FIXING PROCESS FOR PHOTOELECTROPHORETIC IMAGING

Original Filed March 20, 1970 2 Sheets-Sheet 1

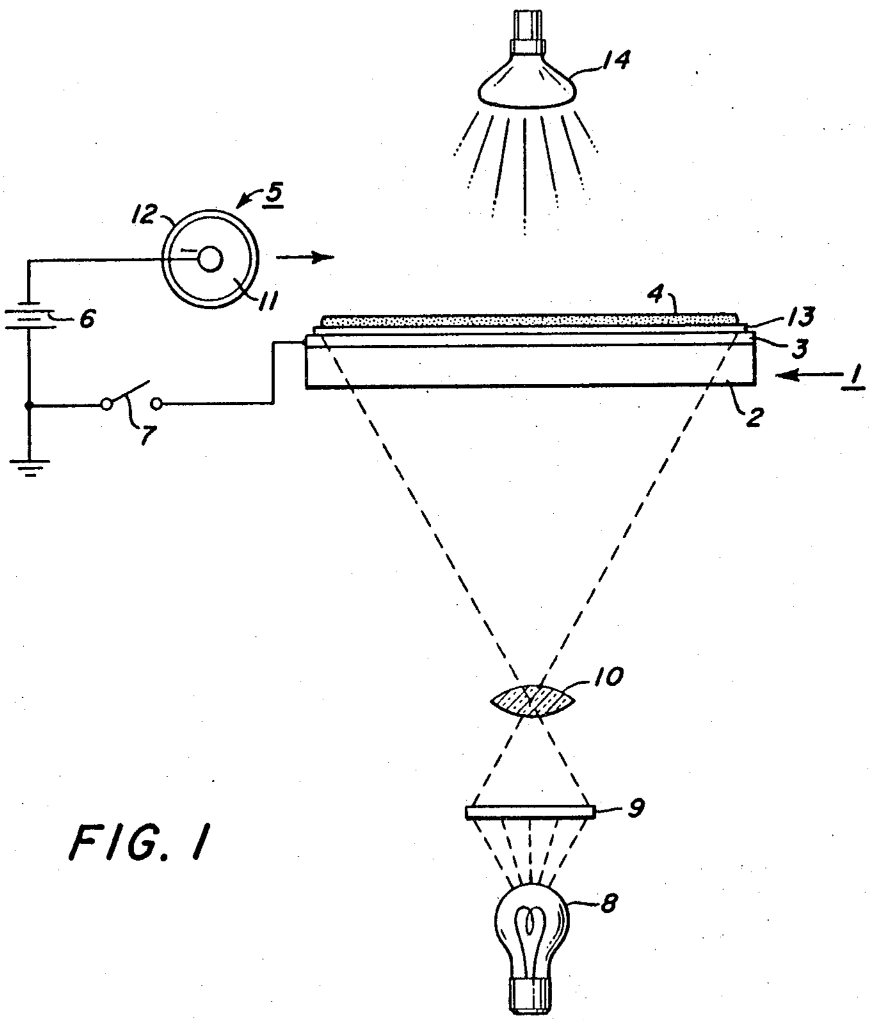

FIG. 1

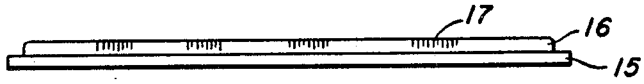

FIG. 2

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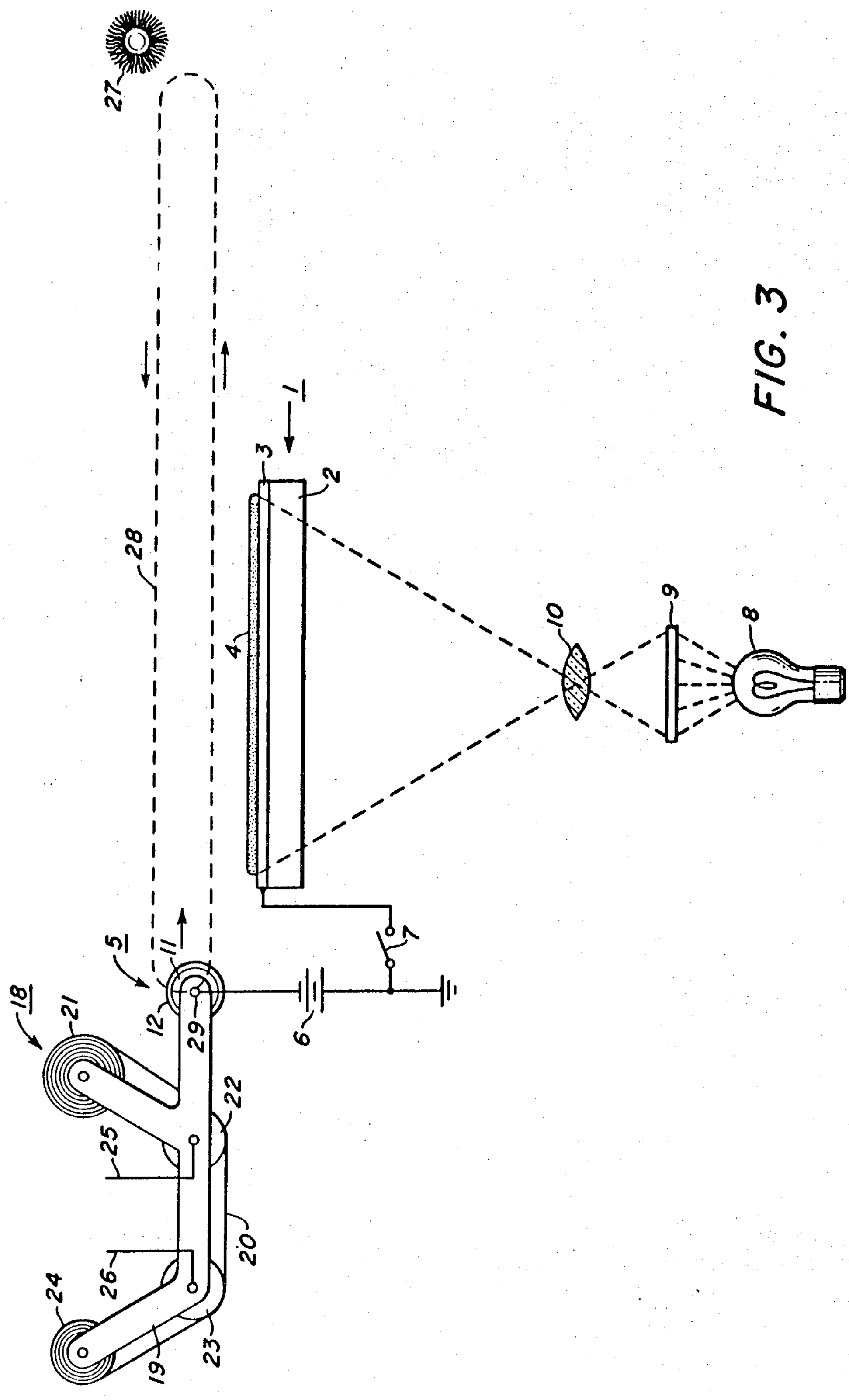

FIG. 3

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3,705,797

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FIXING PROCESS FOR PHOTOELECTROPHORETIC IMAGING

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Continuation of application Ser. No. 808,921, Mar. 20, 1969, which is a continuation-in-part of applications Ser. No. 459,860, May 28, 1965, Ser. No. 677,706 and Ser. No. 677,707, both Oct. 24, 1967, all now abandoned. This application Feb. 18, 1970, Ser. No. 12,364

Int. Cl. G03g *13/22*

U.S. Cl. 96—1 R 9 Claims

ABSTRACT OF THE DISCLOSURE

Methods and apparatus for photoelectrophoretic imaging utilizing thermo-adhesive layers for image transfer and fixing.

BACKGROUND OF THE INVENTION

This invention relates in general to imaging systems and more specifically, to an improved electrophoretic imaging system. This application is a continuation of application Ser. No. 808,921 filed Mar. 20, 1969. Ser. No. 808,921 was a continuation of application Ser. Nos. 459,860 filed May 28, 1965, Ser. No. 677,706 and 677,707 filed Oct. 24, 1967, all of the above now abandoned.

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 copending applications Ser. Nos. 384,737 now U.S. Pat. 3,384,565, issued May 21, 1968; 384,681 now U.S. Pat. 3,384,566, issued May 21, 1968 and 384,680 now U.S. Pat. 3,383,993, issued May 21, 1968, all filed July 23, 1964. In such an imaging system, variously colored light-absorbing particles are suspended in a nonconductive liquid carrier. The suspension is placed between electrodes, subjected to a potential difference and exposed to an image. 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, through interaction with one of the electrodes. In a monochromatic system, particles of a single color may be 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 only to light of a specific wave-length or narrow range of wave-lengths are used. Particles used in this system must have both intense and pure colors and be highly photosensitive.

After the exposure and particle migration steps are completed, the electrodes are separated and the carrier liquid is allowed to evaporate. This leaves images on one or both of the electrodes made up of selectively deposited particles. The carrier liquid may contain a small proportion of a wax or other binder which would serve to bind the particles together in the images. However, if more than a very small amount of binder material is used, undesirable interference with the imaging process takes place. Thus, the images are at this time in a fragile and easily damaged condition. It has been suggested that a transparent sheet be laminated over the images, or a transparent binder resin be sprayed over the images to form a protective coating. While, when carefully done, these techniques will protect the image, the image is often damaged during the application of the protective material. These protective techniques are not suitable for a mechanized system. Also, when it is desired to transfer the image from the electrode material to a receiving sheet, the dangers of smudging or otherwise damaging an unfixed image is very great. Thus, there is a continuing need for a better system for fixing the particulate image formed on the electrode surface and/or for permitting transfer of said image to a receiving sheet.

SUMMARY OF THE INVENTION

It is, therefore, an object of this invention to provide a method of fixing a particulate electrophoretic image which overcomes the above-noted disadvantages.

It is another object of this invention to provide a method of protecting an electrophoretic image from damage.

It is another object of this invention to provide a method of transferring an electrophoretic image to a receiving sheet.

It is still another object of this invention to provide an electrophoretic imaging method capable of producing images which may be handled without damage thereto.

It is still another object of this invention to provide a continuous method of forming electrophoretic images and protecting them against abrasion damage.

The foregoing objects and others are accomplished in accordance with this invention by providing a thermo-adhesive layer, which when brought into contact with an electrophoretic particulate image in a softened state, will permit the particles to be embedded in the layer and be permanently held by the layer when it is permitted to reharden. This thermo-adhesive layer may be coated on the electrode upon which an image is to be formed. After the image is formed, the thermo-adhesive layer is softened and the image is pressed against the layer, thereby embedding the particles therein. When the thermo-adhesive is permitted to reharden, the particles are permanently set therein and are thereby protected against damage from contact with other objects. Alternatively, the thermo-adhesive layer may be coated on a receiving sheet. After an electrophoretic image is formed on an electrode, the receiving sheet is heated to soften the thermo-adhesive and brought into contact with the image. The adhesive picks up the particulate image and when rehardened provides a protective image matrix.

BRIEF DESCRIPTION OF THE DRAWINGS

The advantages of this improved electrophoretic imaging process and apparatus will become apparent upon consideration of the following detailed disclosure of the invention; especially when taken in conjunction with the accompanying drawings wherein:

FIG. 1 shows a side view of a simple exemplary system for carrying out the process of this invention wherein a thermo-adhesive image fixing layer is coated on the imaging electrode;

FIG. 2 shows a simple exemplary receiving sheet capable of accepting transfer of an electrophoretic particulate image from an imaging electrode; and,

FIG. 3 shows a continuous system for forming, transferring and fixing electrophoretic particulate images.

Referring now to FIG. 1, 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." On the surface of injecting electrode **1** is a layer of a thermo-adhesive material **13**, coated on, for example, a conductive cellophane film; layer **13** is capable of becoming tacky when heated by infrared lamp schematically shown at **14.**

Over the solidified thermo-adhesive layer **13** is coated a thin layer **4** of finely divided photosensitive particles dispersed in an insulating liquid carrer. The term "photosenstive," for the purposes of this application, refers to the properties of a particle which, once attracted to the injecting electrode, will migrate away from it under the influence of an applied electric field when it is exposed to activating electromagnetic radiation. For a detailed theoretical explanation of the apparent mechanism of operation of the imaging process, see the above-mentioned copending applications Serial Nos. 384,737 now U.S. Pat. 3,384,565; 384,681 now U.S. Pat. 3,384,566; and 384,680 now U.S. Pat. 3,383,993, the disclosures of which are incorporated herein by reference. Adjacent to the liquid suspension **4** is a second electrode **5**, hereinafter called the "blocking electrode," which is connected to one side of the potential source **6** through a 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** and thermo-adhesive layer **13** 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 the potential source **6**. The core is covered with a layer of a blocking electrode material **12**, which may be, for example, baryta paper. The pigment suspension is exposed to the image to be reproduced while a potential is applied across the blocking and injecting electrodes 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 the blocking electrode, leaving behind a pigment image on the surface of the thermo-adhesive layer **13** which is a duplicate of the original transparency **9**. Particles adhering to the surface of a blocking electrode **5** may be cleaned therefrom and the exposure steps repeated, if desired. The additional steps of exposing and cleaning the blocking electrode have been found to increase color purity and color balance. After exposure, the relatively volatile carrier liquid evaporates off, leaving behind the pigment image. At this time, the pigment pigment image is very susceptible to damage by contact with any object since it consists of, in effect, loosely held particles. The image may be fixed or set by heating the thermo-adhesive layer **13**, as by infrared lamp **14**. Heating the thermo-adhesive layer causes it to become tacky as further explained below and causes the image particles to be embedded therein. The thermo-adhesive layer is allowed to harden and may then be stripped from the injecting electrode and handled without fear of smudging or other damage from surface contact.

In an alternative process, the pigment suspension **4** may be coated directly on the NESA glass surface **13**, omitting the intermediate thermo-adhesive layer **13**. After the imaging steps are carried out as discussed above, a particulate image remains on the NESA glass surface. The particulate image may then be transferred to a receiving sheet such as is shown in FIG. 2. This receiving sheet comprises a base layer **15** which may be paper, cellophane, or other suitable materials. On this base layer is coated a layer of a thermo-adhesive material **16**, the same as layer **13** discussed above. This layer may be made tacky by heating by contact, or by infrared radiation, and be brought into contact with the particulate image on the electrode **1**. Or, the solidified thermo- adhesive layer may be brough into contact with the particulate image and then heated. In either case, the particles become embedded in the thermo-adhesive layer and are fixed therein when the layer is allowed to reharden. The

receiving sheet is then removed from the injecting electrode and may be handled in any conventional manner.

FIG. 3 shows an exemplary system for continuously forming a photoelectrophoretic image, transferring the image to a receiving sheet and fixing the image thereon. In the embodiment of this figure, the transparent electrode **1** and the blocking electrodes are the same as in the above-discussed embodiment of FIG. 1. Here, however, a tractor **18** is coupled to the blocking electrode **5** to automatically transfer and fix the positive image formed on the NESA glass surface **3**. The tractor **18** comprises a frame **19** which supports the blocking electrode **5** and image transfer means for movement across the imaging surface. The transfer means consists of a continuous web **20** of transfer material, e.g. paper, on which is coated a thermo-adhesive layer. The web is mounted on supply roller **21** and is adapted to pass in contact with heating guide roller **22** and cooling guide roller **23** on its way to take-up roller **24**. Heating guide roller **22** is provided with internal heating means capable of heating the thermo-adhesive layer above its softening temperature as it passes said roller. This heating means, schematically shown at **25** may comprise any conventional means, e.g. a pipe admitting steam to the interior of the roller, electrical resistance heating means connected to a power supply or Peltier junction heating means connected to a power supply. Cooling guide roller **23** is supplied with coolant sufficient to cool the thermo-adhesive below its softening point as it passes said roller. The cooling means, schematically shown at **26**, may comprise any conventional means, e.g. cooling water piped to the roller, or Peltier junction means connected to a power supply. In operation, a tri-mix **4** is applied to the transparent electrode **1**. The tri-mix is exposed to an image and the tractor and blocking electrode are moved from left to right across the imaging surface. As the blocking electrode passes the imaging surface, unwanted particles migrate to the blocking electrode surface leaving a positive particulate image on the NESA glass surface **3**. As the tractor reaches the NESA surface, web **20** contacts the NESA surface **3** without relative movement with respect thereto. The thermo-adhesive surface on web **20** is softened by heat applied at roller **22**. The particulate image becomes embedded in the softened surface of the thermo-adhesive layer. Then as the thermo-adhesive passes cooling roller **23**, it is rehardened and wound up on takeup roll **24**. When the tractor and blocking electrode reach the end of their travel, brush **27** cleans unwanted pigment from the surface of blocking electrode **5**. The tractor is then raised slightly and returned to the starting position without again contacting the transparent electrode surface. The dashed line **28** schematically indicates the path taken by axle **29** of the roller electrode during the imaging and return movements. As can be seen, the device is capable of continuously forming, transferring, fixing and storing photoelectrophoretic images.

The thermo-adhesive layers **13** and **16** as described above may comprise any suitable materials. The only requirements are that they be solid at room temperatures and be capable of softening and becoming tacky at reasonably elevated temperatures. When the layer is to be coated directly on the injecting electrode surface, there is the further requirement that they have proper conductive characteristics. Where the thermo-adhesive layer is coated on the blocking electrode surface the layer should have a resistivity between 10^8 and 10^{15} ohm centimeters. The thermoadhesive layer preferably comprises a binder material and a thermo-solvent for the binder. The "thermo-solvent" comprises a material that is solid at room temperature and melts slightly above room temperature, thereby causing the binder-solvent layer to be tacky and permit particles in contact therewith to be embedded therein. Where the final image is to be viewed by projection, the thermo-adhesive layer should be transparent.

Any suitable mixture of binder resin and thermo-solvent may be used in the process of this invention. Optimum results have been obtained with mixtures of Vinylite VYNS, a vinyl chloride-vinyl acetate copolymer, available from Union Carbide Corporation, and Santolite MHP, an aryl sulfonamide-formaldehyde copolymer available from Monsanto; polyvinylpyrrolidone-vinylacetate copolymer and Santolite MHP, and Vinylite VYHO and Aroclor 4465, a blend of chlorinated biphenyls and chlorinated triphenyls. Best results have been obtained with, and therefore, the preferred formulation is, a mixture of 1 part by weight EXON-470, a vinylchloride-vinylacetate copolymer available from Firestone, about 1 part by weight Santicizer 1-H, a sulfonimide resin available from Monsanto, dissolved in about 10 parts acetone. This formulation is preferably coated to a thickness of about 0.5 mil and dried. Any other suitable mixture of binder resin and thermo-solvent may be used. Typical binder resin materials include polyethylenes, polystyrenes, copolymers of vinylchloride and vinylacetate, copolymers of vinylpyrrolidione and vinylacetate, polyvinyl methacrylates, polyvinyl propylene, polyvinylchloride, cellulose acetate, chlorinated rubber, and mixtures and copolymers thereof. Typical thermo-solvents having melting points slightly above room temperature include (with melting temperatures in parentheses) triphenyl phosphate (48° C.); dicyclohexyl phthalate (63° C.); diphenyl phthalate (69° C.); Aroclor 5442 (46–52° C.) a chlorinated polyphenyl available from Monsanto; Santicizer 3 (58° C.), N-ethyl-p-toluenesulfonamide, available from Monsanto; Santolite MHP (62° C.) a sulfonamideformaldehyde resin available from Monsanto; Santicizer 1-H (82° C.) N-cyclohexyl-p-toluenesulfonamide, available from Monsanto; acenaphthene (94° C.) acetanilide (113° C.); o-acetoacetotoluidide (105° C.); o-acetotoluidide (101° C.); o-chloroacetoacetanilide (103° C.); 2-chloro-4-nitroaniline (106° C.); p-dibromobenzene (87° C.); p,p'-methylenedianiline (93° C.); alpha-naphthol (95° C.); beta-naphthol (85° C.); 2-naphthylamine (110° C.), m-nitroaniline (112° C.); 4-nitrobiphenyl (97° C.); sorbitol hexoacetate (98° C.); 2,4-toluenediamine (97° C.) and mixtures thereof.

Any suitable ratio of binder to thermo-solvent may be used. Ratios of from 0.5 to 4 parts binder resin for each part of thermo-solvent may be used. For optimum results, a ratio of binder resin to thermo-solvent of about 1:1 is preferred. This ratio may vary depending upon the particular binder resin and thermo-solvent selected. The thickness of the thermo-adhesive layer should preferably be between 0.1 and 4 mils. The optimum balance between effective transfer and economy of materials has been found to occur with a thermo-adhesive thickness of about 0.5 mil.

DESCRIPTION OF PREFERRED EMBODIMENTS

The following examples further specifically define the present invention with respect to the use of thermo-adhesive layers to fix and/or transfer electrophoretic images. The parts and percentages are by weight unless otherwise indicated. The examples below are intended to illustrate various preferred embodiments of the electrophoretic image fixing and transferring process of this invention.

All of the following Examples I–IV are carried out in an apparatus of the general type illustrated in FIG. 1. In different examples, however, the thermo-adhesive layer may be placed on the NESA glass substrate, or on the blocking electrode surface, or on a separate receiving sheet such as shown in FIG. 2. The NESA glass surface is connected in series with a switch, a potential source, and a conductive center of a roller having a coating of baryta paper on its surface. The roller is approximately

2½ inches in diameter and is moved across the plate surface at about 1.45 centimeters per second. The plate employed is roughly 3 inches square and is exposed with a light intensity of 8,000 foot candles as measured on the uncoated NESA glass surface. Unless otherwise indicated, 7 percent by weight of the indicated pigments in each example are suspended in Sohio Odorless Solvent 3440, a kerosene fraction available from Standard Oil of Ohio, and the magnitude of the applied potential is 2,500 volts. Exposure is made with a 3,200° K, lamp through a multicolor "Kodachrome" transparency.

EXAMPLE I

Equal parts by weight of Vinylite VYNS, a vinylchloride vinylacetate copolymer available from Union Carbide Corporation and Santolite MHP, an aryl sulfonimide formaldehyde copolymer available from Monsanto are mixed in an acetone solvent and coated to a thickness of about 10 microns on a cellophane film which is placed on the NESA glass substrate. A tri-mix comprising a cyan pigment, Monolite Fast Blue GS, the alpha form of metal free phthalocyanine, C.I. No. 74100, available from Arnold Hoffman Co.; a magenta pigment, Vulcan Fast Red BBE toner 35-2201, 3,3'-dimethoxy-4,4'-biphenyl-bis (1" - phenyl-3"-methyl-4"-azo-2"-pyrazolin-5"-one), C.I. No. 21200, available from Collway Colors Company; and a yellow pigment, Indofast Yellow Toner, flavanthrone, C.I. No. 70600, available from Harmon Colors Company, is dispersed in about 100 parts Sohio Odorless Solvent 3440, and coated onto the thermo-adhesive coated cellophane layer on the NESA glass surface. A potential is imposed across the suspension, and an image is formed as discussed above. After the image is formed, residual carrier is allowed to evaporate and the coated cellophane surface is heated until the thermo-adhesive softens with an infrared lamp. A roller having a fluorocarbon coated surface is rolled across the thermo-adhesive layer to press the particulate image into the thermo-adhesive surface. The heating is then stopped and the thermo-adhesive surface is allowed to reharden. An excellent image results, with a tough surface resistant to abrasion damage.

EXAMPLE II

Equal portions of a vinyl pyrrolidone-vinylacetate copolymer is mixed with Santolite MHP in an acetone solvent condition. The solution is coated onto a baryta paper blocking electrode surface to a thickness of about 10 microns and allowed to harden thereon. A tri-mix is prepared, comprising a cyan pigment, Cyan Blue GTNF, the beta form of copper phthalocyanine, C.I. No. 74160, available from Collway Colors Company; a magenta pigment, Quindo Magenta RV-6803, a quinacridone pigment available from Harmon Color Co.; and a yellow pigment Algol Yellow GC, 1,2,5,6-di(C,C'-diphenyl)-thiazole-anthraquinone, C.I. No. 67300, available from General Dyestuffs Co., in about 100 parts Sohio Odorless Solvent 3440. This tri-mix is coated onto the NESA glass substrate and an image is produced as in Example I. The blocking electrode is cleaned of residual unwanted pigments, then the surface is heated until it becomes tacky. Then the roller is rolled across the NESA surface picking up and embedding the image particles therefrom. The thermo-adhesive is allowed to re-harden. The baryta paper carrying the pigment containing adhesive layer is removed from the roller electrode and examined. An image of excellent quality with a tough, abrasion resistant surface is seen.

EXAMPLE III

Equal proportions of Vinylite VYNS and Aroclor 4465, a blend of chlorinated bi-phenyls and chlorinated triphenyls, available from Monsanto, is dissolved in acetone and the solution is coated onto a paper sheet to a thickness of about 5 microns. A tri-mix is prepared as in Example II above and is coated on the NESA glass substrate. An image is produced thereon as in Example I above. The thermo-adhesive coated sheet is heated by placing it on a platen held at about 90° C. When the thermo-adhesive has reached a tacky state, it is removed from the platen and pressed down on the particulate image on the NESA sheet. Upon removal of the sheet and rehardening of the thermo-adhesive, an excellent image is seen with a hard abrasion resistant surface.

EXAMPLE IV

Equal proportions of EXON 470, and Santicizer 1–H are dissolved in acetone and the soltuion is coated onto a long paper web and allowed to dry. The web is wound on a roller and placed in a device such as shown in FIG. 3. A tri-mix prepared as in Example I is coated onto the NESA glass electrode. An image is produced as in Example I. Unwanted pigment particles migrate to the blocking electrode and are removed therewith, leaving a particulate image remaining on the NESA surface. As the tractor passes over the NESA plate, the particulate image is pressed into the heated thermo-adhesive surface. As the paper web is wound past the cooled roller, the thermo-adhesive hardens, resulting in a tough image surface which can be wound onto the takeup roller. The imaging operations may then be repeated without delay.

Although specific components and proportions have been stated in the above description of preferred embodiments of the thermo-adhesive layer, other suitable materials, as listed above, may be used with similar results. In addition, other materials may be added to the mixture to synergize, enhance, or otherwise modify its properties. For example, whiteners may be added to the thermo-adhesive to brighten the image, especially where an inexpensive grade of paper is used for transfer. Where the thermo-adhesive layer is used on the injecting electrode, additives may be included to increase the conductivity of the layer as desired.

Other modifications and ramifications of the present invention will occur to those skilled in the art upon a reading of the disclosure. These are intended to be included within the scope of this invention.

What is claimed is:

1. The method of photoelectrophoretic imaging comprising the steps of:

(a) forming a layer of an imaging suspension comprising electrically photosensitive particles in a substantially insulating carrier liquid on a transparent conductive electrode;

(b) providing a solid layer comprising a resinous binder and a thermo-solvent for said resinous binder on a second electrode; said solid layer having a resistance of between 10^8 and 10^{15} ohm centimeters;

(c) exposing said suspension to a pattern of electromagnetic radiation while contacting the free surface of said imaging suspension with said second electrode and applying a potential difference between said second electrode and said transparent conductive electrode until an image is formed on said second electrode;

(d) heating said solid layer until said layer is at least partially softened; and,

(e) cooling said layer to fix said image.

2. The method of claim 1 wherein said resinous binder thermo-solvent layer comprises from about 0.5 to about 4 parts resin based on one part thermo-solvent.

3. The method of claim 1 wherein said resinous binder thermo-solvent layer has a thickness of from about 0.1 mil to about 4 mils.

4. The method of claim 1 wherein said resinous binder is selected from the group consisting of a vinylchloride-vinylacetate copolymer and a vinylpyrrolidone-vinylacetate copolymer.

5. The method of claim 1 wherein said thermo-solvent is selected from the group consisting of an aryl sulfonamide-formaldehyde copolymer and a blend of chlorinated biphenyls and chlorinated triphenyls.

6. The method of photoelectrophoretic imaging comprising the steps of:

(a) providing a solid layer comprising a resinous binder and a thermo-solvent for said resinous binder on a transparent conductive electrode, said layer having a thickness of from about 0.1 mil to about 4 mils;

(b) forming a layer of an imaging suspension comprising electrically photosensitive particles in a substantially insulating carrier liquid on said solid layer;

(c) exposing said imaging suspension to a pattern of electromagnetic radiation while contacting the free surface of said imaging suspension with a second electrode and applying a potential difference between said second electrode and said transparent conductive electrode until an image is formed on said solid layer;

(d) heating said solid layer until said solid layer is at least partially softened; and,

(e) cooling said solid layer to fix said image.

7. The method of claim 6 wherein said resinous binder thermo-solvent layer comprises from about 0.5 to about 4 parts resin based on one part thermo-solvent.

8. The method of claim 6 wherein said resinous binder is selected from the group consisting of a vinylchloride-vinylacetate copolymer, and a vinylpyrrolidone-vinylacetate copolymer.

9. The method of claim 6 wherein said thermo-solvent is selected from the group consisting of an aryl sulfonamide-formaldehyde copolymer and a blend of chlorinated biphenyls and chlorinated triphenyls.

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U.S. Cl. X.R.

96—1.3; 117—17.5; 204—181