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⑤④ Charge control resin particles and a method of manufacturing the same.

⑤⑦ Charge control resin particles are spheroidal particles which are formed by dispersion polymerization and mainly composed of a copolymer consisting of at least one kind of water-soluble monomer having an anionic group of sulfonic, carboxylic or phosphoric type and at least one kind of oil-soluble monomer, the copolymer having a weight-average molecular weight of 3,000 to 50,000 and the resin particles having a volume median size of 1  $\mu\text{m}$  or more with the ratio  $D_{25}/D_{75}$  of the 25 volume percent integrated diameter  $D_{25}$  to the 75 volume percent integrated diameter  $D_{75}$  being within the range of 1 to 1.4.

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## CHARGE CONTROL RESIN PARTICLES AND A METHOD OF MANUFACTURING THE SAME

The present invention relates to resin particles for use as electric charge control agents in toner for developing an electrostatic image used in electrophotography, and to a method of manufacturing the same.

Generally, the toner for developing electrostatic images used in the field of electrophotography or the like is formed from resin particles of 5 to 20  $\mu\text{m}$  in size consisting of colorants, charge control agents, etc. dispersed in binding resin particles. The charge control agents used to confer triboelectrification ability on the resin particles include nigrosine, monoazo dyes, or metallic complexes of salicylic acid or naphthoic acid, etc. However, since most of these charge control agents lack compatibility with binding resins, the charge control agents become dispersed unevenly in the resin particles, preventing uniform conferring of frictional chargeability. In view of such difficulty, there has been proposed a method wherein charge control resins having polar functional groups are mixed into binding resins to confer frictional chargeability to the toner. For such charge control resins, compatibility with binding resins is required, and generally, a copolymer is used which consists of a water-soluble monomer having a polar group, such as acrylonitrile and styrene sulfonic acid sodium salt., and an oil-soluble monomer capable of forming a polymer having compatibility with the binding resins.

Bulk polymerization, suspension polymerization, solution polymerization, emulsion polymerization, and dispersion polymerization are used for manufacturing the charge control resins. Since the compatibility is generally low between a water-soluble monomer having a polar group and an oil-soluble monomer, a uniform polymer system cannot be formed in bulk polymerization or in suspension polymerization, the resulting copolymer having a large quantity of monomeric units having a polar group and thus lacking compatibility with binding resins. In solution polymerization, it is possible to make the polymer system uniform before polymerization by choosing an appropriate solvent, but as the polymerization progresses, the polymer precipitates, making the composition of the final copolymer non-uniform. In emulsion polymerization, it is possible to control the compositions of the monomers in the copolymer, but since the molecular weight of the copolymer is very high, the compatibility with binding resins is low. Furthermore, in emulsion polymerization, the problem is that the size of the obtained particles is small, thus making it difficult to precipitate the particles. In dispersion polymerization, uniform polymerization is possible, but the size distribution of the obtained particles is broad and the molecular weight tends to become high, thus leaving room for improvement.

The charge control resin particles of the present invention, comprise spheroidal resin particles formed by dispersion polymerization and composed mainly of a copolymer of at least one water-soluble monomer having an anionic group of sulfonic, carboxylic or phosphoric type and at least one kind of oil-soluble monomer, wherein the copolymer has a weight-average molecular weight of 3,000 to 50,000, the resin particles having a volume median size of 1  $\mu\text{m}$  or more with the ratio  $D_{25}/D_{75}$  of the 25 volume percent integrated diameter  $D_{25}$  to the 75 volume percent integrated diameter  $D_{75}$  being within the range of 1 to 1.4.

It is preferred that the anionic group be a sulfonic group or an alkali metal salt thereof. A preferred monomer is then styrene sulfonic acid sodium salt.

It is further preferred that the oil-soluble monomer comprises both styrene and an acrylic monomer.

The ratio of the water-soluble monomer to the oil-soluble monomer forming the copolymer is preferably in the range from 70 : 30 to 99 : 1 by weight.

The present invention also provides a method for manufacturing charge control resin particles, wherein at least one water-soluble monomer having an anionic group of sulfonic, carboxylic or phosphoric type and at least one oil-soluble monomer are dispersed and polymerized in an aqueous medium containing water and a water-miscible organic solvent in the presence of a dispersion stabilizer and a chain transfer agent and the resulting copolymer consisting of the water-soluble monomer and the oil-soluble monomer is collected in the form of particles, in which method a nucleus is formed by reaction of the oil-soluble monomer and the water-soluble monomer; and the thus formed nucleus is grown, the temperature being higher in the nucleus growing step than in the nucleus forming step.

In a preferred embodiment, the mixing ratio of the water and the water-miscible organic solvent in the aqueous medium is in the range from 40 : 60 to 5 : 95 by weight.

In another preferred embodiment, the temperature is 60 to 80°C in the nucleus forming step and 70 to 90°C in the nucleus growing step.

Thus, the invention described herein makes possible one or more of the objectives of (1) providing charge control resin particles having a narrow size distribution and formed from a resin having a relatively low molecular weight; (2) providing charge control resin particles comprising a polymer with a low molecular weight and having a large particle size in the dispersion liquid; (3) providing charge control resin particles having good compatibility with a binding resin or with a monomer forming the binding resin and therefore capable of forming toner with the charge control resin uniformly dispersed in the binding resin;

(4) providing charge control resin particles having uniform chargeability over the entire toner particles and therefore capable of forming toner having excellent charge characteristics; and (5) providing a method capable of readily producing the charge control resin particles having the above-mentioned excellent characteristics.

In the polymer system of the present invention, reaction progresses in solution polymerization in the early stage of polymerization, to form a copolymer comprising monomers having a composition ratio that matches their reactivity ratio. As the polymerization further progresses, the copolymer tries to precipitate, but since a dispersion stabilizer exists in the polymer system, it is believed that some of copolymers combine to form a relatively stable nucleus (dispersed particle). It is believed that since this nucleus is a copolymer formed from an oil-soluble monomer and a water-soluble monomer, the unreacted oil-soluble monomer and water-soluble monomer in the continuous phase are simultaneously absorbed and the polymerization progresses after formation of the nucleus, thus forming a copolymer having a relatively uniform composition. Particularly, when styrene and an acrylic monomer are used simultaneously as the oil-soluble monomer, the particle size is relatively large in the dispersion liquid and particles are readily precipitated therefrom. Furthermore, by using these monomers, the copolymer forming the particles has a relatively low molecular weight.

In the present invention, since a chain transfer agent exists in the reactive system, the molecular weight of the copolymer is kept low.

It is believed that since the temperature is relatively low in the nucleus forming step and relatively high in the nucleus growing step, stable formation of the nucleus is facilitated and the incorporation of the monomers into the nucleus and the polymerization reaction are effectively performed, thus forming charge control resin particles having a low molecular weight and a uniform size distribution.

The charge control resin particles have a large volume median size of 1  $\mu\text{m}$  or more, and therefore, the handling thereof, including separation from a mixture liquid, is facilitated. Also, since they are formed from a low molecular weight copolymer having a weight-average molecular weight of 3,000 to 50,000, the resin particles are evenly and uniformly dissolved within a binding resin such as a styrene copolymer or in monomers forming the binding resin. Furthermore, since the degree of dispersion expressed by the ratio  $D_{25}/D_{75}$  of the 25 volume percent integrated diameter  $D_{25}$  to the 75 volume percent integrated diameter  $D_{75}$  of the resin particles is within the range of 1 to 1.4 and the particle size distribution is sharp, the toner particles with the charge control resin particles dispersed therein are provided with uniform chargeability over the entire particles and exhibit excellent charge

characteristics. Also, when producing toner by suspension polymerization using the charge control resin particles, since the viscosity of suspended oil does not increase and the polarity of the oil is uniform, toner particles having an extremely sharp size distribution can be obtained.

The oil-soluble monomer used for the present invention is a monomer capable of forming a polymer having a good compatibility with binding resins. Hydrophobic monomers such as vinyl aromatic monomers, acrylic monomers such as vinyl ester monomer, vinyl ether monomer, and olefinic monomers are used. The vinyl aromatic monomers include, for example, styrene,  $\alpha$ -methylstyrene, vinyltoluene,  $\alpha$ -chlorostyrene, o-, m-, p-chlorostyrene, p-ethylstyrene, divinylbenzene. The acrylic monomers include, for example, methyl acrylate, ethyl acrylate, butyl acrylate, 2-ethylhexylacrylate, cyclohexyl acrylate, phenyl acrylate, methyl methacrylate, hexyl methacrylate, 2-ethylhexyl methacrylate,  $\beta$ -hydroxyethyl acrylate,  $\gamma$ -hydroxypropyl acrylate,  $\delta$ -hydroxybutyl acrylate,  $\beta$ -hydroxyethyl methacrylate, and ethyleneglycol dimethacrylate. The vinyl ester monomers include, for example, vinyl formate, vinyl acetate, and vinyl propionate. The vinyl ether monomers include, for example, vinyl-n-butyl ether, vinyl phenyl ether, and vinyl cyclohexyl ether. The olefinic monomers include, for example, ethylene, propylene, isobutylene, butene-1, pentene-1, and 4-methylpentene-1. Two or more of these monomers can be used simultaneously.

The water-soluble monomer used for the present invention has an anionic group of sulfonic, carboxylic or phosphoric type. These acid radicals may be either in the form of salts such as sodium salt and other alkali metal salts, ammonium salt, amine salt, etc., or in the form of free acids. Monomers having a sulfonic group or its alkali metal salt are particularly desirable. Such monomers include, for example, styrenesulfonic acid, styrenesulfonic acid sodium salt, 2-acrylamide-2-methylpropanesulfonic acid, 2-phosphoxypropylmethacrylate, 2-phosphoxyethylmethacrylate, 3-chloro-2-phosphoxypropylmethacrylate, acrylic acid, methacrylic acid, fumaric acid, crotonic acid, tetrahydroterephthalic acid, itaconic acid, maleic acid, etc. The styrenesulfonic acid sodium salt is most preferable.

The composition al ratio of the water-soluble monomer to the oil-soluble monomer should be determined in such a manner as to obtain good compatibility with the binding resin and good charge characteristics. Generally the ratio should be in the range of 70 : 30 to 99 : 1, and preferably 80 : 20 to 98 : 2, by weight.

With an oil-soluble monomer comprising styrene and an acrylic monomer, the mixing ratio of styrene and acrylic monomer is preferably in the range of 10 : 0.1 to 10 : 3, by weight. If the amount of an acrylic

monomer exceeds the above range, the particles tend to adhere, thereby causing difficulty in handling thereof. Moreover, other radical polymeric monomers can be included as needed. The amount of these monomers used is preferably 20 percent by weight or less.

If the water-soluble monomer is styrenesulfonic acid sodium salt the mixing ratio of the oil-soluble monomer to styrenesulfonic acid sodium salt is preferably in the range of 70 : 30 to 99 : 1, by weight.

In the present invention, known chain transfer agents are used which include, for example, octyl thioglycolate.

Polymeric dispersion stabilizers are desirable for use as the dispersion stabilizer, and include, for example, polyacrylic acid, salts of polyacrylic acid, polymethacrylic acid, salts of polymethacrylic acid, (meth)acrylic acid-(meth)acrylic acid ester copolymer, acrylic acid-vinylether copolymer, methacrylic acid styrene copolymer, carboxymethyl cellulose, polyethylene oxide, polyacrylamide, methyl cellulose, ethyl cellulose, hydroxyethyl cellulose, polyvinyl alcohol, etc. Nonionic or anionic surfactant may also be used. The amount of the dispersion stabilizer added is preferably 1 to 20 percent by weight, with respect to the monomers.

The water-miscible organic solvents which can be used include, for example, lower alcohols such as methanol, ethanol, isopropanol, etc.; ketones such as acetone, methyl ethyl ketone, methyl butyl ketone, etc.; ethers such as tetrahydrofuran, dioxane etc.; esters such as ethyl acetate, etc.; and amides such as dimethylformamide, etc.

The mixing ratio of the water to the water-miscible organic solvent depends on the kind of the monomer and the kind of the organic solvent. Generally the ratio should be within the range of 40 : 60 to 5 : 95 by weight, and preferably within the range of 30 : 70 to 10 : 90 by weight. The composition ratio is preferably determined so as to have a uniform solution phase within this range. The amount of the medium used is 0.5 to 50 times by weight with respect to a monomer, particularly 5 to 25 times by weight being preferable.

When practicing the method of the present invention, the charge control resin particles may be manufactured, for example, in the following manner. The oil-soluble monomer, water-soluble monomer, dispersion stabilizer, and if necessary, a polymerization initiator, are dissolved in a solution of water and a water-miscible organic solvent, and the chain transfer agent is further added. After stirring the mixture for 0.5 to 4 hours at a temperature of 60 to 80°C, the temperature of the mixture is raised to 70 to 90°C and the liquid is then stirred for 2 to 20 hours. At this time, it is desirable that this process is conducted in the presence of an inert gas to suppress polymerization termination by oxygen. The resulting polymer is filtered, washed with water or other suitable solvent as necessary, and

dried to obtain the desired resin particles.

Any hydrophobic polymerization initiator is used as the polymerization initiator. For such initiators, azo compounds such as azobisisobutyronitrile, etc. and peroxides such as cumene hydroperoxide, di-*t*-butylperoxide, benzoyl peroxide, lauroyl peroxide, etc. are used. The mixing ratio of the initiator is not specifically limited, but should generally be 0.1 to 20 percent by weight with respect to the monomer. Instead of using such initiators, the polymerization can be initiated by using gamma rays or an accelerated electron beam. Alternatively, the polymerization may be initiated by using photosensitizers in combination with ultraviolet rays.

The thus obtained charge control resin particles have a relatively large average size of 3 to 10  $\mu\text{m}$  in a dispersion liquid, so the precipitation thereof from the mixture is facilitated. The particles are polymers having a weight of 3,000 to 50,000. Since the molecular weight is relatively low, the resin particles have good compatibility with the binding resin such as a styrene copolymer or with monomers forming the binding resin, and therefore, a toner with the charge control resin particles uniformly dispersed in the binding resin can be readily obtained. Furthermore, since the ratio  $D_{25}/D_{75}$  of the 25 volume percent integrated diameter  $D_{25}$  to the 75 volume percent integrated diameter  $D_{75}$  is within the range of 1 to 1.4 and the particle size distribution is extremely monodisperse, the toner particles with the charge control resin particles dispersed therein are provided with uniform chargeability over the entire particles and exhibit excellent charge characteristics. According to the present invention, charge control resin particles having the above-mentioned excellent characteristics can be readily manufactured.

The following Examples illustrate the present invention.

#### Example 1

Ten grams of polyacrylic acid were dissolved in a solution consisting of 675 g of isopropyl alcohol and 225 g of water, to which 90 g of styrene, 10 g of styrene sulfonic acid sodium salt, 5 g of octyl thioglycolate, and 7.5 g of azobisisobutyronitrile were added. The thus prepared liquid was placed in a 3-liter separable flask for 2 hours at 70°C while the liquid was being stirred in a 150 rpm stream of nitrogen, after which the temperature was raised to 80°C to perform a reaction for 10 hours, after which polymerization was completed. The obtained emulsion was observed under an optical microscope to note the formation of particles having a median size of about 6.0  $\mu\text{m}$ . The ratio  $D_{25}/D_{75}$  was 1.37 and a very sharp particle size distribution was noted. The weight-average molecular weight measured by gel permeation chromatography (GPC) was  $1.18 \times 10^4$ . It was poss-

ible to collect 100% of the particles in a short time using a qualitative filter.

Ten parts by weight of the obtained styrene-styrenesulfonic acid sodium salt copolymer, 80 parts by weight of styrene-acrylic binder resin, and 10 parts by weight of carbon black were melted and kneaded, and the thus kneaded mixture was crushed and classified to obtain toner having an average particle size of 10  $\mu\text{m}$ . The toner was mixed and stirred with ferrite carrier to prepare developer, which was then subjected to testing on a Mita Industrial Co., Ltd. Model DC-1205 copier to examine splashing and fogging of toner, changes in image density, etc. under high temperature and high humidity (35°C, RH85%) and low temperature and low humidity (10°C, RH45%) conditions. The result showed no problems. Further, the developer showed a high charge amount under the above conditions and also exhibited an extremely good charging-up. The charge distribution of the developer was measured on a toner charge distribution measuring instrument, which showed a sharp charge distribution with no uncharged or reverse charged toner whatsoever noted.

#### Example 2

Ten grams of polymethacrylic acid were dissolved in a solution consisting of 700 g of isopropyl alcohol and 300 g of water, to which 90 g of styrene, 10 g of styrenesulfonic acid sodium, 3 g of octyl thioglycolate, and 7 g of 2,2'-azobis-2,4-dimethylvaleronitrile were added. The thus prepared liquid was placed in a 3-liter separable flask for 2 hours at 65°C while the liquid was being stirred in a 150 rpm stream of nitrogen, after which the temperature was raised to 75°C to perform a reaction for 10 hours, the polymerization being thus completed. The obtained emulsion was observed under an optical microscope to note the formation of particles having a size of about 4.2  $\mu\text{m}$ . The ratio  $D_{25}/D_{75}$  was 1.39, and the weight-average molecular weight measured on a GPC was  $1.25 \times 10^4$ . It was possible to collect 100% of the particles in a short time using a qualitative filter.

Ten parts by weight of the obtained styrene-styrenesulfonic acid sodium salt copolymer, 80 parts by weight of n-butylacrylate, 10 parts by weight of grafted carbon black (MA-100, manufactured by Mitsubishi Kasei), 1.5 parts by weight of low molecular weight polypropylene (BISCALL 550P, manufactured by Sanyo Kasei), and 4 parts by weight of azobisisobutyronitrile were dissolved in this order in 60 parts by weight of styrene. At this time, the styrene-styrenesulfonic acid sodium salt copolymer was quickly dissolved, without flocculating, when charged into the styrene. The mixture was charged into an aqueous solution of 1.5% calcium phosphate (pH adjusted to 11 by sodium hydroxide) and dispersed for suspension therein using a TK-Homomixer (man-

ufactured by Tokushu Kikou Kogyo), after which the mixture was polymerized for 5 hours while being stirred in a normal manner at 80°C in a stream of nitrogen. The thus produced particles were filtered, washed, and dried to obtain the toner. The thus obtained toner was spheroidal, had good flowability, and had sharp particle size and charge amount distributions. The toner was evaluated in the same manner as in Example 1, and yielded good results in all evaluations.

#### Example 3

Ten grams of polyacrylic acid were dissolved in a solution consisting of 675 g of ethanol and 225 g of water, to which 90 g of styrene, 10 g of styrenesulfonic acid sodium, 5 g of octyl thioglycolate, and 10 g of benzoyl peroxide were added. The thus prepared liquid was placed in a 3-liter separable flask for 2 hours at 60°C while the liquid was being stirred in a 150 rpm stream of nitrogen, after which the temperature was raised to 80°C to perform a reaction for 10 hours, the polymerization being thus completed. The obtained emulsion was observed under an optical microscope to note the formation of particles having an average size of 4.6  $\mu\text{m}$ . The ratio  $D_{25}/D_{75}$  was 1.39, and the weight-average molecular weight measured on a GPC was  $1.50 \times 10^4$ .

Toner was prepared in the same manner as in Example 2. The solubility of the styrene-styrenesulfonic sodium salt copolymer in the styrene was good. The obtained toner was evaluated in the same manner as in Example 1, and yielded good results in all evaluations.

#### Comparative Example 1

The composition was the same as used was in Example 1 except that octyl thioglycolate the chain transfer agent and the polymerization was performed for 10 hours at a temperature of 70°C without raising the temperature during the polymerization.

The obtained styrene-styrenesulfonic sodium salt copolymer in the emulsion had an average particle size of 1.8  $\mu\text{m}$ , thus forming particles of a small size. The ratio  $D_{25}/D_{75}$  was 1.55 and the particle size distribution was broad. The weight-average molecular weight measured on a GPC was  $10.88 \times 10^4$ . The copolymer had poor compatibility with monomers such as styrene, and it took time to dissolve the copolymer in the monomer.

#### Comparative Example 2

The composition was the same as in Example 2 although the temperature was not raised during the polymerization but kept at 80°C throughout the polymerization.

The obtained styrene-styrenesulfonic sodium salt copolymer contained in the emulsion coarse agglomerate which accounted for 3% of the entire particles. The average size of the particles was 7.2  $\mu\text{m}$ . The ratio  $D_{25}/D_{75}$  was 1.70 and the particle size distribution was very broad. The weight-average molecular weight was  $8.3 \times 10^4$ . Toner was prepared and evaluated in the same manner as in Example 2. When used, fogging was observed due to incomplete charging of toner particles, which was particularly noticeable under high temperature and high humidity conditions.

### Comparative Example 3

The composition was the same as that of Example 3 except that the temperature was not raised during the polymerization but was kept at 75°C throughout the polymerization.

The styrene-styrenesulfonic sodium salt copolymer particles contained in the emulsion particles were generally less than several  $\mu\text{m}$ , but there were particles of 10 to 20  $\mu\text{m}$  size, showing a very broad particle size distribution.

### Claims

1. Charge control resin particles which are spheroidal resin particles formed by dispersion polymerization and composed mainly of a copolymer formed from at least one water-soluble monomer having an anionic group of sulfonic, carboxylic or phosphoric type and at least one oil-soluble monomer,  
the copolymer having a weight-average molecular weight of 3,000 to 50,000, the resin particles having a volume median size of 1  $\mu\text{m}$  or more with the ratio  $D_{25}/D_{75}$  of the 25 volume percent integrated diameter  $D_{25}$  to the 75 volume percent integrated diameter  $D_{75}$  being within the range of 1 to 1.4.
2. Charge control resin particles according to Claim 1, wherein the anionic group is a sulfonic group or an alkali metal salt thereof.
3. Charge control resin particles according to Claim 2, wherein the water-soluble monomer is styrenesulfonic acid sodium salt.
4. Charge control resin particles according to any preceding claim, wherein the oil-soluble monomer comprises both styrene and an acrylic monomer.
5. Charge control resin particles according to any preceding claim, wherein the ratio of the water-

soluble monomer to the oil-soluble monomer forming the copolymer is in the range from 70 : 30 to 99 : 1 by weight.

6. A method for manufacturing charge control resins particles, wherein at least one water-soluble monomer having an anionic group of sulfonic, carboxylic or phosphoric type and at least one oil-soluble monomer are dispersed and polymerized in an aqueous medium containing water and a water-miscible organic solvent in the presence of a dispersion stabilizer and a chain transfer agent, and the resulting copolymer consisting of the water-soluble monomer and the oil-soluble monomer is collected in the form of particles, the method including the steps of :  
forming a nucleus by reaction of the oil-soluble monomer and the water-soluble monomer ; and  
growing the thus formed nucleus ;  
the temperature being higher in the nucleus growing step than in the nucleus forming step.
7. A method for manufacturing a charge control resin according to Claim 6, wherein the mixing ratio of the water and the water-miscible organic solvent in the aqueous medium is in the range of 40 : 60 to 5 : 95 by weight.
8. A method for manufacturing a charge control resin according to claim 6 or 7, wherein the temperature is 60 to 80°C in the nucleus forming step and 70 to 90°C in the nucleus growing step.



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# EUROPEAN SEARCH REPORT

Application Number

EP 90 31 3047

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.5)
Y	EP-A-0330287 (MITA INDUSTRIAL CO., LTD.) * abstract * * page 3, lines 31 - 35 * * page 4, lines 41 - 53 * * page 7, line 55 - page 8, line 5 * ---	1-8	G03G9/087
Y	GB-A-2083051 (KONISHIROKU PHOTO INDUSTRY CO., LTD.) * page 2, lines 15 - 31 * * page 3, lines 1 - 35 * * page 4, lines 13 - 60 * page 6 examples 16, 17* * page 7, lines 5 - 7 * page 7 example 18* ---	1-8	
Y	GB-A-2070036 (KONISHIROKU PHOTO INDUSTRY CO., LTD.) * abstract * * page 2, lines 1 - 12 * * page 3, lines 11 - 14 * * page 4, line 56 - page 5, line 46 * ---	1-8	
A	PATENT ABSTRACTS OF JAPAN vol. 11, no. 253 (P-606)(2700) 18 August 1987, & JP-A-62 59964 (RICOH CO., LTD.) 16 March 1987, * the whole document * -----	1-8	TECHNICAL FIELDS SEARCHED (Int. Cl.5)  G03G9
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 12 APRIL 1991	Examiner HINDIAS E.
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application I : document cited for other reasons & : member of the same patent family, corresponding document			

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