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(54) **An electrophotographic copying method and insulating paper for use therein.**

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Description

This invention relates to an electrophotographic copying method for providing copies in which the light and dark areas of images and background of an original are formed in a reverse manner and also relates to black insulating paper for electrophotography which can be used in such method. In particular, this invention relates to a copying method of such type in which an ordinary positive-image copying apparatus is used.

Among electrophotographic copying systems, positive-image copying systems and negative-image copying systems are employed. In a positive-image copying system, which is usually used, a positive original having dark images, letters or the like, and a bright background which represents the area of the original excluding the area in which the images are formed is used, latent images corresponding to the images of the original are formed on a photosensitive member, and then developed by a black toner. The developed images are transferred to transfer paper, for example a white paper, and then fixed thereon, so that a positive copy having dark images and a bright background as in the original is obtained. In a negative-image copying system, by using a negative original such as a microfilm having bright images and a dark background or by forming images of light beams emitted from a laser or LED array, a latent image of the background is formed, and thereafter the areas of a photosensitive member other than that of the latent image are developed by a black toner, the developed toner images are transferred to transfer paper, for example a white paper, and fixed thereon, whereby the light and dark areas of images and of background are reversed to obtain a positive copy.

Most copying apparatus are designed to employ only the positive-image copying system, in other words, produce only a positive copy having a bright background from a positive original having a bright background. Therefore, these copying apparatus cannot produce a positive copy having a bright background from a negative original having a dark background. In producing some slides and pamphlets or catalogs, negatives having a dark background and bright images are used. When such a negative original is copied by a conventional positive-image copying system, the obtained copy is negative and hence it is difficult to read. Therefore, a technique by which a positive copy can be obtained from such a negative original is required.

In recent years, copying apparatus which employ both the positive-image copying system and the reversed image copying system have been proposed. Such copying apparatus can be roughly classified into the following two categories. The first system uses a photosensitive member which can be charged positively or negatively, a charger which can positively or negatively charge the photosensitive member, and a toner having a triboelectrification characteristic of a predetermined polarity. In this system, when positive-image copying is to be done, development is conducted while the photosensitive layer is charged with the polarity opposite to that of the toner, and when reverse image copying is to be done, development is conducted while the photosensitive layer and the toner are charged with the same polarity, and a bias voltage is applied to the developing region. Because the materials useful in manufacturing a photosensitive member which can be positively or negatively charged are few, the sensitivity and durability of the member are restricted. Moreover, it is extremely difficult to control the density and image quality of both positive images and reversed images so as to be uniform, because the sensitivity and other electrophotographic characteristics of a photosensitive member when being charged positively are greatly different from those of the member when being charged negatively.

The other method uses a photosensitive member which can be charged with a predetermined polarity and a toner which can be charged positively or negatively. In this method, the photosensitive member is charged, and exposed to the images of an original to form, on the member, latent images which correspond to the images of the original. Thereafter, when positive images are to be formed, the latent images are developed by a toner which has been charged with a polarity opposite to that of the latent images, without applying a bias voltage to the developing region, and, when reversed images are to be formed, the background area (the area other than the areas in which the latent images are formed) is developed by a toner which has been charged with a polarity the same as that of the photosensitive member, while applying a bias voltage to the developing region, whereby reverse development is conducted. However, with this method, two kinds of toners must be used for developing positive images and reversed images, and, according to the situation, electrical control such as application of a bias voltage to a developing region must be conducted. For these reasons, it is difficult to construct and control a copying apparatus having both a positive-image copying system and a reversed-image copying system.

Therefore, an object of this invention disclosed herein is to provide an electrophotographic copying method which can produce reversed images from an original without requiring a special device or a control means.

The electrophotographic copying method of this invention, which overcomes the above-discussed and numerous other disadvantages and deficiencies of the prior art, comprises : using a copying apparatus having a positive-image copying system ; forming toner images with colored insulating paper of a color different to that of said toner ; and fixing the transferred images to the paper ; in which method the colors of the paper and the toner are selected so as to form reverse images.

In preferred practice, the colored toner is a white toner. Moreover, the colored paper is preferably obtained by forming an area of a desired color on a transfer paper by use of an electrophotographic copying apparatus.

Alternatively, there can be used as the colored paper a black insulating paper with a surface specific resistance of $10^9\ \Omega$ or more and a volume specific resistance of $5 \times 10^9\ \Omega \cdot$ cm or more.

This invention thus also provides black insulating paper for electrophotography to which toner images produced by a colored toner, the color of which is paler than that of said black insulating paper, can be transferred and fixed to form reverse images by using a positive-image electrophotographic copying apparatus, in which method the surface specific resistance of said paper is $10^9\ \Omega$ or more and the volume specific resistance of said paper being $5 \times 10^9\ \Omega \cdot$ cm or more.

In one preferred embodiment, said black insulating paper is obtained by mixing an insulating black pigment and a beaten pulp in a step in which mixing of said pulp with one or more of inter alia, a sizing agent, a filler agent and dyes, takes place. Alternatively, said black insulating paper may be obtained by applying an insulating transparent resin to conductive black paper or by impregnating conductive black paper with the resin. The black insulating paper may also be obtained by laminating an insulating transparent film on conductive black paper. Indeed, according to a further preferred procedure, said black insulating paper is obtained by printing or applying an insulating black pigment to white paper, or impregnating white paper with the pigment.

The insulating black pigment when used is preferably a black toner having preferably a volume mean particle size of 5 μm or less. The insulating black pigment may be obtained by coating a conductive black pigment with a transparent insulating resin. The conductive black pigment itselt is preferably selected from carbon black, graphite, cyanine black, $CuCr_2O_4$ and $CuFe_2O_4$-$CuMn_2O_4$.

This invention meets the aforementioned object and provides an electrophotographic copying method for easily obtaining an easy to read positive copy from a negative original. Even if documents produced by a printer or a facsimile apparatus of a thermal transfer type are lost, the copying method of this invention can produce positive images by copying a transfer film having negative images which have been used in producing the documents, thereby allowing the documents to be reproduced. This invention also provides an electrophotographic copying method which can obtain a positive copy from a negative original, and, therefore, is applicable in the fields of design, advertising and printing. The electrophotographic copying method of this invention can easily and reliably produce reversed images on an ordinary white paper.

The black insulating paper of this invention is suitable for use in the above-mentioned methods. The black insulation paper for electrophotography can be made by a simple process and gives a clear image without the danger of wrinkling, curling or the like. Finally, the black insulating paper of this invention is very economical.

The invention will now be described in greater detail, reference being made to the accompanying drawings, wherein :

FIGURE 1 is a graph showing the relationship between the surface specific resistance of a transfer paper and the transfer efficiency when electrophotographic copying is conducted using the transfer paper ;

FIGURE 2 is a graph showing the relationship between the surface specific resistance of a transfer paper and the occurrence of voids in toner images when electrophotographic copying is conducted using the transfer paper ;

FIGURE 3 shows a side sectional view of one example of a copying apparatus which may be used in performing the method of this invention ; and

FIGURE 4 shows a perspective view of portions of the apparatus shown in Figure 3.

The electrophotographic copying method of this invention can be performed with a conventional electrophotographic copying apparatus which has a positive-image copying system. According to this invention, when a positive copy having dark images and a bright background is obtained from a negative original having bright images and a dark background with, for example, an electrophotographic copying apparatus having a positive-image copying system, a colored toner and colored paper are used, the color of the toner being of a relatively low density, and the color of the paper being of a density higher than that of said colored toner. The electrophotographic copying apparatus in which the colored toner and colored paper are set operates in a usual manner as follows : a latent image formed on the photosensitive member, corresponding to the background of the original, is developed by the colored toner the color of which is of a relatively low density, the resulting toner image being transferred to the colored paper the color of which is of a density higher than that of said colored toner, after which the toner image on the colored paper is fixed, thereby obtaining a copy in which the light and darkness of the original are reversed. In this way, according to this invention, a reversed copy can be obtained without a special device or electrical control.

When a positive copy is obtained from a negative original, a white toner is suitably used as the colored toner for developing latent images. Colored paper to which toner images formed by a colored toner are to be transferred and fixed can be obtained by forming a region of a desired color on the paper with a colored toner (for example, a black toner).

A specific example of the method of this invention in which colored paper having a desired-color region thereon formed by a black toner is used as the colored paper, and a white toner is used as the colored toner will be described below.

As the copying apparatus, an ordinary positive-image copying apparatus is used. As the colored toner, for example, a black toner which is used in a conventional copying apparatus is used. First, a completely black original is placed on a document table, alternatively nothing is placed on the table with the table cover kept open. Then, copying is done under usual conditions for copying with a black toner. In this way, colored paper (i.e., white paper one face of which is coated in its entirety with the black toner fixed on it) is obtained. White transfer paper useful in a copying apparatus of the electrostatic transfer type has insulating properties sufficient for being electrostatically transferred. Therefore, with colored paper obtained by transferring a black toner containing carbon (which is conductive) to the whole of one surface of white transfer paper, the two sides of the paper are electrically insulated from each other. When reversed images are to be formed on only one part of colored paper, one part of a white transfer paper is colored in black by using a special original which has been partially colored in black, or, using a copying apparatus having a size-variable function and a special original which is black in its entirety, the desired part of the white transfer paper is colored in black by reducing or magnifying the size of the images of the original.

Once colored paper is formed in this way, the toner to be used in the copying apparatus is changed from black toner to white toner. Then, the colored paper is set as transfer paper in the copying apparatus, a negative original from which a reversed copy is to be formed is placed on the document table, and thereafter copying is conducted. As a result of the above, a positive copy is obtained in which the area thereof corresponding to the background of the original is colored in white by the white toner and the areas corresponding to the images of the original are colored in black by the black-colored paper.

Many recent copying apparatus have a developing unit which can be replaced. In such copying apparatus, a plurality of developing units each of which contains a toner of a color different from each other can be used, and copies by a different color can be obtained by using each of the units. When the method of this invention is to be performed using such copying apparatus, two developing units are prepared, one containing a black toner and the other containing a white toner. Then, as described above, colored paper is made using the developing unit containing a black toner, and thereafter the developing unit is replaced by the other developing unit containing a white toner, and, for example, a positive original which is to be copied is placed on a document table. Under these conditions, when the colored paper with the black toner fixed thereon is supplied again to the copying apparatus, white toner images corresponding to the images of the original are transferred on the colored paper and fixed to the colored paper to form a negative copy in which the images are bright and the background is dark.

Recently, copying apparatus which have two developing units and develop two colors have been developed. These two developing units contain, for example, a black toner and a red toner, respectively, so that it is possible to obtain copies having black images or red images according to the image of an original or to the purpose of the copies. When such two-color copying apparatus are used in performing the method of this invention, a black toner is placed in one of the two developing units and a white toner in the other one, enabling a reversed copy to be easily obtained as described in the above.

According to the method of the present invention, a positive copy having dark images and a bright background can be obtained from a negative original having bright images and a dark background, and also a negative copy having bright images and a dark background can be obtained from a positive original having dark images and a bright background. When a positive copy is to be obtained from a negative original, the colored paper which constitutes the background of a copy is not limited to being white, and may be formed using a toner of a pale color such as yellow, or light blue, and as the colored toner which constitutes images such as letters in a positive copy, in addition to a black toner, a toner of a dark color such as red, blue, or green can be used. When a negative copy is formed from a positive original, the colored toner which constitutes images of the copy is composed of a white toner or a toner of a pale color such as yellow, or light blue, the colored toner which constitutes the background is composed of a black toner or a toner of a dark color such as red, blue, or green.

In the method of this invention, as the colored paper, in addition to a transfer paper having a colored region formed by a colored toner as described above, it is also possible to use a black insulating paper which has a surface specific resistance of $10^9\ \Omega$ or more and a volume specific resistance of $5 \cdot 10^9\ \Omega \cdot$ cm or more, because of the reasons mentioned below.

In electrophotography, the amount of the electric resistance of a transfer paper greatly affects the transfer efficiency of toner images or latent images and also the quality of the copied images. In other words, if the electric resistance of a transfer paper is not sufficiently high, charges from a transfer corotron or a bias roll flow into the ground through the transfer paper, which prevents the formation of an electric field sufficient for transfer,

and, therefore, the transfer efficiency is reduced. Also, if the electric resistance of a transfer paper is too low, charges will leak via the transfer paper to lower the level of the transferring field, thereby producing voids in the toner images. The relationship between the surface specific resistance (Ω) of a transfer paper and the transfer efficiency (%) is shown in Figure 1. The relationship between the surface specific resistance (Ω) of a transfer paper and the occurrence (%) of voids in toner images is shown in Figure 2. As apparent from Figures 1 and 2, if the surface specific resistance decreases below 10^9 Ω, the transfer efficiency markedly decreases, and the occurrence of voids in the toner images sharply increases. If the volume specific resistance decreases below 5×10^9 Ω · cm, the transfer efficiency decreases and the occurrence of voids in toner images rapidly increases.

In view of these phenomena, black insulating paper having a surface specific resistance of 10^9 Ω or more and a volume specific resistance of 5×10^9 Ω · cm or more can be used as colored paper in the method of this invention. If black insulating paper having a surface specific resistance of less than 10^9 Ω or a volume specific resistance of less than 5×10^9 Ω · cm is used in copying, the transfer efficiency will decrease and voids in the toner images will occur, and, therefore, resulting copies will be unclear.

The black insulating paper can be made, for example, by mixing an insulating black pigment (e.g., a black toner) with pulp in a known process of manufacturing paper. In the process of manufacturing paper, pulp is suspended in water, and then beaten. The beaten pulp is mixed with a sizing agent, filler agent, dyes, etc., and then made into paper. The paper is rolled or cut to form rolled paper or cut paper. In the mixing step, the insulating black pigment is added together with a sizing agent, filler agent, dyes, etc. into the beaten pulp. The insulating black pigment can be obtained by coating a conductive black pigment such as carbon black, graphite, cyanine black, $CuCr_2O_4$ and $CuFe_2O_4$-$CuMn_2O_4$ with insulating transparent resin. Examples of the insulating transparent resin are polystyrene ; styrene copolymers such as styrene-butadiene copolymer and styrene-acrylic copolymer ; polyethylene ; ethylene copolymers such as ethylene-vinyl acetate copolymer, and ethylene-vinyl alcohol copolymer ; phenolic resin ; epoxy resin ; allylphtalate resin ; polyamide resin ; polyester resin ; and maleic acid resin. The conductive black pigment can be coated by the same method as is used in manufacturing conventional toners for electrophotography. In this method, a conductive black pigment and an insulating transparent resin are mixed in a mixer, and then the mixture is heated and fused in a heated roll mill to be kneaded. The kneaded product is pulverized in a feather mill or a jet mill, and the resulting powder is classified to obtain insulating black pigment of the desired particle size. It is preferable to conduct the classification so that the volume mean particle size of obtained pigment is 10 μm or less. If an insulating black pigment having a mean particle size larger than 10 μm is used in electrophotography, a clear copy will not be obtained.

An example of the insulating black pigment is a black toner. When a black toner is used as the insulating black pigment, the black toner can be prepared by a method for manufacturing a conventional toner for electrophotography as mentioned above. The above-mentioned insulating transparent resin can be used also as a binder resin. The black toner thus obtained preferably has a volume mean particle size of 5 μm or less. In a conventional process for manufacturing toner, finely pulverized black toner (with a volume mean particle size of 5 μm or less) can be separately obtained, and the use of the separated black toner as the insulating black pigment is economical.

The black insulating paper can be obtained also by applying an insulating transparent resin to conductive black paper or by impregnating the resin into the paper. As the insulating transparent resin, the same resin which is used in coating the conductive black pigment can be used. The resin is, in general, dissolved in an appropriate solvent, and then the solution is applied to or impregnated into conductive black paper. The black insulating paper can be obtained also by laminating an insulating transparent film on conductive black paper. Useful examples of the insulating transparent film are films made from polyethylene terephthalate, poly-4-methylpentene-1, polyolefin resin or the like. Alternatively, the insulating transparent film can be made by forming a film of any of the above-mentioned resins on both sides of conductive black paper, using a laminating method such as an extrusion laminating method.

The black insulating paper can also be made by printing or applying an insulating black pigment on white paper or by impregnating such a pigment into white paper. As the insulating black pigment, a black toner can be suitably used. A black toner having a volume mean particle size of 5 μm or less is preferably used. An example of the white paper is conventional transfer paper which is used in an electrophotographic copy. There can be used as the insulating black pigment other than a black toner, the same material as the insulating black pigment which is mixed with pulp in the above-mentioned method of preparing black insulating paper. The insulating black pigment also can be prepared by coating a conductive black pigment such as carbon black, graphite, etc., with an insulating transparent resin, as described above. As the insulating transparent resin, the resin such as those listed above can be used.

Even if the black insulating paper obtained in this way is used in a conventional electrophotographic copying process, problems of bad transferring will not arise. Therefore, the black insulating paper can be used also in

the method of this invention in which a reversed copy is formed with a conventional copying apparatus. An example of the method of this invention in which said black insulating paper is used is described below.

First, by exposing a positive original having dark images and a bright background, latent images corresponding to the images of the original are formed on a photosensitive member, and the latent images are developed by a white toner. Then, the white toner images are electrostatically transferred onto the black insulating paper, and thermally fixed thereto. As a result, white images corresponding to the images of the original are formed on the black insulating paper, i.e., a negative copy is obtained. In the electrophotographic copying process, no special operations other than using a white toner are necessary.

When a white toner is used as a colored toner in the electrophotographic copying method of this invention, the white toner fixed to the colored paper must cover or conceal the colored paper. It is preferable that such a white toner has a concealment index of 5 or more (as evaluated by the method described below) so that the color of the colored transfer paper (e.g., colored in black) is sufficiently concealed.

Method for evaluating the concealment index of a white toner :

Using a black toner, an original, one face of which is black in its entirety, is copied onto white transfer paper so that the reflection density of the obtained black paper is 1.5. To measure the reflection density, for example, a reflection density meter TC-6D (made by Tokyo Denshoku) is used. Then, images are formed by a white toner on the colored transfer paper to which a black toner has been fixed. When the reflection density of the images formed by the white toner is X, the concealment index is evaluated by the following formula which has been proposed by the present inventors :

$$\text{Concealment index} = 1.5/X.$$

A white toner useful as a colored toner in this invention should have a concealment index of 5 or more as calculated above. Such a white toner may contain a binding resin, white coloring agent, and any other compounding ingredients such as an antistatic agent and a release agent. Their composition ratios are not restricted unless they exert a bad influence on the tone of the toner. In general, it is preferable that the weight ratio of a white coloring agent and a binding resin is in the range of 2 : 100 to 35 : 100. If the portion of the white coloring agent is below this range, the concealment index of the white toner obtained against a colored paper will be insufficient. If the portion is above this range, the triboelectrification property of the white toner will be poor, and spent toners will be more readily generated, thereby reducing the life span of the developing agent.

Useful Examples of the white coloring -agent for the white toner are zinc white, titanium dioxide, tin oxide, antimony white, zinc sulfide, zinc oxide, barium carbonate, clay, silica, white carbon, talc, alumina, baryte, etc.

Examples of the binding resin for the white toner are various resin materials which are known for use as binding resins for toners in electrophotography.

An antistatic agent which can be used for the white toner is any appropriate known material which does not produce a deleterious effect on the tone of a white toner.

Examples of antistatic agents which inhibit positive charging are organic compounds having basic nitrogen such as basic dyes, aminopyrine, pyrimidine compounds, polynuclear polyamine compounds, aminosilanes, and filler agents the surface of which is treated by these substances. Examples of antistatic agents which inhibit negative charging are compounds containing carboxyl groups such as a metal chelate of an alkylsalicylic acid. These antistatic agents are preferably used in a 1 to 10% proportion by weight in a toner.

Examples of release agents for the white toner are silicone oil, olefin resins of low molecular weight, and various kinds of waxes.

The white toner can be obtained by the following processes : the above-mentioned components are fused and kneaded, and after cooling the product is pulverized and classified. Also, the white toner can be obtained by dispersing the above-mentioned components in a resin solution and by applying it in a spray formulation whereby a granulate is produced.

When, using the electrophotographic copying method of this invention, a reversed copy is obtained from an original which is a transparent sheet with images formed thereon, it being possible to place the original on a document table and expose it from the rear side (which is opposite the face of the table) so as to provide a copy having excellent reproducibility. In other words, the exposure is conducted in the direction perpendicular to the surface of the original, whereby the clarity of the images can be improved.

Figure 3 shows an example of a copying apparatus which can be used in performing this method. The copying apparatus is provided with a photosensitive drum (hereinafter, referred as "drum") 10 which is disposed in the center of the apparatus and can rotate in the direction shown by an arrow A. In the area surrounding the drum 10, the following devices are disposed successively : a charger 20 for uniformly charging the drum 10 ;

an exposure system 30 for exposing an original 37 placed on a document table 35 to form latent images corresponding to images of the original ; a surface exposure device 80 which is disposed at the rear side of the original 37, independently from the exposure system 30 ; a developing device 40 for developing toner images from the latent images formed on the drum 10 ; a transfer device 50 for transferring the toner images on the drum 10 to transfer paper ; a separator 55 for separating the transfer paper from the drum 10 ; and a cleaning device 60 for removing residual toner on the drum 10 after the transfer of the toner images to the transfer paper. The surface exposure device 80 disposed at the rear side of the original 37 comprises, as shown in Figure 4, a wide exposure surface 81 and a glass screen 82. The device 80 is mounted so that the exposure surface 81 covers the original 37. After the toner images on the transfer paper are fixed by a fixing device 70, the transfer paper is discharged.

When using the apparatus, the exposure of the images is conducted perpendicularly from the rear side of the original 37 so that the light corresponding to the original 37 is transmitted perpendicularly. The transmitted light is introduced to the drum 10 via a movable lens 32 and mirrors 33 to form latent images. For this reason, the obtained images are clear.

The following Examples illustrate this invention.

Example 1

(1) Preparation of a white toner :

Binding agent: styrene-acrylic resin
·····100 parts by weight

Coloring agent: titanium oxide (median volume particle size: 0.3μ, anatase type)
·····20 parts by weight

Antistatic agent: zinc complex of alkylsalicylic acid (Bontron E84 made by Orient Kagaku)
·····3 parts by weight

Releasing agent: polypropylene of low molecular weight (Biscol 550P made by Sanyo Kasei; Mw 15000)
·····2 parts by weight

The above-listed agents were mixed for 5 minutes in a Henshel mixer FM10B (made by Mitsui Miike). Then the mixture was fused and kneaded in a twin-screw extruder PCM-30 (made by Ikegai Tekko) and allowed to stand for cooling. The kneaded product was coarsely pulverized by a cutting mill, then finely pulverized by a jet pulverizer, and classified to obtain a white toner having a volume mean particle size of 11 to 12 μm.

(2) Preparation of a white developer :

Seven parts by weight of the white toner prepared above and 93 parts by weight of an iron powder carrier (STV25T made by Nippon Teppun) were mixed in a bowl mill to obtain a white developer.

(3) Formation of a reversed copy :

The white developer was charged into a copying apparatus (DC-111 made by Mita Industrial Co., Ltd.). As transfer paper, three kinds of paper which were colored in black, red and navy blue, respectively were used. A negative original having bright images and a dark background was placed on the document table, and copying operations were conducted. Clear positive reversed copies were obtained in which the background of the original were developed by the white toner and the images were displayed by the color (black, red or navy blue) of the colored paper. The values of the reflection density of the images developed by the white toner and those

of the background displayed by the surface of the colored paper are shown in the following table.

Reflection density

	Background	Image
Black	1.50	0.15
Red	0.70	0.11
Navy blue	1.20	0.13

Example 2

(1) Preparation of black insulating paper for electrophotography :

Styrene-acrylic copolymer (Priolite ACL made by Goodyear) 100 parts by weight

Carbon black (Printex L made by Degusa) 20 parts by weight

The above-listed materials were premixed in the Henshel mixer of Example 1. Then the mixture was fused and kneaded in the twin-screw extruder of Example 1 and allowed to stand for cooling. The kneaded product was coarsely pulverized by a cutting mill, then finely pulverized by anultrasonic jet mill PJM-100 to obtain an insulating black pigment having a volume mean particle size of 8 μm.

The insulating black pigment was mixed, into beaten pulp together with a sizing agent, filler agent, dyes, etc. Then, by a well known process of paper manufacture, black insulating paper was made.

The black insulating paper obtained had a surface specific resistance of $1.5 \times 10^{10}\ \Omega$, and a volume specific resistance of $2.1 \times 10^{11}\ \Omega \cdot$ cm.

(2) Formation of a reversed copy :

Using the black insulating paper as transfer paper, the white toner made in in Example 1, and a usual positive-image copying apparatus (DC-111 made by Mita Industrial Co., Ltd.), white images were formed on the black insulating paper. The resulting white images were clear, and no wrinkles or curls were observed in the images.

Example 3

(1) Preparation of black insulating paper for electrophotography :

The same procedures as those of Example 2 were conducted except that 20 parts by weight of carbon black and 2.5 parts by weight of polypropylene of low molecular weight (Biscol 550P made by Sanyo Kasei) were used to obtain a black toner having a volume mean particle size of 9 μm.

The black toner was mixed into beaten pulp, together with a sizing agent, filler agent, dyes etc.

Then, by a well-known process of paper manufacture, black insulating paper was made.

The obtained black insulating paper had a surface specific resistance of $1.5 \times 10^{10}\ \Omega$, and a volume specific resistance of $2.6 \times 10^{11}\ \Omega \cdot$ cm.

(2) Formation of a reversed copy :

Using the black insulating paper as transfer paper, the white toner made in Example 1, and a conventional

positive-image copying apparatus (DC-111 made by Mita Industrial Co., Ltd.), white images were formed on the black insulating paper. The resulting white images were clear, and no wrinkles or curls were observed in the images.

Example 4

(1) Preparation of black insulating paper for electrophotography :

Black toner obtained in the same manner as that in Example 3 was further classified by an alpine classifier to obtain a black toner having a volume mean particle size of 3.5 μm.

The black toner was mixed into beaten pulp, together with a sizing agent, filler agent, dyes etc.

Then, by a well-known process of paper manufacture, black insulating paper was made.

The black insulating paper obtained had a surface specific resistance of $1.8 \times 10^{10}\ \Omega$, and a volume specific resistance of $3.2 \times 10^{11}\ \Omega \cdot$ cm.

(2) Formation of a reversed copy :

Using the black insulating paper as transfer paper, the white toner made in Example 1, and a conventional positive-image copying apparatus (DC-111 made by Mita Industrial Co., Ltd.), white images were formed on the black insulating paper. The resulting white images were clear, and no wrinkles or curls were observed in the images.

Example 5

(1) Preparation of black insulating paper for electrophotography :

A commercially available conductive black paper in which carbon black was dispersed and which had, after controlling the moisture at 20°C and 60% RH, a surface specific resistance of 1.5 to $2 \times 10^{6}\ \Omega$ and a volume specific resistance of $1.2 \times 10^{8}\ \Omega \cdot$ cm was coated with a methanol solution of polyvinyl butyral (20% by weight) so that a layer of 2 to 10 μm in thickness was formed on both surfaces of the conductive black paper.

The black insulating paper obtained had a surface specific resistance of $1.2 \times 10^{10}\ \Omega$, and a volume specific resistance of $1.8 \times 10^{11}\ \Omega \cdot$ cm.

(2) Formation of a reversed copy :

Using the black insulating paper as transfer paper, the white toner made in Example 1, and a conventional positive-image copying apparatus (DC-111 made by Mita Industrial Co., Ltd.), white images were formed on the black insulating paper. The resulting white images were clear, and no wrinkles or curls were observed in the images.

Example 6

(1) Preparation of black insulating paper for electrophotography :

The same procedures as those of Example 5 were carried out except that a methanol solution of polyvinyl pyrrolidone (20% by weight) was used instead of the methanol solution of polyvinyl butyral (20% by weight) to obtain black insulating paper.

The black insulating paper obtained had a surface specific resistance of $8.0 \times 10^{9}\ \Omega$, and a volume specific resistance of $1.5 \times 10^{11}\ \Omega \cdot$ cm.

(2) Formation of a reversed copy :

Using the black insulating paper as transfer paper, the white toner made in Example 1, and a conventional positive-image copying apparatus (DC-111 made by Mita Industrial Co., Ltd.), white images were formed on the black insulating paper. The resulting white images were clear, and no wrinkles or curls were observed in the images.

Example 7

(1) Preparation of black insulating paper for electrophotography :

The same procedures as those of Example 5 were carried out except that a methanol solution of a copolymer of acrylic ester and methacrylic ester (20% by weight) was used instead of the methanol solution of polyvinyl butyral (20% by weight) to obtain black insulating paper.

The black insulating paper obtained had a surface specific resistance of $3.0 \times 10^{10}\ \Omega$, and a volume specific resistance of $5.2 \times 10^{11}\ \Omega \cdot$ cm.

(2) Formation of a reversed copy :

Using the black insulating paper as transfer paper, the white toner made in Example 1, and a conventional positive-image copying apparatus (DC-111 made by Mita Industrial Co. Ltd.), white images were formed on the black insulating paper. The resulting white images were clear, and no wrinkles or curls were observed in the images.

Example 8

(1) Preparation of black insulating paper for electrophotography :

The insulating black pigment obtained in Example 2 was dissolved in toluene in a concentration of 25% by weight. The solution obtained was applied to conventional white transfer paper for electrophotography in a thickness of 5 μm.

The black insulating paper obtained had a surface specific resistance of $4.8 \times 10^{10}\ \Omega$, and a volume specific resistance of $5.6 \times 10^{11}\ \Omega \cdot$ cm.

(2) Formation of a reversed copy :

Using the black insulating paper as transfer paper, the white toner made in Example 1, and a conventional positive-image copying apparatus (DC-111 made by Mita Industrial Co., Ltd.), white images were formed on the black insulating paper. The resulting white images were clear, and no wrinkles or curls were observed in the images.

Example 9

(1) Preparation of black insulating paper for electrophotography :

The insulating black pigment obtained in Example 3 was dissolved in toluene in a concentration of 30% by weight. The solution was applied to conventional white transfer paper for electrophotography in a thickness of 5 μm.

The black insulating paper obtained had a surface specific resistance of $1.3 \times 10^{10}\ \Omega$, and a volume specific resistance of $2.4 \times 10^{11}\ \Omega \cdot$ cm.

(2) Formation of a reversed copy :

Using the black insulating paper as transfer paper, the white toner made in Example 1, and a conventional positive-image copying apparatus (DC-111 made by Mita Industrial Co., Ltd.), white images were formed on the black insulating paper. The resulting white images were clear, and no wrinkles or curls were observed in the images.

Example 10

(1) Preparation of black insulating paper for electrophotography :

The black toner obtained in Example 4 was dissolved in toluene in a concentration of 30% by weight. The solution was applied to a conventional white transfer paper for electrophotography in a thickness of 5 μm.

The black insulating paper obtained had a surface specific resistance of $1.4 \times 10^{10}\ \Omega$, and a volume specific

resistance of 2.8×10^{11} Ω · cm.

(2) Formation of a reversed copy :

Using the black insulating paper as transfer paper, the white toner made in Example 1, and a conventional positive-image copying apparatus (DC-111 made by Mita Industrial Co., Ltd.), white images were formed on the black insulating paper. The resulting white images were clear, and no wrinkles or curls were observed in the images.

Example 11

The same procedures as those of Example 1 were carried out, except that a transparent negative film (a transparent sheet) was used and the exposure was done from the rear side of the film to form reversed images. The images obtained were clear and excellent in reproducibility.

Claims

1. An electrophotographic copying method comprising using a copying apparatus having a positive-image copying system ; forming toner images with a colored toner ; transferring the toner images to a colored insulating paper of a color different to that of said toner ; and fixing the transferred images to the paper, characterised by selecting the colors of the paper and the toner so as to form reverse images.

2. An electrophotographic copying method according to claim 1, wherein said colored toner is a white toner.

3. An electrophotographic copying method according to claim 1 or 2, wherein said colored paper is obtained by forming an area of a desired color on a transfer paper by use of an electrophotographic copying apparatus.

4. An electrophotographic copying method according to claim 1, 2 or 3, wherein said colored paper is black insulating paper with a surface specific resistance of 10^9 Ω or more and a volume specific resistance of 5×10^9 Ω · cm or more.

5. An electrophotographic copying method according to claim 4, wherein said black insulating paper has been obtained by mixing an insulating black pigment and a beaten pulp in a step in which mixing of the pulp with one or more of a sizing agent, a filler agent and a dye takes place.

6. An electrophotographic copying method according to claim 4, wherein said black insulating paper has been obtained by applying an insulating transparent resin to conductive black paper or by impregnating conductive black paper with the resin.

7. An electrophotographic copying method according to claim 4, wherein said black insulating paper has been obtained by laminating an insulating transparent film on conductive black paper.

8. An electrophotographic copying method according to claim 4, wherein said black insulating paper has been obtained by printing or applying an insulating black pigment to white paper, or by impregnating white paper with the pigment.

9. An electrophotographic copying method according to claim 5 or 8, wherein said insulating black pigment is a black toner.

10. An electrophotographic copying method according to claim 9, wherein said black toner has a volume mean particle size of 5 μm or less.

11. An electrophotographic copying method according to claim 5 or 8, wherein said insulating black pigment is obtained by coating a conductive black pigment with a transparent insulating resin.

12. An electrophotographic copying method according to claim 11, wherein said conductive black pigment is selected from carbon black, graphite, cyanine black, $CuCr_2O_4$ and $CuFe_2O_4$-$CuMn_2O_4$.

13. Black insulating paper to which toner images produced by a colored toner, the color of which is paler than that of said black insulating paper, can be transferred and fixed to form reverse images by using a positive-image electrophotographic copying apparatus, characterised by the surface specific resistance of said paper being 10^9 Ω or more and the volume specific resistance of said paper being 5×10^9 Ω · cm or more.

14. Black insulating paper according to claim 13, wherein said paper has been obtained by mixing an insulating black pigment and a beacen raw-material pulp in a step in which mixing of said raw-material pulp with one or more of a sizing agent, a filler agent and a dye takes place.

15. Black insulating paper according to claim 13, wherein said paper has been obtained by applying an insulating transparent resin to conductive black paper or by impregnating conductive black paper with the resin.

16. Black insulating paper according to claim 13, wherein said paper has been obtained by laminating an insulating transparent film on conductive black paper.

17. Black insulating paper according to claim 13, wherein said paper has been obtained by printing or applying an insulating black pigment to white paper, or by impregnating white paper with the pigment.

18. Black insulating paper according to claim 14 or 17, wherein said insulating black pigment is a black toner.

19. Black insulating paper according to claim 18, wherein said black toner has a volume mean particle size of 5 µm or less.

20. Black insulating paper according to claim 14 or 17, wherein said insulating black pigment is obtained by coating a conductive black pigment with a transparent insulating resin.

21. Black insulating paper according to claim 20, wherein said conductive black pigment is selected from carbon black, graphite, cyanine black, $CuCr_2O_4$ and $CuFe_2O_4$-$CuMn_2O_4$.

Ansprüche

1. Elektrophotografisches Kopierverfahren, welches ein Verwenden eines Kopierapparates, der ein positiv-abbildendes Kopiersystem hat, ein Formen von Tonerbildern mit einem Farbtoner, ein Transferieren der Tonerbilder auf ein farbiges Isolierpapier einer zur Farbe des Toners verschiedenen Farbe und ein Fixieren der transferierten Bilder auf das Papier umfaßt, gekennzeichnet durch ein Auswählen der Farben des Papiers und des Toners, um Umkehrabbildungen zu formen.

2. Elektrophotografisches Kopierverfahren nach Anspruch 1, bei dem der Farbtoner ein weißer Toner ist.

3. Elektrophotografisches Kopierverfahren nach Anspruch 1 oder 2, bei dem das farbige Papier durch Formung eines Bereiches einer gewünschten Farbe auf einem Transferpapier durch Verwendung eines elektrophotografischen Kopierapparates erhalten wird.

4. Elektrophotografisches Kopierverfahren nach Anspruch 1, 2 ober 3, bei dem das farbige Papier schwarzes Isolierpapier mit einem spezifischen Oberflächenwiderstand von $10^9\ \Omega$ oder mehr und einem spezifischen Volumenwiderstand von $5 \times 10^9\ \Omega \cdot$ cm oder mehr ist.

5. Elektrophotografisches Kopierverfahren nach Anspruch 4, bei dem das schwarze Isolierpapier durch Mischung eines schwarzen Isolierpigmentes und eines Zellstoffblattes in einem Schritt erhalten wurde, in dem ein Vermischen des Zellstoffes mit einem oder mehrerèn aus einem Appreturmittel, einem Füllmittel und einem Farbstoff stattfindet.

6. Elektrophotografisches Kopierverfahren nach Anspruch 4, bei dem das schwarze Isolierpapier durch Auftragung eines transparenten Isolier-Kunstharzes auf leitendes schwarzes Papier oder durch Imprägnierung leitenden schwarzen Papieres mit dem Kunstharz erhalten wurde.

7. Elektrophotografisches Kopierverfahren nach Anspruch 4, bei dem das schwarze Isolierpapier durch Laminieren eines transparenten Isolierfilmes auf leitendes schwarzes Papier erhalten wurde.

8. Elektrophotografisches Kopierverfahren nach Anspruch 4, bei dem das schwarze Isolierpapier durch Bedrucken oder Auftragen eines isolierenden schwarzen Pigmentes auf weißes Papier oder durch Imprägnieren weißen Papieres mit dem Pigment erhalten wurde.

9. Elektrophotografisches Kopierverfahren nach Anspruch 5 oder 8, bei dem das schwarze Isolierpigment ein schwarzer Toner ist.

10. Elektrophotografisches Kopierverfahren nach Anspruch 9, bei dem der schwarze Toner eine mittlere Partikelgröße von 5 µm oder weniger hat.

11. Elektrophotografisches Kopierverfahren nach Anspruch 5 oder 8, bei dem das schwarze Isolierpigment durch Beschichten eines leitenden schwarzen Pigmentes mit einem transparenten Isolier-Kunstharz erhalten wird.

12. Elektrophotografisches Kopierverfahren nach Anspruch 11, bei dem das leitende schwarze Pigment aus Ruß, Graphit, schwarzem Cyanin, $CuCr_2O_4$ und $CuFe_2O_4$-$CuMn_2O_4$ ausgewählt wird.

13. Schwarzes Isolierpapier, auf welches durch einen Farbtoner, dessen Farbe heller ist als die des schwarzen Isolierpapieres, produzierte Tonerbilder transferiert und fixiert werden können, um Umkehrbilder mittels Verwendung eines positiv-abbildenden elektrophotografischen Kopierapparates zu formen, dadurch gekennzeichnet, daß der spezifische Oberflächenwiderstand des Papiers $10^9\ \Omega$ oder mehr ist und daß der spezifische Volumenwiderstand des Papiers $5 \times 10^9\ \Omega \cdot$ cm oder mehr ist.

14. Schwarzes Isolierpapier nach Anspruch 13, dadurch gekennzeichnet, daß das Papier durch Mischung eines schwarzen Isolierpigmentes und eines Rohmaterial-Zellstoffblattes in einem Schritt erhalten wurde, in dem ein Vermischen des Rohmaterial-Zellstoffes mit einem oder mehreren aus einem Appreturmittel, einem Füllmittel und einem Farbstoff stattfindet.

15. Schwarzes Isolierpapier nach Anspruch 13, dadurch gekennzeichnet, daß das Papier durch Auftragung eines transparenten Isolier-Kunstharzes auf leitendes schwarzes Papier oder durch Imprägnierung leitenden

schwarzen Papieres mit dem Kunstharz erhalten wurde.

16. Schwarzes Isolierpapier nach Anspruch 13, dadurch gekennzeichnet, daß das Papier durch Laminieren eines isolierenden transparenten Filmes auf leitendes schwarzes Papier erhalten wurde.

17. Schwarzes Isolierpapier nach Anspruch 13, dadurch gekennzeichnet, daß das Papier durch Aufdrucken oder Auftragen eines schwarzen Isolierpigmentes auf weißes Papier oder durch Imprägnierung weißen Papies mit dem Pigment erhalten wurde.

18. Schwarzes Isolierpapier nach Anspruch 14 oder 17, dadurch gekennzeichnet, daß das schwarze Isolierpigment ein schwarzer Toner ist.

19. Schwarzes Isolierpapier nach Anspruch 18, dadurch gekennzeichnet, daß der schwarze Toner eine mittlere Partikelgröße von 5 µm oder weniger hat.

20. Schwarzes Isolierpapier nach Anspruch 14 oder 17, dadurch gekennzeichnet, daß das schwarze Isolierpigment durch Bedecken eines leitenden schwarzen Pigmentes mit einem transparenten Isolier-Kunstharz erhalten wird.

21. Schwarzes Isolierpapier nach Anspruch 20, dadurch gekennzeichnet, daß das leitende schwarze Pigment aus Ruß, Graphit, schwarzem Cyanin, $CuCr_2O_4$ und $CuFe_2O_4$-$CuMn_2O_4$ ausgewählt ist.

Revendications

1. Une méthode de copie électrophotographique comprenant l'utilisation d'un appareil à copier présentant un système de copie d'images positives, la formation d'images de toner avec du toner de couleur, le report des images de toner sur un papier isolant de couleur d'une couleur différente de celle dudit toner, et la fixation des images reportées sur le papier, caractérisée par la sélection des couleurs du papier et du toner, de manière à former des images inversées.

2. Une méthode de copie électrophotographique suivant la revendication 1, dans laquelle ledit toner de couleur est un toner blanc.

3. Une méthode de copie électrophotographique suivant la revendication 1 ou 2, dans laquelle ledit papier de couleur est obtenu en formant une zone d'une couleur désirée sur un papier à report à l'aide d'un appareil à copier électrophotographique.

4. Une méthode de copie électrophotographique suivant la revendication 1, 2 ou 3, dans laquelle ledit papier de couleur est du papier isolant noir à résistance spécifique superficielle de $10^9\ \Omega$ ou plus et à résistance spécifique en volume de $5 \times 10^9\ \Omega \cdot$ cm ou plus.

5. Une méthode de copie électrophotographique suivant la revendication 4, dans laquelle ledit papier isolant noir a été obtenu par mélange d'un pigment noir isolant et d'une pulpe battue dans une étape au cours de laquelle a lieu le mélange de la pulpe avec un ou plusieurs agents pris parmi un agent d'encollage, une charge et un colorant.

6. Une méthode de copie électrophotographique suivant la revendication 4, dans laquelle ledit papier isolant noir a été obtenu par application d'une résine transparente isolante sur du papier noir conducteur ou par imprégnation du papier noir conducteur avec de la résine.

7. Une méthode de copie électrophotographique suivant la revendication 4, dans laquelle ledit papier isolant noir a été obtenu par laminage d'un film transparent isolant sur du papier noir conducteur.

8. Une méthode de copie électrophotographique suivant la revendication 4, dans laquelle ledit papier isolant noir a été obtenu par impression ou application d'un pigment noir isolant sur du papier blanc, ou par imprégnation de papier blanc avec le pigment.

9. Une méthode de copie électrophotographique suivant la revendication 5 ou 8, dans laquelle ledit pigment noir isolant est un toner noir.

10. Une méthode de copie électrophotographique suivant la revendication 9, dans laquelle ledit toner noir a une taille de particule moyenne en volume de 5 µm ou moins.

11. Une méthode de copie électrophotographique suivant la revendication 5 ou 8, dans laquelle ledit pigment noir isolant est obtenu par revêtement d'un pigment noir conducteur avec une résine isolante transparente.

12. Une méthode de copie électrophotographique suivant la revendication 11, dans laquelle ledit pigment noir conducteur est sélectionné parmi le noir de carbono, le graphite, le noir de cyanine, $CuCr_2O_4$ et $CuFe_2O_4$-$CuMn_2O_4$.

13. Papier isolant noir sur lequel peuvent être reportées et fixées des images de toner produites par un toner de couleur, dont la couleur est plus pâle que celle dudit papier isolant noir, pour former des images inversées à l'aide d'un appareil à copier électrophotographique à images positives, caractérisé en ce que la résistance spécifique superficielle dudit papier est de $10^9\ \Omega$ ou plus et que la résistance spécifique en volume dudit

papier est de $5 \times 10^9\ \Omega \cdot$ cm ou plus.

14. Papier isolant noir suivant la revendication 13, dans lequel ledit papier a été obtenu par mélange d'un pigment noir isolant et d'une pulpe brute battue dans une étape au cours de laquelle a lieu le mélange de ladite pulpe brute avec un ou plusieurs agents pris parmi un agent d'encollage, une charge et un colorant.

15. Papier isolant noir suivant la revendication 13, dans lequel ledit papier a été obtenu par application d'une résine transparente isolante sur du papier noir conducteur ou par imprégnation du papier noir conducteur avec de la résine.

16. Papier isolant noir suivant la revendication 13, dans lequel ledit papier a été obtenu par laminage d'un film transparent isolant sur du papier noir conducteur.

17. Papier isolant noir suivant la revendication 13, dans lequel ledit papier a été obtenu par impression ou application d'un pigment noir isolant sur du papier blanc, ou par imprégnation de papier blanc avec le pigment.

18. Papier isolant noir suivant la revendication 14 ou 17, dans lequel ledit pigment noir isolant est un toner noir.

19. Papier isolant noir suivant la revendication 18, dans lequel ledit toner noir a une taille de particule moyenne en volume de 5 µm ou moins.

20. Papier isolant noir suivant la revendication 14 ou 17, dans lequel ledit pigment noir isolant est obtenu par revêtement d'un pigment noir conducteur avec une résine isolante transparente.

21. Papier isolant noir suivant la revendication 20, dans lequel ledit pigment noir conducteur est sélectionné parmit le noir de carbone, le graphite, le noir de cyanine, $CuCr_2O_4$ et $CuFe_2O_4$-$CuMn_2O_4$.

FIG. 1

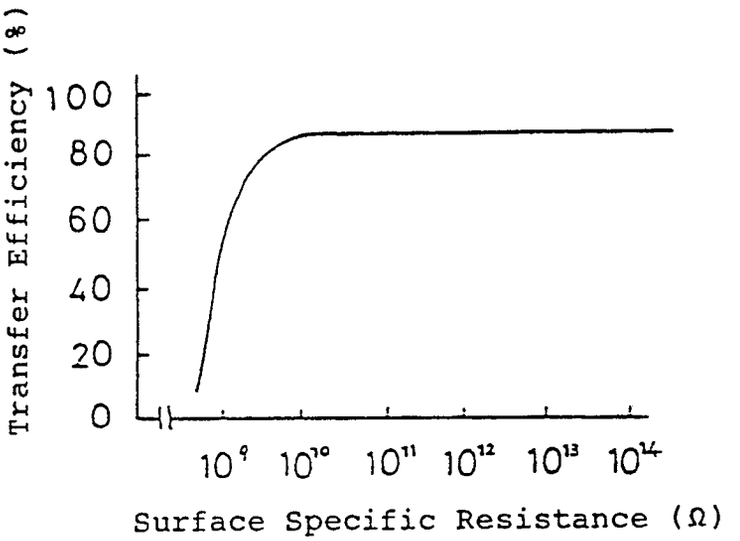
Transfer Efficiency (%)
100
80
60
40
20
0
10^9 10^{10} 10^{11} 10^{12} 10^{13} 10^{14}
Surface Specific Resistance (Ω)

FIG. 2

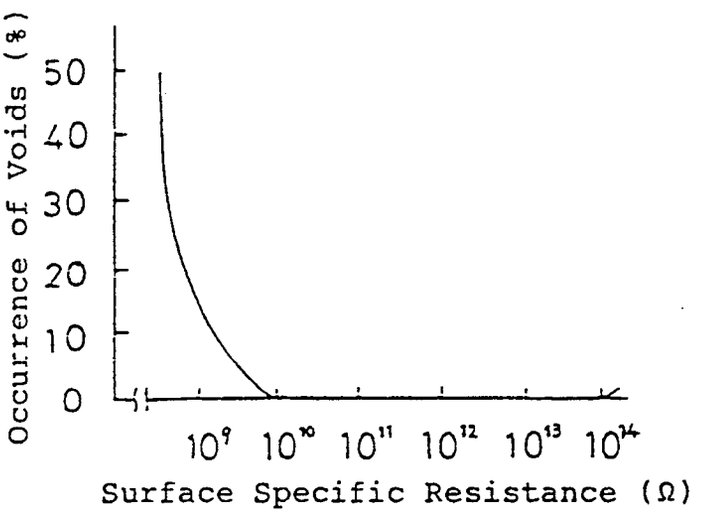
Occurrence of Voids (%)
50
40
30
20
10
0
10^9 10^{10} 10^{11} 10^{12} 10^{13} 10^{14}
Surface Specific Resistance (Ω)

FIG. 3

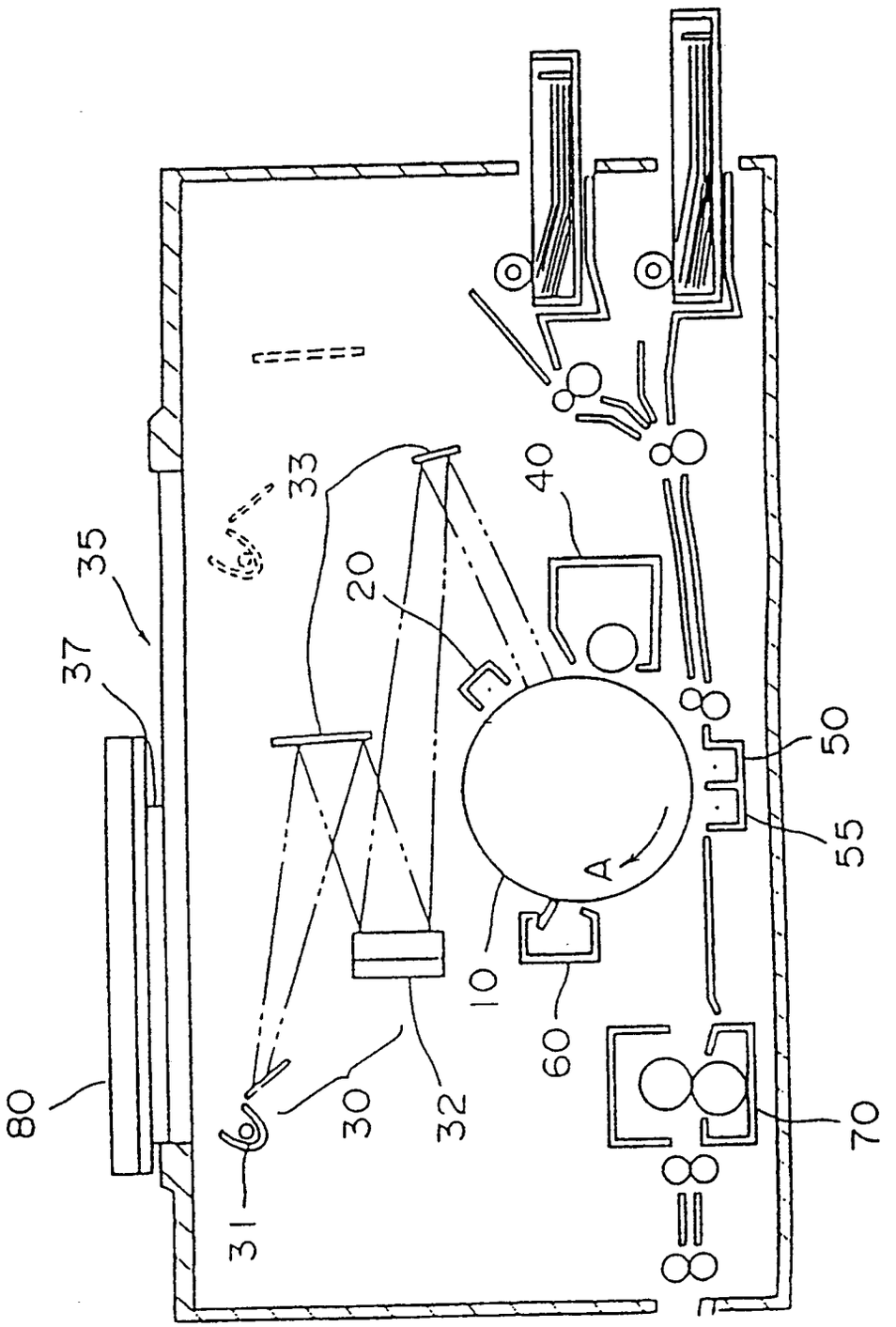

FIG. 4