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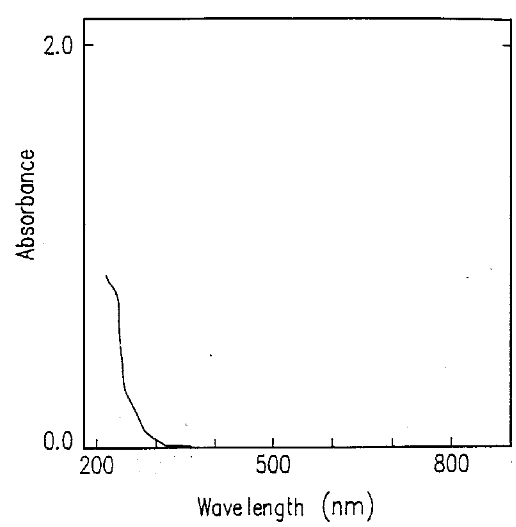
(54) **Image forming method**

(57) This invention provides an image forming method using a transfer belt for transferring a toner image, wherein toner includes toner particles which contain a binding resin and particles of a magnetic powder dispersed in the binding resin, the magnetic powder being contained in a ratio of 0.1 to 5 parts by weight with respect to 100 parts of the binding resin, the binding resin is formed of a composition containing a resin having an anionic group, and an extracted solution obtained by extracting the toner with methanol has substantially no absorption peak in the range of 280 to 350 nm, and has a substantially zero absorbance in the range of 400 to 700 nm.

FIG.1



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Description

1. Field of the Invention:

The present invention relates to an image forming method using a transfer belt for transferring a toner image to a transfer medium. In particular, the present invention relates to an image forming method which uses toner for a two-component developer, the toner having a satisfactory transfer efficiency and providing a long life to the developer to reproduce images at a desired density for a long period of time despite the elimination of a charge control agent, and is preferably used in an electrophotographic image forming apparatus including a transfer belt system such as an electrostatic copying machine and a laser beam printer.

2. Description of the Related Art:

Image forming apparatuses such as laser printers, copying machines, and plain paper facsimile apparatuses utilizing an electrophotographic method generally use corona discharge in order to transfer a toner image formed on a photosensitive body onto a transfer medium such as a paper sheet. The transfer system using corona discharge has a poor transfer .efficiency under high humidity. The transfer system using corona discharge also tends to generate defective transfer due to stains of a corona wire and creases of the transfer paper sheet. Further, corona discharge generates environmentally hazardous substances such as ozone and NO_x. For these reasons, a transfer system which does not use corona discharge has been desired.

One such transfer system is described in, for example, Japanese Laid-Open Patent Publication No. 4-345183. An image forming apparatus described in the above-mentioned publication includes a transfer and transportation unit located opposed to a photosensitive body. The transfer and transportation unit includes a driving roller, a subordinate roller provided parallel to the driving roller with a space from the driving roller, a transfer and transporting belt extended between the driving roller and the subordinate roller, and a transfer roller located opposed to the photosensitive body with the transfer and transportation belt interposed therebetween. In such an image forming apparatus, image transfer is performed in the following manner. By applying a high voltage to the transfer roller, the toner which is forming a toner image on the photosensitive body is absorbed onto the paper sheets which are sequentially supplied between the photosensitive body and the transfer belt. Such a system using a transfer belt improves the transfer efficiency under high humidity without generating environmentally hazardous substances.

Such a system using a transfer belt requires toner which has a satisfactory and stable chargeability and satisfactory cleaning characteristics. Examples of undesirable toner include toner which has a significantly insufficient charge amount and toner which has a polarity inverse to the polarity required for development. (In order to develop an electrostatic latent image having, for example, a positive potential, the toner should have a negative potential; but sometimes toner has a partially positive potential.) In the case where such undesirable toner is used, the toner scatters in a developing area and causes "fog"; i.e., the toner is attached to a non-image area of a surface of the photosensitive body. By the conventional transfer method using corona discharge, the toner which is attached to the non-image area of the photosensitive body is not transferred to the transfer paper sheet as long as the fog is restricted to a certain level. By a system using a transfer belt, image transfer is performed while the transfer belt and the photosensitive body are in a constant pressure-contact state. Accordingly, although the ease of transfer is improved, a small amount of toner attached to the non-image area of the photosensitive body is transferred to the transfer paper sheet, resulting in fog.

The system using a transfer belt involves a further problem. The transfer belt has a width corresponding to the size of a maximum paper sheet usable in the image forming apparatus. In the case where a transfer paper sheet having a smaller width than the transfer belt is used, toner attached to a non-image area of the photosensitive body, especially a non-image area outside the width of the transfer paper sheet, is directly transferred and thus attaches to the transfer belt. In order to remove the toner attached to the transfer belt, a cleaning device is necessarily provided. However, after the developer is used continuously for a long period of time, toner components strongly stick to the transfer belt. In such a case, the toner cannot be removed by the cleaning device. Thus, satisfactory transfer is impaired, and the reverse surface of a transfer paper sheet to which an image is not transferred is contaminated with toner (hereinafter, such a phenomenon will be referred to as "reverse surface contamination"). For these reasons, clean duplicates cannot be obtained.

Convententional toner contains a metal complex as a charge control agent in order to improve the chargeability. Since the metal complex is situated on the surfaces of toner particles, toner is attached to surfaces of carrier particles as a spent when the toner is mixed and stirred with the carrier, and the surface of the photosensitive body is contaminated when the mixture of the toner and the carrier slides on the surface as a magnetic brush. Accordingly, after the developer is used continuously for a long period of time, the developing characteristic of the toner is gradually changed. More specifically, the metal complex generates fog and causes the toner to scatter. The transfer belt is also contaminated in the same manner as the photosensitive body and the carrier.

Under the circumstances, toner used in an image forming apparatus using a transfer belt for image transfer is required to contain no substance contaminating the carrier or transfer belt, to have a sufficient chargeability, and to prevent to scatter and fog.

SUMMARY OF THE INVENTION

The image forming method of this invention using a transfer belt for transferring a toner image, which overcomes the above-discussed and numerous other disadvantages and deficiencies of the prior art, wherein toner includes toner particles which contain a binding resin and particles of a magnetic powder dispersed in the binding resin, the magnetic powder being contained in a ratio of 0.1 to 5 parts by weight with respect to 100 parts of the binding resin, the binding resin is formed of a composition containing a resin having an anionic group, and an extracted solution obtained by extracting the toner with methanol has substantially no absorption peak in the range of 280 to 350 nm, and has a substantially zero absorbance in the range of 400 to 700 nm.

In a preffered embodiment, the magnetic powder is contained in a ratio of 0.5 to 3 parts by weight with respect to 100 parts of the binding resin.

In a preffered embodiment, the toner particles have a volume-based average particle diameter of 5 through 15 μm, and spacer particles having a volume-based average particle diameter of 0.05 through 1.0 μm are attached onto surfaces of the toner particles.

Thus, the invention described herein makes possible the advantages of (1) providing an image forming method using a transfer belt for image transfer which does not generate reverse surface contamination; (2) providing an image forming method using a transfer belt for image transfer which effectively prevents contamination of the transfer belt; and (3) providing an image forming method using a transfer belt for image transfer which stably forms images having substantially no fog with a sufficient density and a satisfactory transfer efficiency for a long period of time.

These and other advantages of the present invention will become apparent to those skilled in the art upon reading and understanding the following detailed description with reference to the accompanying figures.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure **1** is a graph showing absorbance of a methanol extracted solution of toner used in the present invention in the range of 200 to 700 nm;

Figure **2** is a graph showing absorbance of a methanol extracted solution of toner having an azo dye - chrome complex as a charge control agent in the range of 200 to 700 nm;

Figure **3** is a graph showing absorbance of a methanol extracted solution of toner having a salicylic acid - metal complex as the charge control agent in the range of 200 to 700 nm;

Figure **4** is a graph showing absorbance of a methanol extracted solution of carrier in a two-component magnetic developer used for a long time in which toner has an azo dye - chrome complex as the charge control agent and chargeability of carrier is unstabilized by a spent in the range of 200 to 700 nm;

Figure **5** is a graph showing a relationship between shaking time and a spent ratio obtained with regard to two kind of a two-component magnetic developer, one comprising toner having a charge control agent and magnetic carrier and another comprising toner having no charge control agent and magnetic carrier;

Figure **6** is a graph showing a relationship between shaking time and quantity of charge of toner obtained with regard to two kind of a two-component magnetic developer, one comprising toner having a charge control agent and magnetic carrier and another comprising the toner having no charge control agent and magnetic carrier;

Figure **7** is a graph showing a relationship between an amount of spent of carrier and content of a charge control agent in a toner particle;

Figure **8** is a graph showing a relationship between shaking time and amount of spent obtained in the case where each component contained in a toner particle and magnetic carrier are individually mixed and shaken;

Figure **9** illustrates a mechanism of a charge failure caused by a spent in a conventional two-component magnetic developer;

Figure **10** is a schematic view of an image forming apparatus for forming an image using a method according to the present invention;

DESCRIPTION OF THE PREFERRED EMBODIMENTS

Toner used in the present invention has no charge control agent, such as a dye of an azo compound - metal complex and an oxycarboxylic acid - metal complex, at all. Therefore, a spent caused by a charge control agent, which will be described in detail below, scarcely occurs in the toner used in the method of the present invention, resulting in realizing a high quality copied image for a long period of time.

Further, in the case where toner containing spacer particles is used, the area of contact between toner particles and an organic photoconductor (OPC) body is reduced. Accordingly, attachment of a composition containing a resin included in the toner other than charge control agent (CCA) to a surface of a photoconductive layer of the OPC body can be effectively avoided.

Since the toner used in the method of the present invention has no charge control agent, it is impossible to detect any charge control agent, i.e., a dye type compound, from the toner by any chemical or physical method. For example, such a compound cannot be detected in the toner used in the.method of the present invention by any chemical reaction. Alternatively, absorption peaks owing to such a compound cannot be detected in an organic solvent extracted solution of the toner used in the method of the present invention. For example, when the toner used in the method of the present invention is extracted with an organic solvent such as methanol, the extracted solution has substantially no absorption peak in the range of 280 to 350 nm, and has substantially zero absorbance in the range of 400 to 700 nm. Herein, "to have substantially no absorption peak" means, in an extracted solution obtained by extracting 0.1 g of the toner used in the method of the present invention with 50 ml of methanol, absorption peaks are not detected at all, or if detected, values of the absorbance peaks are 0.05 or less. Similarly, "to have substantially zero absorbance" means that values of the absorbance of the extracted solution obtained by extracting 0.1 g of the toner used in the method of the present invention with 50 ml of methanol are 0.05 or less.

In the present invention, instability of the charge of the toner due to a lack of a charge control agent is compensated for as follows. First, a polymer having an anionic group is used as a binder resin of a toner particle, and secondly, magnetic powder is contained in the toner particle at a predetermined proportion. In the toner used in the method of the present invention, in order to further enhance the function of the toner, the binder resin is made of a composition containing a resin including a low molecular weight polymer and a high molecular weight polymer, and both the polymers have an anionic group. This results in further decreasing charge failure of the toner. Furthermore, spacer particles having a desired particle diameter are attached on the surfaces of the toner particles, if necessary, thereby increasing the transfer efficiency of the toner.

The above-mentioned characteristics of the toner used in the method of the present invention will be described in detail.

Figure **1** shows an UV-visible spectrum of a methanol extracted solution of the toner used in the method of the present invention in the range of 200 to 700 nm. As is shown in this spectrum, the extracted solution has no peak, which is otherwise formed because of a charge control agent. Specifically, the solution has substantially no absorption peak in the range of 280 to 350 nm, and the absorbance in the range of 400 to 700 nm is substantially zero. To the contrary, in an absorbance curve of a methanol extracted solution of toner having an azo dye - chrome complex as a charge control agent shown in Figure 2, absorption peaks are found in the range of 400 to 700 nm, in particular, 550 to 570 nm. Further, in the UV-visible spectrum of a methanol extracted solution of toner having a salicylic acid - metal complex as a charge control agent shown in Figure **3**, an absorption peak is found in the range of 280 to 350 nm.

It is because the charge control agent is present on the surfaces of the toner particles at a rather high concentration that the methanol extracted solution of the toner having the charge control agent has absorption peaks due to the charge control agent.

A carrier included in a developer which has insufficient chargeability owing to occurrence of a spent is extracted with methanol, and then the UV-visible spectrum of the extracted solution is measured to find absorption peaks in the range of 400 to 700 nm derived from a charge control agent. For example, the developer comprising the toner having an azo dye - chrome complex, whose UV-visible spectrum is shown in Figure **2**, was used for a long period of time to cause a spent therein. Then, UV-visible spectrum of a methanol extracted solution of the carrier in this developer was measured to give the spectrum shown in Figure **4**. As is shown in Figure **4**, absorption peaks are found at the same position as the spectrum in Figure **2**. It is conventionally understood that a spent is caused because a binder resin in the toner is attached to the surface of a carrier particle to form a resin film. The comparison between the absorbance curves in Figures **2** and **4**, however, reveals that one of the major causes of a spent is the transfer of the charge control agent from the toner particles to the carrier particles.

The present inventors conducted the following experiments in order to find out more about the relationship between a charge control agent and a spent: First, toner comprising toner particles containing 1.5 wt% of the azo dye - chrome complex was mixed with a carrier to obtain a developer. The toner and the carrier was shaken for a predetermined period of time. Figure **5** shows a relationship between the shaking time and amount of an attachment on the surfaces of the carrier particles. In Figure **5**, the amount of attachment is indicated as a spent ratio, that is, a percentage based on a total weight of the carrier particles bearing the attachment. Furthermore, Figure **6** shows the relationship between the shaking time and the amount of charge of the toner. The same procedure was repeated with regard to a developer comprising toner having no charge control agent and carrier. The experimental results of this developer are also shown in Figures **5** and **6**, wherein the results obtained by the developer including the toner having the charge control agent are plotted with black circles, and those by the developer including the toner having no charge control agent are plotted with white circles. It is apparent from Figures **5** and **6** that a larger amount of attachment is formed on the carrier particles as the spent and the charge amount of the toner has a greater decrease in the developer including the toner particle having the charge control agent than in the developer including the toner particle having no charge control agent.

Next, the weight of toner components attached on the surfaces of the carrier particles as the spent was measured with time. The results are shown in a graph of Figure **7**, wherein the abscissa indicates a measured amount of the spent and the ordinate indicates the content of the charge control agent in the toner particle. The broken line in Figure **7** indicates the amount of the charge control agent calculated in assuming that the toner components attached as the spent are identical to the components in the toner particles. Figure **7** reveals that a large amount of the charge control agent is deposited to be attached on the surfaces of the carrier particles at the initial stage. In Figure **7**, as amount of the spent increases, the measured values approximate the calculated values. This is because they are experimental results obtained in a close system having no supply of fresh toner. Therefore, when toner is exchanged as in a copying machine, the difference between the measured values and the calculated values would be much larger.

Furthermore, the present inventors measured the weight of the attachment on the surfaces of the carrier particles resulting from mixing the carrier with each of the toner components, that is, a charge control agent, a binder resin, carbon black as a coloring agent and wax, so as to find out the relationships between the respective toner components and the spent. The results are shown in Figure **8** as a variation with time in the amount of the attachment (i.e., amount of the spent), wherein the results obtained from the mixture with the charge control agent is plotted with white circles, those from the carbon black with black circles, those from the binder resin with squares, and those from the wax with triangles. It is apparent from Figure **8** that the charge control agent causes the largest amount of attachment due to the spent.

Based on the above-mentioned facts, the charge failure caused by the spent in a conventional two-component magnetic developer is explained as follows referring to Figure **9**. In the initial stage of the usage of a developer, a carrier particle **1** is positively charged and a toner particle **2** is negatively charged as is shown in an upper portion of Figure **9**. In this case, the toner particle works as a negative toner particle **21**. When this developer is continued to be used, a component including the charge control agent as a main component in the toner particle is attached on the surface of the carrier particle **1**. Attachment **201**, which is the spent, is negatively charged. The negatively charged attachment **201** leads to the formation of a toner particle having positive charge, that is, a reversely charged toner particle **22**. The reversely charged toner particle **22** is formed on the surface of the carrier particle **1** as is shown in a lower portion of Figure **9**, resulting in scattering of the toner and decreasing the transfer efficiency of the toner.

As described above, preferably, the toner does not have a charge control agent not only because the agent can include a heavy metal but also because the agent is the main cause of the spent, scatter of the toner and of a decrease in the transfer efficiency of the toner. Accordingly, the toner used in the method of the present invention has no charge control agent at all.

The instability of charge of the toner due to the lack of the charge control agent, in particular, the insufficiency in charge amount of the toner is compensated by using a binder resin having an anionic group as mentioned above. The insufficiency in charge amount of the toner particles can be supplemented because the binder resin has a negative charge in itself owing to the anionic group included therein. Since the anionic group is bonded to the main chain of the binder resin, it would never move onto the surface of the carrier particle as the charge control agent does, and hence it never causes the spent. On the contrary, charge around the surface of the toner particle caused by the anionic group of the binder resin is not so large that the electrostatic attraction between the toner particle and the carrier particle owing to the Coulomb force is insufficient when they are conveyed as a magnetic brush for development. Therefore, in a rapid copying operation, the toner cannot be sufficiently prevented from scattering because of insufficient coupling with the carrier particles. The scattered toner stains the inner wall of the copying machine, and can cause so-called a fog on a copied image.

In order to overcome such disadvantages, the toner used in the method of the present invention includes magnetic powder at a predetermined proportion, that is, 0.1 to 5 parts by weight on the basis of 100 parts by weight of the binder resin. The insufficiency in the charge amount of the toner particles can be thus compensated for. The magnetic powder contained in the toner particle causes magnetic attraction between the toner particle and the carrier particle. This magnetic attraction between the toner particle and the carrier particle together with electrostatic attraction prevents the toner from scattering.

The content of the magnetic powder in toner particles is in the range of 0.1 to 5 parts by weight per 100 parts by weight of the binder resin as described above. When the content is less than 0.1 parts by weight, the magnetic attraction between the toner particle and the carrier particle is insufficient, resulting in insufficient coupling with the carrier particle and causing toner scattering or fog forming on a copied image. Furthermore, the density of the copied image is low because of the insufficient charge amount. When the contents exceeds 5 parts by weight, the magnetic attraction between the carrier particle and the toner particle becomes so strong that the toner is not sufficiently attached onto an electrostatic latent image, resulting in decreasing the density of the copied image.

Several attempts have been made to improve the resolution of a copied image and the like by including (inclusively adding) magnetic powder as a toner component. For example, Japanese Laid-Open Patent Publication No. 56-106249 discloses a toner particle including 10 wt% of ferrite, and Japanese Laid-Open Patent Publication No. 59-162563 discloses a toner particle including 5 through 35 wt% of a magnetic fine particle. In either case, however, the content of the magnetic powder is excessive, and hence, the density of the copied image is low. Japanese Laid-Open Patent Publication No. 3-67268 discloses toner to which 0.05 to 2 wt% of magnetic powder is externally added. In this case, since the magnetic powder is not included in the toner particle, the powder is likely to be ununiformly attached onto the surface of the toner particle, resulting in insufficient magnetic attraction between the toner particle and the carrier particle. Furthermore, in either of the above-mentioned toners, the spent can be disadvantageously caused because a charge control agent is contained therein.

According to the present invention, the developing characteristic of the toner is controlled without using a metal complex having a charge control function. Thus, generation of a spent on the surfaces of the carrier particles and contamination of the photosensitive body are effectively prevented. Therefore, the developing characteristic is stabilized for a long period of time, thus effectively preventing scattering of the toner to the surface of the photosensitive body and generation of the fog. Further, transfer of the toner to the transfer belt is also prevented. Since the toner does not contain any substance which easily contaminates the transfer belt, the toner attached to the transfer belt is easily removed by the cleaning device.

According to the present invention, spacer particles having a particle diameter of 0.05 μm to 1.0 μm are attached preferably to surfaces of the toner particles in order to improve the transfer efficiency and to further prevent contamination of the transfer belt with the toner. The spacer particles can improve the fluidity of the toner particles and also form a space between the photosensitive body and the toner particles when the toner is attached to the electrostatic latent image on the photosensitive body. Thus, even if the charge amount of the toner is increased after the developer is used continuously for a long period of time, the toner is easily transferred from the photosensitive body to the transfer paper sheet. Thus, the transfer efficiency is improved. Moreover, the spacer particles form a space between the toner particles and the transfer belt, thus facilitating cleaning of the transfer belt.

A fine particle having a particle diameter of approximately 0.015 μm is used to enhance fluidity of a conventional toner. Such a small particle cannot form a sufficient gap between the photosensitive body and the toner particles, and cannot work as the spacer particle for the aforementioned purposes.

Now, preferable resins to be used as the binder resin in the toner used in the method of the present invention will be described. Herein, a "lower alkyl group" indicates alkyl having 1 to 5 carbon atoms.

(Binder resin of a toner particle included in the present depeloper)

The binder resin of the toner particle used in the present invention comprises a composition including a polymer having an anionic group. Such a binder resin is obtained by polymerizing a monomer having an anionic group or a mixture of the monomer having an anionic group with other monomers. The obtained resin can be a homopolymer or a copolymer.

The binder resin used in the toner is preferably a copolymer, such as a randam copolymer, a block copolymer and a grafted copolymer, obtained from a monomer having an anionic group and other monomers.

Examples of the monomer having an anionic group include monomers having a carboxylic acid group, a sulfonic acid group or a phosphoric acid group, and a monomer having a carboxylic acid group is generally used. Examples of the monomer having a carboxylic acid group include ethylenically unsaturated carboxylic acids such as acrylic acid, methacrylic acid, crotonic acid, maleic acid and fumaric acid; monomers that can form a carboxylic acid group such as maleic anhydride; and lower alkyl halfester of dicarboxylic acid such as maleic acid and fumaric acid. Examples of the monomer having a sulfonic acid group include styrene sulfonic acid and 2-acrylamido-2-methylpropane sulfonic acid. Examples of the monomer having a phosphoric acid group include 2-phosphono(oxy)propylmethacrylate, 2-phosphono (oxy) ethylmethacrylate, 3-chloro-2-phosphono(oxy) propylmethacrylate.

Such a monomer having an anionic group can be a free acid, a salt of an alkaline metal such as sodium and potassium, a salt of an alkaline earth metal such as calcium and magnesium, and a salt such as zinc.

The monomer having no anionic group used to prepare the binder resin is selected so that the resultant binder resin has a sufficient fixability and chargeability required of toner, and is one or a combination of an ethylenically unsaturated monomer. Examples of such a monomer include ethylenically unsaturated carboxylic acid ester, monovinyl arene, vinyl ester, vinyl ether, diolefin and monoolefin.

The ethylenically unsaturated carboxylic acid esters are represented by the following Formula (I):

$$CH_2=C(R^1)-COOR^2 \qquad (I)$$

wherein R^1 is a hydrogen atom or a lower alkyl group; and R^2 is a hydrocarbon group having 11 or less carbon atoms or a hydroxyalkyl group having 11 or less carbon atoms.

Examples of such ethylenically unsaturated carboxylic acid esters include methyl acrylate, ethyl acrylate, butyl acrylate, 2-ethylhexyl acrylate, cyclohexyl acrylate, phenyl acrylate, methyl methacrylate, hexyl methacrylate, 2-ethylhexyl methacrylate, β-hydroxyethylacrylate, γ-hydroxypropylacrylate, δ-hydroxybutylacrylate and β-hydroxyethylmethacrylate.

The monovinyl arenes are represented by the following Formula (II):

$$CH_2=C(R^3)-\phi-R^4 \qquad (II)$$

wherein R^3 is a hydrogen atom, a lower alkyl group or a halogen atom; R^4 is a hydrogen atom, a lower alkyl group, a halogen atom, an alkoxy group, an amino group or a nitro group; and ϕ is a phenylene group.

Examples of such monovinyl arene include styrene, α-methylstyrene, vinyltoluene, α-chlorostyrene, o-chlorostyrene, m-chlorostyrene, p-chlorostyrene and p-ethylstyrene.

The vinyl esters are represented by the following Formula (III):

$$CH_2=CH-OC(=O)R^5 \qquad (III)$$

wherein R^5 is a hydrogen atom or a lower alkyl group.

Examples of such vinyl esters include vinyl formate, vinyl acetate and vinyl propionate.

The vinyl ethers are represented by the following Formula (IV):

$$CH_2=CH\text{-}O\text{-}R^6 \qquad (IV)$$

wherein R^6 is a monovalent hydrocarbon group having 11 or less carbon atoms.

Examples of such vinyl ethers include vinyl methyl ether, vinyl ethyl ether, vinyl n-butyl ether, vinyl phenyl ether and vinyl cyclohexyl ether.

The diolefins are represented by the following Formula (V):

$$CH_2=C(R^7)-C(R^8)=CH-R^9 \qquad (V)$$

wherein R^7, R^8 and R^9 are independently a hydrogen atom, a lower alkyl group or a halogen atom.

Examples of such diolefins include butadiene, isoprene and chloroprene.

The monoolefins are represented by the following Formula (VI):

$$CH_2=C(R^{10})-R^{11} \qquad (VI)$$

wherein R^{10} and R^{11} are independently a hydrogen atom or a lower alkyl group.

Examples of such monoolefins include ethylene, propylene, isobutylene, 1-butene, 1-pentene and 4-methyl-1-pentene.

Specific examples of the polymer having an anionic group, that is, a (co)polymer obtained through the polymerization of the aforementioned monomers, include styrene-acrylic acid copolymers, styrene-maleic acid copolymers and ionomer resins. Furthermore, a polyester resin having an anionic group can be also used. The polymer having an anionic group preferably includes the anionic group at a proportion for attaining an acid value of 2 through 30, and preferably 5 through 15, when the anionic group is present as a free acid. When part or the entire anionic group is neutralized, the anionic group is preferably contained at such a proportion that the acid value would be in the aforementioned range in assuming that it is present as a free acid. When the acid value, i.e., the concentration of the anionic group, of the polymer or the composition is below the aforementioned range, the chargeability of the resultant toner is insufficient. When it exceeds the range, the resultant toner disadvantageously has a hygroscopic property. A preferable binder resin is a copolymer obtained from the monomer having an anionic group and at least one of the ethylenically unsaturated carboxylic acid ester represented by Formula (I) as an indispensable components, and any of the monomers represented by Formulae (II) through (VI) as an optional component to be used if necessary. One or a combination of two or more of the aforementioned monomers is used for preparing the binder resin.

The binder resin used in the invention is made of the composition including the aforementioned polymers, and the composition can further include a polymer having no anionic group as well. In this case, the proportion of the anionic group in the entire composition is preferably within the aforementioned range.

(Magnetic powder)

The magnetic powder contained in (inclusively added to) the toner particles can be any magnetic powder used in a conventional one-component type developer. Examples of the material for the magnetic powder include triiron tetroxide (Fe_3O_4), maghemite (γ-Fe_2O_3), zinc iron oxide ($ZnFe_2O_4$), yttrium iron oxide ($Y_3Fe_5O_{12}$), cadmium iron oxide ($CdFe_2O_4$), gadolinium iron oxide ($Gd_3Fe_5O_{12}$), copper iron oxide ($CuFe_2O_4$), lead iron oxide ($PbFe_{12}O_{19}$), nickel iron oxide ($NiFe_2O_4$), neodyum iron oxide ($NdFeO_3$), barium iron oxide ($BaFe_{12}O_{19}$), magnesium iron oxide ($MgFe_2O_4$), manganese iron oxide ($MnFe_2O_4$), lanthanum iron oxide ($LaFeO_3$), iron (Fe), cobalt (Co) and Nickel (Ni). Particularly preferable magnetic powder is made from triiron tetroxide (magnetite) in the shape of fine particles. The particle of preferable magnetite is in the shape of a regular octahedron with a particle diameter of 0.05 through 1.0 μm. Such a magnetite particle can be subjected to a surface treatment with a silane coupling agent or a titanium coupling agent. The particle diameter of the magnetic powder contained in the toner particle is generally 1.0 μm or smaller, and preferably in the range between 0.05 and 1.0 μm.

The content of the magnetic powder in the toner particle is in the range of 0.1 to 5 parts by weight, more preferably 0.5 to 4 parts by weight, and most preferably 0.5 to 3 parts by weight per 100 parts by weight of the binder resin. When the content is too small, the toner can be scattered during the development or a fog can be formed on a copied image.

(Inner additives in the toner particle)

The toner particle contains, as described above, the binder resin and the magnetic powder as indispensable components, and can optionally include some inner additive generally used for a toner, if necessary.

Examples of such additives include a coloring agent and a release agent.

As the coloring agent, the following pigments can be used:

- Black pigment:
  carbon black, acetylene black, lampblack, aniline black;

- Extender:
  barite powder, barium carbonate, clay, silica, white carbon, talc, alumina white.

Such a pigment is contained in the toner particle in the range of 2 to 20 parts by weight, and preferably 5 to 15 parts by weight per 100 parts by weight of the binder resin.

As the release agent, various wax and olefin resins can be used as in a conventional toner. Examples of the olefin resin include polypropylene, polyethylene, and propylene-ethylene copolymers, and polypropylene is particularly preferred.

(Preparation of the toner)

The toner particles in the toner included in a two-component type developer of the present inventioncan be produced by any ordinary method for toner particles such as crushing and classification, fusing granulation, spray granulation and polymerization, and are generally produced by the crushing and classification method.

For example, the components for the toner particles are previously mixed in a mixer such as a Henschel mixer, kneaded with a kneader such as a biaxial extruder, and then cooled. The resultant is crushed and classified to give toner particles. The particle diameter of the toner particle is generally in the range between 5 and 15 μm and preferably between 7 and 12 μm in the volume-base averaged particle diameter (a medium size measured with a Coulter counter).

It is possible to improve the fluidity of the toner by attaching, as an outer additive, a fluidity enhancer such as hydrophobic vapor depositioned silica particles onto the surfaces of the toner particles, if necessary. The primary particle diameter of the fluidity enhancer such as the silica particles is generally approximately 0.015 μm, and such a fluidity enhancer is added to the toner in the range of 0.1 to 2.0 percent by weight on the basis of the weight of the entire toner, i.e., the total weight of the toner particles and the fluidity enhancer.

Furthermore, spacer particles having a larger particle diameter than that of the fluidity enhancer are preferably added in the present invention. As the spacer particles, any of organic and inorganic inactive particles with a particle diameter of 0.05 through 1.0 µm, more preferably 0.07 through 0.5 µm can be used. Examples of the material for such inactive particles include silica, alumina, titanium oxide, magnesium carbonate, an acrylic resin, a styrene resin and magnetic materials. The spacer particle can not only work as a fluidity enhancer but also increase the transfer efficiency as described above. As the spacer particle, the same type of magnetic powder as included in the toner particle, in particular, triiron tetroxide (magnetite) in the shape of fine particle is preferably used. The magnetic powder, when used as the spacer particles, effectively suppresses the scattering of the toner as described above. The content of the spacer particles is 10 percent by weight or less, more preferably in the range of 0.1 to 10 percent by weight, and most preferably 0.1 to 5 percent by weight on the basis of the total weight of the toner. When the spacer particles are excessively included in toner, the density of a copied image is insufficient. When the magnetic powder is used as the spacer particles, the total amount of the magnetic powder togethher with that contained in the toner particles is preferably 10 parts by weight or less per 100 parts by weight of the binder resin. When it is excessively included, the density of a copied image can be decreased.

When the fluidity enhancer and the spacer particles are added to the toner particles, the following production method is preferred. The fluidity enhancer and the spacer particles are first sufficiently mixed with each other, and then the obtained mixture is added to the toner particles, and then is sufficiently unbound. Thus, the spacer particles can be attached onto the surfaces of the toner particles. To "be attached" herein means both to be held in contact with the surface of the toner particle and to be partly embedded in the toner particle. In this manner, the toner used in the method of the present invention is produced.

(Preperation of the developer)

A two-component type developer is prepared by mixing the above-mentioned toner and magnetic carrier.

Ferrite particle, in particular spherical soft ferrite particle which contains at least one metal selected from the group of Cu, Mg, Mn and Ni in addition to Fe, such as sintered Cu-Zn-Mg-ferrite particle, is preferably used as the magnetic carrier. A surface of the carrier particle may be coated or not coated with organic materials. Preferably, the surface of the carrier is coated with silicone resin (polysiloxane), fluorine contained resin, epoxy resin, amino resin, uretane resin. The particle diameter of the carrier particle is in the range of 30 to 200 µm, and preferably 50 to 150 µm. The carrier particle has a saturation magnetization in the range between 30 and 70 emu/g, and preferably 45 and 65 emu/g. The mixing ratio of the carrier and the toner is generally 98:2 through 90:10, and preferably 97:3 through 94:6, by weight.

(Image forming method)

Next, an image forming method according to the present invention will be described.

Figure **10** is a schematic view of an image forming apparatus usable for conducting an image forming method according to the present invention.

An image forming apparatus **2** includes an image carrier **4** formed of a photosensitive drum provided rotatably. The image carrier **4** is surrounded by a corona charger **5**, a developing device **6**, a transfer belt unit **8** including a transfer belt **86**, a cleaning unit **10**, and a removing lamp **12** provided in this order in the direction of arrow **A**. The image forming apparatus **2** further includes an optical system having a lamp **14**, a first mirror **16**, a second mirror **18**, a third mirror **20**, a lens **22**, and a fourth mirror **24.** A document placed on a transparent document table (not shown) is irradiated by light emitted from the lamp **14**, and the light reflected by the document is projected on the image carrier **4** via the first mirror **16**, the second mirror **18**, the third mirror **20**, the lens **22**, and the fourth mirror **24**, to form an electrostatic latent image.

The image forming apparatus **2** includes a paper supply unit **26** for supplying a transfer paper sheet to the transfer belt unit **8**. The paper supply unit **26** includes a paper cassette **28** for accommodating transfer paper sheets, a paper supply roller **30**, a pair of feeding rollers **32**, a guide path **34**, a pair of transport rollers **36**, another guide path **38**, and a pair of resist rollers **40**. A pair of binding rollers **42** and a pair of delivery rollers **44** are provided downstream the paper transportation direction with respect to the transfer belt unit **8**.

The image forming apparatus **2** operates in the following manner.

While the image carrier **4** rotates in the direction of arrow **A**, the corona charger **5** charges the surface of a photosensitive layer on the image carrier **4** to a predetermined polarity at a substantially uniform level. The document placed on the transparent document table (not shown) is irradiated by light emitted from the lamp **14**. The light reflected by the document travels to the surface of the image carrier **4** via the first mirror **16**, the second mirror **18**, the third mirror **20**, the lens **22**, and the fourth mirror **24**. Thus, an area of the surface of the image carrier **4** corresponding to the image of the document is exposed to light, thereby forming an electrostatic latent image. The electrostatic latent image is developed into a toner image by the developing device **6**.

A transfer paper sheet accommodated in the paper cassette **28** of the paper supply unit **26** is supplied by the paper supply roller **30** and is transported to the transfer belt unit **8** via the pair of feeding rollers **32**, the guide path **34**, the pair of transport rollers **36**, the guide-path **38**, and the pair of resist rollers **40**. Then, the toner image is transferred on the transfer paper sheet while the transfer paper sheet is passing between the image carrier **4** and the transfer belt **86** of the transfer belt unit **8**. The image is then bound by the binding roller **40**, and the transfer paper sheet is delivered outside by the pair of delivery rollers **42**.

After that, the toner is removed from the image carrier **4** by the cleaning unit **10**, and the charge on the surface of the image carrier **4** is removed by light emitted from the removing lamp **12**.

(Transfer belt)

An image forming method according to the present invention is usable in an apparatus including a multiple layer transfer belt or a single-layer transfer belt.

A multiple layer transfer belt includes, for example, a conductive layer having a resistance of 10^8 Ωcm or less and an insulation layer having a resistance of 10^{10} Ωcm or more. A single layer transfer belt includes, for example, an insulation layer having a resistance of 10^{10} Ωcm or more. The insulation layer of each type of transfer belt is formed of an elastic material such as polyurethane rubber or polychloroprene rubber. The transfer belt is also formed by treating a surface of a transfer belt formed of such a material with a release agent such as tetrafluoroethylene to have a resistance in the above-mentioned range. By such surface treatment, the toner attached to the transfer belt can be removed more easily.

Examples

The present invention will now be described by way of examples. It is noted that the invention is not limited to these examples.

(Example 1)

<Preparation of toner>

Table 1 Components of toner particle

component	Parts by weight
Binder resin [a]	100
Coloring agent: Carbon black	8
Magnetic powder: Magnetite	2

[a] The binder resin used in this example was a styrene-acrylic copolymer having a carboxyl group (wherein the weight ratio among styrene, butyl methacrylate, acrylic acid was 70:28:2).

The above listed components were fused and kneaded with a biaxial extruder, and the resultant was crushed with a jet mill, and classified with a pneumatic classifier to give toner particles with an average particle diameter of 10.0 μm.

To the obtained toner particles were added 0.3 part by weight of hydrophobic silica fine powder with an average particle diameter of 0.015 μm as a fluidity enhancer on the basis of 100 parts by weight of the toner particles. The resultant mixture was mixed with a Henschel mixer for two minutes to give toner.

(Example 2)

To the toner particle obtained in Example 1 were added 0.5 parts by weight of acrylic resin particles with an average particle diameter of 0.3 μm and 0.3 part by weight of hydrophobic silica fine powder with an average particle diameter of 0.015 μm on the basis of 100 parts by weight of the toner particles. The resultant mixture was mixed with a Henschel mixer for two minutes to give toner.

(Comparative Example 1)

The same procedure was repeated as in Example 1 except that an azo dye - chrome complex was further added to components listed in Table 1 of Example 1.

[Evaluation of the method for developing an electrostatic latent image]

Obtained toners in Examples 1, 2 and Comparative example 1 and carrier were homogeneously mixed to give a two-component type developer having a toner concentration of 3.5 wt%. Then, the obtained developers were evaluated with regard to the following items.

For the tests described below, a copying machine (manufactured by Mita Industrial Co., Ltd.; brand name: DC-4685) was used after being modified to include a transfer belt unit as shown in Figure **10**. A single layer endless belt used in the transfer belt unit was obtained by surface-treating a belt formed of polychloroprene rubber with tetrafluoroethylene to have a total resistance of 10^{11} Ωcm. For transfer, a voltage of 2.5 kV was applied. The photosensitive body was formed of selenium, and image formation was performed under such a condition as to make the surface potential after charged 800 V.

(a) Image density (I.D.):

A copying operation was continued by using an original bearing characters with a black area ratio of 8% until 50,000 copies were made with regard to the developers of Examples 1 and 2 and Comparative Example 1. The density of a black portion in a copied image on every 5000 copies was measured by a reflection densitometer (manufactured by Tokyo penshoku Co., Ltd.; TC-6D), and the average density was taken as an image density (I.D.). An original used for sampling every 5000 copies had a black area ratio of 15% including a black solid portion. The results obtained from the developers of Examples 1 and 2 and Comparative Example 1 are listed in Table 2.

(b) Fog density (F.D.):

A copying operation was continued by using an original bearing characters with a black area ratio of 8% until 50,000 copies were made with regard to the developers of Examples 1 and 2 and Comparative Example 1. The density of a white portion in a copied image on every 5000 copies was measured by the reflection densitometer (manufactured by Tokyo Denshoku Co., Ltd.; TC-6D). A difference between the thus measured density and the density of paper to be used for the copying operation (base paper) measured by the reflection densitometer was calculated, and the maximum difference was taken as a fog density (F.D.). An original used for sampling every 5000 copies had a black area ratio of 15% including a black solid portion. The results obtained from the developers of Examples 1 and 2 and Comparative Example 1 are listed in Table 2.

(c) Charge amount:

A copying operation was continued by using an original bearing characters with a black area ratio of 8% until 50,000 copies were made with regard to the developers of Examples 1 and 2 and Comparative Example 1. During this copying operation, after making every 5,000 copies, the charge amount of 200 mg of the developer was measured by a blowoff type powder charge amount measuring device (manufactured by Toshiba Chemical Co., Ltd.), and the average of the.charge amount per 1 g of the toner was calculated based on the measured value. The results obtained from the developers of Examples 1 and 2 and Comparative Example 1 are listed in Table 2.

(d) Transfer efficiency:

The amount of toner in a toner hopper in the copying machine was measured at first, and a predetermined number of copies were made. Then, the amount of the toner left in the toner hopper was measured. From a difference between the amounts of the toner before and after the copying operation, a consumed amount of the toner was calculated. At the same time, the amount of the toner collected in a cleaning process during the copying operation was also measured as a collected amount. Based on these amounts, the transfer efficiency of the toner was calculated by using Equation (i) as below. An original used in the copying operation bore characters with a black area ratio of 8%.

$$\text{Transfer efficiency (\%)} = \frac{\text{(Consumed amount)-(Collected amount)}}{\text{(Consumed amount)}} \quad \text{(i)}$$

With regard to the developers of Examples 1 and 2 and Comparative Example 1, 50,000 copies were made, and the results obtained from these developers are listed in Table 2.

(e) Amount of attachment on the surface of the carrier particle due to the spent:

A copying operation was conducted by using an original bearing characters with a black area ratio of 8%. After making 50,000 copies, the developer was tested as follows: The developer was placed on a screen of 400 mesh, and sucked from the below with a blower, thereby separating the toner and the carrier. Five g of the carrier remained on the screen was charged in a beaker, to which toluene was added. Thus, the toner component attached onto the surfaces of the carrier particles due to the spent was dissolved. Then, the toluene solvent was discarded with the carrier attracted upon the bottom of the beaker with a magnet. This procedure was repeated several times until the resultant toluene solution became transparent. Then, the resultant carrier was heated with an oven to evaporate the toluene attached thereto, and the weight of the obtained residue was measured. A difference between the weight of the carrier charged in the beaker at first (i.e., 5 g in this case) and the weight of the residue after evaporating the toluene was taken as the amount of the toner components attached onto the surfaces of the carrier particles due to the spent (i.e., the spent amount). The spent amount is indicated as the weight in mg of the toner components attached to 1 g of the carrier. The results obtained from the developers of Examples 1 and 2 and Comparative Example 1 are listed in Table 2.

(f) Toner scattering:

A copying operation was continued by using an original bearing characters with a black area ratio of 8% until 50,000 copies were made. Then, the toner scattering state in the copying machine was visually observed and evaluated. The results obtained from the developers of Examples 1 and 2 and Comparative Example 1 are listed in Table 2. In these tables, ○ indicates that the toner was not scattered; and × indicates that the toner was scattered.

(g) transfer belt filming:

A copying operation was performed by using an original bearing characters with a black area ratio of 8%. After making 50,000 copies, the surface of the transfer belt in the copying machine was visually observed. The results are listed in Table 2, wherein ○ indicates that no filming was observed; and × indicates that a filming was observed.

(h) reverse surface contamination

A copying operation was performed by using an original bearing characters with a black area ratio of 8%. After making 50,000 copies, the reverse suface which is not image forming side of a copy paper was visually observed and evaluated. The results obtained from the developers of Examples 1 and 2 and Comparative Example 1 are listed in Table 2. In these tables, ○ indicates that the reverse surface contamination was not observed; and × indicates that the the reverse surface contamination was observed.

Table 2

Toner component and Evaluation of developers of Example 1 and 2 and Comparative Examples.			
	Example 1	Example 2	Comparative Example 1
Toner component (parts by weight)			
Bainder resin	100	100	100
Carbon black	8	8	8
Magnetic powder	2	2	2
Charge control agent	none	none	2
Additive 1 (silica;0.015 μm)	0.3	0.3	0.3
Additive 2 (acrylic resin;0.30 μm)	none	0.5	none
Results of evaluation			
I.D.	1.368	1.379	1.369
F.D.	0.005	0.004	0.015
Transfer efficiency (%)	77.3	80.2	62.3

Continuation of the Table on the next page

Table 2 (continued)

Toner component and Evaluation of developers of Example 1 and 2 and Comparative Examples.			
	Example 1	Example 2	Comparative Example 1
Toner scattering	○	○	×
Spent amount (mg)	0.55	0.5	1.83
Charge amount (μC/g)	-21.4	-20.3	-15.9
Transfer belt filming	○	○	×
Reverse surface contamination	○	○	×

[Review of the evaluation]

The toners produced in Examples 1 and 2 were stable in a satisfactory state in the image density, the fog density, and the charge amount, from the start of the copying until after 50,000 images were reproduced.

Especially, the toners produced in Examples 1 and 2 were less in the generation of the spent and also less in the reduction in the charge amount, as compared with the toner produced in comparative example 1. As a result, the toner produced in Examples 1 and 2 did not scatter or cause the filming to the transfer belt. Even after the developer containing either one of the toners in Examples 1 and 2 is used continuously for a long period of time, reverse surface contamination due to the filming to the transfer belt did not occur. In the case of using the toner produced in comparative example 1, reverse surface contamination was generated from approximately the 30,000th duplicate.

According to the present invention, a developing method using toner containing no charge control agent which is a main cause of the spent during copying is provided. Toner particles of the toner according to the present invention contain a binder resin having an anionic group and also contain a magnetic powder at a predetermined ratio. When necessary, spacer particles having a predetermined particle diameter are attached to the surfaces of the toner particles. Accordingly, the toner has a sufficient chargeability, does not scatter during copying, and has a sufficient transfer efficiency. Due to such advantages, images having a necessary density can be reproduced stably for a long period of time.

Further according to the present invention, a developer containing the above-described toner is used. Therefore, contamination of the transfer belt and reverse surface contamination of the transfer medium such as a paper sheet can be restricted. Thus, satisfactory images can be reproduced for a long period of time.

The toner according to the present invention is preferably used in an electrophotographic image forming apparatus including a transfer belt system such as an electrostatic copying machine or a plain paper facsimile apparatus.

Various other modifications will be apparent to and can be readily made by those skilled in the art without departing from the scope and spirit of this invention. Accordingly, it is not intended that the scope of the claims appended hereto be limited to the description as set forth herein, but rather that the claims be broadly construed.

Claims

1. An image forming method using a transfer belt for transferring a toner image , wherein:

   toner includes toner particles which contain a binding resin and particles of a magnetic powder dispersed in the binding resin, the magnetic powder being contained in a ratio of 0.1 to 5 parts by weight with respect to 100 parts of the binding resin,

   the binding resin is formed of a composition containing a resin having an anionic group, and

   an extracted solution obtained by extracting the toner with methanol has substantially no absorption peak in the range of 280 to 350 nm, and has a substantially zero absorbance in the range of 400 to 700 nm.

2. An image forming method according to claim 1, wherein the magnetic powder is contained in a ratio of 0.5 to 3 parts by weight with respect to 100 parts of the binding resin.

3. An image forming method according to claim 1, wherein the toner particles have a volume-based average particle diameter of 5 through 15 μm, and spacer particles having a volume-based average particle diameter of 0.05 through 1.0 μm are attached onto surfaces of the toner particles.

FIG.1

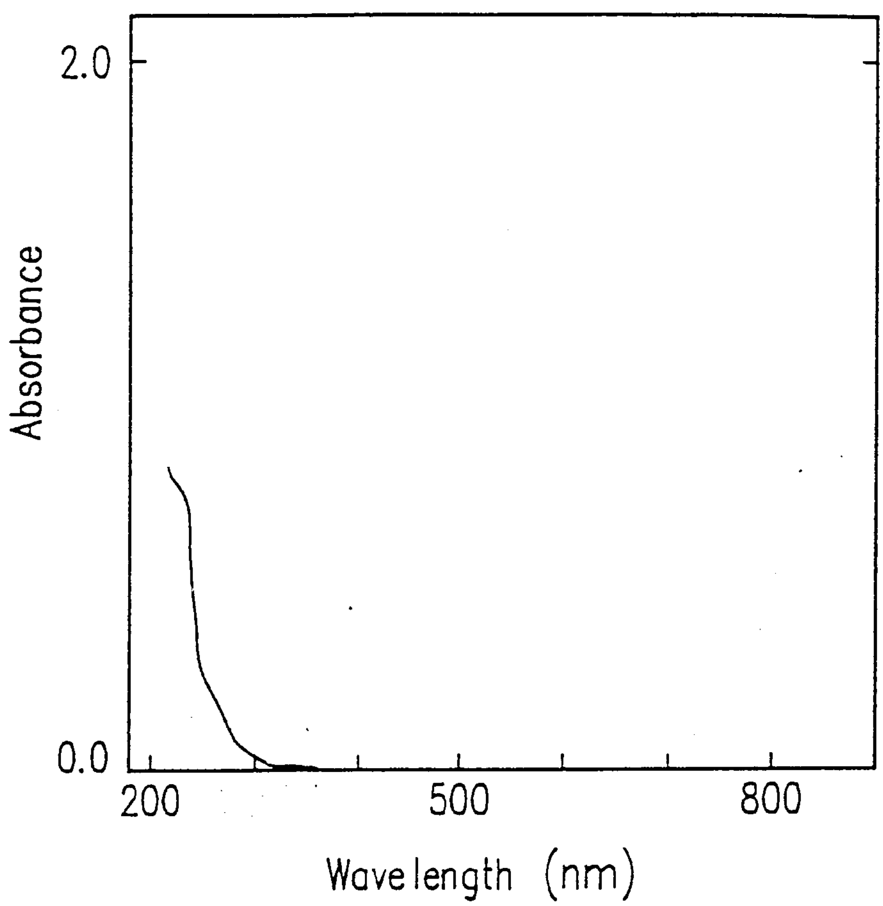


FIG. 2

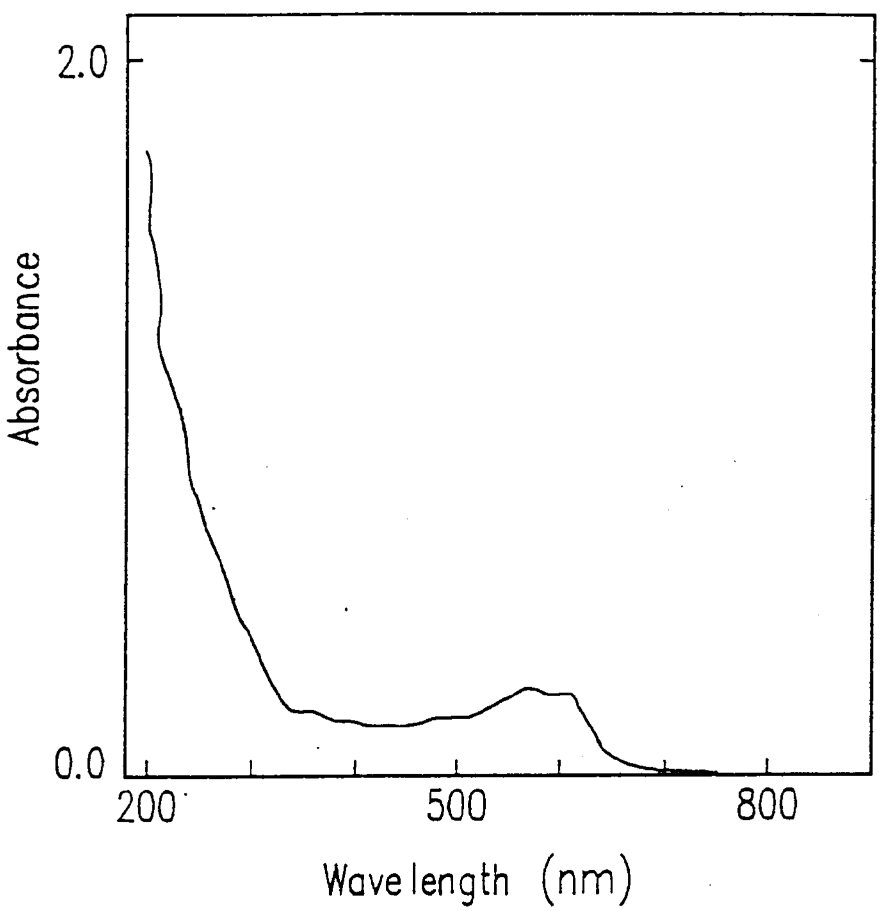


FIG. 3

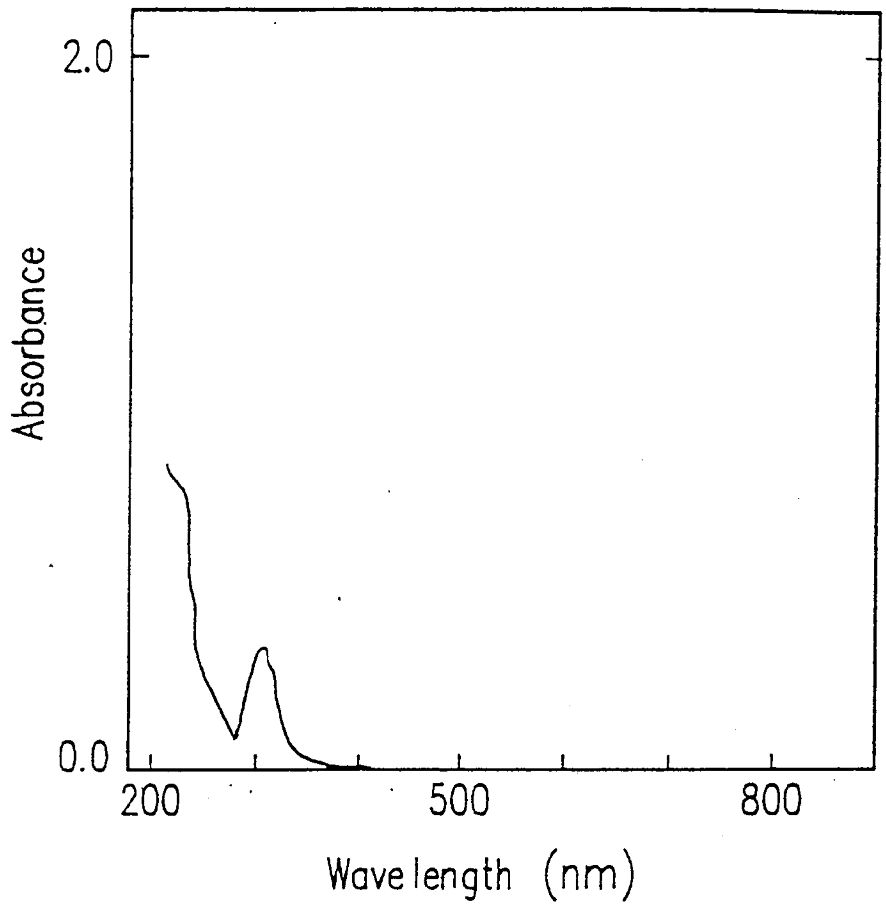


FIG. 4

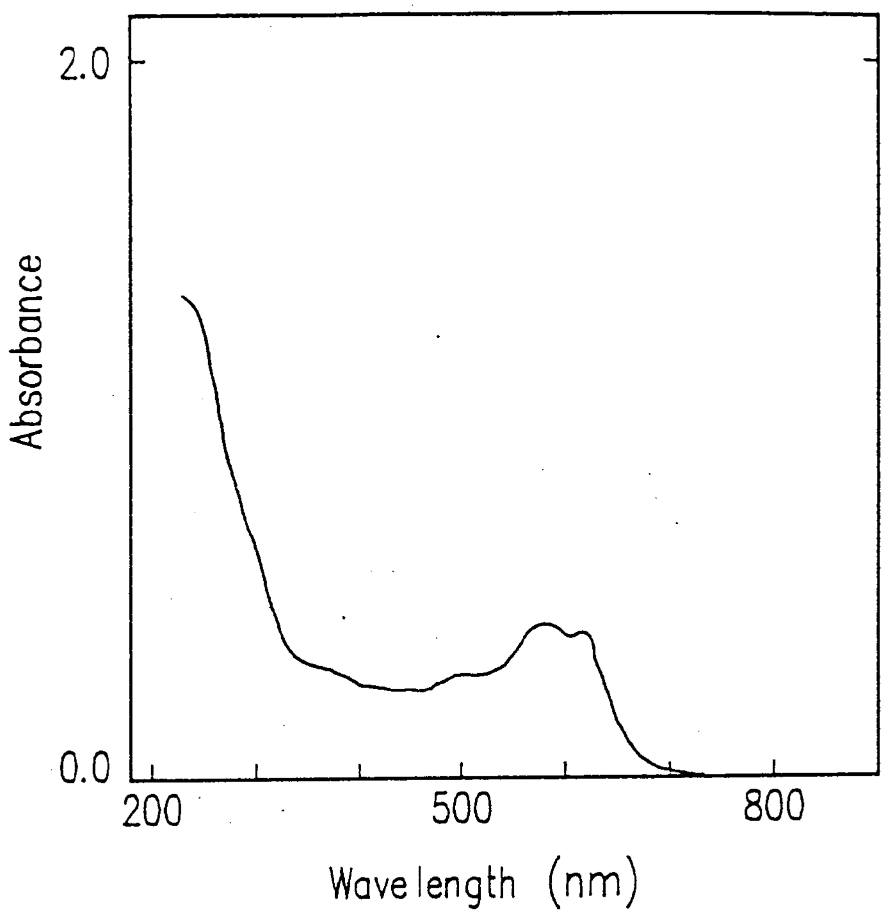


FIG. 5

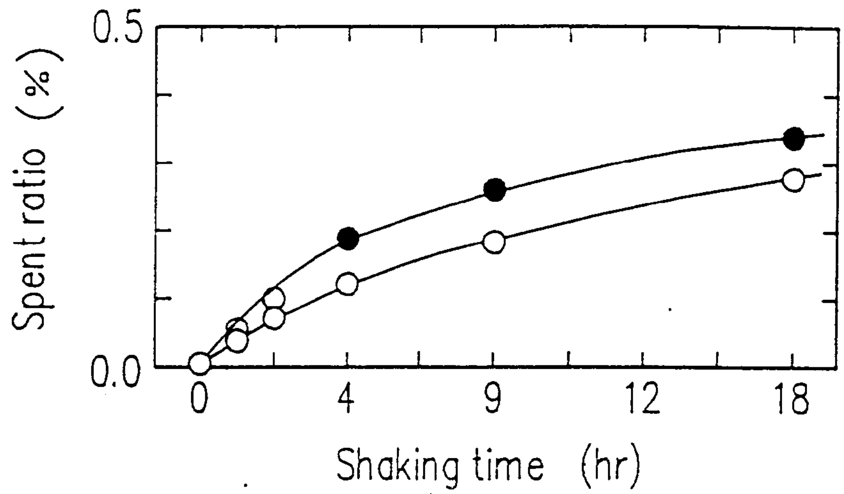

0.5
0.0
Spent ratio (%)
0
4
9
12
18
Shaking time (hr)

FIG. 6

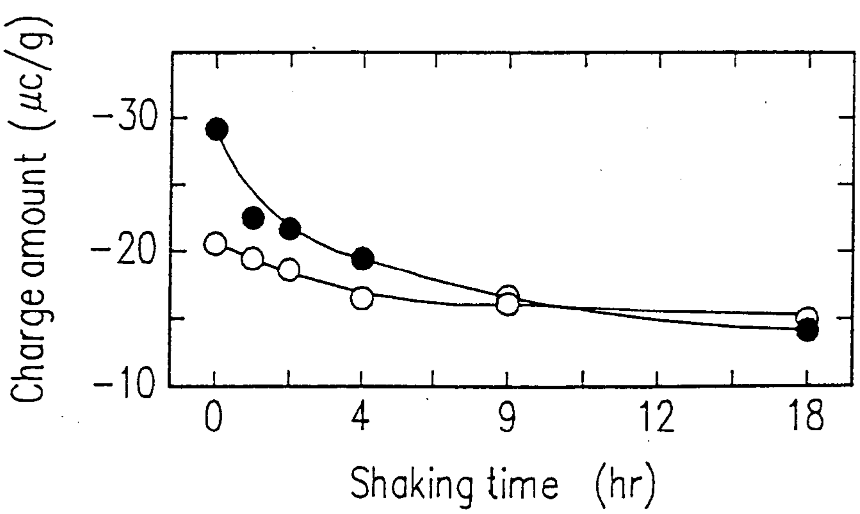

Charge amount (μc/g)
-30
-20
-10
0
4
9
12
18
Shaking time (hr)

FIG. 7

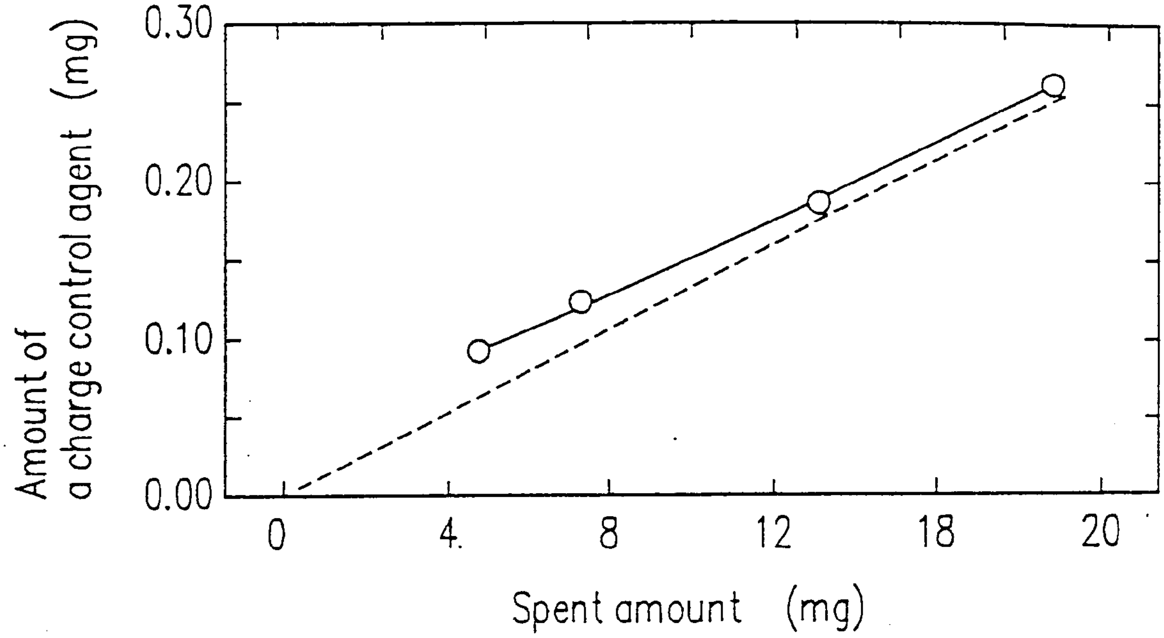

Amount of
a charge control agent (mg)
0.30
0.20
0.10
0.00
0
4.
8
12
18
20
Spent amount (mg)

FIG. 8

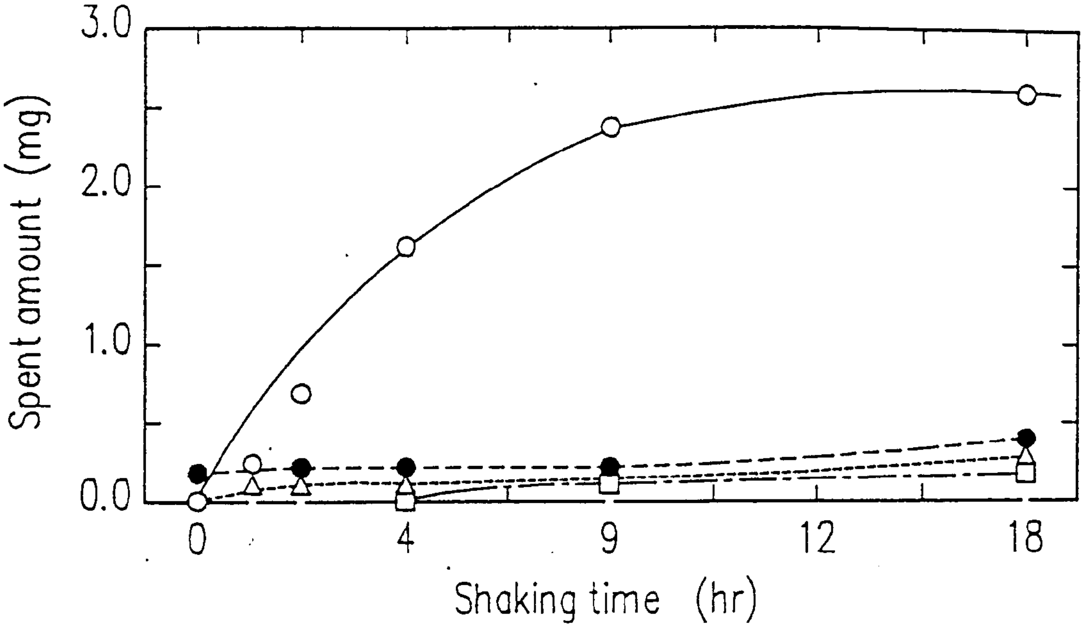


FIG.9

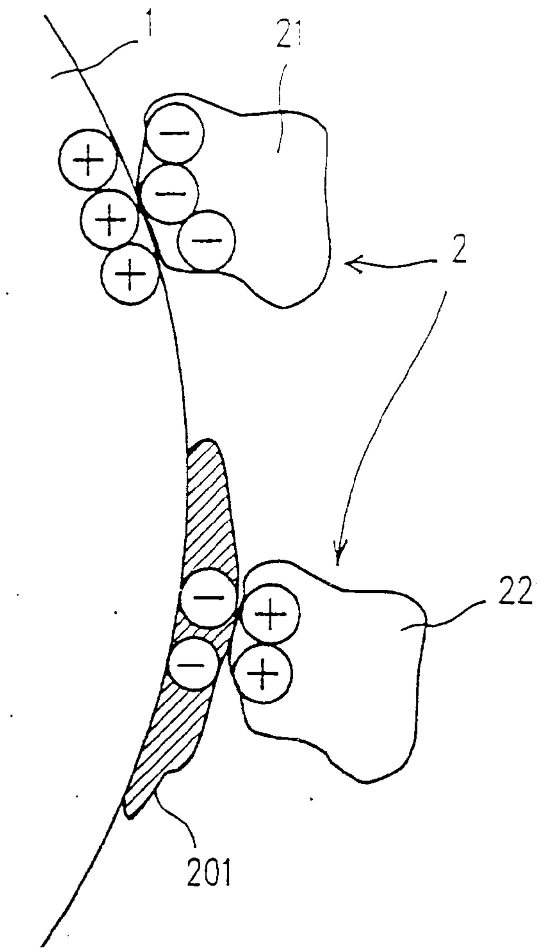
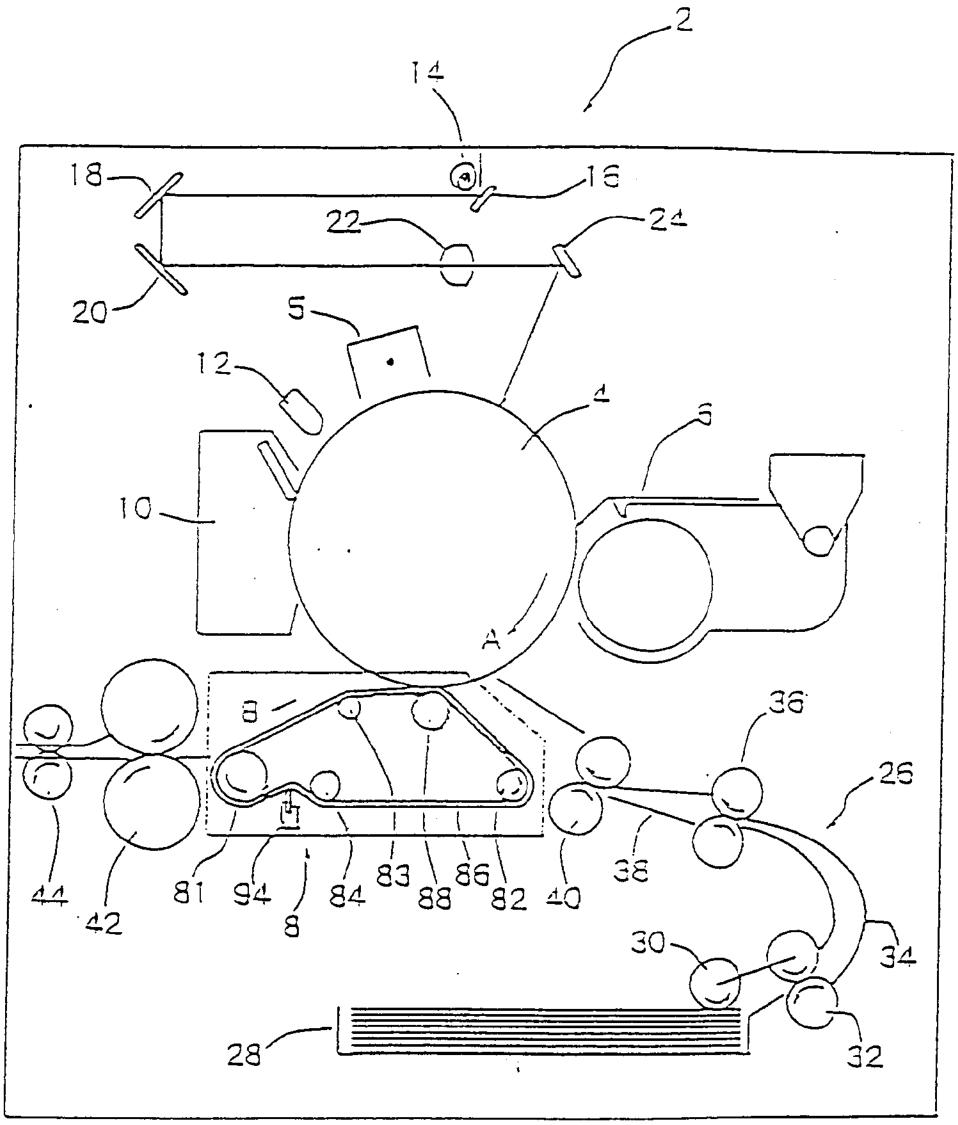


FIG.10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EUROPEAN SEARCH REPORT

Application Number
EP 95 30 6129

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
A	EP-A-0 357 042 (TDK) * claims 1-6; examples 1-4 * ---	1-3	G03G9/087 G03G9/083 G03G21/10
A	PATENT ABSTRACTS OF JAPAN vol. 9 no. 120 (P-358) [1843] ,24 May 1985 & JP-A-60 004950 (CANON) 11 January 1985, * abstract * ---	1-3	
A	EP-A-0 407 604 (MITA) * page 12, line 7 - page 17, line 10; claim 4; examples 1-6 * * page 26, line 11 - line 22 * ---	1-3	
A	DATABASE WPI Section Ch, Week 9335 Derwent Publications Ltd., London, GB; Class G06, AN 93-275420 & JP-A-05 188 640 (HITACHI) , 30 July 1993 * abstract * ---		TECHNICAL FIELDS SEARCHED (Int.Cl.6)
P,X	EP-A-0 643 337 (MITA) * page 15, line 55 - page 16, line 1; claims 1,2 * -----	1-3	G03G

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	8 January 1996	Vanhecke, H

CATEGORY OF CITED DOCUMENTS

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