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METHOD OF REMOVING ACCUMULATED CHARGES IN PHOTOELECTROPHORETIC IMAGING

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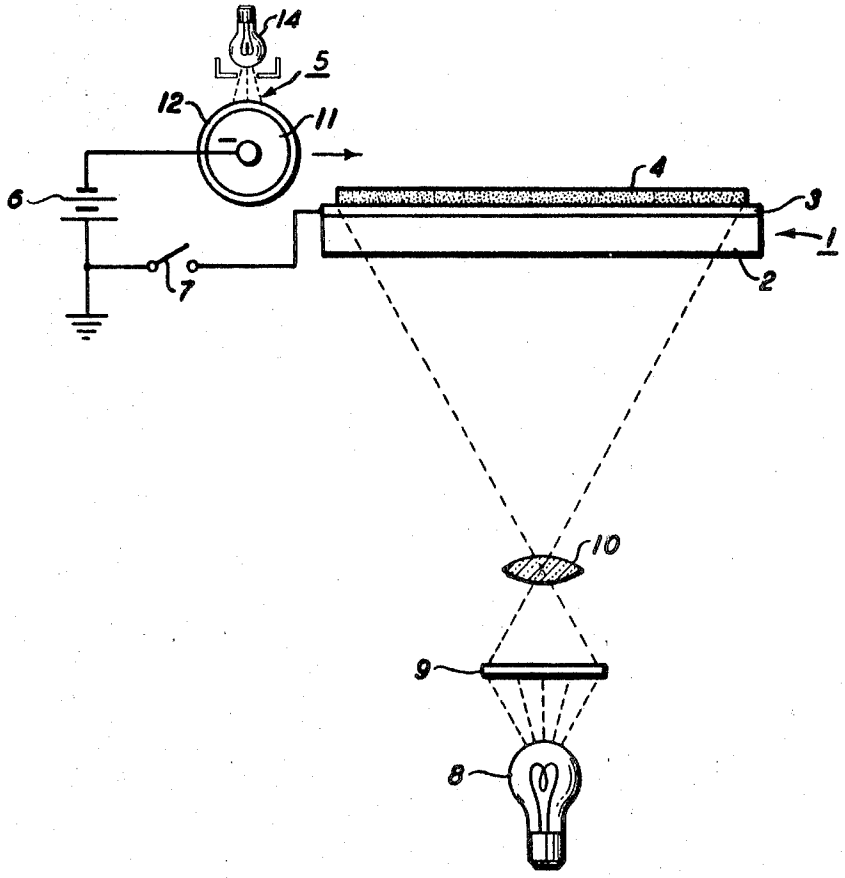

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METHOD OF REMOVING ACCUMULATED CHARGES IN PHOTOELECTROPHORETIC IMAGING

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ABSTRACT OF THE DISCLOSURE

Accumulation of undesired potential on the blocking electrode in a photoelectrophoretic imaging system during successive imaging operations is prevented by incorporating a photoconductive layer in the blocking electrode. The photoconductive layer is discharged by flood illumination between imaging cycles.

BACKGROUND OF THE INVENTION

This invention relates in general to imaging systems. More specifically, the invention concerns an improved photoelectrophoretic imaging system.

There has been recently developed an electrophoretic imaging system capable of producing color images which utilizes electrically photosensitive particles. This process is described in detail and claimed in U.S. Pats. 3,384,566, to H. E. Clark, 3,384,565, to V. Tulagin et al., and 3,383,-993, to Shu-Hsiung Yeh. In such an imaging system, variously colored light absorbing particles are suspended in a non-conducting liquid carrier. The suspension is placed between electrodes, one of which is generally conductive, called the "injecting" electrode and the other of which is generally insulating and called the "blocking" electrode. One of these electrodes is at least partially transparent to activating electromagnetic radiation. The suspension is subjected to a potential difference between the electrodes across the suspension and exposed to an image through said transparent electrode. As these steps are completed, selective particle migration takes place in image configuration, providing a visible image at one or both of the electrodes. An essential component of the system is the suspended particles which must be electrically photosensitive and which apparently undergo a net change in charge polarity upon exposure to activating electromagnetic radiation when within interaction range of one of the electrodes. In a monochromatic system, particles of a single color are used, producing a single colored image equivalent to conventional black-and-white photography. In a polychromatic system, the images are produced in natural color because mixtures of particles of two or more different colors which are each sensitive to light of a specific wavelength or narrow range of wavelengths are used.

This system, using a conductive injecting electrode, a substantially insulating blocking electrode and photosensitive particles dispersed in an insulating carrier liquid between the electrodes has been found to be capable of producing excellent images. Most of the polymeric materials having desirable physical properties for use in blocking electrodes have volume resistivity greater than 10^{15} ohm-cms. However, it has been found that where the blocking electrode surface is made of a highly insulating material, which e.g., has a resistivity of 10^{11} ohm-centimeters or greater, image quality progressively decreases. In particular, where several images are made using the same electrodes in rapid succession, image quality has been found to fall off progressively. While a great many substantially insulating materials are operative when used as a surface for the blocking electrode, i.e., will produce satisfactory images, very few materials have been found which consistently produce high quality images under cyclic imaging conditions due to the accumulation of undesirable charges. The reason for this may be understood by a consideration of the time constant τ in seconds for the discharge of static electricity from an insulator having a dielectric constant K and resistivity ρ in ohm-cm., which may be calculated to be $\tau=8.85\times10^{-14}$ $K\rho$ where the numerical constant has the units seconds cm./ohm. Practical blocking electrodes have dielectric constants between 2 and 200 or more, typically about 10. For a resistivity of $\rho=10^{13}$ ohm-cm. therefore the time constant for 63% discharge of accumulated static is 0.885 K, or about 1.8 to 180 seconds, typically about 9 seconds. For $\rho=10^{14}$ ohm-cm. these times become ten times longer. Since commercial machines must frequently cycle every 0.1 to 1 second, it is necessary to discharge the blocking layer at least this rapidly between cycles. It is necessary to provide electrode layers which are blocking, and therefore insulating, at the imaging station, but which may be rendered temporarily sufficiently conductive, with, e.g., ρ less than about 10^{11} ohm-cm. and preferably 10^{7}–10^{9} ohm-cms. to allow rapid discharge between cycles.

Where the resistivity of the blocking electrode is less than about 10^{6} ohm-cm. there is a tendency for particles which should move to the blocking electrode surface and adhere thereto to exchange charge with the electrode and be reflected back toward the injecting electrode thereby degrading the quality of the image on both the blocking and injecting electrode surfaces.

Many of the physical properties of those materials which are capable of producing excellent images are not ideal. For example, conventional blocking electrode polymers such as Mylar polyester and Tedlar poly (vinyl fluoride) are too highly resistive having resistivities of about 10^{17} and 10^{13} ohm-cms. respectively. Baryta paper surface blocking electrodes produce excellent images; however, the baryta paper surface is humidity sensitive and is not easily cleaned between imaging steps. The limited number of materials known to be useful do not have as wide a range of humidity resistance, cleanability, surface smoothness, abrasion resistance and cost as would be desirable. Thus, there is a continuing need for improved blocking electrode materials and for methods of using the blocking electrode which would permit utilization of a wider range of materials on the blocking electrode surface. It is also desirable to provide a blocking electrode surface which may be easily purged of charge accumulation on its surface during successive imaging cycles.

SUMMARY OF THE INVENTION

It is, therefore, an object of this invention to provide an electrophoretic imaging system which overcomes the above-noted disadvantages.

It is another object of this invention to provide an electrophoretic imaging system capable of utilizing a wide range of blocking electrode materials.

It is another object of this invention to provide a photoelectrophoretic imaging system capable of producing a plurality of images in rapid succession of uniformly high quality.

It is another object of this invention to provide a blocking electrode material which may be readily purged of accumulated charge on its surface.

The above objects, and others are accomplished by providing a photoelectrophoretic imaging system in which the blocking electrode incorporates a photoconductive insulating layer, which is insulating during the photoelectrophoretic imaging step. To eliminate charge buildup the photoconductive layer is then exposed to flood illumination which allows the accumulated charge to pass through the photoconductive layer to a conductive electrode which transports the charge away. It was discovered that such blocking electrodes perform well in cyclic photoelectrophoretic imaging. In contrast, prior blocking electrode materials having surfaces which were highly insulating were found to perform in less than optimum manner, due to the buildup of undesirable electrostatic charge on its surface. This problem was especially great where several images were made in rapid succession since undesired charge continues to build up on the surface of the blocking electrode causing steady degradation of successive images.

The photoconductive blocking electrode may comprise any suitable photoconductive layer. The photoconductor may be homogeneous or a mixture of two or more materials. The photoconductor may be organic or inorganic. The photoconductive layer may comprise photoconductive particles dispersed in insulating or photoconductive insulating binders and may be overcoated. Preferably, the photoconductor is not sensitive to the wavelengths of light used to cause particle migration in the imaging suspension. For example, the photoconductive layer may comprise a homogeneous layer of polyvinyl carbazole which is sensitive to ultraviolet radiation but is comparatively insensitive to visible light. Of course, if electrically photosensitive particles are used which are sensitive to infrared or ultravolet radiation, it would then be desirable to have the photoconductive blocking electrode be insensitive to such radiation. It is not essential, however, that the blocking electrode photoconductive layer be insensitive to the wavelengths of light used to expose the imaging suspension since the imaging suspension will itself act as an efficient light filter. If two or more imaging passes per image are made however, the imaging suspension becomes less dense allowing possible photodischarge of the photoconductive blocking electrode, which is undesirable during imaging. Alternatively, one may choose the photoresistivity of the photoconductive blocking layer to be sufficiently low, compared to that of the electrically photoresistive imaging particles, that a minor exposure during the imaging step is ineffective in reducing resistance. In this case a suitably greater, but still practical, discharge exposure must be applied.

BRIEF DESCRIPTION OF THE DRAWINGS

The advantages of this improved method of imaging will become apparent upon consideration of the detailed disclosure of the invention especially when taken in conjunction with the accompanying drawings wherein the figure is a side sectional view of a simple electrophoretic imaging system utilizing a photoconductive blocking electrode.

Referring now to the figure there is seen a transparent electrode generally designated **1** which in this exemplary instance is made up of a layer of optically transparent glass **2** overcoated with a thin optically transparent layer **3** of tin oxide commercially available under the name NESA glass. This electrode will hereafter be referred to as the "injecting" electrode. Coated on the surface of injecting electrode **1** is a thin layer **4** of finely divided electrically photosensitive particles dispersed in an insulating liquid carrier. Electrically photosensitive for the purposes of this disclosure refers to the properties of a particle which, once attracted near the injecting electrode, will migrate away from it under the influence of an applied electric field when it is exposed to actinic electromagnetic radiation. For a detailed theoretical explanation of the apparent mechanism of operation of the imaging process, see U.S. Pat. No. 3,384,565 issued May 21, 1968 to V. Tulagin et al. and U.S. Pat. No. 3,384,566 issued May 21, 1968 to H. E. Clark the disclosures of which are incorporated herein by reference. Adjacent to liquid suspension **4** is a second electrode **5**, hereinafter called the blocking electrode which is connected to one side of potential source **6** through switch **7**. The opposite side of potential source **6** is connected to the injecting electrode **1** so that when switch **7** is closed an electric field is applied across the liquid suspension **4** between electrodes **1** and **5**. An image projector made up of a light source **8**, a transparency **9**, and a lens **10** is provided to expose the dispersion **4** to a light image of the original transparency **9** to be reproduced. Electrode **5** is made in the form of a roller having a conductive central core **11** connected to potential source **6**. Core **11** is covered with a layer of a photoconductive insulating material **12** which may be any suitable photoconductive insulating material as discussed further below. The particle suspension is exposed to the image to be reproduced while a potential is applied across electrodes **1** and **5** by closing switch **7**. Roller **5** is caused to roll across the top surface of injecting electrode **1** with switch **7** closed during the period of image exposure. This light exposure causes exposed pigment particles originally attracted to electrode **1** to migrate through the liquid and adhere to the surface of blocking electrode **5**, leaving behind a particulate image on the injecting electrode surface which is a duplicate of the original transparency **9**. After exposure, the relatively volatile carrier liquid evaporates off, leaving behind the particulate image. This particulate image may then be fixed in place, as for example, by placing a lamination over its top surface or by virtue of a dissolved binder material in the carrier liquid such as paraffin wax or other suitable binder that comes out of the solution as the carrier liquid evaporates. In an alternative, the particulate image remaining on the injecting electrode may be transferred to another surface and fixed thereon. This system can produce either monochromatic or polychromatic images depending upon the type and number of pigments suspended in the carrier liquid and the color of light to which the suspension is exposed in the process.

Where the above imaging steps are repeated, with cleaning of the blocking electrode but without discharging the blocking electrode and reapplication of the particle suspension onto the injecting electrode between imaging operations, it has been found that there is a steady decrease in the image quality in successive copies. It has been found that this gradual decrease in image quality is due to the accumulation of undesired electrostatic charge on the surface of the blocking electrode. In accordance with this invention before or after the blocking electrode has been cleaned of unwanted particles, photoconductive layer **12** is exposed to a source **14** of actinic electromagnetic radiation which allows the accumulated charge to pass through layer **12** to central core **11**.

The roller blocking electrode configuration shown in the drawing is of course merely representative and any other suitable configuration may be used. Typically, the blocking electrode may be in the form of a moveable or stationary flat plate, or in the form of a belt entrained over rollers.

Photoconductive layer **12** may comprise any suitable photoconductor. Preferably the photoconductor responds substantially only to radiation in the ultraviolet region so that visible light does not cause the blocking electrode to become conductive. Typical photoconductive materials having these properties include: Poly (N-vinyl carbazole); poly (9-vinyl anthracene); poly (acenaphthylene); poly

(2-vinyl quinoline); poly (3-vinyl pyrene); all of which are homogeneous photoconductors whose sensitivity may be augmented by complexing with suitable Lewis acids as described by H. Hoegl in the Journal of Physical Chemistry, 69, 755 (1965); poly (triphenylamine) as described in U.S. Pat. 3,265,496; poly (N-propenyl carbazole) as described in U.S. Pat. No. 3,341,472; and mixtures thereof.

Typical Lewis acids which may be used as sensitizers include tetrachorophthalic anhydride; 2,4,7 trinitro 9-dicyanomethyl fluorene; anthraquinone; boron trifluoride and mixtures thereof.

Binder dispersions or solutions of polycyclic aromatic or heterocyclic compounds, such as the amino aryl 1,3,4-oxadiazoles as described in U.S. Pat. 3,189,447; triazines as described in U.S. Pat. 3,130,046; amino aryl 1,3,4-triazoles as described in U.S. Pat. 3,112,197 and triarylamines as described in Canadian Pat. 678,931; and mixtures thereof. These compounds may also be Lewis acid sensitized.

Homogeneous layers of inorganic photoconductors such as sulfur; zinc sulfide; zinc oxide; and mixtures thereof.

Binder dispersions of inorganic photoconductors such as sulfur; zinc sulfide; zinc oxide; zinc cadmium sulfide; cadmium sulfide; cadmium sulfide selenide and mixtures thereof.

Photoconductive charge transfer complexes of normally non-photoconductive aromatic polymers of the Lewis base type with monomeric Lewis acids; for example, aromatic polycarbonates, epoxies, polyphenyleneoxide, melamine, phenol-aldehyde, phenoxides, silicones, polyurethanes, polysulfones, and mixtures thereof.

In a monochromatic system, particles of a single color are dispersed in the carrier liquid and exposed to a black-and-white image. A single color image results, corresponding to black-and-white photography. In a polychromatic system, the particles are selected so that those of different colors respond to different wavelengths in the visible spectrum corresponding to their principal absorption bands. Also, the pigments should be selected so that their spectral response curves cannot have substantial overlap, thus allowing for color separation and subtractive multicolor image formation. In a typical substractive multicolor system, the particle dispersion should include cyan colored particles sensitive mainly to red light, magenta particles sensitive mainly to green light and yellow particles sensitive mainly to blue light. When mixed together in a carrier liquid, these particles produce a black appearing liquid. When one or more of the particles are caused to migrate from the injecting electrode towards the blocking electrode, they leave behind particles which produce a color equivalent to the color of the impinging light. Thus, for example, red light exposure caused the cyan colored particles to migrate leaving behind the magenta and yellow particles which combine to produce red in the final image. In the same manner, blue and green colors are reproduced by the removal of yellow and megenta, respectively. When white light impinges upon the mix, all particles migrate leaving behind the color of the white or transparent substrate. No exposure leaves behind all pigments which combine to produce a black image. This is an ideal technique of substractive color imaging in that the particles may be each composed of a single component and, in addition, they perform the dual functions of final image colorant and photosensitive medium. Typical photosensitive pigments include those described in copending application Ser. No. 473,607, filed July 21, 1965, now abandoned; U.S. Pat. No. 3,384,488, issued May 21, 1968 to V. Tulagin et al.; copending Ser. No. 445,240, filed Apr. 2, 1969, now U.S. Pat. No. 3,384,632, and U.S. Pat. No. 3,357,989, issued Dec. 12, 1967 to J. F. Byrne et al.

Although various electrode spacings may be employed, spacings of less than 1 mil and extending down even to the point where the electrodes are pressed together as in the case of the roller electrode configuration constitute a particularly preferred form of the invention in that they produce better resolution and, in polychrome imaging, superior color separation results than is produced with wider spacings. This improvement is believed to take place because of the high field strength across the suspension during imaging. It is also possible to provide a transparent blocking electrode with the discharge lamp being located inside the electrode.

DESCRIPTION OF THE PREFERRED EMBODIMENT

The following examples further specifically define the present invention with respect to the elimination of undesired surface charge buildup on the blocking electrode in photoelectrophoretic imaging systems. Parts and percentages are by weight unless otherwise indicated. The examples below are intended to illustrate various preferred embodiments of the photoelectrophoretic imaging system of the present invention. All the following examples are carried out in an apparatus of the general type illustrated in the figure with the addition of means to clean particles from the blocking electrode surface not shown. The imaging suspension comprises the desired electrically photosensitive particles in an insulating carrier liquid and is coated on a NESA glass substrate through which exposure is made. The NESA glass surface is connected in series with a switch, a potential source, and the conductive center of a roller having a coating of a photoconductive insulating blocking electrode material on its surface. The roller is approximately 2½ inches in diameter and is moved across the plate surface at about 4 centimeters per second. The plate employed is roughly 3 inches square and is exposed to light of an intensity of about 2,000 foot-candles as measured on the uncoated NESA glass surface.

EXAMPLE I.—PRIOR ART

A blocking electrode in roller configuration is prepared consisting of a metal core having Mylar polyester film on its surface. A trimix is prepared consisting of about 7 parts of electrically photosensitive pigments dispersed in about 100 parts of Sohio Odorless Solvent 3440, (a mixture of kerosene fractions). The photosensitive pigments comprise equal portions of a cyan pigment, Monolite Fast Blue GS the alpha form of metal free phthalocyanine available from the Arnold Hoffman Company; a magenta pigment, Watchung Red B, an azo pigment available from E. I. du Pont and a yellow pigment, N - 2'' - pyridyl - 8,13 - dioxodinaphtho - (2,1-b; 2'3'-d)-furan-6-carboxamide, prepared by the method disclosed in copending application Ser. No. 421,281, filed Dec. 28, 1964, now U.S. Pat. No. 3,447,922. This dispersion is coated onto the NESA injecting electrode to a thickness of about 3 mils, a negative potential of about 3,000 volts is imposed on the core of the blocking electrode roller and the roller is rolled across the injecting electrode surface while the suspension is exposed to an image from a conventional Kodachrome transparency. When the roller is passed beyond the injecting electrode, potential application and image exposure is stopped. The blocking electrode surface is then cleaned manually using absorbent cotton moistened with Sohio Odorless Solvent 3440. The image produced on the injecting electrode is observed to be of excellent quality and good color balance. The image is transferred electrostatically to a receiving sheet. The roller electrode is then returned to the starting position and the coating, imaging, and transfer steps are repeated seven additional times. The average time between imaging steps is about 3 minutes. The images produced by the successive imaging steps are then compared. It is observed that there is a gradual loss in color density, primarily in blue areas, in succeeding images. A shift toward magenta is observed and progressive overall drastic loss in contrast is observed. This loss of contrast can be avoided by discharging the Mylar by means of an alcohol-moistened cotton pad, and allowing the Mylar

to dry between cycles. This shows that the problem is due to static accumulation on the blocking electrode.

EXAMPLE II

A photoconductive blocking electrode is prepared as follows: about 0.8 gram of a polyester of carbonic acid and bis (4-hydroxphenyl) 2,2-propane (available from General Electric Company as Lexan Polycarbonate Resin grade 125, color 111 powder) is put into a 50 ml. beaker containing about 7.5 grams of dichloromethane and one gram of cyclohexanone. The mixture is agitated until the resin is dissolved. About 0.2 gram of 2,4,7 trinitrofluorenone is then added and again stirred to dissolve all of the materials.

The solution is coated on the conductive metal roller so that when dry the resulting dry layer is approximately 8 microns thick. After drying the coated roller is used as the blocking electrode.

The imaging operation of Example I is repeated except that after each cleaning step the blocking electrode is exposed to U.V. radiation from a 10 watt BLB phosphor blacklight fluorescent bulb in a reflector mount placed 2" above the roller. In this case the time between imaging cycles is approximately 2 seconds. Each of the eight images produced is of substantially equal high color quality.

EXAMPLE III

The experiment of Example II is repeated except that the blocking electrode has a 10 micron layer of polyvinylcarbazole, sensitized by 10 wt. percent of tetrachloro phthalic anhydride, in place of the polycarbonate photoconductive layer. Each of the eight images produced is of substantially equal high color quality. Cycling time may be reduced to 0.5 second.

EXAMPLE IV

The experiment of Example II is repeated except that the photoconductive surface is formed by coating a solution of one gram of 2,5 - bis - [4" - diethylaminophenyl-(1')]-1,3,4-oxadiazole, 0.1 gram of 2,4,7 trinitrofluorenone, and 2 grams of a phenolic resin of the Novolak type in 30 grams sof methylenechloride and drying. Each of the eight images produced is of substantially equal high color quality. Cycling time is about 2 seconds.

EXAMPLE V

A blocking electrode is prepared as in Example II. In this example the tri-mix is replaced with a suspension of 7 parts of the "X"-form of metal free phthalocyanine prepared as shown in U.S. Pat. 3,357,989 in Sohio 3440. Imagewise exposure is adjusted to 200 foot candles and the roller is rolled across the suspension at about 50 cm./sec. For monochrome imaging the blocking electrode is polarized 3000 v. positive relative to the injecting electrode. The imaging process of Example II is then repeated producing 8 negative monochrome images of substantially equal density and contrast on the photoconductive blocking electrode. A positive image is formed on the surface of the injecting electrode. Either image may be transferred if desired.

EXAMPLE VI

The experiment of Example V is repeated except the imaging suspension is deposited on the photoconductive blocking electrode instead of on the NESA electrode and imaged. A positive image is formed on the NESA electrode and a negative image is formed on the photoconductive blocking electrode. Successive pairs of images are of substantially equal high quality.

EXAMPLE VII

A one micron thick continuous polycrystalline layer of N - 2" - pyridyl - 8,13 - dioxodinaphtho - (2,1-b; 2',3'-d)-furan-6-carboxamide, is deposited on a flexible belt of 5 mil thick cartridge brass. It is overcoated with a 10 micron layer of poly (N-vinyl carbazole) as described above. The blocking electrode belt is tracked over a roller equivalent in position to that shown in FIG. 1 and a second roller, two diameters distant from the first. Three BLB lamps, with proper baffles, are mounted over the second roller and used to discharge the charges accumulated on the poly (N-vinyl carbazole). The pigment acts as sensitizer for the discharge of charge on the adjacent PVK layer. The length of the belt extends the time available for discharge by a factor of about 2.5 fold.

EXAMPLE VIII

The experiment of Example V is repeated except that instead of applying a D.C. potential between the injecting electrode and the electrode behind the blocking layer, the blocking electrode is first illuminated then corona charged, each cycle, to a positive potential of 2000 volts. The injecting electrode is grounded. Each of the eight images produced is of substantially equally high density and contrast. Positive images are formed on the NESA, withs negative images formed on the photoconductive blocking electrode.

EXAMPLE IX

The experiment of Example VIII is repeated except that the potential on the photoconductive blocking electrode is a negative 2000 volts. Negative images are formed on the NESA electrode and positive images are formed on the photoconductive blocking electrode. Successive pairs of images are of substantially equal quality.

Although specific components and proportions have been described in the above examples, other suitable materials, as listed above, may be used with similar results. In addition, other materials may be added to the photoconductive layers to synergize, enhance or otherwise modify their properties. For example, the photoconductive layers may be dye or impurity sensitized in known ways to alter their spectral response. Other modifications and ramifications of the present invention will occur to those skilled in the art upon a reading of the present disclosure. These are intended to be included within the scope of the invention.

What is claimed is:

1. A method of photoelectrophoretic imaging comprising the steps of:
   - (a) providing a first electrode having a photoconductive insulating layer, and a second at least partially transparent electrode;
   - (b) placing on at least one of said electrodes a layer of a suspension of electrically photosensitive particles each of said particles dispersed in an insulating carrier liquid, comprising a pigment, said pigment being both the primary electrically photosensitive component and colorant for the particles;
   - (c) exposing said suspension to an image of electromagnetic radiation to which the suspension is sensitive;
   - (d) imposing an electric field between said first and second electrodes across said suspension until at least one image is formed;
   - (e) exposing said first electrode to electromagnetic radiation to which said photoconductive insulating layer is sensitive to eliminate charges accumulated thereon; and,
   - (f) repeating steps (b) through (d) at least one additional time.

2. The method of claim 1 and further including the step of cleaning said first electrode at least subsequent to step (d).

3. The method of claim 1 and further including the step of transferring at least one of said images to an image receiving member.

4. The method of claim 1 wherein said photoconductive layer is overcoated with a layer of poly (N-vinyl carbazole).

5. The method of claim 1 wherein said photoconductive layer is exposed to ultraviolet radiation and said imaging suspension is exposed to visible light.

6. The method of claim 1 wherein said photoconductive layer is responsive to ultraviolet radiation.

7. The method of claim 1 wherein said photosensitive particles comprise cyan particles primarily responsive to red light, magenta particles primarily sensitive to green light and yellow particles primarily responsive to blue light.

8. The method of claim 1 wherein the surface of said first electrode is corona charged prior to being brought into contact with said imaging suspension.

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