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RECEIVED BY VICKING
CLERK, U.S. DISTRICT COURT
NORTHERN DISTRICT OF CALIFORNIA

6 Attorneys for Plaintiff CopyTele, Inc.

7
8 UNITED STATES DISTRICT COURT
9 NORTHERN DISTRICT OF CALIFORNIA

NC

10 COPYTELE, INC., a Delaware Corporation,

C No. 13 0378

11 Plaintiff,

COMPLAINT FOR PATENT
INFRINGEMENT

12 v.

JURY TRIAL DEMANDED

13 E INK HOLDINGS, INC., a Taiwanese
corporation; and E INK CORPORATION, a
14 Delaware corporation,

15 Defendants.

16 Plaintiff CopyTele, Inc. ("CopyTele") files this Complaint for patent infringement against
17 Defendants E Ink Holdings, Inc. and E Ink Corporation (collectively, "E Ink") and alleges as
18 follows:

19 NATURE OF THE ACTION

20 1. This is an action for patent infringement under the patent laws of the United States,
21 Title 35, United States Code.

22 PARTIES

23 2. Plaintiff CopyTele, Inc. is a Delaware corporation with its principal place of
24 business at 900 Walt Whitman Road, Melville, New York 11747.

25 3. On information and belief, defendant E Ink Holdings Inc. is a Taiwanese
26 corporation, having its principal place of business at 1, 3 Li-Hsin Rd, 1 Hsinchu Science Park,
27 Hsinchu 300 Taiwan, Republic of China. E Ink Holdings Inc. manufactures electronic products,
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1 including electrophoretic displays, in Taiwan and China and, directs those products to the United
2 States, including California, through established distribution channels involving various third
3 parties, knowing that these third parties will use their respective nationwide contacts and
4 distribution channels to import into, sell, offer for sale, and use these products in California and
5 elsewhere in the United States.

6 4. On information and belief, defendant E Ink Corporation is a domestic subsidiary of
7 E Ink Holdings Inc. that directly and indirectly imports into, sells, and offers for sale its products
8 in California and elsewhere in the United States. E Ink Corporation is a Delaware corporation,
9 having its principal place of business at 733 Concord Avenue, Cambridge, Massachusetts 02138.
10 E Ink Corporation also maintains corporate offices in San Mateo, California. E Ink Corporation
11 markets and sells E Ink Holdings Inc.'s products throughout the United States.

12 JURISDICTION AND VENUE

13 5. This action arises under the patent laws of the United States, 35 U.S.C. § 1, *et seq.*,
14 including without limitation §§ 271, 281, 283, 284 and 285. This Court has subject matter
15 jurisdiction over this case for patent infringement under 28 U.S.C. §§ 1331 and 1338(a).

16 6. This Court has personal jurisdiction over Defendants E Ink. On information and
17 belief, both Defendants do business in the State of California and the Northern District of
18 California, and are selling and offering to sell, and have within a reasonable period prior to the
19 filing of this action, sold and offered to sell their products, including their electrophoretic display
20 products, to customers in this State and in this District, either directly or indirectly. Additionally,
21 or in the alternative, E Ink has placed its products, including electrophoretic display products, into
22 the stream of commerce, knowing or reasonably expecting that such products will be used, sold or
23 offered to be sold in this State and in this District. E Ink has intentionally established distribution
24 channels to offer its products for sale and to sell its products, including the infringing products, in
25 this State and in this District.

26 7. Venue is proper in the Northern District of California pursuant to 28 U.S.C. §§
27 1391(b)-(c) and 1400(b) because E Ink resides in this District within the meaning of 28 U.S.C. §
28

1 1391(c), and because a substantial part of the events or omissions giving rise to the claims
2 asserted herein occurred in this District.

3 **BACKGROUND**

4 8. United States Patent No. 5,964,935 (“the ’935 patent”), entitled “Initiator-treated
5 pigment particles and method for preparing same,” was duly and legally issued by the United
6 States Patent and Trademark Office on October 12, 1999, naming Jing Hong Chen and Frederic
7 E. Schubert as the inventors and CopyTele, Inc. as the assignee. Plaintiff CopyTele, Inc. is the
8 assignee and sole owner of all rights, title and interest in the ’935 patent. A true and correct copy
9 of the ’935 patent is attached hereto as Exhibit A.

10 9. United States Patent No. 6,113,810 (“the ’810 patent”), entitled “Methods of
11 preparing electrophoretic dispersions containing two types of particles with different colors and
12 opposite charges,” was duly and legally issued by the United States Patent and Trademark Office
13 on September 5, 2000, naming Wei-Hsin Hou and Frederic E. Schubert as the inventors and
14 CopyTele, Inc. as the assignee. Plaintiff CopyTele, Inc. is the assignee and sole owner of all
15 rights, title and interest in the ’810 patent. A true and correct copy of the ’810 patent is attached
16 hereto as Exhibit B.

17 10. United States Patent No. 6,194,488 (“the ’488 patent”), entitled “Method for
18 making polymer-coated pigment particles using initiator-treated pigments,” was duly and legally
19 issued by the United States Patent and Trademark Office on February 27, 2001, naming Jing
20 Hong Chen and Frederic E. Schubert as the inventors and CopyTele, Inc. as the assignee.
21 Plaintiff CopyTele, Inc. is the assignee and sole owner of all rights, title and interest in the ’935
22 patent. A true and correct copy of the ’488 patent is attached hereto as Exhibit C.

23 11. E Ink markets and sells one or more models of electrophoretic displays
24 (collectively, “EPD displays”), including without limitation, the Pearl, SURF, and Vizplex
25 models.

FIRST CAUSE OF ACTION
(Infringement Of The '935 Patent)

1
2 12. Plaintiff refers to and incorporates by reference paragraphs 1 through 10, above, as
3 though fully set forth herein.

4 13. E Ink directly or indirectly infringes, in violation of 35 U.S.C. § 271, one or more
5 claims of the '935 patent, literally and/or under the doctrine of equivalents, by selling, offering
6 for sale, and/or using EPD displays in the United States and/or importing EPD displays into the
7 United States. E Ink has infringed, actively induced infringement of, and/or contributorily
8 infringed the '935 patent by using, causing to be used, making, causing to be made, importing,
9 causing to be imported, offering to sell, causing to be offered for sale, selling, and causing to be
10 sold products that are made by a method that infringes a claim of the '935 patent in this judicial
11 district and elsewhere in the United States.

12 14. As a result of E Ink's infringement of the '935 patent, Plaintiff has suffered
13 damages, and is entitled to recover an amount subject to proof at trial, together with interest and
14 costs as fixed by this Court pursuant to 35 U.S.C. § 284.

15 15. On information and belief, E Ink will continue to infringe the '935 patent unless
16 enjoined by this Court.

17 16. E Ink's infringement of Plaintiff's exclusive rights under the '935 patent will
18 continue to damage Plaintiff, causing irreparable harm for which there is no adequate remedy at
19 law, unless enjoined by this Court. Considering the balance of hardships between CopyTele and
20 E Ink, a remedy in equity is warranted. The public interest would not be disserved by a
21 permanent injunction.

22 17. E Ink is infringing the '935 Patent with knowledge of CopyTele's patent rights and
23 without a reasonable basis for believing its conduct is lawful. E Ink's infringement is willful and
24 deliberate, and will continue unless enjoined by this Court, making this an exceptional case and
25 entitling CopyTele to increased damages and reasonable attorneys' fees pursuant to 35 U.S.C. §§
26 284 and 285.

SECOND CAUSE OF ACTION
(Infringement Of The '810 Patent)

1
2 18. Plaintiff refers to and incorporates by reference paragraphs 1 through 10, above, as
3 though fully set forth herein.

4 19. E Ink directly or indirectly infringes, in violation of 35 U.S.C. § 271, one or more
5 claims of the '810 patent, literally and/or under the doctrine of equivalents, by selling, offering
6 for sale, and/or using EPD displays in the United States and/or importing EPD displays into the
7 United States. E Ink has infringed, actively induced infringement of, and/or contributorily
8 infringed the '810 patent by using, causing to be used, making, causing to be made, importing,
9 causing to be imported, offering to sell, causing to be offered for sale, selling, and causing to be
10 sold products that are made by a method that infringes a claim of the '810 patent in this judicial
11 district and elsewhere in the United States.

12 20. As a result of E Ink's infringement of the '810 patent, Plaintiff has suffered
13 damages, and is entitled to recover an amount subject to proof at trial, together with interest and
14 costs as fixed by this Court pursuant to 35 U.S.C. § 284.

15 21. On information and belief, E Ink will continue to infringe the '810 patent unless
16 enjoined by this Court.

17 22. E Ink's infringement of Plaintiff's exclusive rights under the '810 patent will
18 continue to damage Plaintiff, causing irreparable harm for which there is no adequate remedy at
19 law, unless enjoined by this Court. Considering the balance of hardships between CopyTele and
20 E Ink, a remedy in equity is warranted. The public interest would not be disserved by a
21 permanent injunction.

22 23. E Ink is infringing the '810 Patent with knowledge of CopyTele's patent rights and
23 without a reasonable basis for believing its conduct is lawful. E Ink's infringement is willful and
24 deliberate, and will continue unless enjoined by this Court, making this an exceptional case and
25 entitling CopyTele to increased damages and reasonable attorneys' fees pursuant to 35 U.S.C. §§
26 284 and 285.

THIRD CAUSE OF ACTION
(Infringement Of The '488 Patent)

1
2 24. Plaintiff refers to and incorporates by reference paragraphs 1 through 10, above, as
3 though fully set forth herein.

4 25. E Ink directly or indirectly infringes, in violation of 35 U.S.C. § 271, one or more
5 claims of the '488 patent, literally and/or under the doctrine of equivalents, by selling, offering
6 for sale, and/or using EPD displays in the United States and/or importing EPD displays into the
7 United States. E Ink has infringed, actively induced infringement of, and/or contributorily
8 infringed the '488 patent by using, causing to be used, making, causing to be made, importing,
9 causing to be imported, offering to sell, causing to be offered for sale, selling, and causing to be
10 sold products that are made by a method that infringes a claim of the '488 patent in this judicial
11 district and elsewhere in the United States.

12 26. As a result of E Ink's infringement of the '488 patent, Plaintiff has suffered
13 damages, and is entitled to recover an amount subject to proof at trial, together with interest and
14 costs as fixed by this Court pursuant to 35 U.S.C. § 284.

15 27. On information and belief, E Ink will continue to infringe the '488 patent unless
16 enjoined by this Court.

17 28. E Ink's infringement of Plaintiff's exclusive rights under the '488 patent will
18 continue to damage Plaintiff, causing irreparable harm for which there is no adequate remedy at
19 law, unless enjoined by this Court. Considering the balance of hardships between CopyTele and
20 a. E Ink, a remedy in equity is warranted. The public interest would not be disserved by a
21 permanent injunction.

22 29. E Ink is infringing the '488 Patent with knowledge of CopyTele's patent rights and
23 without a reasonable basis for believing its conduct is lawful. E Ink's infringement is willful and
24 deliberate, and will continue unless enjoined by this Court, making this an exceptional case and
25 entitling CopyTele to increased damages and reasonable attorneys' fees pursuant to 35 U.S.C.
26 §§ 284 and 285.

PRAYER FOR RELIEF

1
2 Plaintiff respectfully requests that the Court rule in its favor and against Defendants, and
3 that the Court grant Plaintiff the following relief:

- 4 a. An adjudication that E Ink infringes one or more claims of the '935, '810, and
5 '488 patents, literally and/or under the doctrine of equivalents;
- 6 b. An adjudication that E Ink's infringement of the '935, '810, and '488 patents has
7 been willful;
- 8 c. Damages according to proof;
- 9 d. Costs;
- 10 e. Pre-judgment and post-judgment interest;
- 11 f. If one or more of E Ink's acts of infringement are found to be willful from the time
12 that E Ink became aware of the infringing nature of its actions, which is, at the
13 latest, the time of the filing of Plaintiff's Complaint for Patent Infringement, an
14 award of treble damages for the period of such infringement pursuant to 35 U.S.C.
15 § 284;
- 16 g. A permanent injunction pursuant to 35 U.S.C. § 283, enjoining E Ink from further
17 acts of infringement;
- 18 h. A determination that this case is exceptional and an award to Plaintiff of its
19 reasonable attorneys' fees and costs in accordance with 35 U.S.C. § 285; and
- 20 i. Such other and further relief as the Court deems just and proper.
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JURY DEMAND

Pursuant to Federal Rule of Civil Procedure 38, Plaintiff hereby requests a jury trial.

Dated: January 28, 2013

LIEFF CABRASER HEIMANN & BERNSTEIN, LLP

By: Eric B. Fastiff
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Attorneys for Plaintiff CopyTele, Inc.





US005964935A

United States Patent [19]

Chen et al.

[11] Patent Number: 5,964,935

[45] Date of Patent: Oct. 12, 1999

[54] **INITIATOR-TREATED PIGMENT PARTICLES AND METHOD FOR PREPARING SAME**

[75] Inventors: **Jing Hong Chen, Bethlehem, Pa.; Frederic E. Schubert, Shoreham, N.Y.**

[73] Assignee: **Copytele, Inc., Huntington Station, N.Y.**

4,655,897	4/1987	DiSanto et al.	359/296
4,732,830	3/1988	DiSanto et al.	430/20
5,190,586	3/1993	Mizuguchi et al.	106/499
5,281,561	1/1994	Lin	106/31.65
5,380,362	1/1995	Schubert	106/493
5,403,518	4/1995	Schubert	252/572
5,411,656	5/1995	Schubert	345/107

[21] Appl. No.: **08/916,895**

[22] Filed: **Aug. 22, 1997**

[51] Int. Cl.⁶ **C09C 1/62**

[52] U.S. Cl. **106/401; 106/400; 106/403; 106/413; 106/417; 106/429; 106/445; 106/447; 106/460; 106/471; 106/476; 106/487; 106/491; 106/493; 106/499; 428/403; 428/407**

[58] Field of Search **106/400, 401, 106/403, 413, 417, 429, 445, 447, 460, 471, 476, 487, 491, 493, 499; 428/403, 407**

OTHER PUBLICATIONS

Murau, et al., "The Understanding and Elimination of Some Suspension Instabilities in an Electrophoretic Display", *Jour. of App. Phy.* vol. 49, No. 9., Sep. 1978.

Primary Examiner—Mark L. Bell
Assistant Examiner—Scott L. Hertzog
Attorney, Agent, or Firm—Arthur L. Plevy

[57] ABSTRACT

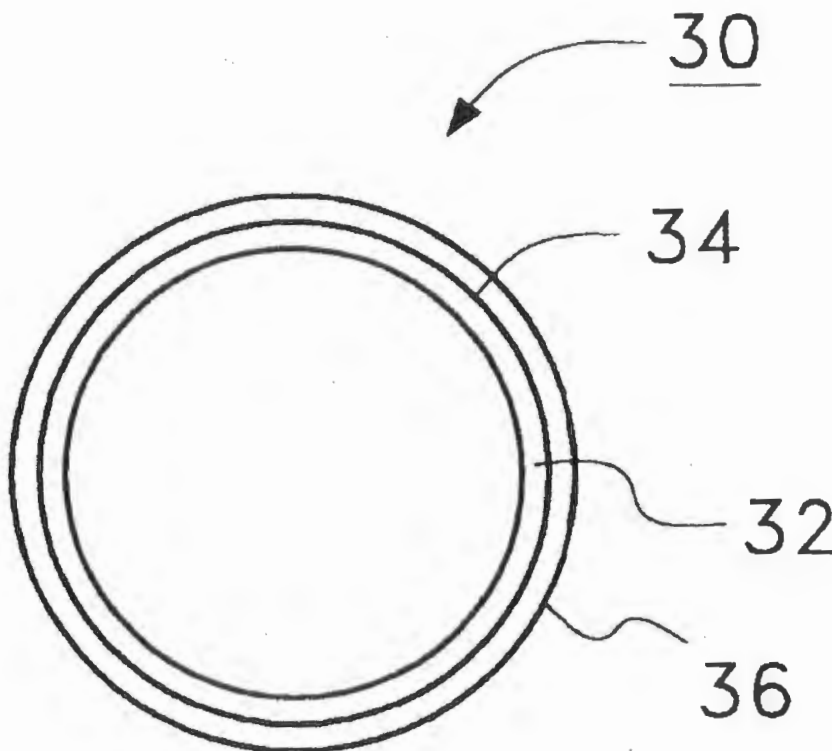
Initiator-treated pigment particles for use in making polymer coated pigment particles, are made up of a plurality of pigment particles. Each of the pigment particles has an outer surface that is covered with a coating of initiator material. The initiator material operates as a nucleus for initiating polymerization on the surface of the particle. The initiator-treated pigment particles are made by providing a plurality of pigment particles and applying a coating of initiator material to the outer surface of each pigment particle.

[56] **References Cited**

U.S. PATENT DOCUMENTS

4,093,534	6/1978	Carter et al.	359/296
4,285,801	8/1981	Chiang	252/62.53
4,298,448	11/1981	Muller et al.	359/296

20 Claims, 1 Drawing Sheet



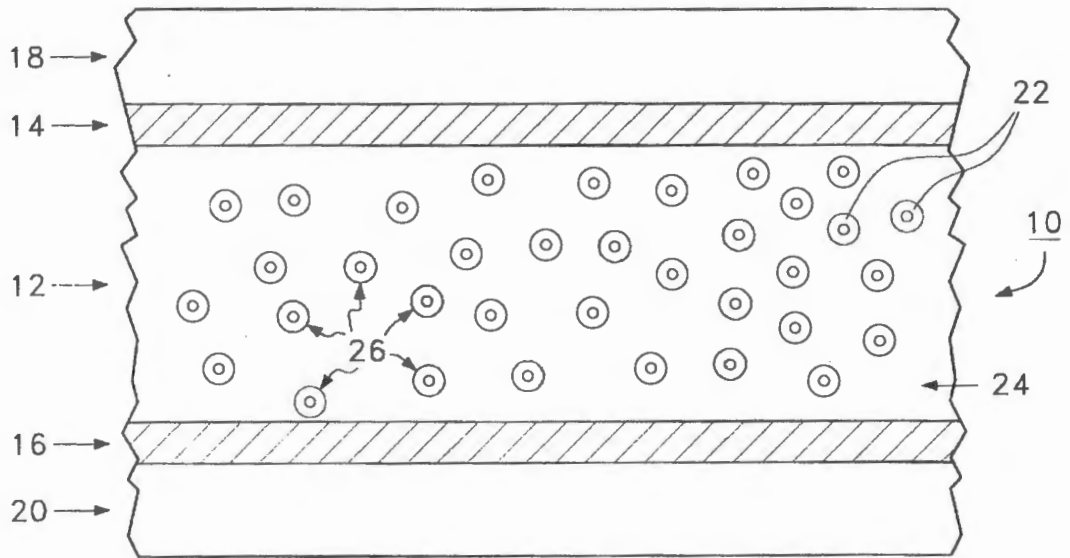


Fig. 1 (Prior Art)

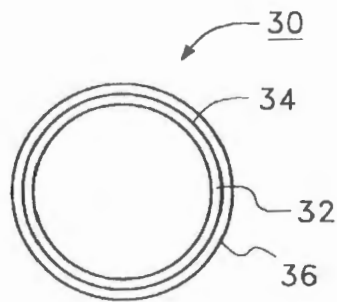


Fig. 2

INITIATOR-TREATED PIGMENT PARTICLES AND METHOD FOR PREPARING SAME

FIELD OF THE INVENTION

The present invention relates generally to coating of pigment particles with polymers and more particularly, to pigment particles pretreated with polymerization initiators and method for preparing same.

BACKGROUND OF THE INVENTION

The electrophoretic effect is well known in the art as evidenced by the many patents and articles which describe the effect. As one of ordinary skill in the art will recognize, the electrophoretic effect operates on the principle that certain particles suspended in a medium can be electrically charged and thereby caused to migrate through the medium to an electrode of opposite charge. Electrostatic printing and electrophoretic image displays (EPID) use the electrophoretic effect to produce desired images. For an example of devices using the electrophoretic effect, reference is made to U.S. Pat. No. 4,732,830, issued to Frank J. DiSanto et al., on Mar. 22, 1988, entitled ELECTROPHORETIC DISPLAY PANELS AND ASSOCIATED METHODS, and assigned to Cotypele, Inc., the assignee herein.

In typical EPIDs, dielectric particles are suspended in a fluid medium that is either clear or of an optically-contrasting color as compared with the dielectric particles. To effect the greatest optical contrast between the particles and the suspension medium, it is desirable to have either light-colored particles suspended in a dark medium or dark-colored particles, preferably black, suspended in a backlighted clear medium. A variety of pigment particle and dispersion medium compositions are known in the art. See, for example, U.S. Pat. No. 4,298,444, issued to K. Muller, et al., on Nov. 3, 1981, entitled ELECTROPHORETIC DISPLAY.

It has been found difficult to produce black and other very dark colored electrophoretic particles that are dielectric, of uniform size, and have a density matching that of a common suspension medium. As a result, EPIDs commonly use readily-manufactured light colored electrophoretic pigment particles suspended in media which contain dark color dyes. Such EPIDs are exemplified in U.S. Pat. Nos. 4,655,897 to DiSanto et al.; 4,093,534 to Carter et al.; 4,298,448 to Muller et al.; and 4,285,801 to Chiang. The use of a yellow pigment particle is disclosed in the following patents, all of which issued to Frederic E. Schubert and are assigned to Cotypele, Inc., the assignee herein: U.S. Pat. No. 5,380,362, issued Jan. 10, 1995, entitled SUSPENSION FOR USE IN ELECTROPHORETIC IMAGE DISPLAY SYSTEMS; U.S. Pat. No. 5,403,518, issued Apr. 4, 1995, entitled FORMULATIONS FOR IMPROVED ELECTROPHORETIC DISPLAY SUSPENSIONS AND RELATED METHODS; and U.S. Pat. No. 5,411,656, issued May 2, 1995, entitled GAS ABSORPTION ADDITIVES FOR ELECTROPHORETIC SUSPENSIONS. These three patents are incorporated herein by reference.

The selection of the electrophoretic particles used in the EPID is very important in determining performance of the EPID and the quality of the image produced. Ideally, electrophoretic particles should have an optimum charge-to-mass ratio, which is dependent upon the particle size and surface charge; the optimum charge-to-mass ratio is desirable to obtain good electrostatic deposition at high velocity as well as rapid reversal of particle motion when voltages change.

There are advantages to polymerically coating electrophoretic particles for use in the EPID or other electrophoretic devices. A polymer coating applied to the particles can have the effect of enhancing the ability of the particles to scatter light. When polymer-coated particles impinge upon a transparent screen electrode (thereby displacing the fluid medium from the screen), a brighter color and sharper image is produced (as compared with when uncoated particles are used). Additionally, it naturally is desirable to use electrophoretic particles that are stable and resistant to interaction with the suspension medium to improve the efficiency and life-time of the EPID; the suspension medium may comprise, for example, aromatic or aliphatic solvents, including benzenes, toluenes, hydrocarbon oil, nonane, decane, or the like, which may interact over time with some typical uncoated pigment particles used in EPIDs. Polymer-coated pigment particles produce a harder and more solvent-resistant composite when compared to uncoated particles.

Furthermore, polymer-coated electrophoretic particles can be less apt to adhere to surfaces within the electrophoretic cell. Over recent years, attention has been directed to dispersion stabilization by adsorbed polymers on particle surfaces. See, for example, an article by P. Murau and B. Singer, appearing in Vol 49, No. 9, of the Journal of Applied Physics (1978), entitled "The Understanding and Elimination of Some Suspension Instabilities in an Electrophoretic Display." See also U.S. Pat. No. 5,403,518, issued to Schubert, referenced above, and U.S. Pat. No. 4,285,801, issued to Anne A. Chiang on August 25, 1981, entitled ELECTROPHORETIC DISPLAY COMPOSITION.

It has been discovered that fluoro-polymers are advantageous for use as pigment particle coatings, as they have low critical surface tensions which tend to produce anti-stick properties. See, for example, an article by M. Hudlicry and A. E. Pavlath, appearing in Vol. 187, page 983, of ACS Monograph (1995), entitled "Properties of Fluorinated Compounds II." The lower surface tensions of perfluorinated polymers, when used as a pigment particle coatings, leads to less interaction between the pigment particle surfaces and the solvents comprising the suspension medium in which the particles are dispersed. The stabilizing polymer molecule hydrocarbon chains are more apt to extend out into the solvent and not sit on the particle surface because of its anti-stick character. This enhances the efficiency of the stabilizing mechanism.

There are, however, manufacturing problems associated with coating pigment particles. These problems involve the inability to coat all the pigment particles in a batch with a polymer and the formation of a percentage of polymer particles that have no pigment core.

Accordingly, a need exists for a method which enables all the pigment particles in a batch to be polymer coated while preventing the formation of polymer particles having no pigment core.

SUMMARY OF THE INVENTION

Initiator-treated pigment particles which are for use in manufacturing polymer-coated pigment particles. The initiator-treated pigment particles comprise a plurality of pigment particles each having an outer surface which is covered with a coating of initiator material that operates as a nucleus for initiating polymerization on the surface of the particle.

The method used to make initiator-treated pigment particles described above comprise the steps of providing a plurality of pigment particles and applying a coating of initiator material to the outer surface of each pigment particle.

BRIEF DESCRIPTION OF THE DRAWING

For a detailed understanding of the present invention, reference should be made to the following detailed description taken in conjunction with the accompanying drawings wherein:

FIG. 1 is a diagrammatic, cross-sectional view of an EPID having particles in accordance with the present invention and

FIG. 2 is cross-sectional view through an initiator-treated particle of the present invention.

DETAILED DESCRIPTION OF THE INVENTION

As will become apparent, the initiator-treated pigment particles of the present invention are especially useful for making electrophoretic particles used in electrophoretic image displays (EPIDs). Accordingly, the initiator-treated pigment particles of the present invention will be described in connection with their use in making electrophoretic particles. It should be understood, however, that the initiator-treated pigment particles are also useful in many different applications where dispersions containing charged particles are desired, such as paint, ink, and electrostatic toner.

The initiator treated particles of this invention have the advantage of being at least as stable as or more stable than the pure initiator material. This is possible because the individual initiator molecules are distributed in a thin surface layer on the pigment particle as will be explained further on. While individual molecules can still undergo deactivation reactions on the surface of the particle, reactions will not propagate through the sample as they can in the bulk material. Providing pigment particles that are all polymer-coated will enhance the operation of an EPID since, all the pigment particles in the EPID will have the enhanced ability to scatter light to produce a brighter color; are harder, more solvent-resistant composites; and are adhesion-resistant with regard to surfaces within the electrophoretic cell.

To fully appreciate the initiator-coated pigment particles of the present invention, a brief description of an EPID is provided as follows with reference to FIG. 1 which depicts a cross-sectional view through a segment of a simple EPID 10 enlarged to show a single pixel. As will be recognized by a person skilled in the art, an EPID comprises an anode 14 and a cathode 16 with a volume of an electrophoretic dispersion 12 disposed therebetween. The anode 14 and the cathode 16 are deposited upon glass plates 18, 20 in the form of a thin layer of indium-tin-oxide (ITO) or a like compound. The ITO layer is deposited in such a manner as to be substantially transparent when viewed through the glass plates 18, 20.

The electrophoretic dispersion 12 is comprised of a plurality of polymer-coated dielectric electrophoretic particles 22 suspended in a fluid medium 24. When a sufficient electrical bias is applied between the anode 14 and the cathode 16, the particles 22 migrate to either the cathode 16 or anode 14, depending on polarity, and they displace the dark color medium adjacent the ITO layer, thereby creating a bright pixel. Reversing the voltage produces a dark pixel.

Referring now to FIG. 2, a cross-sectional view through an initiator-treated particle 30 is shown. The particle 30 comprises a core pigment particle 32 having an outer surface 34 which is coated by a layer 36 of initiator material. The outer surface 34 coated with the initiator material 36, serves as a nucleus for polymerization to initiate on the outer

surface 34. Accordingly, the initiator-treated pigment particles of the present invention are produced prior to polymerization of the pigment particles.

The initiator-treated particles are prepared according to the present invention by the following process. First, pigment particles are selected, cleaned and dried using conventional techniques. The pigment particles are then dispersed in a solvent containing a dispersing agent and an initiator material. The dispersing agent disperses and thus, separates and suspends the particles in the solvent so that all the particles can be coated with the initiator. This produces a mixture of pigment, solvent, and initiator.

Next, the mixture is sonicated for approximately 20 minutes using a Sonicor Model 1150 Ultrasonic Probe or like device. After sonication, the mixture is placed in an evaporator and the solvent is evaporated at room temperature. Once the solvent has evaporated, all that remains are the initiator-treated particles of the present invention. This process typically yields greater than 95% pigment particles which are initiator-treated.

Table I below provides an exemplary formulation for making the initiator-treated particles of the present invention:

TABLE I

Materials	Weight (grams)
Pigment	1.2-1.4
Initiator	0.02-0.10
Wetting Agent (for pigment)	3.0-5.0
Solvent	30.0-35.0

Pigment particles which can be treated in accordance with the present invention comprise light and dark colored, inorganic pigments, organic pigments, and various particles produced from polymers, dyes and the like. Examples of light and dark colored inorganic and organic pigments include titanium dioxide, zinc oxide, zinc sulfide, silica, calcium silicate, alumina hydrate, carbon black, iron oxide black, lamp black, Zn Fe Cr brown Spinel, Magnesium Ferrite, Chreen Spinel, Cr oxide Green, Diarylide Yellow, Arylide Yellow, Hansa Yellow, Benzidine Yellow, Perinone Orange, Diarylide Orange, Ultramarine Blue, Indanthrone Blue, Ultramarine Blue Dioxazine Violet, Quinacridone Violet, Anthraquinoid Red, and Perylene Red and the like.

The polymerization initiator used in the present invention is selected from azo-initiators, photo-initiators, organic peroxides and the like. A preferred polymerization initiator is 4,4'-Azobis (4-cyanovaleric acid). Other polymerization initiators include 2,2"-azobis (2-methyl-butyroneitrile), benzoyl peroxide, lauroyl peroxide, octanoyl peroxide, and the like.

The solvent used in the process of the present invention is selected from various aromatic hydrocarbons, aliphatic hydrocarbons, dioxane and dichloromethane. The aromatic hydrocarbon solvents preferably include benzene, toluene, xylene, or tertiary butyl benzene. The aliphatic hydrocarbon solvents preferably include hexane or decane.

The dispersing agent used in the process of the present invention is selected from alkyl hydrocarbons or commercial dispersing agents. The preferred dispersing agents are alkyl hydrocarbons with functional groups —OH and —NH₂.

The initiator-treated particles of the present invention are useful in coating pigment particles with one or more solid coatings of polypentafluorostyrene, polystyrene-divinyl benzene and the like. Moreover, the initiator-treated particles of the present invention can be used to produce porous polymer-coated pigment particles.

It will be understood that the embodiments described herein are merely exemplary and that a person skilled in the art may make many variations and modifications to the described embodiments utilizing functionally equivalent elements to those described. Any variations or modifications to the invention described hereinabove are intended to be included within the scope of the invention as defined by the appended claims.

What is claimed is:

1. Initiator-treated pigment particles for use in manufacturing polymer-coated pigment particles, comprising:

a plurality of pigment particles each having an outer surface which is covered with a coating of initiator material that operates as a nucleus for initiating polymerization on said surface of said particle.

2. The initiator-treated pigment particles of claim 1, wherein said plurality of pigment particles are selected from the group consisting of inorganic pigment particles, organic pigment particles, polymer pigment particles, and dye pigment particles.

3. The initiator-treated pigment particles of claim 1, wherein said coating initiator-material is selected from the group consisting of azo-initiators, photo-initiators, and organic peroxides.

4. The initiator-treated pigment particles of claim 1, wherein said polymer-coated pigment particles comprise dielectric electrophoretic particles.

5. A method of making initiator-treated pigment particles for use in manufacturing polymer-coated pigment particles, said method comprising the steps of:

providing a plurality of pigment particles, each of said particles having an outer surface and

applying a coating of initiator material to said outer surface of each of said plurality of pigment particles, said coating of initiator material operating as a nucleus for initiating polymerization on said surface of said particle.

6. The method of making initiator-treated pigment particles according to claim 5, wherein said initiator-material is selected from the group consisting of azo-initiators, photo-initiators, and organic peroxides.

7. The method of making initiator-treated pigment particles according to claim 5, wherein said plurality of pigment particles are selected from the group consisting of inorganic pigment particles, organic pigment particles, polymer pigment particles, and dye pigment particles.

8. The method of making initiator-treated pigment particles according to claim 5, wherein said step of applying includes the step of dispersing said plurality of pigment particles in a solvent to form a mixture.

9. The method of making initiator-treated pigment particles according to claim 8, wherein said solvent is selected from the group consisting of aromatic hydrocarbons, aliphatic hydrocarbons, dioxane and dichloromethane.

10. The method of making initiator-treated pigment particles according to claim 8, wherein said solvent includes a dispersing agent for dispersing said plurality of pigment particles in said solvent.

11. The method of making initiator-treated pigment particles according to claim 10, wherein said dispersing agent comprises an alkyl hydrocarbon.

12. The method of making initiator-treated pigment particles according to claim 11, wherein said alkyl hydrocarbon has a functional group selected from the group consisting of —OH functional groups and —NH₂ functional groups.

13. The method of making initiator-treated pigment particles according to claim 8, wherein said solvent includes said initiator-material.

14. The method of making initiator-treated pigment particles according to claim 8, wherein said step of applying further includes the step of sonicating said mixture.

15. The method of making initiator-treated pigment particles according to claim 14, wherein said step of applying further includes the step of evaporating said solvent from said mixture after said step of sonicating.

16. The method of making initiator-treated pigment particles according to claim 8, wherein said step of applying further includes the step of evaporating said solvent from said mixture.

17. A formulation for initiator-treated pigment particles that are used for manufacturing polymer-coated pigment particles, comprising:

a solvent;

a wetting agent for pigment;

a polymerization initiator dissolved in said solvent; and

a plurality of pigment particles dispersed in said solvent with the assistance of said wetting agent.

18. The formulation of claim 17, wherein said solvent is selected from the group consisting of aromatic hydrocarbons, aliphatic hydrocarbons, dioxane and dichloromethane.

19. The formulation of claim 17, wherein said initiator-material is selected from the group consisting of azo-initiators, photo-initiators, and organic peroxides.

20. The formulation of claim 17, wherein said plurality of pigment particles are selected from the group consisting of inorganic pigment particles, organic pigment particles, polymer pigment particles, and dye pigment particles.

* * * * *





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United States Patent [19]

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Hou et al.

[45] Date of Patent: **Sep. 5, 2000**

[54] **METHODS OF PREPARING ELECTROPHORETIC DISPERSIONS CONTAINING TWO TYPES OF PARTICLES WITH DIFFERENT COLORS AND OPPOSITE CHARGES**

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Colloids and Surfaces, 6 (1983) p. 101, Luckham et al., no month available.
Journal of Chemical Society, Farada 1, 76 (1980), p., 655 no month available.

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[21] Appl. No.: **08/383,667**

[22] Filed: **Feb. 3, 1995**

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Related U.S. Application Data

[63] Continuation of application No. 08/065,871, May 21, 1993, abandoned.

[51] Int. Cl.⁷ **H01B 3/00**

[52] U.S. Cl. **252/572; 313/483; 359/296**

[58] Field of Search **252/572, 73, 77, 252/79; 313/483; 204/299 R; 359/296**

[57] ABSTRACT

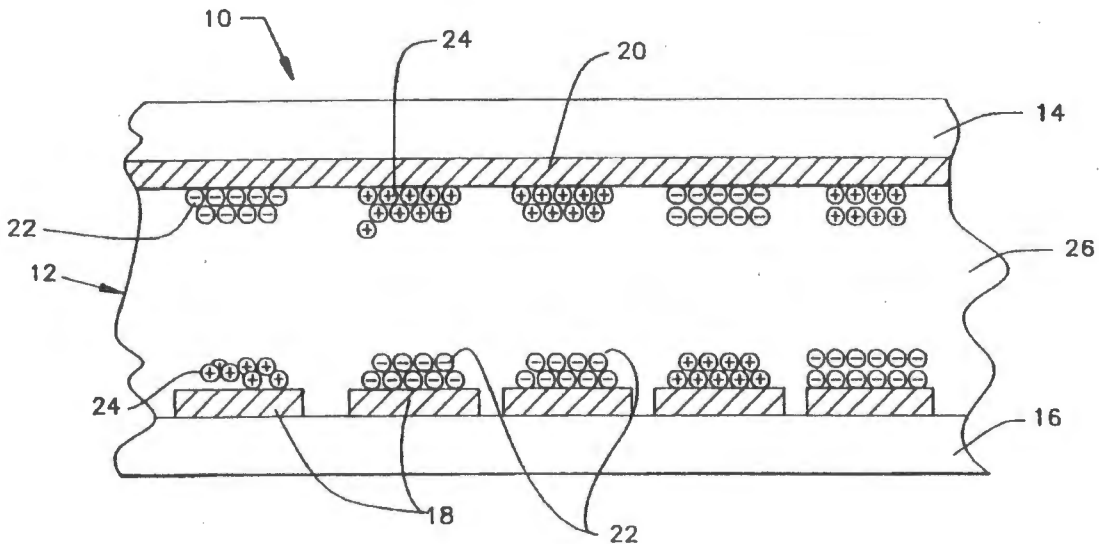
A dielectric dispersion for use in an electrophoretic display includes a dielectric fluid, a first plurality of particles of a first color having a surface charge of a selected polarity dispersed within the dielectric fluid and a second plurality of particles of a second color having a surface charge of opposite polarity to that of the first plurality and a steric repulsion thereto preventing coagulation of the first and second pluralities. In one embodiment, the first and second plurality of particles are each formed by separate two stage dispersion polymerization reactions. Each set of particles is formed with unique secondary and functional monomers. Corresponding charge control agents are added to the dispersion to establish opposite polarities on the respective particles.

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18 Claims, 1 Drawing Sheet



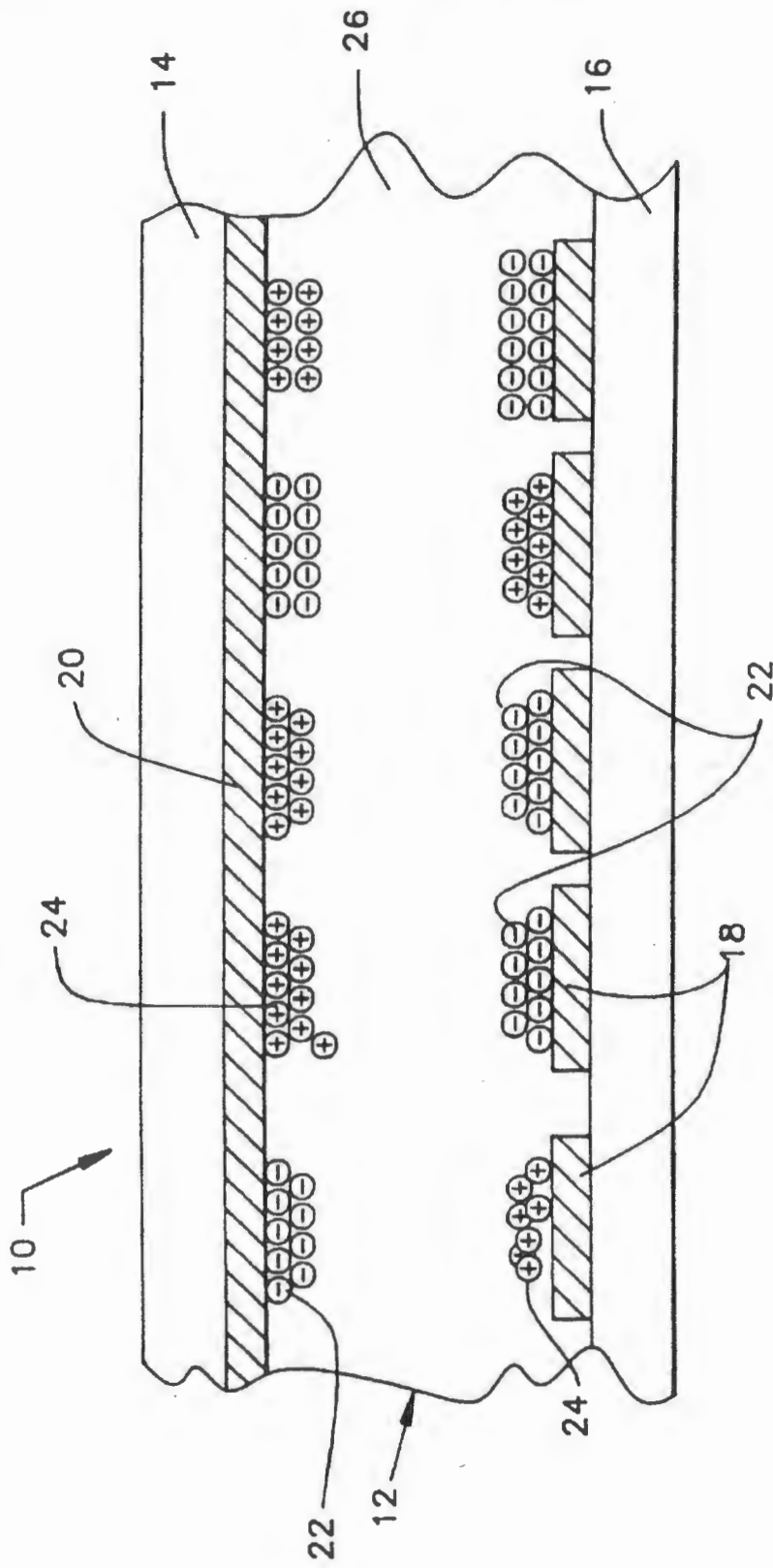


FIG. 1

**METHODS OF PREPARING
ELECTROPHORETIC DISPERSIONS
CONTAINING TWO TYPES OF PARTICLES
WITH DIFFERENT COLORS AND OPPOSITE
CHARGES**

This is a continuation of application Ser. No. 08/065,871, filed on May. 21, 1993, now abandoned entitled Methods of Preparing Electrophoretic Dispersions Containing Two Types of Particles with Different Colors and Opposite Charges.

FIELD OF THE INVENTION

The present invention relates to dielectric particles and electrophoretic dispersions containing them for use in electrophoretic image displays, electrostatic printing or the like, and a corresponding method of producing same. More particularly, the present invention relates to the formation of stable electrophoretic dispersions which contain two types of particles having different colors and opposite charges.

BACKGROUND OF THE INVENTION

The electrophoretic effect is well known and the prior art is replete with a number of patents and articles which describe the effect. As will be recognized by a person skilled in the art, the electrophoretic effect operates on the principle that certain particles, when suspended in a medium, can be electrically charged and thereby to migrate through the medium to an electrode of opposite charge. Electrostatic printing and electrophoretic image displays (EPID) utilize the electrophoretic effect to produce desired images.

In prior art EPIDs, colored dielectric particles are suspended in a fluid medium of an optically contrasting color as compared to the dielectric particles. The colored electrophoretic particles are then selectively caused to migrate to, and impinge upon, a transparent screen, thereby displacing the fluid medium from the screen surface and creating the desired image. EPIDs commonly use readily manufactured light colored electrophoretic particles suspended in media which contains dark color dyes. Such EPIDs are exemplified in U.S. Pat. Nos: 4,655,897 to DiSanto et al., 4,093,534 to Carter et al., 4,298,448 to Muller et al., and 4,285,801 to Chaing. The dark medium can not be completely displaced by the suspended particles because there is always some dark liquid left on the screen or around the particle surface. The resulting images therefore suffer some loss of contrast.

An alternative way of creating contrasting images is to have a dispersion that contains both black and white dielectric particles having opposite charges. When an electric field is applied, the black and white particles are caused to migrate in opposite directions due to their opposite polarity. This creates the desired image with black and white contrast. Since the black and white dielectric particles have opposite charges, they will completely separate and migrate in opposite directions under the applied electric field, without leaving any black particles on the white image or any white particles on the black image. This produces highly contrasted black and white images. Ideally, the black and white dielectric particles must have optimum electrophoretic mobilities and be thermodynamically stable in the same medium in order to have reasonable imaging speed and to prevent flocculation due to the electrostatic attraction between the oppositely-charged particles.

In nature, as well as in industry, aggregation of oppositely-charged particles in dispersion has been shown in a large variety of fields such as soil conditioning, water

purification, filtration and adhesion or aggregation of biological cells. Good examples are given in Colloids and Surface, 6, 83(1983); 6, 101(1983) and in Journal of Chemical Society Faraday, 76, 665(1980), in which the particles adsorption isotherms of small positively-charged polystyrene particles onto large negatively-charged polystyrene particles have been reported. In the prior art, it has proven difficult to produce an electrophoretic dispersion containing oppositely-charged particles without flocculation, particularly in non-aqueous systems. Theoretically, with the addition of a proper polymeric stabilizer, it is possible to stabilize two oppositely-charged particles in the same medium if the steric repulsion provided from the absorbed polymer layer can overcome the electrostatic attraction between the two oppositely-charged particles. However, very few surfactants have been found to be able to produce enough steric repulsion to stabilize oppositely-charged particles without trading off their electrophoretic mobility. Accordingly, there is no known electrophoretic image display, which gives a highly contrasted black and white image.

SUMMARY OF THE INVENTION

The problems and disadvantages associated with conventional dielectric dispersions are overcome by the present invention which includes a dielectric fluid, a first plurality of particles of a first color having a surface charge of a selected polarity dispersed within the dielectric fluid and a second plurality of particles of a second color having a surface charge of opposite polarity to that of the first plurality and a steric repulsion thereto preventing coagulation of the first and second pluralities. A method in accordance with the present invention for making a dielectric dispersion includes providing a dielectric fluid; preparing a first plurality of particles of a first color and having a surface charge of a selected polarity; preparing a second plurality of particles of a second color having a surface charge of opposite polarity to that of the first plurality and a steric repulsion thereto preventing coagulation of said first and second pluralities; and dispersing the first plurality of particles and second plurality of particles in the dielectric fluid. A charge control agent is added to the dielectric fluid.

BRIEF DESCRIPTION OF THE FIGURES

FIG. 1, the sole figure, is a diagrammatic, cross-sectional view of an EPID having particles in accordance with a first embodiment of the present invention.

**DETAILED DESCRIPTION OF THE
INVENTION**

In the present invention, electrophoretic dispersions containing two types of particles having different colors e.g., black and white, and opposite charges, which are thermodynamically stable, are produced by properly selecting certain surfactants which selectively charge the different types of particles positively and negatively, and also provide enough steric repulsion to prevent the flocculation of the oppositely-charged particles without trading off their electrophoretic mobilities.

Products in accordance with this invention can also be used in electrostatic printing for color images. In the prior art, conventional electrostatic printing produces color images by a multi-step printing process in which each step of printing only forms one color. Combinations of different color images from each single printing process, thereby, produce a desired color image. Since the formation of each color image needs to go through every single printing cycle,

it usually takes a long time to produce a multi-colored image. In this invention, electrophoretic dispersions containing two different color particles with opposite charges are capable of producing two color images in one printing cycle resulting in energy and time saved.

Although the present invention's electrophoretic dispersions can be used in many different applications where dispersions containing different color particles with opposite charges are desired, such as paint, ink and electrostatic toner, it is especially suitable for use in connection with electrophoretic image displays (EPIDs). Accordingly, the present invention will be described in connection with a typical EPID.

Referring to FIG. 1, there is shown a cross sectional view of a segment of a simple electrophoretic image display 10 magnified to show a single pixel. As will be recognized by a person skilled in the art, an EPID may contain a volume of an electrophoretic dispersion 12 disposed between an anode 14 and a cathode 16. The anode 14 and cathode 16 are deposited upon glass plates 18, 20 in the form of a thin layer of indium-tin-oxide (ITO) or a like compound. The ITO layer is deposited in such a manner as to be substantially transparent when viewed through the glass plates 20, 18.

In accordance with an embodiment of the present invention, the electrophoretic dispersion 12 is comprised of white dielectric electrophoretic particles 22 suspended in a dark color medium 24. The electrophoretic particles 22 have a density substantially equivalent to that of the fluid medium 24 so as to remain randomly disperse.

Referring again to FIG. 1, in the embodiment shown, the electrophoretic dispersion 12 is comprised of black dielectric particles 22 and white dielectric particles 24 suspended in a clear medium 26. When a selected electrical bias is applied between a cathode line 14 and an anode line 16, the black dielectric particles 22 and white dielectric particles 24 separate and migrate in opposite directions to the cathode line 14 and anode line 16 due to the opposite polarity of the black and white particles, thereby creating an image with optical contrast between the black electrophoretic particles 22 and the white electrophoretic particles 24. The vertical dashed arrows 28 show the direction of motion.

As indicated above, the production of an electrophoretic dispersion containing two types of particles with different colors and opposite charges is highly desirable to produce black and white contrasting images. However, a major problem associated therewith is creating a thermodynamically stable electrophoretic dispersion with oppositely-charged particles. The present invention white electrophoretic particles 22 and black electrophoretic particles 24 may be formed from crosslinked polymer particles using a two stage dispersion polymerization technique with and without staining with a metal oxide, respectively. Since the surface functionalities of the electrophoretic particles can be well controlled during the polymerization, the black and white particles can be made with acidic and basic (or basic and acidic) surface characteristics, respectively and can be charged negatively and positively (or positively and negatively). In general, the polymeric materials are dielectric and have densities close to 1 g/cm³, therefore the final electrophoretic particles are non-conductive and can easily match the specific gravity of many suspension fluids. In addition, the electrophoretic particles are highly crosslinked polymer particles, which have excellent heat and solvent resistance. As a result, they can be used in a wide range of organic solvents and at high temperatures during cell sampling. In addition to the synthetic black and white polymeric particles, other light color inorganic or organic pigments such as titanium dioxide, zinc oxide, silica, Dairylide Yellow, perinone orange, Ultramarine Blue and the like, can also be used to form contrast images with the black synthetic polymer particles.

To form electrophoretic dispersions containing two types of particles with different colors and opposite charges, surfactants are selected to charge the two different types of particles positively and negatively and to provide a strong steric repulsion between the two oppositely-charged particles to form a thermodynamically stable dispersion. As will be recognized by a person skilled in the art, surface charging for a non-aqueous dispersion is dependent upon the surface functionality of the particles and the charge control agent selected. The surface functionality of particles can be controlled during the formation of particles or be modified after the formation of particles by chemical or physical means.

PROCESS ONE

In an exemplary embodiment of the present invention, the electrophoretic dispersion is made to contain both positively-charged black particles and negatively-charged white particles (or negatively-charged black particles and positively-charged white particles) in a dielectric medium. The black and white dielectric particles are each separately prepared by a two-stage dispersion polymerization technique. The polymerization recipes for producing the positively-charged and negatively-charged polymer particles are listed in Table I and Table II, respectively.

TABLE I

Materials	Weight (g)
Methanol	100
divinylbenzene	5
styrene	5
2,2'-azobisisobutyronitrile	0.5
poly(vinyl pyrrolidone)	2
Acrylamide	0.5

TABLE II

Materials	Weight (g)
Methanol	100
divinylbenzene	5
styrene	5
2,2'-azobisisobutyronitrile	0.5
poly(acrylic acid)	2
methacrylic acid	0.5

Although prepared separately, the oppositely charged dielectric particles are formed by essentially the same overall process. Namely, prior to use, the inhibitors of the monomer styrene and the crosslinker divinylbenzene are removed by washing with 10% NaOH aqueous solution several times, drying with calcium carbonate overnight at 0° C. and then passing through a column containing an appropriate inhibitor remover. The inhibitor-free monomers and crosslinker are stored in a freezer for later use. In this, and subsequent processes, the styrene is of the type commercially available from Fisher Scientific, Inc. and the divinylbenzene is of the type commercially produced by Aldrich Chemical Co.

The initiator, 2,2'-azobisisobutyronitrile (AIBN), is made by Kodak Co. and the stabilizers, poly(vinyl pyrrolidone) (PVP) and poly(acrylic acid), are made by GAF Co. and Aldrich Co. respectively. The second stage monomers, acrylamide and methacrylic acid, and the dispersion medium, methanol, are of the types commercially available from Fisher Scientific, Inc.

The styrene and divinylbenzene are mixed with methanol and charged to a closed container containing the initiator and stabilizer which are carefully weighed. The closed container

is purged with nitrogen by bubbling through the solution for a certain time. The container is then warmed and agitated for a desired reaction time. In one preferred embodiment, the mixture is tumbled at thirty revolutions per minute for eight hours at sixty degrees celsius. After eight hours of tumbling, the second stage monomer, either methacrylic acid or acrylamide, is injected into the container which continues to tumble at the same reaction condition for another desired reaction time. The final product made by the two stage dispersion polymerization process is highly crosslinked poly(styrene-co-divinylbenzene) particles with polyacrylamide grafted on the surface in the case of acrylamide as the second stage monomer or, in this case of methacrylic acid, poly(methacrylic acid) is grafted on the surface. The final particles are uniform in size and vary from 0.2 to 2 μm depending upon the reaction media used, i.e. mixtures of methanol and xylene produce larger particles. The particle size being dependent upon the ratio of the mixed solvents. The final particles are dielectric with good whiteness and have densities close to 1 g/cm^3 .

To form dark particles, one or the other of the polymer particles produced from the preceding process are separated from the dispersion medium by centrifuging and decanting the dispersion medium. They are then mixed and tumbled with a two weight percent osmium tetroxide aqueous solution at room temperature for a desired reaction time. The osmium tetroxide reacts with, and stains, the residual double bonds of the poly(styrene-co-divinylbenzene) particles, thereby resulting in highly crosslinked polymer particles having a desired degree of blackness, that can be used as the present invention black dielectric particles 22. It should be understood that in place and stead of the osmium tetroxide, ruthenium tetroxide or other metal oxides may also be used.

By varying the polymerization recipe of Table I and Table II and by varying other reaction parameters of the method of manufacture, the surface characteristics of the white and black particles produced can be selectively altered for particle charging. The surface functionality of the final particles can be varied by introducing different basic functional monomers, such as vinyl acetate, methyl methacrylate, acrylonitrile, N-(isobutoxymethyl) acrylamide, dimethylaminopropylmethacrylamide, and the like, at the second stage polymerization to produce poly(styrene-co-divinylbenzene) particles with basic surface characteristics which are suitable for positive charging in dielectric media. Alternatively, by introducing different acidic functional monomers, such as acrylic acid, sodium styrene sulfonate, maleic acid, chlorostyrene, vinyl alcohol, and the like, at the second stage polymerization, poly(styrene-co-divinylbenzene) particles with acidic surface characteristics which are suitable for developing negatively-charged particles in dielectric media may be produced.

As will be recognized by a person skilled in the art, the other ingredients, such as the stabilizer, initiator, monomer, crosslinker, and the ratio of the monomer and the crosslinker in Table I and Table II can also be varied to effect other properties of the final particles, such as molecular weight and glass transition temperature for different applications.

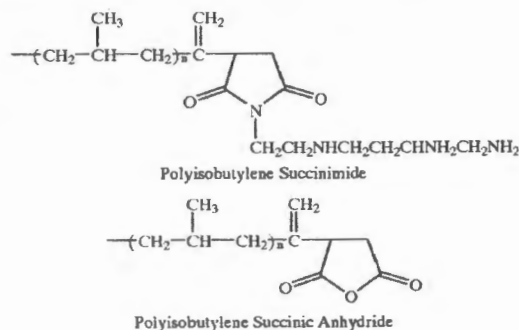
As will be recognized by a person skilled in the art, highly crosslinked polymer particles having surfaces with controlled acidic moieties or basic moieties, made by emulsion polymerization, miniemulsion polymerization, microemulsion polymerization, suspension polymerization, precipitation, seeded emulsion polymerization or seeded dispersion polymerization, could be used as the white or as the black (after being stained with the metal oxide) electrophoretic particles.

After the polymerization process, each of the final polymer particle products are transferred from their respective dispersion mediums to a desired dielectric medium by a

washing process. This involves mixing the final product with a solvent which is mutually soluble in both the dispersion medium and the desired dielectric medium, centrifuging the mixture to separate the particles and the liquids and decanting the supernatant to receive the concentrated particles. The washing process is repeated several times until the dispersion medium (methanol) is completely removed. The solvent washed particles are then redispersed in a dielectric medium with the addition of a desired charge control agent. The particles made from the Recipe I (Table I) exhibiting basic surface functionality, which can be charged positively by a positive charge control agent, can be used either as white positively-charged particles if not stained with a metal oxide, or as black positively-charged particles after being stained with a metal oxide. Similarly, the particles made from Recipe II (Table II) exhibiting acidic surface functionality, which can be charged negatively by a negative charge control agent, can be used as white negatively-charged particles if not stained with a metal oxide or as black negatively-charged particles after being stained with a metal oxide.

The preferred dielectric media for the final dispersion are non-polar solvents such as tetrachloroethylene, carbon tetrachloride, pentane, octane, decane, cyclohexane, benzene, xylene, sec-butylbenzene, Isopars, and the like, or a mixture of these liquids. The solvents used for washing the particles should be mutually soluble in both the dispersion medium and the dielectric medium. Examples are ethanol, propanol, butanol, acetone, tetrahydrofuran, ketones, ethers, esters, and the like.

The preferred surfactants in the present invention are functionalized polymeric (or oligomeric) compounds which not only function as charge control agents for particle charging but also function as steric stabilizers to prevent coagulation. In accordance with the present invention, the molecular weight of the surfactants ranges from a few thousand to hundreds of thousands. Simultaneously using two different surfactants having similar molecular weight and chemical structure but different end groups, is preferred. The surfactants used for negative charging preferably have basic end groups; with the surfactants used for positive charging preferably having acidic end groups. Examples for negative and positive charging are polyisobutylene succinimide and polyisobutylene succinic anhydride, respectively. The chemical structure of these two surfactants are given below:



Alternatively, mixtures of charge control agents and steric stabilizers are also useful in producing a thermodynamically stable dispersion containing positively-charged and negatively-charged particles. The desirable charge control agents for positive charging are surfactants with acidic character such as cupric naphthenate, zirconium octoate, zinc octoate, calcium octoate, polyvinyl alcohol, polyacrylic

acid, polymethacrylic acid, polyvinyl chloride, and the like. The desirable negative charge control agents, however, are surfactants with a basic character such as barium petronate, barium sulfonate, barium dinolynphthalenesulfonate, metal oxides, polyvinyl pyridine, pyridine, lecithin, polyvinyl acetate, polyethylene oxide, polymethyl methacrylate, polyacrylamide, polyesters, polyethers, and the like. The preferred stabilizers or co-stabilizers used with the charge control agents are typically high molecular weight compounds including homopolymers, copolymers, graft polymers, block copolymer, natural high molecular weight compounds, and the like. Specific examples of suitable stabilizers are poly(12-hydroxystearic acid)-graft-poly(methyl methacrylate-methacrylic acid), polystyrene-copoly(vinyl pyridine), poly(vinyl alcohol)-co-polyethylene, polyisobutylene-co-polyacrylic acid, polyisobutylene-copolyamide, and the like.

After preparing the black and white electrophoretic dispersions individually, the positively-charged white dispersion is then mixed with the negatively-charged black dispersion or the positively-charged black dispersion is mixed with the negatively-charged white dispersion to form a thermodynamically stable electrophoretic dispersion containing both black and white dielectric particles with opposite charges. The mixed dispersion is gray in color, however, it forms a black and white contrasting image after applying an electric field in an electrophoretic image display.

As will be recognized by a person skilled in the art, the ratio of the black and white electrophoretic dispersions can be varied to produce a desired degree of black and white contrast in EPIDs.

PROCESS TWO

In the following description of alternative embodiments in Processes Two through Seven, the preferred dielectric media, solvents for washing the particles, surfactants, stabilizers, charge control agents, and co-stabilizers are the same as those described in Process One above unless otherwise noted. In a second, alternative embodiment of the present invention, the black electrophoretic dispersions with positively-charged or negatively-charged particles made by Process One are mixed with a negatively-charged or positively-charged titanium dioxide dispersion to form an electrophoretic dispersion which is able to produce black and white contrasting images.

The titanium dioxide particles are either pre-treated with alumina to form a basic surface or pre-treated with silica to form an acidic surface, which can be positively charged with a positive charge control agent or be negatively charged with a negative charge control agent respectively, and mixed with the negatively-charged or positively-charged black dispersion to produce black and white contrasting images in EPIDs.

PROCESS THREE

In a third alternative embodiment of the present invention, electrophoretic dispersions containing positively-charged black particles and negatively-charged white particles or negatively-charged black particles and positively-charged white particles are made to produce black and white contrasting images in EPIDs.

The black electrophoretic dispersions with positively-charged or negatively-charged particles are made by Process One and the white electrophoretic dispersions are mixtures of the white electrophoretic dispersions made by Process One and titanium dioxide dispersions.

As in Process Two, the titanium dioxide particles are either pre-treated with alumina to form a basic surface or pre-treated with silica to form an acidic surface, which can be positively charged with a positive charge control agent or be negatively charged with a negative charge control agent respectively. The particles are then mixed with the positively or negatively-charged white dispersion made by the Process One yielding a fluid which produces black and white contrasting images in EPIDs.

PROCESS FOUR

In a fourth alternative embodiment of the present invention, the black electrophoretic dispersions with positively-charged or negatively-charged particles made by Process One are mixed with a yellow electrophoretic dispersion to form an electrophoretic dispersion which is able to produce black and yellow contrasting images.

The yellow particles are either organic or inorganic pigments such as Dairylide Yellow, Hansa Yellow, Benzidine Yellow and the like. The yellow pigments can be modified to produce a basic or acidic surface, which can be positively charged with a positive charge control agent or be negatively charged with a negative charge control agent respectively, and mixed with the negatively-charged or positively charged black dispersion to produce black and yellow contrasting images in EPIDs.

PROCESS FIVE

In a fifth alternative embodiment of the present invention, the black electrophoretic dispersions with positively-charged or negatively-charged particles made by Process One are mixed with a light-color electrophoretic dispersion to form an electrophoretic dispersion which is able to produce black and light color contrasting images.

The light-color particles are either organic or inorganic pigments such as titanium dioxide, zinc oxide, silica, zinc sulfide, calcium silicate, alumina hydrate, Dairylide Yellow, Arylide Yellow, Diarylide Orange, Perinone Orange, Ultramarine Blue and the like. The light color pigments are further modified to produce a basic surface or acidic surface, which can be positively charged with a positive charge control agent or be negatively charged with a negative charge control agent respectively, and mixed with the negatively-charged or positively charged black dispersion to produce black and yellow contrasting images in EPIDs.

As will be recognized by a person skilled in the art, the ratio of the black and light-colored electrophoretic dispersions can be varied to produce a desired degree of black and light-color contrast in EPIDs.

PROCESS SIX

In a sixth alternative embodiment of the present invention, the black electrophoretic dispersions with positively-charged or negatively-charged particles made by Process One are mixed with a mixture of light-color electrophoretic dispersions to form an electrophoretic dispersion which is able to produce black and a desired color contrasting images.

The light-color particles are either organic or inorganic pigments such as the white polymeric particles made the Process One, Titanium dioxide, zinc oxide, silica, zinc sulfide, calcium silicate, alumina hydrate, Dairylide Yellow, Arylide Yellow, Diarylide Orange, Perinone Orange, Ultramarine Blue and the like. The light color pigments are be further modified to same surface characters and mixed

together, which can be positively charged with a positive charge control agent or be negatively charged with a negative charge control agent respectively, and mixed with the negatively-charged or positively-charged black dispersion to produce black and a desired color contrasting images in EPIDs.

PROCESS SEVEN

In a seventh alternative embodiment of the present invention, a dark color electrophoretic dispersion with positively-charged or negatively-charged particles is mixed with a mixture of light-color electrophoretic dispersion to form an electrophoretic dispersion which is able to produce dark and desired color contrasting images.

The dark-color particles are either organic or inorganic pigments such as carbon black, iron oxide black, lamp black, Zn Fe Cr brown Spinel, Magnesium Ferrite, Chreen Spinel, Cr oxide Green, Indanthrone Blue, Ultramarine Blue Dioxazine Violet, Quinacridone Violet, Anthraquinoid Red, Perylene Red and the like. The light-colored particles are either organic or inorganic pigments such as the white polymeric particles made by Process One, titanium dioxide, zinc oxide, silica, zinc sulfide, calcium silicate, alumina hydrate, Dairylyde Yellow, Arylide Yellow, Diarylyde Orange, Perinone Orange and the like. The light-color pigments are further modified to same surface characters and mixed together, which can be positively charged with a positive charge control agent or be negatively charged with a negative charge control agent respectively, and mixed with the negatively-charged or positively charged dark-color dispersion to produce a desired color contrasting images in EPIDs.

As will be recognized by a person skilled in the art, the ratio of the dark-color and light-color electrophoretic dispersions can be varied to produce a desired degree of dark-color and light-color contrast in EPIDs.

All equivalents, variations and modifications that can be applied to the described present invention by a person skilled in the art, are intended to be included within the scope of this invention as defined by the appended claims.

What is claimed is:

1. A dielectric dispersion, comprising:

(a) a dielectric fluid;

(b) a first plurality of particles of a first color having a surface charge of a selected polarity dispersed within said dielectric fluid; and

(c) a second plurality of particles of a second color which contrasts substantially with said first color, having a surface charge of opposite polarity to that of said first plurality of particles; and

(d) means for preventing coagulation of said first and second plurality of particles, wherein said means includes a charge control agent for positively charging said first plurality of particles, a second charge control agent for negatively charging said second plurality of particles, and a stabilizer for thermodynamically stabilizing said first plurality of particles and said second plurality of particles, wherein said stabilizer is selected from the group consisting of homopolymers, copolymers, graft polymers, block polymers, and natural high molecular weight compounds.

2. The dispersion of claim 1, wherein said first and second plurality of particles have basic and acidic surface functionalities, respectively.

3. The dispersion of claim 2, wherein said charge control agents are each of comparable molecular weight and structure but have differing end groups.

4. The dispersion of claim 3, wherein said first charge control agent is selected from the group consisting of cupric naphthenate, zirconium octoate, zinc octoate, calcium octoate, polyvinyl alcohol, polyacrylic acid, polymethacrylic acid, and polyvinyl chloride.

5. The dispersion of claim 4, wherein said second charge control agent is selected from the group consisting of polyisobutylene succinimide, barium petronate, barium sulfonate, barium dinonylnaphthalensulfonate, metal oxides, polyvinyl pyridine, pyridine, lecithin, polyvinyl acetate, polyethylene oxide, polymethyl methacrylate, polyacrylamide, polyesters and polyethers.

6. The dispersion of claim 5, wherein one of said two pluralities of particles is dark colored, the other said plurality of particles is light colored and said dielectric fluid is clear.

7. The dispersion of claim 6, wherein at least one of said two pluralities of particles are polymer particles with an outer layer of a different polymer grafted onto the surface thereof.

8. The dispersion of claim 7, wherein both said first and second pluralities of particles are polymer particles with an outer layer of a different polymer grafted onto the surface thereof.

9. The dispersion of claim 8, wherein one of said two pluralities of particles is dyed black by exposure to a metal oxide.

10. The dispersion of claim 9, further including a third plurality of titanium dioxide particles.

11. The dispersion of claim 10, wherein said titanium dioxide particles are pretreated with alumina to form a basic surface thereon for positive surface charging.

12. The dispersion of claim 10, wherein said titanium dioxide particles are pretreated with silica to form an acidic surface thereon for negative surface charging.

13. The dispersion of claim 7, wherein said first plurality of particles are polymer particles with an outer layer of a different polymer grafted onto the surface thereof and dyed by exposure to a metal oxide and said second plurality of particles are yellow pigments selected from the group consisting of dairylyde yellow, hansa yellow and benzidine yellow.

14. The dispersion of claim 7, wherein said first plurality of particles are polymer particles with an outer layer of a different polymer grafted onto the surface thereof and dyed by exposure to a metal oxide and said second plurality of particles are light colored pigments selected from the group consisting of titanium dioxide, zinc oxide, silica, zinc sulfide, calcium silicate, alumina hydrate, dairylyde yellow, arylide yellow, diarylyde orange, perinone orange and ultramarine blue.

15. The dispersion of claim 6, wherein said dark colored particles are selected from the group consisting of carbon black, iron oxide black, lamp black, ZnFeCr brown spinel, magnesium ferrite, chreen spinel, Cr oxide green, indanthrone blue, ultramarine blue, dioxazine violet, quinacridone violet, anthraquinoid red and perylene red and said light colored particles are selected from the group consisting of poly(styrene-co-divinylbenzene)/polyacrylamide, poly(styrene-co-divinylbenzene)/poly(methacrylic acid), titanium dioxide, zinc oxide, silica, zinc sulfide, calcium silicate, alumina hydrate, dairylyde yellow, arylide yellow, diarylyde orange and perinone orange.

16. The dispersion of claim 1, wherein said first charge control agent is a surfactant comprising polyisobutylene succinic anhydride and said second charge control agent is a surfactant comprising polyisobutylene succinimide.

17. The dispersion of claim 16, wherein said first charge control agent charges said first plurality of particles positive

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and said second charge control agent charges said second plurality of particles negative.

18. The dispersion of claim 1, wherein said dielectric fluid is selected from the group consisting of tetrachloroethylene,

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carbon tetrachloride, pentane, octane, decane, cyclohexane, benzene, xylene, sec-butylbenzene and Isopars.

* * * * *

EXHIBIT C



US006194488B1

(12) **United States Patent**
Chen et al.

(10) **Patent No.:** **US 6,194,488 B1**
(45) **Date of Patent:** **Feb. 27, 2001**

(54) **METHOD FOR MAKING
POLYMER-COATED PIGMENT PARTICLES
USING INITIATOR-TREATED PIGMENTS**

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(US)

(73) **Assignee:** **Copytele, Inc.**, Huntington Station, NY
(US)

(*) **Notice:** Subject to any disclaimer, the term of this
patent is extended or adjusted under 35
U.S.C. 154(b) by 0 days.

(21) **Appl. No.:** **09/259,769**

(22) **Filed:** **Mar. 1, 1999**

Related U.S. Application Data

(63) Continuation of application No. 08/916,855, filed on Aug.
22, 1997, now Pat. No. 5,932,633.

(51) **Int. Cl.⁷** **C08K 9/04**

(52) **U.S. Cl.** **523/211; 523/205; 524/190;**
522/60; 522/62; 430/32; 430/34; 430/37

(58) **Field of Search** **523/205, 211;**
522/60, 62; 524/190; 430/32, 34, 37

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Ingersoll PC

(57) **ABSTRACT**

A method of making polymer-coated pigment particles,
involving the steps of providing a plurality of pigment
particles, each of which has an outer surface coated with a
layer of material that operates as a nucleus for initiating
polymerization on the outer surface of each of the pigment
particles. The pigment particles are mixed with a reaction
mixture containing a solvent and a monomer material,
wherein the layer of material coating each of the pigment
particles causes the monomer material to polymerize on the
outer surface of substantially each of the pigment particles
to form a polymer coating thereon.

12 Claims, 1 Drawing Sheet

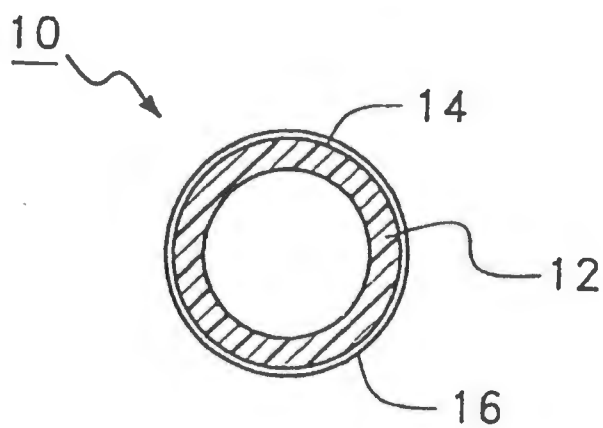


Fig. 1

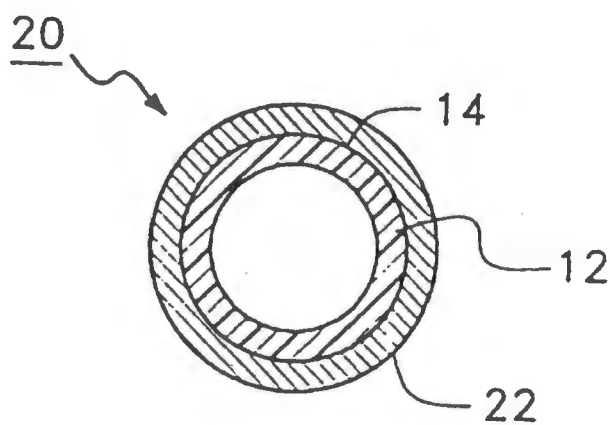


Fig. 2

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METHOD FOR MAKING POLYMER-COATED PIGMENT PARTICLES USING INITIATOR-TREATED PIGMENTS

RELATED PENDING APPLICATIONS

This application is a continuation of Ser. No. 08/916,855 filed Aug. 22, 1997, now U.S. Pat. No. 5,932,633.

Copytele, Inc., the assignee herein, is record owner of U.S. patent application Ser. No. 08/803,716 entitled POLYMERIC-COATED DIELECTRIC PARTICLES AND FORMULATION AND METHODS FOR PREPARING THE SAME filed on Feb. 21, 1997 by Chen et al. now U.S. Pat. No. 5,783,614.

FIELD OF THE INVENTION

The present invention relates generally to pigments and more particularly, to a method of making polymer-coated pigment particles using initiator-treated pigment particles.

BACKGROUND OF THE INVENTION

Pigment particles are used in a variety of applications. For example, dielectric pigment particles suspended in a fluid medium are used in electrophoretic image displays (EPIDs) and electrostatic printers, both of which operate according to the electrophoretic effect. The electrophoretic effect is well known in the art as evidenced by the many patents and articles which describe the effect. As one of ordinary skill in the art will recognize, the electrophoretic effect operates on the principle that pigment particles suspended in the medium can be electrically charged and thereby caused to migrate through the medium to an electrode of opposite charge. For an example of devices using the electrophoretic effect, reference is made to U.S. Pat. No. 4,732,830, issued to Frank J. DiSanto et al., on Mar. 22, 1988, entitled ELECTROPHORETIC DISPLAY PANELS AND ASSOCIATED METHODS, and assigned to Copytele, Inc., the assignee herein.

In typical EPIDs, the dielectric pigment particles are suspended in a fluid medium that is either clear or of an optically-contrasting color as compared with the dielectric pigment particles. To effect the greatest optical contrast between the pigment particles and the fluid suspension medium, it is desirable to have either light-colored pigment particles suspended in a dark medium or dark-colored pigment particles, preferably black, suspended in a backlighted clear medium. A variety of pigment particle and dispersion medium compositions are known in the art. See, for example, U.S. Pat. No. 4,298,444, issued to K. Muller, et al., on Nov. 3, 1981, entitled ELECTROPHORETIC DISPLAY.

It has been found difficult to produce black and other very dark-colored pigment particles that are dielectric, of uniform size, and have a density matching that of a common suspension medium. As a result, EPIDs commonly use readily-manufactured light colored dielectric pigment particles suspended in fluid medium which contain a dark color dye. Such EPIDs are exemplified in U.S. Pat. No. 4,655,897 to DiSanto et al.; U.S. Pat. No. 4,093,534 to Carter et al.; U.S. Pat. No. 4,298,448 to Muller et al.; and U.S. Pat. No. 4,285,801 to Chiang. The use of a yellow pigment particle is disclosed in the following patents, all of which issued to Frederic E. Schubert and are assigned to Copytele, Inc., the assignee herein: U.S. Pat. No. 5,380,362, issued Jan. 10, 1995, entitled SUSPENSION FOR USE IN ELECTROPHORETIC IMAGE DISPLAY SYSTEMS; U.S. Pat. No. 5,403,518, issued Apr. 4, 1995, entitled FORMULATIONS

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FOR IMPROVED ELECTROPHORETIC DISPLAY SUSPENSIONS AND RELATED METHODS; and U.S. Pat. No. 5,411,656, issued May 2, 1995, entitled GAS ABSORPTION ADDITIVES FOR ELECTROPHORETIC SUSPENSIONS.

The selection of the dielectric pigment particles used in the EPID (electrophoretic particles) is very important in determining performance of the EPID and the quality of the image produced. Ideally, the electrophoretic particles should have an optimum charge-to-mass ratio, which is dependent upon the particle size and surface charge; the optimum charge-to-mass ratio is desirable to obtain good electrostatic deposition at high velocity as well as rapid reversal of particle motion when voltages change.

There are advantages to polymerically coating the pigment particles used in EPIDs or other electrophoretic-effect devices. A polymer coating applied to the pigment particles can have the effect of enhancing the ability of the pigment particles to scatter light. When polymer-coated pigment particles impinge upon a transparent screen electrode (thereby displacing the fluid medium from the screen), a brighter color and sharper image is produced (as compared with when uncoated pigment particles are used). Additionally, it naturally is desirable to use pigment particles that are stable and resistant to interaction with the fluid suspension medium to improve the efficiency and lifetime of the EPID; the fluid suspension medium may comprise, for example, aromatic or aliphatic solvents, including benzenes, toluenes, hydrocarbon oil, nonane, decane, or the like, which may react with some typical pigment particles used in EPIDs. Polymer-coated pigment particles can produce a harder and more solvent-resistant composite when compared to uncoated pigment particles.

Furthermore, polymer-coated pigment particles can be less apt to adhere to surfaces within the electrophoretic cell. Over recent years, attention has been directed to dispersion stabilization by way of adsorbed polymers on particle surfaces. See, for example, an article by P. Murau and B. Singer, appearing in Vol. 49, No. 9, of the Journal of Applied Physics (1978), entitled "The Understanding and Elimination of Some Suspension Instabilities in an Electrophoretic Display." See also U.S. Pat. No. 5,403,518, issued to Schubert, referenced above, and U.S. Pat. No. 4,285,801, issued to Anne A. Chiang on Aug. 25, 1981, entitled ELECTROPHORETIC DISPLAY COMPOSITION.

It has been discovered that fluoro polymers are advantageous for use as pigment particle coatings, as they have low critical surface tensions which tend to produce anti-stick properties. See, for example, an article by M. Hudlicry and A. E. Pavlath, appearing in Vol. 187, page 983, of ACS Monograph (1995), entitled "Properties of Fluorinated Compounds II." The lower surface tensions of perfluorinated polymers, when used as a pigment particle coatings, leads to less interaction between the pigment particle surfaces and the solvents comprising the fluid suspension medium in which the pigment particles are dispersed. The stabilizing polymer molecule hydrocarbon chains are more apt to extend out into the solvent and not sit on the particle surface because of its anti-static character. This enhances the efficiency of the stabilizing mechanism.

There are many different methods for preparing polymer-coated particles. For example, thirty different methods are reported in an article by R. E. Sparks, et al., edited by M. A. El-Nokaly et al., *Polymeric Delivery Systems* ACS Symposium Series #520, Washington D.C. (1993). All of these methods produce a three component mixture comprising

polymer-coated particles, uncoated particles and pure polymer particles (polymer particles having no pigment particle core for example).

Accordingly, a need exists for an improved method of manufacturing polymer-coated pigment particles which yields substantially no uncoated pigment particles and substantially no polymer particles having no pigment particle core.

SUMMARY OF THE INVENTION

A method of making polymer-coated pigment particles, comprising the steps of providing a plurality of pigment particles, each of which have an outer surface coated with a layer of material that operates as a nucleus for initiating polymerization on the outer surface of each of the pigment particles. The pigment particles are mixed with a reaction mixture containing a solvent and a monomer material, wherein the layer of material coating each of the pigment particles causes the monomer material to polymerize on the outer surface of substantially each of the pigment particles to form a polymer coating thereon.

BRIEF DESCRIPTION OF THE DRAWINGS

For a detailed understanding of the present invention, reference should be made to the following detailed description taken in conjunction with the accompanying drawings wherein:

FIG. 1 is a cross-sectional view through an initiator-treated particle used in the method of the present invention and

FIG. 2 is a cross-sectional view through a polymer-coated particle prepared in accordance with the method of the present invention.

DETAILED DESCRIPTION OF THE INVENTION

As will become apparent, the method described herein for manufacturing polymer-coated particles is especially useful for making polymer-coated pigment particles that are used in electrophoretic image displays (EPIDs) and like electrophoretic-effect devices. It should be understood, however, that the method of the present invention is also useful for making other types of polymer-coated pigment and like particles that are used in many different applications such as paint, ink, and electrostatic toner.

The key component of the method of the present invention is in the use of initiator-treated pigment particles to produce polymer-coated pigment particles. Initiator-treated pigment particles are described in copending U.S. Patent Application Serial No. entitled INITIATOR-TREATED PIGMENT PARTICLES AND METHOD FOR PREPARING SAME Jing Hong Chemby filed on Aug. 22, 1997, now U.S. Pat. No. 5,964,935, and assigned to Copy-tele, Inc., the assignee herein.

Referring to FIG. 1, a cross-sectional view through an initiator-treated pigment particle 10 is depicted. Each initiator-treated pigment particle 10 comprises a core pigment particle 12 whose outer surface 14 is coated by a layer 16 of material which initiates polymerization on the outer surface 14 of the core pigment particle 12. In other words, the layer 16 of initiator material serves as a nucleus for polymerization to initiate on the outer surface 14 of the core pigment particle 12. The layer 16 of initiator material is selected from azo-initiators, photo-initiators, organic peroxides and the like.

An exemplary embodiment of the method of the present invention is as follows. First, an appropriate type of initiator-treated pigment particles is selected. This is based upon the desired type of core pigment particles to be polymer-coated. For example, if the desired core pigment particles to be polymer-coated are Diarylide yellow pigment particles, then a Diarylide yellow pigment particles which have been initiator-treated are selected. If none are readily available, the appropriate initiator-treated particles can be produced as follows.

The selected core pigment particles to be polymer-coated are cleaned and dried using conventional techniques. The pigment particles are then dispersed in a solvent containing a pigment wetting agent and dissolved initiator material. The wetting agent aids in dispersing the pigment particles in the solvent so that all the pigment particles are coated with the initiator material. This produces a mixture of pigment particles, solvent, and initiator material.

Next, the mixture is sonicated for approximately 20 minutes using a Sonicor Model 1150 Ultrasonic Probe or like device. After sonication, the mixture is placed in an evaporator and the solvent is evaporated at room temperature which leaves the desired initiator-treated pigment particles.

Table I below provides an exemplary formulation for making initiator-treated particles which are used in the method of the present invention:

TABLE I

Materials	Weight(grams)
Pigment	1.2-1.4
Initiator	0.02-0.10
Wetting Agent for pigment	3.0-5.0
Solvent	30.0-35.0

Light and dark colored, inorganic pigments, organic pigments, and various particles produced from polymers, dyes and the like can be initiator-treated and thus, used in the method of the present invention. Examples of light and dark colored inorganic and organic pigments include titanium dioxide, zinc oxide, zinc sulfide, silica, calcium silicate, alumina hydrate, carbon black, iron oxide black, lamp black, Zn Fe Cr brown Spinel, Magnesium Ferrite, Chreen Spinel, Cr oxide Green, Diarylide Yellow, Arylide Yellow, Hansa Yellow, Benzidine Yellow, Perinone Orange, Diarylide Orange, Ultramarine Blue, Indanthrone Blue, Ultramarine Blue Dioxazine Violet, Quinacridone Violet, Anthraquinoid Red, and Perylene Red and the like.

The polymerization initiator material described in Table I is selected from azo-initiators, photo-initiators, organic peroxides and the like. A preferred polymerization initiator material is 4,4'-Azobis (4-cyanovaleic acid). Other polymerization initiator materials include 2,2"-azobis (2-methylbutyronitrile), benzoyl peroxide, lauroyl peroxide, octanoyl peroxide, and the like.

The solvent described in Table I is selected from various aromatic hydrocarbons, aliphatic hydrocarbons, dioxane and dichloromethane. The aromatic hydrocarbon solvents preferably include benzene, toluene, xylene, or tertiary butyl benzene. The aliphatic hydrocarbon solvents preferably include hexane or decane.

The wetting agent described in Table I is selected from alkyl hydrocarbons with functional groups. The preferred wetting agents are alkyl hydrocarbons with functional groups —OH and —NH₂.

Getting back to the method of the present invention, once the initiator-treated pigment particles have been selected or prepared as described above, they are mixed with a solvent that contains the desired monomer for polymer coating according to the exemplary recipe described below in Table II.

TABLE II

Materials	Weight(grams)
Initiator Treated Particles	1.45-2.0
Monomer	1.0-6.0
Solvent	25.0-35.0

Polymerization is initiated on the surface of each initiator-treated pigment particle via the initiator coating, by placing the reaction mixture in a conventional shaker device which contains zirconium oxide beads. The reaction mixture is shaken for approximately two hours. After shaking, the zirconium oxide beads are removed by screening and the reaction mixture is placed in a conventional temperature-controlled tumbler reactor and tumbled at a rotation rate of approximately 40 rpm for approximately six hours at 60° C. and then tumbled about 30 more hours at 70° C. to produce the desired polymer-coated pigment particles.

The polymer-coated pigment particles are then washed with hexane and alcohol to remove any soluble impurities, using a Soxhlet extractor or like apparatus. After washing, the polymer-coated pigment particles are dried in a vacuum oven. The success of the method of the present invention has been ascertained by scanning electron microscopy, particle size analysis and particle density measurements.

FIG. 2 depicts a cross-sectional view through a polymer-treated pigment particle 20 produced by the method of the present invention. As shown, the polymer-coated pigment particle 20 comprises the core pigment particle 12 shown in FIG. 1. However, the layer of initiator material 16 shown in FIG. 1 has been replaced by a polymer coating 22 in FIG. 2 via the polymerization reaction which was initiated on the surface 14 of the core pigment particle 12 by the layer of initiator material 16.

Tables III, IV, and V respectively describe exemplary recipes for making pentafluoropolystyrene coated pigment particles, polystyrene/divinyl benzene coated pigment particles, and porous polymer treated pigment particles, using the method of the present invention. By varying the recipes given below, or by varying other reaction parameters, the physical characteristics of the polymer-coated pigment particles can be selectively altered as desired for a given application.

TABLE III

Materials	Weight(grams)
Initiator Treated Particles	1.20-1.50
Pentafluorostyrene	0.5-2.0
Hydrocarbon Solvent	20.0-30.0

TABLE IV

Materials	Weight(grams)
Initiator Treated Particles	1.20-1.50
Styrene	0.10-2.0

TABLE IV-continued

Materials	Weight(grams)
Divinylbenzene	0.50-6.0
Hydrocarbon Solvent	25.0-35.0

TABLE V

Materials	Weight(grams)
Initiator Treated Particles	1.20-1.50
Styrene	0.0-2.0
Divinylbenzene	0.5-6.0
Diluent	0.3-4.0
Hydrocarbon Solvent	25.0-35.0

In the recipe of Table III, 2,3,4,5,6-Pentafluorostyrene is selected as the monomer for preparing the Pentafluorostyrene coated pigment particles. When this monomer is polymerized, it produces a near-perfluorinated polymer coating that functions as an effective coating for the pigment particles, since perfluorinated polymers have low critical surface tensions.

The Styrene selected as the monomers in the recipes of Tables IV and V is commercially available from Fisher Scientific, Inc. The divinylbenzene selected as the crosslinker in the recipes of Tables IV and V is commercially produced by Dow Chemicals, Inc.

The Hydrocarbon solvents used in the recipes of Tables III, IV, and V are preferably selected from decane or nonane. Other like hydrocarbon solvents can be used also.

For non-aqueous systems, dispersion agents would be used in the above recipes, selected from alkyl hydrocarbons with functional groups —OH and —NH₂. For aqueous systems, surfactants would be used in the above recipes, selected from sodium lauryl sulfate, sodium dodecyl sulfate, Dowfax surfactants, Igepal surfactants, Aerosol surfactants, Pluronic surfactants, Cantrez surfactants, Arlacel surfactants, Tetronic surfactants, poly(vinylalcohol), poly(ethylene oxide), and the like.

The Diluent used in the recipe of Table V is n-hexanol or the like.

The method of the present invention has the advantages of yielding only polymer-coated pigment particles and substantially no uncoated pigment particles and substantially no polymer particles having no pigment particle core. Accordingly, the method of the present invention requires no special separation and purification techniques because only the polymer-coated pigment particles are formed.

Further, the method of the present invention requires no special polymerization techniques, as it has been used successfully in emulsion, dispersion, suspension and inverse emulsion polymerization formats, to produce polymer-coated pigment particles. Moreover, the method of the present invention can be used to form polymer coatings on pigment particles from addition and condensation polymerizations.

Still further, the method of the present invention can be used to polymer-coat many different types of pigment particles. Inorganic polymer pigment particles, metal-organic polymer pigment particles and ceramic pigment particles are examples of some of the different types of pigment particles that can be coated using the method of the present invention.

It will be understood that the embodiments described herein are merely exemplary and that a person skilled in the

art may make many variations and modifications to the described embodiments utilizing functionally equivalent elements to those described. Any variations or modifications to the invention described hereinabove are intended to be included within the scope of the invention as defined by the appended claims.

What is claimed is:

1. A formulation which provides polymer-coated pigment particles that are used in an electrophoretic-effect device, comprising:

a solvent;

a monomer; wherein said monomer is dissolved in said solvent; and

a plurality of pigment particles, wherein said particles are dispersed in said solvent, wherein each of said particles has an outer surface coated with a layer of material which causes said monomer to polymerize on said outer surface of substantially each of said pigment particles thereby forming a polymer coating thereon.

2. The formulation of claim 1, wherein said solvent is a hydrocarbon solvent.

3. The formulation of claim 2, wherein hydrocarbon solvent is selected from the group consisting of decane and nonane.

4. The formulation of claim 1, wherein said layer of material is selected from the group consisting of azo-initiators, photo-initiators, and organic peroxides.

5. The formulation of claim 1, wherein said plurality of pigment particles are selected from the group consisting of inorganic pigment particles, organic pigment particles, polymer pigment particles, and dye pigment particles.

6. The formulation of claim 1, further comprising a cross-linker dissolved in said solvent.

7. The formulation of claim 1, wherein said monomer is selected from the group consisting of pentafluorostyrene and styrene.

8. A formulation for polymer-coated pigment particles that are used in an electrophoretic-effect device, comprising:

a solvent;

a cross linker dissolved in said solvent;

a monomer dissolved in said solvent; and

a plurality of pigment particles dispersed in said solvent, each of said particles having an outer surface coated with a layer of material which causes said monomer material to polymerize on said outer surface of substantially each of said pigment particles to form a polymer coating thereon.

9. The formulation of claim 8, wherein said cross-linker comprises divinyl/benzene.

10. The formulation of claim 8, further comprising a diluent dissolved in said solvent.

11. The formulation of claim 10, wherein said diluent comprises ethyl alcohol.

12. A formulation for polymer-coated pigment particles that are used in an electrophoretic-effect device, comprising:

a solvent selected from the group consisting of nonane and decane;

a monomer dissolved in said solvent; and

a plurality of pigment particles dispersed in said solvent, each of said particles having an outer surface coated with a layer of material which causes said monomer material to polymerize on said outer surface of substantially each of said pigment particles to form a polymer coating thereon.

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