one of the preceding
  Claims wherein said silver halide emulsion layers having
  different wavelength ranges of sensitivity are a bluesensitive silver halide emulsion layer capable of forming
  a yellow dye image, a green-sensitive silver halide emulsion
  layer capable of forming a magenta dye image, and a redsensitive silver halide emulsion layer capable of forming
  a cyan dye image.

