

PASSIVE MATRIX ELECTROPHORETIC DISPLAY MATERIALS

TECHNICAL FIELD

[0001] The invention relates to the complete synthesis of a passively-driven (segmented or passive matrix) electrophoretic display materials, including the manufacture of the separate electrophoretic dispersion components, the combination of these components into a combined electrophoretic fluid that exhibits the phenomenon of voltage-induced charge switching (VICS), the encapsulation of the electrophoretic fluid, and the processing of the encapsulated electrophoretic fluid for display purposes.

BACKGROUND

[0002] An electrophoretic display (EPD) is a non-emissive device that exploits the electrophoretic movement of charged pigment particles suspended in a dielectric solvent to display information. Principles of operation were originally proposed in 1969, and significant developments have been made since. The display usually comprises two oppositely-facing plates, each of which is coated with an electrically conductive layer, that are separated by a known distance (e.g., 50 μm). One or both of the plates (and their respective electrically conductive layers) can be transparent. An electrophoretic suspension, usually composed of charged pigment particles dispersed in a dielectric solvent, is then injected, enclosed, or encapsulated between the two plates, and the plates are sealed and connected to driving circuitry. By driving an electrophoretic display device on a pixel-by-pixel basis, an image or a character can be displayed by a multiplicity of pixels.

[0003] The physicochemical theory of electrophoretic fluids is briefly described here. The electrophoretic fluid in conventional electrophoretic display devices is usually synthesised by first suspending specially-designed pigment particles in a liquid medium with low dielectric constant, followed by adding a charge-inducing agent(s) and/or a dispersing agent(s) into the pigment-liquid mixture in order to induce a charge on the pigment particles and adequately disperse the pigment particles. Generally, it is theorised that the formation of charge at the particle surface stems from the modification of the particle's zeta potential as a consequence of dissociation occurring at the particle surface. The dispersion of the particles is thought to result from steric repulsion between particles as a consequence of adsorption of the dispersing

agent onto the particle surface. However, it is well known to those familiar with the art that neither of these theories satisfactorily explains all observed electrokinetic phenomena, and research is ongoing.

[0004] The functioning of an electrophoretic display is briefly described here. When a voltage is applied between the two electrodes, the charged pigment particles present in the electrophoretic suspension fluid migrate to the electrode of opposite polarity to the particle charge (i.e., positive particles move to the negative electrode and vice versa, while neutral particles remain stationary). Depending on the movement direction (which is based on the polarity of the electric field), the viewer of the display either observes the colour of the pigment that migrates to the transparent electrode surface/plane or observes the colour of the oppositely charged pigment, stationary pigment, or dyed solvent. Importantly, most EPD displays are known to be 'bistable', in that the pigment particles remain in place after being moved, thus the display may be configured to consume no power once the image or information has been presented. As such, EPD displays can have very low power consumption, increasing their appeal for static and/or portable applications where minimising energy use is critical.

[0005] In terms of the electrophoretic fluid used in EPDs, there are potentially several different types of electrophoretic suspensions that are suitable. Single particle systems are usually comprised of one positively- or negatively-charged pigment suspended in a coloured (dyed) dielectric fluid. Dual-particle systems use two pigments that are different in colour (e.g., black and white) and possess different charge characteristics (i.e., positive, negative, or neutral), so that the charged particles migrate to the corresponding electrode depending on the polarity of the electric field, while neutral particles (if used) remain unaffected by the application of the electric field or are displaced by the movement of electrophoretically-mobile particles. In multi-particle systems, several types of pigment particles are used that have both different charge characteristics and different behaviours under the influence of an external electric field (e.g., two pigments that are positively charged and thus move toward the negative electrode, but exhibit different electrophoretic mobility due to different physical characteristics such as colloid size or surface charge). Multi-particle systems can be used to display more than two colours (e.g., red, black, and white).

[0006] The charge characteristics of electrophoretic pigment particles and their sensitivity to

the applied electric field are usually dependent on the nature, concentration, and functionality of both the pigments used and the charge-inducing agents used in the system, and this is especially important if a threshold agent is added to suppress or enhance the charge characteristics of the system. It is hypothesised that the charge-inducing agent in an electrophoretic fluid acts as an electrolyte in the mixture and forms an electric double layer at the interface between the electrophoretic particles in the dispersion and the surrounding solvent, while the threshold agent, if used, suppresses the mobility of the particle to a certain extent. The explanation given for this phenomenon is that the charge density on the surface of the particles is dependent on the degree of dissociation of the counter-ions when the particles are subject to an electric field. The explanation further conjectures that, when both the surface of the electrophoretic pigment particles are rich in electron donors and the solvent comprises an electron acceptor, the pigment particles develop a positive charge, and vice versa. The charge characteristics of the electric double layer on the particle surface resulting from the electron donor/acceptor mechanism are less sensitive to the changes in applied voltage or field strength when compared with the charge characteristics or potential of the electric double layer resulting from a proton donor/acceptor mechanism. Displays engineered to exploit this phenomenon can be driven using a multiplicity of driving schemes (some of which are described below) through careful combination of a strongly voltage-sensitive surface charge effect or agent with a separate voltage-insensitive surface charge effect or agent of opposite polarity.

[0007] Importantly to this invention, a new and previously unobserved phenomenon is utilised, and is briefly described here. The observed phenomenon is that the charge characteristics of a pigment particle can be changed based on a change in voltage magnitude and not sign, i.e. a positively charged particle becomes negatively charged above an equilibrium voltage, and vice versa. This phenomenon is titled voltage-induced charge switching (VICS), and proposed mechanisms are the focus of ongoing research. It is recognised that the mechanism for this phenomenon is different to the mechanism for charge suppression described above. The phenomenon of VICS utilises mixed surfactant systems, including mixed micelles, which generally exhibit significant changes in their physical properties when compared with single-component surfactant systems. Generally, mixed surfactant systems are usually encountered in applications such as separation, dispersion, and detergency. Importantly, the interactions between surfactants in a mixed system, as well as the composition of the individual surfactants, can strongly affect the phase behavior of surfactant

mixtures. Particular attention has been given to the group of surfactants known as room-temperature ionic liquids (RTILs). The study of RTILs has been limited to date, with only a few studies involving detailed research and analysis being undertaken. For example, one study has identified that the electrostatic interactions between single-component charged surfactants are negligible in ionic liquids because of strong charge screening.

[0008] Once a suitable form of electrophoretic fluid has been synthesised, the next step in creating an EPD is to either isolate the fluid into small compartments/cells (usually sized between 10 to >100 μm , but usually <500 μm) or encapsulate the fluid, which mitigates issues such as display sealing, particle migration, and instability. With these methods, the electrophoretic suspension is ideally not in physical contact with the front and back electrodes, in order to prevent problems such as electroplating and undesired movement of particles, and this separation also enhances the stability and thus longevity of the EPD. However, appropriate encapsulation or isolation technologies often require extensive processing (e.g., filtering, drying and re-suspending microcapsules in an appropriate adhesive, or creation of phase-separating curable polymer dispersion), and there continues to be a need to simplify such steps in order to reduce the cost of manufacturing EPDs.

[0009] Display driving methods for EPDs are briefly described here. An EPD may be a segmented display, a passive matrix (PM) display or an active matrix (AM) display, depending on the driving mechanism and circuitry design. Of these mechanisms, the segmented and PM driving schemes are both simpler and cheaper than the AM system. The segmented driving scheme can be used for the presentation of simple information such as numbers and letters, and does not use a pixel matrix; as such, images apart from those defined by each segment cannot be created. A PM system uses row/column electrodes on the top plate and column/row electrodes on the bottom plate, with the top and bottom electrodes being perpendicular to each other. This type of driving mechanism can generate individual pixels at the points where the row and column electrodes overlap. However, there are well-known problems with this driving scheme, namely the phenomenon of crosstalk (described below). An AM display uses a switching mechanism at each pixel in order to have pixel-specific control of the driving voltage, with little to no crosstalk. While this method is known to be the best driving scheme for EPDs (and other displays), the fabrication of pixel-specific switching mechanisms (e.g., using thin-film transistors) is usually considerably more expensive than the fabrication of a PM driving scheme. As such, the PM system is best suited for the cost-

effective production of static, low power, and dynamic displays, so long as issues related to crosstalk can be resolved.

[0010] The phenomenon of crosstalk is briefly described here. With reference to FIG. 1, in a PM driving scheme a voltage V_A is applied to a specific row (X_2 in FIG. 1) and column (Y_2 in FIG. 1) electrode, which provides an electric potential at the pixel (V_A at the point of overlap of electrodes X_2 and Y_2). However, if non-selected electrodes (X_1 , X_3 , Y_1 , and Y_3 in FIG. 1) in the display are left to 'float' (i.e., no voltages are applied to these electrodes), the parts of the display along the selected row and column will also experience an electric potential (V_{float}) that is greater than zero volts but less than the applied voltage at the intersection of the selected electrodes (V_A). One method to control the effect of the induced 'floating' voltage in non-selected pixels is to apply a voltage to the non-selected row and column electrodes in the display (X_1 , X_3 , Y_1 , and Y_3 in FIG. 1); however, by inference, the controlling voltage that can be practically applied to the non-selected row and column electrodes can either be $\frac{1}{3} V_A$ (for detailed justification, refer to U.S. Patent No. 6781745, assigned to Sipix Imaging, Inc.), or the controlling voltage can be $\frac{1}{2} V_A$. If the display material exhibits threshold characteristics (e.g., liquid crystal), crosstalk between pixels will not occur so long as the threshold voltage of the display material is within $\frac{1}{3} V_A$ to $\frac{1}{2} V_A$. For display materials such as electrophoretic dispersion fluids, the phenomenon of crosstalk is problematic; the particles in the fluid are usually electrophoretically mobile at any voltage, and crosstalk may occur along the selected row and column in a PM driving scheme. Some methods have been proposed to manage crosstalk in EPDs, including the addition of gating electrodes, increasing the distance between adjacent cells, and other such methods; ideally, however, a method is needed to induce a threshold voltage in the electrophoretic fluid itself. If the response-voltage characteristics are such that electrophoretic movement of particles is suppressed below a specific voltage, crosstalk can be eliminated. However, many EPD displays made using the electrophoretic materials and techniques currently available typically do not have the required response-voltage characteristics to prevent crosstalk.

[0011] Electrophoretic dispersions with threshold characteristics have been reported previously e.g., Ota et al. (1977), as well as in U.S. Patent Numbers 3612758, 3612758, 4655897, 5177476, 5460688, and 7572394. In these cases and in others, while a threshold voltage was demonstrated, a number of issues were raised and problems encountered, including but not limited to: insufficient response times, complexity in chemical synthesis

(e.g., by requiring synthesis of complex organic molecules c.f., U.S. Patent Number 7572394), complexity in manufacturing processes (e.g., by adding an additional conductor layer or attaching magnetic particles to the pigments c.f., U.S. Patent Number 6239896), redox reactions or electrodeposition at electrode surfaces, lack of image uniformity, or reduced display longevity. As such, while the PM driving system is one of the most cost-effective driving mechanisms, few have been commercially realised e.g., Bridgestone's Quick-Response Liquid Powder Display (QR-LPD). Therefore, there is still a need for a cost effective method for inducing or enhancing the threshold characteristics of an electrophoretic fluid without the trade-offs in image uniformity and display longevity, and without the need for complex chemical synthesis steps or complex driving circuitry.

SUMMARY

[0012] The present invention relates to electrophoretic particles, an electrophoretic dispersion liquid, a means for their encapsulation, and an electrophoretic display device using the encapsulated particle-containing electrophoretic dispersion fluid.

[0013] In a first aspect, the invention refers to the synthesis of a pigment fluid comprising pigment-polymer composite microparticles and/or nanoparticles dispersed in a solvent that possess a low dielectric constant ($\epsilon_r < 5$). The particles may either be suspended or allowed to settle depending on the specific compositions of the pigment, polymer, and solvent.

[0014] In a second aspect, the invention refers to the synthesis of a new type of charge-inducing agent, comprised of a diluted surfactant mixture, which results in particles that exhibit voltage-induced charge switching (VICS). The new charge-inducing agent is made by:

1. diluting a known surfactant that has charge-inducing characteristics in a non-polar or weakly-polar solvent in order to decrease the surfactant viscosity;
2. diluting a known room-temperature ionic liquid (RTIL) to a specific proportion of the quantity of charge-inducing surfactant used in step 1;
3. mixing the solutions of dilute surfactant and dilute RTIL in specific proportions in order to create a single mixture that imparts voltage-induced charge switching (VICS) to pigment particles suspended in a non-polar or weakly-polar solvent.

[0015] In a third aspect, the invention refers to the manufacturing of the pigment fluid

described in the first aspect. The method comprises:

1. synthesising suitable metal oxide pigment microparticles and/or nanoparticles either chemically or mechanically;
2. fusing or reacting the metal oxide pigment microparticles and/or nanoparticles with a suitable polymer to provide specific properties to the microparticles/nanoparticles, namely both the steric stabilisation and the reduction of the specific gravity of the nanoparticles;
3. suspending or mixing the metal oxide pigment microparticles/nanoparticles into a non-polar or weakly-polar solvent;
4. mixing and grinding the solvent, metal oxide pigment microparticles/nanoparticles, and polymer to form a pigment fluid. The mixing involves methods known to those familiar in the art, and can include ball mill grinding, sonication, hand milling with mortar-and-pestle, mechanical grinding, and so on.

[0016] In a fourth aspect, the invention refers to the synthesis of the electrophoretic fluid by addition of a VICS agent described in the second aspect to the to the pigment fluid described in the third aspect, along with an additive to impart colour to the solvent such as an oleophilic, solvent-soluble dye. The method comprises:

1. addition of a known quantity of a VICS agent to the pigment fluid;
2. addition of a known quantity of oleophilic, solvent-soluble dye to the pigment fluid;
3. extended shaking and ultrasonication of the combined mixture to impart the VICS properties of the added agents to the pigment fluid.

[0017] In a fifth aspect, the invention refers to the microencapsulation of the electrophoretic fluid described in the fourth aspect. The method utilises complex coacervation of suitable copolymers, and comprises:

1. dissolving a known quantity of a water-soluble copolymer coacervates in a water bath at a specific temperature under stirring;
2. adding the electrophoretic mixture described in the fourth aspect to the coacervate mixture and emulsifying to initially create dispersed microcapsules of defined size;
3. altering the properties of the mixture by modifying temperature and pH at specific intervals in order to induce coacervation and enhance microcapsule stability;
4. adding a cross-linking agent to the mixture that functions to both strengthen the formed microcapsules and impart self-annealing properties to the microcapsules;

5. rinsing, filtering and sieving the microcapsules for storage as an un-annealed wet slurry.

[0018] In a sixth aspect, the invention refers to the synthesis of a self-annealing microcapsule film that is robust, flexible, and can be placed between display components. The method comprises:

1. partially drying the microcapsule slurry described in the fifth aspect;
2. depositing the partially-dry microcapsules, in air and at room temperature, onto a substrate (e.g., plastic, or display components) via slot-coating, draw-down coating, air blade, and so on.
3. fully drying the microcapsules, in air and at room temperature, to form a self-annealing film that can either be independently manipulated or placed onto display components (front and back plane);
4. alternatively, the partially-dried microcapsules described in the step 1 can be directly printed onto display components (front and back plane) using methods known to those in the art, including inkjet printing, drawdown coating, slot-coating, and so on. The microcapsules can then be fully dried in air and at room temperature.

BRIEF DESCRIPTION OF THE DRAWINGS

[0019] With reference to the detailed description, the invention can be better understood when it is considered in conjunction with the drawings accompanying this application and the non-limited subset of examples provided.

[0020] FIG. 1 is a diagram illustrating the problem of crosstalk in a passive matrix (PM) driving scheme where the non-selected electrodes are 'floating';

[0021] FIG. 2 is a diagram cross-section of a PM display utilising the VICS-modified electrophoretic fluid of the present invention;

[0022] FIG. 3 is a diagram illustrating a method of mitigating crosstalk in a PM driving scheme for a display containing the VICS-modified electrophoretic fluid of the present invention, where the non-selected electrodes are set to half the applied voltage of the selected electrodes;

[0023] FIG. 4 is a graph demonstrating the phenomenon of voltage-induced charged switching (VICS), showing Examples 1, 2, and 7 of the Experimental Section of this application;

[0024] FIG. 5 is a micrograph of the self-annealed microcapsules containing the VICS-modified electrophoretic mixture.

DETAILED DESCRIPTION

[0025] As disclosed in this application, a novel mixture comprising a surfactant and a room temperature ionic liquid is used to impart the characteristic of VICS to particles mixed in a solvent with low dielectric constant. The VICS-modified pigment can then be either mixed with other charge-modified pigments or simply dyed with an oil-soluble or solvent-soluble dye in order to form an electrophoretic fluid suitable for an EPD. The coloured VICS-modified pigment is then encapsulated using a novel microencapsulation approach that self-anneals on drying into a thin film suitable for driving using a PM scheme.

[0026] As used herein, the term "surfactant" refers to different non-limiting classes of charge-inducing agents. Generally, useful surfactants may include, but are not limited to, surfactants comprised of oligomers or polymers that have a functional group capable of forming hydrogen bonding, acid-base interaction, donor acceptor interaction, metal-ligand interaction or Coulombic interaction with the functional group(s) on the surface of particles. Suitable functional groups may include, but are not limited to, -OR, -COOR, -CONRR', -OCONRR', -NRCONR'R'', -SO₂NRR', -NRR', where R, R' and R'' can be hydrogen, alkyl, aryl, alkylaryl, arylalkyl, or a halogenated derivative thereof. Optionally, R, R' or R'' may comprise a heteroatom such as N, S, Si or a metal ion, or an ionic group such as ammonium, carboxylate, pyridinium, phosphate, sulfonate, or sulfate. The molecular weight of the surfactant may range from about 100 to about 1,000,000, preferably from about 300 to about 300,000, more preferably from about 500 to about 100,000. Useful organic amide surfactants include, but are not limited to, polyisobutylene succinimides such as OLOA 1200, OLOA 370, OLOA 11000, polyisobutylene monosuccinimide (T151), and N-vinyl pyrrolidone polymers. Useful organic sulfates and sulfonates include, but are not limited to, ammonium lauryl sulphate, barium sulfonate, calcium dodecyl benzene sulfonate, calcium petroleum sulfonate, dodecylbenzenesulfonic acid sodium salt, neutral or basic calcium dinonylnaphthalene sulfonate, neutral or basic barium dinonylnaphthalene sulfonate, Petronate L (sodium

sulfonate), Petrostep B100 (petroleum sulfonate), and sodium bis(2-ethylhexyl) sulfosuccinate. Other useful surfactants include, but are not limited to, and Alcolac LV30, Solsperse 9000, Solsperse 17000, Span 80 (sorbitan monooleate), Span 85 (sorbitan trioleate), and Unithox 750. Particle dispersion stabilisers may be added to prevent particle flocculation or attachment to encapsulants, including, but not limited to, acetylenic glycols, alkanolamides, alkyl amines, dialkyl oxides, glycol ethers, sorbitol derivatives, and sulfosuccinates. The surfactants used in all embodiments of the present invention is compatible with the solvent in an electrophoretic fluid and may interact with the particle surface to initially generate either positive or negative charge on the surface on the pigment particles.

[0027] The term "room temperature ionic liquid", or RTIL, refers to salts where the cation can be a nitrogen-containing organic compound in tertiary, quaternary, or cyclic arrangement, usually available in salt form, where the cation may be, but is not limited to: alkyl or aryl ammonium, 1-alkyl-3-methylimidazolium, dimethyldinonylammonium, 1-ethyl-3-methylimidazolium ethylsulfate, 1-hexyl-3-methylimidazolium, imidazolium, methyl tridecylammonium, methyl trioctylammonium, piperidinium, pyrrolidinium, pyridinium, and tricaprylammonium. The anion of the salt can also consist of, but is not limited to, halogens from Group 17 of the Periodic Table of Elements such as bromide, chloride, fluoride, and iodide, as well as Brønsted-Lowry conjugate base substituents such as PF_6^- , CH_3COO^- , CF_3COO^- , $(\text{CF}_3\text{SO}_2)_2\text{N}^-$, nitrate, sulphate, as well as aliphatic and aromatic carboxylate anions, including, but not limited to, acetate, benzoic acid, hexanoic acid, as well as thiol-containing anions such as thiosalicylic acid. Among the RTILs having a nitrogen center, quaternary amine salts are particularly useful when the anion of the salt is either a halogen or a Brønsted-Lowry conjugate base. Preferable RTILs can be found in De Los Rios et al., "Ionic Liquids in Separation Technology" (2014), and preferable quaternary salts can be found in "Cationic Surfactants: Organic Chemistry", Surfactant Science Series Vol. 34 (1990). The synthesis of a novel RTIL is also shown in Example 7 in the present application. In one embodiment of the invention, synthesised RTILs are used comprising of methyltrioctylammonium salts where the anion can be acetate, bicarbonate, bisulfate, borohydride, bromide, chloride, hexafluorophosphate, iodide, nitrate, salicylate, thiosalicylate, or other Brønsted-Lowry conjugate base substituents. The RTILs used in all embodiments of the present invention is compatible with the solvent in an electrophoretic

fluid and may interact with the particle surface to initially generate either positive or negative charge on the surface on the pigment particles.

[0028] The term "pigment" refers to any type of particle or dye comprised of soluble compounds or insoluble colloids made of either organic or inorganic components that can impart colour to a solution. The term "particle" as used herein refers to microstructures, nanostructures, and combinations thereof. For example, micro- or nanostructures may be microparticles, microrods, nanoparticles, nanorods, and the like. The term "microstructures" refers to materials with at least one dimension in the micrometer range (between 1 to 100 μm). The term "nanostructures" refers to materials with at least one dimension in the nanometer range (between 1 and 999 nm). Inorganic pigment particles can include metal particles comprised oxide of a metal selected from Group 4 to Group 14 of the Periodic Table of Elements. Metal oxide particles may be metal oxide microstructures or nanostructures. In various embodiments, the metal oxide pigment particles are comprised of an oxide of a metal selected from the group consisting of aluminium, iron, tin, titanium, tungsten, zinc, and combinations thereof. For example, the metal oxide particles may be selected from the group consisting of TiO_2 , ZnO , Fe_2O_3 , Al_2O_3 , and combinations thereof. In specific embodiments, the metal oxide particles comprise or consist predominantly of TiO_2 . Organic pigment particles or dyes can include, but are not limited to, derivatives of anthraquinone, cyanine, diazine, diketopyrrolopyrrole, imine, Indigo, merocyanine, naphthalimide, naphthalocyanine, perylene, porphyrin, phthalocyanine, polyaniline, quinacridone, quinine, quinone, quinacridone, squarylium, triphenylmethane, and xanthene. Ideally, the concentration of the dye or pigment in the electrophoretic fluid can be from about 0.01% to about 10% by weight, preferably about 0.05% to about 5% by weight, or even more preferably from about 0.1% to about 2% by weight. The dyes can be of any colour that provides good contrast to the pigment particles, e.g., white, blue, black, red, green, yellow, or any combination of these colours.

[0029] The pigment particles can be chemically or physically combined with a polymer in order to impart properties suitable for an electrophoretic fluid, including, but not limited to, the properties of lower specific gravity, steric stabilisation, colour enhancement, steric repulsion, buoyancy, increased electrophoretic mobility, enhanced surfactant-particle interaction, bistability, and so forth. The term "polymer" refers to any aliphatic or aromatic hydrocarbon chain where the base monomer can be acrylate, butene, butadiene, bisphenol, ethylene, ester, propylene, siloxane, urethane, and so forth. The surface of the pigment

particles may be coated with a single type of the long-chain polymer stabilisers or several types of polymer stabilisers of different structures. Polymers that have more side chains or have higher lengths are preferred as stabilisers. Such polymers may be formed from base monomers of acrylate or methacrylate such as, but not limited to, hexyl acrylate, lauryl acrylate, lauryl methacrylate, hexyl methacrylate, octyl acrylate, octylmethacrylate, octadecyl acrylate, and octadecyl methacrylate. The polymer stabilisers may either be chemically cross-linked in the presence of a cross-linking agent to form a polymer network over the surface of the pigment particles, or they may be physically fused or bound to the pigment particles through mechanical action and partial dissolution.

[0030] As used in this application, the term "solvent" refers to any non-polar or weakly polar fluid with low dielectric constant (relative permittivity, $\epsilon_r < 5$), which includes various liquid hydrocarbons, organic solvents, silicon oils, halogenated organic solvents, and polymer liquids including linear or branched saturated hydrocarbon liquids having a low molecular weight. Suitable dielectric solvents are solvents of low viscosity, low vapor pressure, and low dielectric constant. Examples of the liquid hydrocarbons may include linear, branched, or cyclic aliphatic and aromatic hydrocarbons where the number of carbons present in each hydrocarbon compound is 30 or less, such as benzene, cycloalkanes, cyclohexane, decane, dimethyl benzene, dodecane, Isopar, n-hexane, naphthalene, nonane, paraffin, tetradecane, and toluene. Halogenated organic solvents may also be used, such as carbon tetrachloride, chloroform, chlorobenzene, dichloromethane, and tetrachloroethylene. Additionally, the dielectric fluid may be prepared by mixing two or more types of organic solvents or oils together in proportions suitable to impart the desired properties. Importantly, the solvent should have good electrophoretic characteristics, such as being non-polar, having high electrical resistivity, having a low dielectric constant, having suitable specific gravity that closely matches other mixture constituents, being able to adequately dissolve any coloured dyes used in the mixture but is also a poor solvent for the microparticles present in the mixture. The solvent should also have good physical characteristics such as low toxicity, high boiling point, suitable viscosity, and so forth. In addition to the electrophoretic pigment particles, a contrasting colourant may be used in the electrophoretic dispersion of the present invention. The contrasting colourant may be formed from dyes or pigments, comprised of substances described above. If the solvent is not coloured by a dye, colour can also be provided by dispersing multiple types of microparticles of different colours in the same suspension.

[0031] The present invention refers, in one aspect, to the synthesis of a mixture of pigment, polymer, and solvent as the starter material for the electrophoretic fluid. The synthesis of a pigment-based electrophoretic fluid usually requires a method of dispersion as one of the first steps, for which a disperser or dispersion process can be used. In general, the pigment particles are added to the suspending solvent, and the resulting mixture is dispersed over time to render the pigment powder into microstructured or nanostructured particles. If necessary, coloured dyes or pigments can be added to the suspension during the dispersion process. Examples of dispersers and dispersion processes include colloid milling, cool milling, cryogenic crushing, ball milling, disk milling, attriting, grinding, milling, microfluidising, homogenisation, and ultrasonic homogenisation. This dispersion process usually requires time, and can take anywhere from as little as 0.5 hours to more than 172 hours (1 week). This dispersion process must be adequate to uniformly disperse the particles in the selected medium or solvent, ensure adequate mixing between components (including processes such as fusing, dissolving, reacting, melting, conjoining, welding, and so forth), ensure reaction steps, if any, are initiated and completed, and charge attribution, if any, takes place. In a preferred embodiment, the time required for the dispersion process is between 4 hours to 24 hours, more preferably between 6 hours to 10 hours.

[0032] The present invention also refers to a method for inducing or enhancing a phenomenon referred to as charge switching (VICS), where the applied voltage dictates the direction of movement of particles within the electrophoretic dispersion. The VICS phenomenon is illustrated in FIG. 2, which shows a cross-section of an example embodiment of the current invention. In FIG. 2, the pigment particles (1) are encapsulated in microcapsules (5), which are in turn layered onto a backplane featuring row electrodes (4) and a frontplane featuring column electrodes (2 and 3). For the purpose of this example, the backplane or the frontplane may be transparent, or both planes may be transparent. Regarding the VICS phenomenon, FIG. 2 further shows that, when a voltage V_A is applied at the intersection of a column electrode (2) and a row electrode (4), and is above the equilibrium voltage V_E , or more preferably twice that of the equilibrium voltage ($V_A = 2 V_E$), the particles move towards the column electrodes (2). In order to mitigate crosstalk, a controlling voltage V_C is also applied to adjacent column electrodes (3) and adjacent row electrodes (not shown) with a value of $\frac{1}{2} V_A$. When V_C is equal to $\frac{1}{2} V_A$, which is in turn equal to V_E , the particles do not move towards either electrode. A VICS-modified display can be easily driven without crosstalk, and

a driving scheme is illustrated in FIG. 3. In FIG. 3, a voltage of V_A is applied at the intersection of the selected electrodes (X_2 and Y_2), while a controlling voltage equal to $\frac{1}{2} V_A$ is applied to the non-selected electrodes (X_1 , X_3 , Y_1 , and Y_3). If $\frac{1}{2} V_A$ is equal to the equilibrium voltage V_E , the voltage used to drive the selected pixel will not affect pixels along the non-selected rows and columns. Furthermore, the direction of movement of particles can be finely tuned using the VICS phenomenon; for example, if it is necessary to move particles adjacent to the selected column and row towards the positive electrode for the purpose of generating an image, V_A can be set to less than $2 V_E$ and V_C to less than V_E , thus mobilising the particles in the adjacent row and column electrodes towards the opposite electrode.

[0033] The VICS agent is comprised of any material that is soluble or dispersible in the solvent mixture, or continuous phase, of an electrophoretic dispersion, and imparts or induces the phenomenon of charge-switching of electrophoretic pigment particles when appropriate voltages are applied. In the context of the present invention, the VICS agent may broadly include any materials which meet the criteria describe above. In one preferred embodiment, a quantity of a surfactant and a quantity of RTIL is combined to form the VICS agent. The method comprises adding the VICS agent to an electrophoretic fluid base that contains electrophoretic pigment particles in a solvent mixture. When preparing the VICS agent, the proportion of surfactant to RTIL can be upwards of 10,000:1, 1,000:1, 100:1, 10:1, or 1:1. In one preferred embodiment, the proportion of surfactant to undiluted RTIL is close to 50:1. The VICS agent is soluble or dispersible in the dielectric solvent or solvent mixture of the electrophoretic dispersion. The concentration of the VICS agent used in the present invention is about 0.01% to about 5% by weight, preferably from about 0.02% to about 2% by weight, based on the total weight of the electrophoretic pigment fluid comprising the pigment particles and solvent or continuous phase. It is believed that the VICS agent has two separate dissociation aspects, where, below an equilibrium voltage, dissociation occurs between one ion pair that is weakly bonded, thus inducing a charge on the surface of the pigment particles of one polarity e.g., negative polarity. When the equilibrium voltage is reached, the dissociation of the weakly bonded ions reaches a maximum, while at the same time dissociation of a more strongly bonded ion pair begins to take place, thus inducing a charge on the surface of the pigment particles of the opposite polarity e.g., positive polarity. Pigment particles move in one direction below the equilibrium voltage, and move in the opposite direction above the equilibrium voltage; however, when the equilibrium voltage is reached or

applied, the movement of pigment particles is neither towards nor away from either electrode, and thus is effectively negated.

[0034] The present invention also refers, in another aspect, to the electrophoretic dispersion produced by the methods described in this application. For example, the electrophoretic dispersion may comprise electrophoretic pigment particles dispersed in a dielectric solvent or solvent mixture and a VICS agent, which is soluble or dispersible in the dielectric solvent or solvent mixture of the electrophoretic fluid and induces the properties of charge-switching at different voltages. The electrophoretic dispersion shows good display performance, in that the dissolved dye used as a highly effective colouring agent produces a background colour for the electrophoretic display medium, but is not necessarily involved in the electrophoretic behavior of the separate and undissolved charged pigment particles when an electric field is applied, and under the action of which only the undissolved charged pigment particles respond to the applied electric field. In a preferred embodiment, only one type of electrophoretic particle is involved in the electrophoretic display, while the colourant is a dissolved pigment that does not necessarily undergo electrophoresis. In this invention, the display may be driven by an up/down switching mode, whereby one electrode plane is transparent thus allowing the viewer to observe either the pigment particles within when they are electrophoretically moved towards the transparent electrode plane, or the dyed solvent when the pigment particles are electrophoretically moved away from the transparent electrode plane.

[0035] To enable deposition of the electrophoretic fluid in a manner suitable for display synthesis, as well as increase the stability of particles in electrophoretic display medium and improve the display performance, the electrophoretic display fluid of this invention can be enclosed in compartments, either as partitions, cells, microcapsules, and the like, each of which is filled with the electrophoretic fluid produced from methods described in this application. When microcapsules are used as enclosures, the methods of microcapsule synthesis can include, but are not limited to, in-situ polymerisation, interfacial polymerisation, single or complex coacervation, phase separation, physical processes such as spray-drying, in-liquid curing, surface deposition, and so forth. As such, the present invention refers, in another aspect, to the microencapsulation of the electrophoretic fluid, comprised of the assembled components of pigment, solvent, and VICS agent, as an internal phase. Encapsulation of the internal phase may be accomplished in a number of different ways, some

of which are detailed in Benita, "Microencapsulation Methods and Industrial Applications" (2006), and Kondo, "Microcapsule Processing and Technology" (1979). When complex coacervation is selected as the microencapsulation procedure, the polymers can be selected from, but are not limited to, one or more of the following compounds: carboxymethyl cellulose, carboxymethyl starch, gelatin, gum arabic, hydroxyethyl cellulose, hydroxypropyl methyl cellulose, melamine, polyvinyl alcohol, polyethylene glycol, soluble starch, soy protein, and urea.

[0036] When the selected microencapsulation technique comprises polymerisation or coacervation, it is often useful to also include a hardening agent or cross-linking agent, in order to stabilise the microcapsules, increase their robustness, and allow for ease of deposition onto display components by e.g., mixing with an adhesive and coating onto a display back-plane or front-plane. There are several hardening agents or cross-linking agents known to those skilled in the art, and can include, but are not limited to, aliphatic and acyclic aldehydes such as acetaldehyde, formaldehyde, glutaraldehyde, glyoxal, 1,3,5-trioxane, as well as co-complexing compounds such as salts of borate, metabisulphate, and tetraborate. Aliphatic and acyclic polyols such as glycol, glycerol, polyethylene glycol, and polyvinyl alcohol are also useful cross-linking, plasticising, or hardening agents for the purpose of microencapsulation.

[0037] According to this invention, microcapsules generally have a shape that is an oblate sphere, with a horizontal diameter from about 10 to 400 μm , preferably from 40 to 200 μm , more preferably from 80 to 150 μm , and a vertical diameter from about 10 to 150 μm , preferably from 20 to 80 μm , more preferably from 30 to 60 μm . In the context of this invention, encapsulation and like terms refer not only to the formation of such articles as are generally referred to as microcapsules, but also to the formation of stable emulsions or dispersions of the electrophoretic material in an encapsulating medium. While most microencapsulation techniques produce discrete microcapsules that can be further processed as a display material e.g., by addition of an adhesive to the microcapsules and further mixing, this invention also includes processes whereby the cross-linking agent used to harden or plasticise the microcapsules also binds the microcapsules together without need for adhesives. The process, termed 'self-annealing microencapsulation', is useful in that the steps required to produce display components, with special regards to the thin film of microencapsulated

electrophoretic fluid that is placed between electrodes, are significantly reduced, and the process of manufacture of this thin film is simplified.

EXPERIMENTAL SECTION

[0038] Methods of fabricating an electrophoretic display that address the well-known problem of crosstalk are provided. In the embodiments disclosed herein, there are two broad steps in the synthesis of the display materials, namely the synthesis of an electrophoretic fluid and the encapsulation of the fluid into a thin film. Regarding the synthesis of the electrophoretic fluid, in embodiments disclosed herein, the fluid is comprised of a mixture of metal oxide/polymer particles in a non-polar solution, along with an agent that engenders the property of VICS mixed directly into the solution, to form an electrophoretic fluid that can be addressed using a passive matrix scheme. Compared to conventional methods of synthesising electrophoretic fluid, where particles usually possess one charge regardless of applied voltage or the electrophoretic movement of particles is suppressed below an equilibrium voltage, the particles in the fluid of the present invention will move in one direction below an equilibrium voltage, and move in the opposite direction above the equilibrium voltage. The process of direct mixing of components allows the creation of the fluid via simple steps.

[0039] Also disclosed herein are examples of the method for synthesising a self-annealing, flexible film comprised of microencapsulated electrophoretic fluid. Creating a self-annealing film eliminates the need for other methods of annealing such as high temperature annealing or the addition of annealing agents and/or adhesives, greatly simplifying production processes.

Example 1. Preparation of electrophoretic fluid (no VICS)

[0040] The steps for synthesising an electrophoretic fluid with no VICS characteristics is described here. The fluid is used for the purpose of providing a comparison, and can be incorporated into a display that uses segments or pre-defined images rather than images formed from pixels.

[0041] In the first step, a fluid base was prepared by first combining 0.749 g of micronised rutile TiO_2 (Sigma Aldrich) with 0.251 g of granular polyethylene (Sigma Aldrich, average molecular weight of 4000) in a ball mill grinding pot containing 25 to 30 Y-stabilised ZrO_2

balls (average diameter of 3 mm). Then, 21.683 g of toluene (Moraka Pte. Ltd) is added to the ball mill grinding pot, and the mixture is set to grind at 600 rpm for 8 hours in a planetary ball mill. The combined ball mill mixture is then removed and stored.

[0042] In the second step, an electrophoretic fluid was prepared. In a separate container, 0.031 g of polyisobutylene monosuccinimide (T151, Wuxi South Petroleum Additive Company) was mixed with a 5.167 g aliquot of the fluid base of the first step. Then, 0.018 g of Sudan Black B (Sigma Aldrich) was added to the aliquot, and the container was shaken vigorously for 1 min. The container was then placed in an ultrasonic water bath (42 kHz, 3 W) that contained cool water (<15 °C) and sonicated for 1 hour. Then, a 130 µL aliquot of this electrophoretic fluid was injected between two ITO-coated glass plates separated by a 80 µm spacer. Electrodes were attached to the ITO-glass plates, and a square-wave voltage was applied to the plates for a range of voltages from 5 V to 80 V. At 20 V, the electrophoretic fluid was mobile in both directions with an estimated electrophoretic mobility of ~250 µm/s (or ~300 ms for particles to move from one electrode plate to the other). The white state reflectance was around 25%, while the black state reflectance was around 6%, as measured by photometry software. The electrophoretic mobility of this preparation is shown in FIG. 4, labeled 'Example 1'. However, after 500 to 1,000 cycles, some stickiness of particles to the electrodes was noticeable, thus slightly degrading the contrast ratio.

Example 2. Preparation of electrophoretic fluid with VICS agent

[0043] The first step of Example 1 was followed regarding the synthesis of the fluid base. In the second step, the following procedure was undertaken. To begin with, a 0.051 g aliquot of a RTIL, specifically methyltrioctylammonium chloride (VWR International Ltd.), was diluted in 5.098 g of toluene. To prepare the VICS agent, a proportional aliquot of the diluted RTIL was added to a proportional aliquot of polyisobutylene monosuccinimide (T151, Wuxi South Petroleum Additive Company) and mixed thoroughly. In order to prepare the electrophoretic fluid, a proportional aliquot of the solution synthesised in the first step of Example 1 was added to the VICS agent, followed by 0.019 g of Sudan Black B (Sigma Aldrich), and the container was shaken vigorously for 1 min. The container was then placed in an ultrasonic water bath (42 kHz, 3 W) that contained cool water (<15 °C) and sonicated for 1 hour.

[0044] To test the effect of the different proportions of VICS agent, T151, and fluid base, a 130 μL aliquot of this electrophoretic fluid was injected between two ITO-coated glass plates separated by a 80 μm spacer. Electrodes were attached to the ITO-glass plates, and a square-wave voltage was applied to the plates for a range of voltages from 5 V to 80 V. The proportions and their respective results are listed in Table 1, below. The behaviour of one particular mixture, Mixture No. 6 in Table 1, is described here. At applied voltages <30 V, the electrophoretic fluid was mobile in both directions with an estimated electrophoretic mobility of between -10 to -50 $\mu\text{m/s}$, that is, the particles took between 8 s to 1.5 s to move from the positive electrode plate to the negative electrode plate when a positive bias was applied, and vice versa. The white state reflectance was around 20% at best, while the black state reflectance was around 10% at best, as measured by photometry software. However, at applied voltages between 30 to 40 V, the particles appeared to undergo charge switching, that is, at these voltages, the direction of the electrophoretic movement of particles was no longer uniformly up or down (towards the front of the display or towards the back of the display), and no discernible white or black states were apparent. Then, at applied voltages between 45 to 80 V, what had been the 'black' state was now prominently the 'white' state, and vice versa, and this was especially apparent at voltages above 60 V. The new white state reflectance was upwards of 50%, while the new black state reflectance was lower than 4%, as measured by photometry software. However, after 500 to 1,000 cycles, some stickiness of particles to the electrodes was noticeable, slightly degrading the contrast ratio. With particular regards to Mixture No. 6 in Table 2, the electrophoretic response to applied voltages is also shown in FIG. 4, labeled 'Example 2'.

Table 1. Electrophoretic mobility of mixtures as described in Example 2

Mixture No.	Dil. RTIL (mg)	T151 (mg)	Fluid base (g)	Electrophoretic mobility (mm/s)		
				@ 10V	@ 30V	@ 50V
1	29	58	5.793	-50	-70	-130
2	51	51	5.098	-30	-10	20
3	74	49	4.910	-10	-10	30
4	92	51	5.099	-10	0	80
5	108	57	5.702	-10	0	100
6	114	58	5.795	-10	0	160
7	112	56	5.611	-10	20	120
8	146	49	4.898	-10	30	90

Example 3. Modification of the VICS-enabled electrophoretic fluid

[0045] In Examples 1 and 2, while the electrophoretic fluid responded appropriately to applied voltages, particle stickiness to electrodes over successive cycles resulted in slight degradation of the contrast ratio between white and black states. To resolve this, the mixture of the first step of Example 1 was modified via the following procedure, with particular regards to the solvents used. To begin with, a mixture of micronised rutile TiO₂ and polyethylene, in a proportion of 75% TiO₂ and 25% polyethylene, was added to the ball mill grinding pot. Then, a proportion of toluene (Moraka Pte. Ltd) was added to the ball mill grinding pot, and the mixture was set to grind at 600 rpm for 6 hours in a planetary ball mill. Then, a proportion of tetrachloroethylene (TCE, VWR International Ltd.) was added to the grinding pot, and the mixture was set to grind at 600 rpm for a further 2 hours. The combined ball mill mixture, used as the fluid base, was then removed and used to create electrophoretic fluids with reagent proportions described in Example 2, Mixture No. 6. The proportions and their respective results are listed in Table 2, below. Mixture No. 3 appeared to show the best contrast ratio, and the electrophoretic particles did not exhibit the noticeable 'stickiness' seen in Examples 1 and 2.

Table 2. Performance of mixtures as described in Example 3

Mixture No.	Toluene (g)	TCE (g)	Reflectance (%)		'Sticky' (1000 cycles)
			White state	Black state	
1	8.67	16.70	43	8	No
2	13.01	8.35	45	6	No
3	13.87	6.68	57	3	No
4	15.61	11.69	53	5	No
5	17.34	-	50	7	Yes

Example 4. Encapsulation of the electrophoretic fluid

[0046] Hardened, non-self-annealing microcapsules were prepared by initially mixing 5.997g urea (VWR International Ltd) and 11.380g formaldehyde (37%) (VWR International Ltd) in 50mL of deionised water in a beaker. The mixture was stirred rapidly for half an hour, after which the pH value was adjusted to 8 using either 10% NaOH (VWR International Ltd) or triethanolamine (VWR International Ltd). The mixture was then heated to 70 °C for 1 hour, after which 25 mL water was added to dilute the product and obtain an urea-formaldehyde

prepolymer. An 8.2 mL aliquot of the electrophoretic dispersion fluid in Example 3 was added to the urea-formaldehyde prepolymer fluid and stirred until the average diameter of the resultant microcapsules were around 100 μm . Then the pH was adjusted to 4 using 10% HCl (VWR International Ltd), and the mixture was stirred continuously for 4 hours prior to cooling and collection of the resultant microcapsules by extensive rinsing, filtering, sieving, and drying.

[0047] To test the microcapsules containing the electrophoretic fluid, the microcapsules were placed between two ITO-coated glass plates, and a square-wave voltage was applied to the bottom and top plates, with the voltages ranging from 5 V to 200 V. At applied voltages <60 V, the encapsulated electrophoretic fluid was not visibly mobile. However, at applied voltages >120 V, electrophoretic movement of particles was visible, with a white state reflectance of 23% and a black state reflectance of 12%, as measured by photometry software.

Example 5. Polymer dispersion encapsulation of electrophoretic fluid

[0048] In Example 4, the electrophoretic dispersion fluid of Example 3 was encapsulated in microcapsules; however, not only was the driving voltage very high, utilising these microcapsules in a display requires further processing, such as mixing with an adhesive and laminating onto front and back planes. An alternative to microencapsulation and sealing is to disperse droplets of electrophoretic fluid in a suitable polymer, such as gelatin. A 10 mL solution of 15% W/W gelatin (VWR International Ltd) was prepared by dissolving 1.5 g gelatin in 10 mL of deionised water with stirring at 450 RPM, at 45 °C. A further 15 mg of sodium dodecyl sulfate (VWR International Ltd) was added to the 15% gelatin solution. Then, 6.0 mL of electrophoretic fluid as prepared in Example 3 was first pre-warmed to 45 °C prior to being added to the gelatin solution, and the mixture was stirred at 150 RPM for 10 minutes. A 0.5 mL aliquot of the mixture was pipetted onto a sheet of ITO-coated glass fitted with plastic spacers at 170 μm thickness. The aliquot was then spread over the glass using a metal blade that tracked the spacers, resulting in an initial 'wet' film that was ~ 170 μm in thickness. The film was then allowed to dry in air at ambient temperature for 2-3 hours, after which the final film thickness reduced to ~ 60 μm , and microscopy indicated that the maximum droplet size in the medium was about 30 μm .

[0049] To test the encapsulated electrophoretic film, a top ITO-coated glass plate was placed onto the film, and a square-wave voltage was applied to the bottom and top plates, with the voltages ranging from 5 V to 200 V. At applied voltages <80 V, the encapsulated electrophoretic fluid was not visibly mobile. However, at applied voltages >80 V, electrophoretic movement of particles was visible, although contrast between discernible black and white states was not adequate for display purposes. At applied voltages >150 V, the white state reflectance was at most 25%, while the black state reflectance was around 9%, as measured by photometry software.

Example 6. Preparation of self-annealing electrophoretic fluid microcapsules

[0050] In Example 4, the electrophoretic fluid of Example 3 was encapsulated in microcapsules; however, utilising these microcapsules in a display requires further processing, such as mixing with an adhesive and laminating onto front and back planes. In Example 5, a polymer dispersion method produced self-annealed encapsulated electrophoretic fluid, but driving voltages were far too high, indicating that the thickness of the gelatin layer encapsulating the droplets is not consistent, and the droplet sizes cannot be easily controlled. In this Example, microencapsulation is used together with an annealing agent to provide the advantages of both approaches.

[0051] Firstly, 0.5 g acacia was dissolved in 50 mL deionised water at 50 °C, with stirring at 400 RPM for 30 min. Then, 0.5 g gelatin was added to the dissolved acacia mix, and stirring continued at 400 RPM for another 30 min. The RPM was then adjusted to 300 RPM. Then, 7.9 mL of the electrophoretic fluid of Example 3 was injected into the solution, with the injection port below the surface of the acacia/gelatin mixture. The mixture was then stirred for 40 minutes. Then the pH of mixture was lowered to 4.4 by adding acetic acid (10% AcOH) dropwise, at a rate of one drop every 30 seconds. When the pH was ~4.4, the temperature of mixture was lowered to 10 °C or less by placing the beaker in an ice water bath. When the temperature was 10 °C or less, 0.6 mL glycerol was added, and stirring continued at <10 °C for one hour. Then the pH was raised to 8.5 by addition of 10% NaOH dropwise at a rate of one drop every 30 seconds, after which the mixture was removed from the ice bath and stirring continued for one hour, with the temperature allowed to rise to room temperature (~20 to 25 °C). The microcapsule mixture was then removed from the hotplate/stirrer, and decanted into a separating funnel for rinsing and sieving. The rinsed, sieved, wet microcapsules were

then spread out onto a plastic sheet using a plastic blade that was spaced at 170 μm using a 'draw-down' system, and then left to dry overnight, after which the average thickness of the film was around 100 μm . A micrograph of the resultant microcapsule film is shown in FIG. 5.

[0052] To test the encapsulated electrophoretic film, a top ITO-coated glass plate was placed onto the film, and a square-wave voltage was applied to the bottom and top plates, with the voltages ranging from 5 V to 200 V. At applied voltages <50 V, the electrophoretic fluid was mobile in both directions with a white state reflectance was around 16% at best, while the black state reflectance was around 10% at best, as measured by photometry software. However, charge-switching was clearly observed at applied voltages between 60 to 90 V, and the white state reflectance was upwards of 38%, while the new black state reflectance was lower than 8%, as measured by photometry software.

Example 7. Preparation of a novel room temperature ionic liquid

[0053] In this Example, methyltrioctylammonium chloride (VWR International Ltd) was selected as a suitable room temperature ionic liquid. Equimolar amounts of methyltrioctylammonium chloride and a selected Brønsted acid, specifically thiosalicylic acid (VWR International Ltd), were transferred to a round-bottomed flask. A sodium hydroxide solution (12 molL^{-1}) was added and the solution was vigorously stirred for 4 hours at 60 $^{\circ}\text{C}$. The ionic liquid phase was separated from the aqueous phase in a separation funnel and washed several times with water. The main water content of the ionic liquid was evaporated for 2 hours at 60 $^{\circ}\text{C}$ and 10 mbar on a rotavapor and subsequently dried at high vacuum at 10^{-2} mbar for 12 hours.

[0054] Then the procedure of Example 3 was followed, except that the novel RTIL preparation described above was used when synthesising the VICS agent. To begin with, the VICS agent was prepared separately by diluting a 0.052 g aliquot of the novel RTIL, specifically methyltrioctylammonium thiosalicylate, with 5.204 g of toluene. Then, in a new container, a 0.114 g aliquot of the diluted RTIL was added to a 0.059 g aliquot of polyisobutylene monosuccinimide (T151, Wuxi South Petroleum Additive Company). Then, a further 5.901 g aliquot of the fluid base described in Example 3 (Mixture No. 3) was added to the new container, followed by 0.017 g of Sudan Black B (Sigma Aldrich), and the

container was shaken vigorously for 1 min. The container was then placed in an ultrasonic water bath (42 kHz, 3 W) that contained cool water (<15 °C) and sonicated for 1 hour.

[0055] To test the effect of the new RTIL in the VICS agent on the preparation, a 130 μ L aliquot of this electrophoretic fluid was injected between two ITO-coated glass plates separated by a 80 μ m spacer. Electrodes were attached to the ITO-glass plates, and a square-wave voltage was applied to the plates for a range of voltages from 5 V to 80 V. Results are shown in FIG. 4, labeled 'Example 7'. In comparison to Example 2, the electrophoretic mobility was generally better than Example 2 in both directions, although the voltage at which charge switching occurred was around the same as Example 2.

CLAIMS

1. A passive matrix electrophoretic display material that is an electronically-addressable film comprising self-annealed microcapsules, each encapsulating an electrophoretic dispersion comprising a) electrophoretic pigment particles and b) a voltage-induced charge switching agent comprising a surfactant and a room temperature ionic liquid, dispersed in a dielectric solvent, wherein depending on magnitude, but independent of polarity, of a voltage applied to the electrophoretic dispersion, each of the electrophoretic pigment particles has a surface charge polarity which is generated and/or varied by interaction with the voltage-induced charge switching agent, and which results in movement of the electrophoretic pigment particles in a direction dictated by the surface charge polarity on the electrophoretic pigment particles upon application of the voltage.
2. The passive matrix electrophoretic display material according to claim 1, wherein the electrophoretic pigment particles comprises a white pigment and a polymer, and wherein weight ratio of the white pigment to polymer is between 50:50 and 90:10, and weight ratio of the white pigment to the dielectric solvent is between 1:100 and 1:10.
3. The passive matrix electrophoretic display material according to claim 1 or 2, wherein the surfactant is a non-ionic, oil-soluble surfactant, and wherein weight ratio of the non-ionic, oil-soluble surfactant to the room temperature ionic liquid is between 1000:1 and 10:1.
4. The passive matrix electrophoretic display material according to any one of claims 1 to 3, wherein the surfactant is polyisobutylene monosuccinimide and the room temperature ionic liquid is methyltrioctylammonium chloride or methyltrioctylammonium thiosalicylate.
5. The passive matrix electrophoretic display material according to any one of claims 1 to 4, wherein concentration of the voltage-induced charge switching agent in the electrophoretic dispersion is in the range of about 0.01 wt% to about 5 wt%.

6. The passive matrix electrophoretic display material according to any one of claims 1 to 5, wherein concentration of the voltage-induced charge switching agent in the electrophoretic dispersion is in the range of about 0.01 wt% to about 2 wt%.
7. The passive matrix electrophoretic display material according to any one of claims 1 to 6, wherein the dielectric solvent comprises more than one solvent, each of which is a non-polar or a weakly polar fluid with relative permittivity of less than 5.
8. The passive matrix electrophoretic display material according to any one of claims 1 to 7, further comprising at least one oil-soluble dye having a contrasting colour to the electrophoretic pigment particles.
9. The passive matrix electrophoretic display material according to any one of claims 1 to 8, wherein the self-annealed microcapsules are formed from one or more cross-linked copolymers.
10. The passive matrix electrophoretic display material according to any one of claims 1 to 9, wherein each of the self-annealed microcapsules has a diameter of between 10 μm to 500 μm .
11. The passive matrix electrophoretic display material according to any one of claims 1 to 10, wherein the electrophoretic pigment particles retain their position in the dispersion for an extended period of time, or indefinitely, upon application of the voltage.
12. The passive matrix electrophoretic display material according to any one of claims 1 to 11, further comprising row electrodes arranged on one face of the passive matrix electrophoretic display material and column electrodes on an opposing face of the passive matrix electrophoretic display material, wherein pixels are formed on the passive matrix electrophoretic display material that is an electronically-

addressable film comprising self-annealed microcapsules, by selectively activating a driving voltage along one or both of the row electrodes and the column electrodes.

13. A method of preparing a passive matrix electrophoretic display material according to any one of claims 1 to 12, the method comprising
 - a) dissolving one or more water-soluble copolymers in a water bath under stirring to form a mixture,
 - b) adding an electrophoretic dispersion to the mixture and emulsifying to form dispersed microcapsules, wherein each of the dispersed microcapsules encapsulates the electrophoretic dispersion,
 - c) adding a cross-linking agent to the resultant mixture for the dispersed microcapsules to form self-annealing microcapsules, and
 - d) depositing the self-annealing microcapsules onto a substrate and forming self-annealed microcapsules to obtain the passive matrix electrophoretic display material that is an electronically-addressable film comprising self-annealed microcapsules.
14. The method according to claim 13, wherein the electrophoretic dispersion is prepared by
 - a) physically or chemically combining pigment particles with a polymer to form modified pigment particles,
 - b) suspending the modified pigment particles in a dielectric solvent to form a suspension,
 - c) mixing and grinding the suspension to obtain a mixture comprising electrophoretic pigment particles dispersed in the dielectric solvent, and
 - d) adding a voltage-induced charge switching agent comprising a surfactant and a room temperature ionic liquid to the mixture to obtain the electrophoretic dispersion, wherein the voltage-induced charge switching agent interacts with the electrophoretic pigment particles to generate and/or vary a surface charge polarity on each of the electrophoretic pigment particles depending on magnitude, but independent of polarity, of a voltage applied to the electrophoretic dispersion, and which results in movement of the electrophoretic

pigment particles in a direction dictated by the surface charge polarity on the electrophoretic pigment particles upon application of the voltage.

15. The method according to claim 13 or 14, wherein depositing the self-annealing microcapsules onto a substrate and forming self-annealed microcapsules comprises applying the self-annealing microcapsules across on the substrate, and forming self-annealed microcapsules by drying the self-annealing microcapsules in air until annealing of the self-annealing microcapsules is completed.
16. The method according to any one of claims 13 to 15, further comprising arranging row electrodes on one face of the passive matrix electrophoretic display material that is an electronically-addressable film comprising self-annealed microcapsules and column electrodes on an opposing face of the passive matrix electrophoretic display material that is an electronically-addressable film comprising self-annealed microcapsules, wherein pixels are formed on the passive matrix electrophoretic display material that is an electronically-addressable film comprising self-annealed microcapsules by selectively activating a driving voltage along one or both of the row electrodes and the column electrodes.