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### **Piezo-Electrophoretic Films and Displays, and Methods for Manufacturing the Same**

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#### **Abstract**

Low voltage piezo-electrophoretic films and display films including low profile piezo-electrophoretic films and displays. In some embodiments, the piezoelectric material of the piezo-electrophoretic films can be selectively patterned with high-voltage electric fields during or after fabrication of the piezo-electrophoretic films. Such films have high contrast ratio and are useful as security markers, authentication films, or sensors. The films are generally flexible. Some films are less than 100  $\mu\text{m}$  in thickness. Some films are less than 50  $\mu\text{m}$  in thickness. Displays formed from the films do not require an external power source.

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## **Background/Summary**

CROSS-REFERENCE TO RELATED APPLICATIONS [0001] This application is a continuation-in-part of U.S. application Ser. No. 18/817,972, filed Aug. 28, 2024 (Publication No. 2024-0419044), which is a continuation-in-part of U.S. application Ser. No. 18/171,719, filed Feb. 21, 2023 (Publication No. 2023-0273495), which claims priority to U.S. Provisional Application No. 63/314,584, filed Feb. 28, 2022. U.S. application Ser. No. 18/817,972 claims priority to U.S. Provisional Application No. 63/579,375, filed Aug. 29, 2023. This application also claims priority to U.S. Provisional Application No. 63/640,556, filed Apr. 30, 2024. [0002] The entire disclosures of the aforementioned applications and all other applications or publications referred to below are incorporated by reference herein in their entireties.

### **FIELD OF THE INVENTION**

[0003] The subject matter disclosed herein relates to electrophoretic displays, and in particular, to thin piezo-electrophoretic displays having an improved contrast ratio, and methods for making the same. The subject matter disclosed herein also relates to low-profile piezo-electrophoretic displays which may be activated or driven without being connected to a power source, and methods for their manufacture.

### **BACKGROUND OF THE INVENTION**

[0004] An electrophoretic display (“EPD”) is a non-emissive device based on the electrophoresis of charged pigment particles dispersed in a solvent or solvent mixture. The display typically comprises two electrodes placed opposing each other which provide an electric field to drive the motion of the charged pigment particles. One of the electrodes is usually transparent. When a voltage difference is imposed between the two electrodes, the pigment particle(s) migrate to one side or the other causing either the color of the pigment particles or the color of the solvent (if colored) being seen from the viewing side.

[0005] Many electrophoretic displays incorporate an electrophoretic fluid that includes a non-polar solvent and one or more sets of charged pigment particles. The particles can have different optical properties (colors), different charges (positive or negative), different charge magnitudes (zeta potentials), and/or different absorptive properties (broadly light-absorbing or broadly light-reflecting, or selectively-absorbing or selectively reflecting). In the instance where there are multiple particle sets with opposite charge polarities, application of an electric field may cause a pigment particle of one set to appear at the viewing surface while the other pigment particle is driven away from the viewing surface.

[0006] Many electrophoretic displays are bi-stable meaning the optical state of such displays persists even after the activating electric field is removed. Bistability is primarily a result of induced dipole charge layers forming around the charged pigments due to complex interactions between the pigments, charge control agents, and free polymers dispersed in the solvent. A bistable display can last for years in the last-addressed optical state before being switched again with the application of a new driving field.

[0007] Driving an electrophoretic display requires a power source such as a battery to provide

power to the display and/or its driving circuitry. For example, a battery may be used to supply power to a driver IC that in turn generates an electric field to energize the display's electrodes. The power source could also be, e.g., a photovoltaic cell, a fuel cell, or a power supply that receives power from a wall outlet. The power source could also be a piezo-electric element which creates charge through physical motion or thermal expansion, as described in U.S. Pat. No. 5,930,026, which is incorporated by reference in its entirety.

[0008] In all of these examples, some type of driving circuitry is required to provide an electrical pathway between the power source and the electrodes. Typically, the circuitry also includes control elements (e.g., switches, transistors, etc.), and a number of discrete components (e.g., resistors, capacitors, etc.).

[0009] In most instances, the circuitry used in conventional displays is complex, but fairly well-known to those skilled in display technology. However, incorporating such circuitry can limit the display's tolerance to mechanical stresses such as flexing and/or twisting. Furthermore, the presence of the additional components typically necessitates an increase in the overall physical dimensions of the fully-assembled display.

[0010] The physical limitations imposed on a display by the addition of a power source and driving circuitry can render such displays unsuitable for an increasing number of applications for which it is desirable to reduce the overall thickness of the display. Accordingly, in an effort to reduce display thickness, some electrophoretic displays utilize a lower-profile piezoelectric element that creates charge in response to mechanical strain or thermal cycling. However, the thickness of a layer of piezoelectric material generally has a direct correlation to the amplitude of the voltage the piezoelectric material is capable of generating in response to mechanical stress. That is, reducing the thickness of the piezoelectric material also reduces the magnitude of the voltage the piezoelectric material generates under stress (and vice versa). Accordingly, in order to generate a voltage potential large enough to cause the charged pigment particles to move by an amount sufficient to achieve acceptable contrast ratio, conventional piezo-electrophoretic displays have typically incorporated a layer of piezoelectric material too thick for such displays to be viable for use in applications requiring them to be durable and substantially unnoticeable when incorporated into thin, low-profile final products such as paper or bank notes.

## SUMMARY OF THE INVENTION

[0011] There is therefore a need for simply-constructed, flexible, durable, and thin electrophoretic displays for applications such as security markers, sensors, and indicators. There is also a need for piezo-electrophoretic displays that are sufficiently thin and durable to be used for such applications requiring low-profile final products while also providing a high contrast ratio.

[0012] According to one aspect of the subject matter disclosed herein, an electro-optic display may include a layer of electrophoretic material; a first conductive layer; and a piezoelectric material positioned between the layer of electrophoretic material and the first conductive layer, the piezoelectric material overlaps with a portion of the layer of electrophoretic material, and a portion of the first conductive layer overlaps with the rest of the electrophoretic material.

[0013] In a first aspect, the invention includes an electrophoretic display film, less than 100  $\mu\text{m}$  thick (top to bottom), comprising a first adhesive layer, an electrophoretic medium layer, a patterned piezo electric layer comprising zones of differential polarization, and a flexible, light-transmissive electrode layer. In some embodiments, the electrophoretic medium layer comprises a plurality of microcapsules containing a non-polar fluid and charged pigment particles that move toward or away from the piezo electric layer when the piezo electric layer is flexed, wherein the microcapsules are coupled to each other with a polymer binder. In some embodiments, the electrophoretic medium layer comprises a plurality of microcells containing a non-polar fluid and charged pigment particles that move toward or away from the piezo electric layer when the piezo electric layer is flexed, wherein the non-polar fluid and charged pigment particles are sealed in the microcells with a scaling layer. In some embodiments, the film is less than 50  $\mu\text{m}$  thick. In some

embodiments, the patterned piezo electric layer comprises polyvinylidene fluoride (PVDF). In some embodiments, the PVDF is poled to create the zones of differential polarization. In some embodiments, the flexible, light-transmissive electrode layer comprises a metal oxide comprising tin or zinc. In some embodiments, the flexible, light-transmissive electrode layer comprises poly(3,4-ethylenedioxythiophene) (PEDOT). In some embodiments, the invention includes an electrophoretic display film assembly comprising a release sheet coupled to an electrophoretic display film as described above, wherein the release sheet is coupled to the first adhesive layer. In some embodiments, a second adhesive layer coupled to the flexible, light-transmissive electrode layer, and a second release sheet coupled to the second adhesive layer.

[0014] In a second aspect, the invention includes a method of making an electrophoretic display film. The method includes the steps of coupling a film of polyvinylidene fluoride (PVDF) to a polymer film comprising acrylates, vinyl ethers, or epoxides to create a piezo-microcell precursor film, coupling the piezo-microcell precursor film to a flexible, light-transmissive electrode layer, coupling the light-transmissive electrode layer to a first release film with a first adhesive layer, embossing the piezo-microcell precursor film to create an array of microcells, wherein the microcells have a bottom, walls, and a top opening, filling the microcells with an electrophoretic medium through the top opening, and sealing off the top opening of the filled microcells with a water-soluble polymer. In some embodiments, the method further comprises applying a primer to the polymer film comprising acrylates, vinyl ethers, or epoxides before coupling the polymer film to the film of polyvinylidene fluoride (PVDF). In some embodiments, the method further comprises coupling the water-soluble polymer to a second release film with a second adhesive layer. In some embodiments, the method further comprises removing the first release film to produce an electrophoretic display film that is less than 100  $\mu\text{m}$  thick. In some embodiments, the electrophoretic medium layer comprises a plurality of microcells containing a non-polar fluid and charged pigment particles that move toward or away from the piezo electric layer when the piezo electric layer is flexed, wherein the non-polar fluid and charged pigment particles are sealed in the microcells with a sealing layer. In some embodiments, the PVDF is poled to create differential zones of polarization. In some embodiments, the flexible, light-transmissive electrode layer comprises a metal oxide comprising tin or zinc. In some embodiments, the flexible, light-transmissive electrode layer comprises poly(3,4-ethylenedioxythiophene) (PEDOT). In some embodiments, the film of polyvinylidene fluoride is patterned with an electric field to create areas of differing polarization. In some embodiments, the method further comprises patterning the completed electrophoretic display film with an electric field to create areas of differing polarization in the film of polyvinylidene fluoride.

[0015] In a third aspect, the invention includes a method of making an electrophoretic display film. The method comprises dispersing a polyvinylidene fluoride (PVDF) solution on a first release to produce a PVDF film less than 10  $\mu\text{m}$  in thickness, coupling the PVDF film to a second release with a conductive adhesive, removing the first release, coupling a polymer film comprising acrylates, vinyl ethers, or epoxides to create a piezo-microcell precursor film, coupling the piezo-microcell precursor film to a flexible, light-transmissive electrode layer, coupling the light-transmissive electrode layer to a first release film with a first adhesive layer, embossing the polymer film comprising acrylates, vinyl ethers, or epoxides to create an array of microcells, wherein the microcells have a bottom, walls, and a top opening, filling the microcells with an electrophoretic medium through the top opening, and scaling off the top opening of the filled microcells with a water-soluble polymer. In some embodiments, the method further comprises applying a primer to the polymer film comprising acrylates, vinyl ethers, or epoxides before coupling the polymer film to the PVDF film. In some embodiments, the method further comprises coupling the water-soluble polymer to a second release film with a second adhesive layer. In some embodiments, the method further comprises removing the first release film to produce an electrophoretic display film that is less than 100  $\mu\text{m}$  thick. In some embodiments, the

electrophoretic medium layer comprises a plurality of microcells containing a non-polar fluid and charged pigment particles that move toward or away from the piezo electric layer when the piezo electric layer is flexed, wherein the non-polar fluid and charged pigment particles are sealed in the microcells with a sealing layer. In some embodiments, the PVDF is poled to create zones of differential polarization. In some embodiments, the flexible, light-transmissive electrode layer comprises a metal oxide comprising tin or zinc. In some embodiments, the flexible, light-transmissive electrode layer comprises poly(3,4-ethylenedioxythiophene) (PEDOT). In some embodiments, the PVDF film is patterned with an electric field to create areas of differential polarization. In some embodiments, the method further comprises patterning the completed electrophoretic display film with an electric field to create areas of differential polarization in the PVDF film.

[0016] In a fourth aspect, an electrophoretic display film, less than 100  $\mu\text{m}$  thick (top to bottom), comprising, a first adhesive layer, a patterned piezo electric layer comprising zones of differential polarization, an electrophoretic medium layer, and a flexible, light-transmissive electrode layer. In some embodiments, the electrophoretic medium layer comprises a plurality of microcapsules containing a non-polar fluid and charged pigment particles that move toward or away from the piezo electric layer when the piezo electric layer is flexed, wherein the microcapsules are coupled to each other with a polymer binder. In some embodiments, the electrophoretic medium layer comprises a plurality of microcells containing a non-polar fluid and charged pigment particles that move toward or away from the piezo electric layer when the piezo electric layer is flexed, wherein the non-polar fluid and charged pigment particles are sealed in the microcells with a sealing layer. In some embodiments, the scaling layer is conductive. In some embodiments, the film is less than 50  $\mu\text{m}$  thick. In some embodiments, the patterned piezo electric layer comprises polyvinylidene fluoride (PVDF). In some embodiments, the PVDF is poled to create differential zones of polarization. In some embodiments, the flexible, light-transmissive electrode layer comprises a metal oxide comprising tin or zinc. In some embodiments, the flexible, light-transmissive electrode layer comprises poly(3,4-ethylenedioxythiophene) (PEDOT). In some embodiments, the invention includes an electrophoretic display film assembly comprising a release sheet coupled to an electrophoretic display film as described above, wherein the release sheet is coupled to the first adhesive layer. In some embodiments, the electrophoretic display film additionally includes a second adhesive layer coupled to the flexible, light-transmissive electrode layer, and a second release sheet coupled to the second adhesive layer.

[0017] In a fifth aspect, the invention includes a method of patterning a piezo-electrophoretic medium film. The method includes coupling a film of polyvinylidene fluoride (PVDF) to a layer of electrophoretic media to create a piezo-electrophoretic medium film, and patterning the piezo-electrophoretic medium film with an electric field. In some embodiments, the electric field is provided by a corona discharge. In some embodiments, the method additionally includes disposing a conductive mask adjacent the piezo-electrophoretic medium film before patterning the piezo-electrophoretic medium film with the corona discharge. In some embodiments, the electric field is provided by a high-voltage write head. In some embodiments, the patterning includes forming regions of differing polarities within the PVDF. In some embodiments, the patterning creates a security marker. In some embodiments, the layer of electrophoretic media comprises a plurality of microcapsules containing a non-polar fluid and charged pigment particles that move toward or away from the piezo electric layer when the piezo electric layer is flexed, wherein the microcapsules are coupled to each other with a polymer binder. In some embodiments, the layer of electrophoretic media comprises a plurality of microcells containing a non-polar fluid and charged pigment particles that move toward or away from the piezo electric layer when the piezo electric layer is flexed, wherein the non-polar fluid and charged pigment particles are sealed in the microcells with a sealing layer.

[0018] In a sixth aspect, the invention includes an electrophoretic display film, less than 100  $\mu\text{m}$

thick (top to bottom), including an adhesive layer, an electrophoretic medium layer, a patterned piezo electric layer comprising zones of differential polarization, and a conductive adhesive layer. In some embodiments, the electrophoretic medium layer comprises a plurality of microcapsules containing a non-polar fluid and charged pigment particles that move toward or away from the piezo electric layer when the piezo electric layer is flexed, wherein the microcapsules are coupled to each other with a polymer binder. In some embodiments, the electrophoretic medium layer comprises a plurality of microcells containing a non-polar fluid and charged pigment particles that move toward or away from the piezo electric layer when the piezo electric layer is flexed, wherein the non-polar fluid and charged pigment particles are sealed in the microcells with a sealing layer. In some embodiments, the sealing layer is conductive. In some embodiments, the film is less than 50  $\mu\text{m}$  thick. In some embodiments, the patterned piezo electric layer comprises polyvinylidene fluoride (PVDF). In some embodiments, the PVDF is poled to create the zones of differential polarization. In some embodiments, the invention includes an electrophoretic display film assembly comprising a release sheet coupled to an electrophoretic display film as described above, wherein the release sheet is coupled to the first adhesive layer. In some embodiments, the invention includes an electrophoretic display film assembly comprising a release sheet coupled to an electrophoretic display film including a conductive adhesive layer, wherein the release sheet is coupled to the conductive adhesive layer.

[0019] In a seventh aspect, the invention includes an electrophoretic display film, less than 100  $\mu\text{m}$  thick (top to bottom), comprising an adhesive layer, a patterned piezo electric layer comprising zones of differential polarization, an electrophoretic medium layer, and a conductive adhesive layer.

[0020] In an eighth aspect, the invention includes a method for making a piezo-electrophoretic display. The method includes depositing a first electrically-conductive adhesive on a first substrate, and depositing a piezoelectric material comprising polyvinylidene fluoride (PVDF) solution on the first electrically-conductive adhesive to produce a piezoelectric layer less than 5  $\mu\text{m}$  in thickness. The method also includes applying a mask to the piezoelectric layer, where the mask includes a plurality of masking portions shielding a first plurality of areas of the piezoelectric layer and a plurality of unmasked portions leaving a second plurality of areas of the piezoelectric layer unshielded. The method also includes polarizing the piezoelectric layer to create a plurality of polarized portions of piezoelectric material corresponding to the second plurality of areas of the piezoelectric layer and plurality of unpolarized portions of piezoelectric material corresponding to the first plurality of areas of the piezoelectric layer. The method also includes removing the mask from the piezoelectric layer, and bonding the piezoelectric layer with a microcell precursor material. The method also includes embossing the microcell precursor material to create a layer of microcells, where the microcells have a bottom, walls, and a top opening. The method also includes filling the microcells with an electrophoretic medium through the top opening, and sealing off the top opening of the filled microcells with a water-soluble polymer to create a sealing layer. The method also includes depositing a second electrically-conductive adhesive on a second substrate, and bonding the sealing layer to the second electrically-conductive adhesive.

[0021] In some embodiments, the method also includes coupling a polymer film comprising acrylates, vinyl ethers, or epoxides to create the microcell precursor material. In some embodiments, the method also includes applying a primer to the microcell precursor material before bonding the piezoelectric layer with the microcell precursor material. In some examples, the primer includes a thermoplastic or thermoset material or a precursor thereof, such as polyurethane, a multifunctional acrylate or methacrylate, a vinylbenzene, a vinyl ether, an epoxide or an oligomers or polymer thereof.

[0022] In some embodiments, the method also includes activating the microcells with a vapor plasma treatment before filling the microcells with the electrophoretic medium. In some embodiments, the electrophoretic medium layer includes a non-polar fluid and charged pigment

particles that move toward or away from the piezoelectric layer when the piezoelectric layer is mechanically stressed, where the non-polar fluid and charged pigment particles are sealed in the microcells with the sealing layer. In some embodiments, the piezoelectric layer is polarized with an electric field. In some embodiments, the electric field is provided by a corona discharge.

[0023] In some embodiments, the first substrate and the second substrate are release films.

[0024] In some embodiments, the method also includes peeling the second substrate from the second electrically-conductive adhesive, and bonding the second electrically-conductive adhesive to a target object. In some embodiments, bonding the second electrically-conductive adhesive to a target object includes hot stamping the second electrically-conductive adhesive to the target object.

[0025] In some embodiments, the method also includes peeling the first substrate from the first electrically-conductive adhesive, and applying a protective coating over the remaining layers of the piezo-electrophoretic display and the target object. In some embodiments, the protective coating comprises a lacquer. In some embodiments, the target object comprises one of paper, a bank note, and a currency bill.

[0026] In a ninth aspect, the invention includes a method for making a piezo-electrophoretic display. The method includes depositing a piezoelectric material including polyvinylidene fluoride (PVDF) solution onto a temporary substrate to produce a piezoelectric layer less than 5  $\mu\text{m}$  in thickness. The method also includes bonding the piezoelectric layer with a first electrically-conductive adhesive on a first substrate, where the temporary substrate is removed from the piezoelectric layer during the bonding process. The method also includes applying a mask to the piezoelectric layer, where the mask includes a plurality of masking portions shielding a first plurality of areas of the piezoelectric layer and a plurality of unmasked portions leaving a second plurality of areas of the piezoelectric layer unshielded. The method also includes polarizing the piezoelectric layer to create a plurality of polarized portions of piezoelectric material corresponding to the second plurality of areas of the piezoelectric layer and plurality of unpolarized portions of piezoelectric material corresponding to the first plurality of areas of the piezoelectric layer. The method also includes removing the mask from the piezoelectric layer, and depositing a second electrically-conductive adhesive onto a second substrate. The method also includes bonding the second electrically-conductive adhesive with a microcell precursor material, and embossing the microcell precursor material to create a layer of microcells, where the microcells have a bottom, walls, and a top opening. The method also includes filling the microcells with an electrophoretic medium through the top opening, and sealing off the top opening of the filled microcells with a water-soluble polymer to create a sealing layer. The method also includes bonding the sealing layer with the piezoelectric layer.

[0027] In some embodiments, the method also includes coupling a polymer film comprising acrylates, vinyl ethers, or epoxides to create the microcell precursor material. In some embodiments, the method also includes applying a primer to the microcell precursor material before bonding the second electrically-conductive adhesive with the microcell precursor material. In some embodiments, the method also includes activating the microcells with a vapor plasma treatment before filling the microcells with the electrophoretic medium.

[0028] In some embodiments, the electrophoretic medium layer comprises a non-polar fluid and charged pigment particles that move toward or away from the piezoelectric layer when the piezoelectric layer is mechanically stressed, where the non-polar fluid and charged pigment particles are sealed in the microcells with the sealing layer. In some embodiments, the piezoelectric layer is polarized with an electric field. In some embodiments, the electric field is provided by a corona discharge.

[0029] In some embodiments, the first substrate and the second substrate are release films.

[0030] In some embodiments, the method also includes peeling the second substrate from the second electrically-conductive adhesive, and bonding the second electrically-conductive adhesive to a target object. In some embodiments, bonding the second electrically-conductive adhesive to a

target object includes hot stamping the second electrically-conductive adhesive to the target object. [0031] In some embodiments, the method also includes peeling the first substrate from the first electrically-conductive adhesive, and applying a protective coating over the remaining layers of the piezo-electrophoretic display and the target object. In some embodiments, the protective coating comprises a lacquer. In some embodiments, the target object includes one of paper, a bank note, and a currency bill.

[0032] The invention also includes a method for integrating a layer of piezoelectric material with an electrode. The method includes providing a first substrate, depositing an electrically-conductive material onto the first substrate, and forming a first electrode on the first substrate from the electrically-conductive material. The method also includes depositing an adhesive material onto the first electrode, forming a tie layer on the first electrode from the adhesive material, and depositing a piezoelectric material including polyvinylidene fluoride (PVDF) solution on the tie layer to produce a piezoelectric layer less than 5  $\mu\text{m}$  in thickness.

[0033] In some embodiments, the first substrate is a release film. In some embodiments, the electrically-conductive material includes a light-transmissive conductive polymer. In some embodiments, the light-transmissive conductive polymer includes poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS). In some embodiments, the light-transmissive conductive polymer further includes a crosslinker. In some embodiments, the light-transmissive conductive polymer includes an aqueous solution comprising PEDOT:PSS.

[0034] In some embodiments, the electrically-conductive material onto the first substrate includes using a Mayer rod, a doctor blade, a slot die, or gravure coating, or a combination thereof. In some embodiments, the polyvinylidene fluoride (PVDF) solution is an aqueous solution.

[0035] In some embodiments, the adhesive material includes a mixture of acrylates, polyurethane, and a solvent based on methyl ethyl ketone. In some embodiments, forming the tie layer on the first electrode includes curing the adhesive material with electromagnetic radiation.

[0036] The invention also includes a method for integrating a layer of microcells with an electrode. The method includes providing a first substrate, depositing an electrically-conductive material onto the first substrate, and forming a first electrode on the first substrate from the electrically-conductive material. The method also includes depositing an adhesive material onto the first electrode, and forming a tie layer on the first electrode from the adhesive material. The method also includes bonding a microcell precursor material to the tie layer, and embossing the microcell precursor material to create a layer of microcells, where the microcells have a bottom, walls, and a top opening. The method also includes filling the microcells with an electrophoretic medium through the top opening, and sealing off the top opening of the filled microcells with a water-soluble polymer to create a sealing layer.

[0037] In some embodiments, the first substrate is a release film. In some embodiments, the electrically-conductive material includes a light-transmissive conductive polymer. In some embodiments, the light-transmissive conductive polymer includes poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS). In some embodiments, the light-transmissive conductive polymer further includes a crosslinker. In some embodiments, the light-transmissive conductive polymer includes an aqueous solution comprising PEDOT:PSS.

[0038] In some embodiments, the electrically-conductive material onto the first substrate includes using a Mayer rod, a doctor blade, a slot die, or gravure coating, or a combination thereof.

[0039] In some embodiments, the method includes applying a primer to the microcell precursor material before bonding the microcell precursor material to the tie layer. In some embodiments, the method includes activating the microcells with a vapor plasma treatment before filling the microcells with the electrophoretic medium. In some embodiments, the primer is in an aqueous solution.

[0040] In some embodiments, the adhesive material includes a mixture of acrylates, polyurethane,



and a solvent based on methyl ethyl ketone. In some embodiments, forming the tie layer on the first electrode includes curing the adhesive material with electromagnetic radiation.

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## Description

### BRIEF DESCRIPTION OF DRAWINGS

[0041] FIG. 1A shows a side view of a piezo-electrophoretic display film of the invention, which includes a star-shaped area of differential polarization. Three exemplary positions, convex, neutral, and concave, are shown from the side. The total thickness of the piezo-electrophoretic display film can be less than 100  $\mu\text{m}$ , e.g., less than 50  $\mu\text{m}$ , e.g., less than 25  $\mu\text{m}$ .

[0042] FIG. 1B shows a top view of a piezo-electrophoretic display film of the invention, which includes a star-shaped area of differential polarization. Three exemplary positions, convex, neutral, and concave, are shown from above. When the piezo-electrophoretic display film is flexed, the area of differential polarization results in the oppositely charged particles appearing at the viewing surface.

[0043] FIG. 2A shows an exemplary thin layer of piezoelectric material on a substrate.

[0044] FIG. 2B exemplifies a method for creating areas of differential polarization in the thin layer of piezoelectric material by using the strong electric fields of a corona discharge. By moving the piezoelectric material closer and further from the discharge, the amount of polarization can be controlled spatially.

[0045] FIG. 2C exemplifies a method for creating areas of differential polarization in the thin layer of piezoelectric material by using the strong electric fields of a corona discharge. A conductive mask is used to pattern the piezoelectric material to create areas of differential polarization.

[0046] FIG. 2D illustrates a polarization (poling) pattern that can be achieved with the methods of FIG. 2B and FIG. 2C.

[0047] FIG. 3A illustrates a side view of a piezo-electric film poled in the A direction.

[0048] FIG. 3B illustrates a top view of a piezo-electric film poled in the A direction.

[0049] FIG. 3C illustrates a side view of a piezo-electric film poled in the G direction using a conductive mask.

[0050] FIG. 3D illustrates a top view of a piezo-electric film poled in the G direction using a conductive mask.

[0051] FIG. 4A shows an exemplary thin layer of a piezo-microcell precursor film on a substrate.

[0052] FIG. 4B exemplifies a method for creating areas of differential polarization in the thin layer of piezoelectric material of a piezo-microcell precursor film by using the strong electric fields of a corona discharge. By moving the piezo-microcell precursor film closer and further from the discharge, the amount of polarization can be controlled spatially.

[0053] FIG. 4C exemplifies a method for creating areas of differential polarization in the thin layer of piezoelectric material of a piezo-microcell precursor film by using the strong electric fields of a corona discharge. A conductive mask is used to pattern the piezoelectric material of the piezo-microcell precursor film to create areas of differential polarization.

[0054] FIG. 4D illustrates a polarization (poling) pattern in a piezo-microcell precursor film that can be achieved with the methods of FIG. 3B and FIG. 3C.

[0055] FIG. 5A is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[0056] FIG. 5B is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[0057] FIG. 5C is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[0058] FIG. 5D is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[0059] FIG. 6A is a schematic cross section of an embodiment of a piezo-electrophoretic display.

[0060] FIG. 6B is a schematic cross section of an embodiment of a piezo-electrophoretic display.

[0061] FIG. 7 details a method for creating a piezo-electrophoretic film or (optionally) display.

[0062] FIG. 8A is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[0063] FIG. 8B is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[0064] FIG. 9A is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[0065] FIG. 9B is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[0066] FIG. 10A is a schematic cross section of an embodiment of a piezo-electrophoretic display.

[0067] FIG. 10B is a schematic cross section of an embodiment of a piezo-electrophoretic display.

[0068] FIG. 10C is a schematic cross section of an embodiment of a piezo-electrophoretic display.

[0069] FIG. 11 details a method for creating a low-profile piezo-electrophoretic film.

[0070] FIG. 12A is a schematic cross section of a piezo-electrophoretic film created with the method shown in FIG. 11.

[0071] FIG. 12B is a schematic cross section of a piezo-electrophoretic display created with the method shown in FIG. 11.

[0072] FIG. 13A is a schematic cross section of an alternative piezo-electrophoretic film created with the method shown in FIG. 11.

[0073] FIG. 13B is a schematic cross section of an alternative piezo-electrophoretic display created with the method shown in FIG. 11.

[0074] FIG. 14 is a flow chart detailing the steps of a method for creating high contrast piezo-electrophoretic films and displays.

[0075] FIG. 15A is a schematic cross section of a piezo-electrophoretic film at step **1440** of the method shown in FIG. 14.

[0076] FIG. 15B is a schematic cross section of a piezo-electrophoretic film after completion of step **1440** of the method shown in FIG. 14.

[0077] FIG. 15C is a schematic cross section of a piezo-electrophoretic film after completion of step **1450** of the method shown in FIG. 14.

[0078] FIG. 15D is schematic cross section of a piezo-electrophoretic film after completion of step **1470** of the method shown in FIG. 14.

[0079] FIG. 15E is a cross section of a piezo-electrophoretic film bonded with a target object after completion of step **1480** of the method shown in FIG. 14.

[0080] FIG. 15F is a cross section of a piezo-electrophoretic film bonded with a target object and coated with a protective coating after completion of step **1480** of the method shown in FIG. 14.

[0081] FIG. 16 shows an enlarged view of a partial cross section of a piezo-electrophoretic display.

[0082] FIG. 17 illustrates an exemplary equivalent circuit of the enlarged cross section shown in FIG. 16

[0083] FIG. 18 is a flow chart detailing the steps of a method for creating high contrast piezo-electrophoretic films and displays.

[0084] FIG. 19A is a schematic cross section of a piezo-electrophoretic film at step **1810** of the method shown in FIG. 18.

[0085] FIG. 19B is a schematic cross section of a piezo-electrophoretic film at step **1830** of the method shown in FIG. 18.

[0086] FIG. 19C is a schematic cross section of a piezo-electrophoretic film after completion of step **1840** of the method shown in FIG. 18.

[0087] FIG. 19D is schematic cross section of a piezo-electrophoretic film after completion of steps **1850** and **1860** of the method shown in FIG. 18.

[0088] FIG. 19E is schematic cross section of a piezo-electrophoretic film after completion of step **1870** of the method shown in FIG. 18.

[0089] FIG. 19F is a cross section of a piezo-electrophoretic film bonded with a target object according to the method shown in FIG. 18.

[0090] FIG. 19G is a cross section of a piezo-electrophoretic film bonded with a target object and coated with a protective coating after completion of step **1880** of the method shown in FIG. 18.

[0091] FIG. 20 is a flow chart detailing the steps of a method for integrating a thin film of

piezoelectric material with an electrode.

## DETAILED DESCRIPTION OF THE INVENTION

[0092] Low-profile piezo-electrophoretic films and displays and methods for manufacturing such films and displays are disclosed herein. In some embodiments, the piezoelectric material of the piezo-electrophoretic films can be patterned with high-voltage electric fields after fabrication of the piezo-electrophoretic films. This feature allows a final user to address the piezoelectric materials with, e.g., a corona discharge at the point of production, which may include, e.g., a bar code or a serial number that is only viewable when the piezo-electrophoretic film is manipulated. The low-profile films and displays described below also achieve a high contrast ratio. The films and displays described herein are generally flexible, and are useful as security markers, authentication films, or sensors. Some films are less than 100  $\mu\text{m}$  in thickness. In some embodiments, the piezo-electrophoretic films are less than 50  $\mu\text{m}$  and foldable without breaking. Displays formed according to the subject matter disclosed herein do not require an external power source.

[0093] The term “electro-optic,” as applied to a material or a display, is used herein in its conventional meaning in the imaging art to refer to a material having first and second display states differing in at least one optical property, the material being changed from its first to its second display state by application of an electric field to the material. Although the optical property is typically color perceptible to the human eye, it may be another optical property, such as optical transmission, reflectance, luminescence or, in the case of displays intended for machine reading, pseudo-color in the sense of a change in reflectance of electromagnetic wavelengths outside the visible range.

[0094] The terms “bistable” and “bistability” are used herein in their conventional meaning in the art to refer to displays comprising display elements having first and second display states differing in at least one optical property, and such that after any given element has been driven, by means of an addressing pulse of finite duration, to assume either its first or second display state, after the addressing pulse has terminated, that state will persist for at least several times, for example at least four times, the minimum duration of the addressing pulse required to change the state of the display element. It is shown in U.S. Pat. No. 7,170,670 that some particle-based electrophoretic displays capable of gray scale are stable not only in their extreme black and white states but also in their intermediate gray states, and the same is true of some other types of electro-optic displays. This type of display is properly called “multi-stable” rather than bistable, although for convenience the term “bistable” may be used herein to cover both bistable and multi-stable displays.

[0095] The term “gray state” is used herein in its conventional meaning in the imaging art to refer to a state intermediate two extreme optical states of a pixel, and does not necessarily imply a black-white transition between these two extreme states. For example, several of the E Ink patents and published applications referred to below describe electrophoretic displays in which the extreme states are white and deep blue, so that an intermediate “gray state” would actually be pale blue. Indeed, as already mentioned, the change in optical state may not be a color change at all. The terms “black” and “white” may be used hereinafter to refer to the two extreme optical states of a display, and should be understood as normally including extreme optical states which are not strictly black and white, for example, the aforementioned white and dark blue states. The term “monochrome” may be used hereinafter to denote a display or drive scheme which only drives pixels to their two extreme optical states with no intervening gray states.

[0096] The term “pixel” is used herein in its conventional meaning in the display art to mean the smallest unit of a display capable of generating all the colors which the display itself can show. In a full color display, typically each pixel is composed of a plurality of sub-pixels each of which can display less than all the colors which the display itself can show. For example, in most conventional full color displays, each pixel is composed of a red sub-pixel, a green sub-pixel, a blue sub-pixel, and optionally a white sub-pixel, with each of the sub-pixels being capable of displaying a range of colors from black to the brightest version of its specified color.

[0097] Several types of electro-optic displays are known. One type of electro-optic display uses an electrochromic medium, for example an electrochromic medium in the form of a nanochromic film comprising an electrode formed at least in part from a semi-conducting metal oxide and a plurality of dye molecules capable of reversible color change attached to the electrode; see, for example O'Regan, B., et al., *Nature* 1991, 353, 737; and Wood, D., *Information Display*, 18 (3), 24 (March 2002). See also Bach, U., et al., *Adv. Mater.*, 2002, 14 (11), 845. Nanochromic films of this type are also described, for example, in U.S. Pat. Nos. 6,301,038; 6,870,657; and 6,950,220. This type of medium is also typically bistable.

[0098] Another type of electro-optic display is an electro-wetting display developed by Philips and described in Hayes, R. A., et al., "Video-Speed Electronic Paper Based on Electrowetting", *Nature*, 425, 383-385 (2003). It is shown in U.S. Pat. No. 7,420,549 that such electro-wetting displays can be made bistable.

[0099] One type of electro-optic display, which has been the subject of intense research and development for a number of years, is the particle-based electrophoretic display, in which a plurality of charged particles moves through a fluid under the influence of an electric field. Electrophoretic displays can have attributes of good brightness and contrast, wide viewing angles, state bistability, and low power consumption when compared with liquid crystal displays.

[0100] An electrophoretic display normally comprises a layer of electrophoretic material and at least two other layers disposed on opposed sides of the electrophoretic material, one of these two layers being an electrode layer. In most such displays both the layers are electrode layers, and one or both of the electrode layers are patterned to define the pixels of the display. For example, one electrode layer may be patterned into elongate row electrodes and the other into elongate column electrodes running at right angles to the row electrodes, the pixels being defined by the intersections of the row and column electrodes. Alternatively, and more commonly, one electrode layer has the form of a single continuous electrode, and the other electrode layer is patterned into a matrix of pixel electrodes, each of which defines one pixel of the display. In another type of electrophoretic display, which is intended for use with a stylus, print head or similar movable electrode separate from the display, only one of the layers adjacent the electrophoretic layer comprises an electrode, the layer on the opposed side of the electrophoretic layer typically being a protective layer intended to prevent the movable electrode damaging the electrophoretic layer.

[0101] Numerous patents and applications assigned to or in the names of the Massachusetts Institute of Technology (MIT) and E Ink Corporation describe various technologies used in encapsulated electrophoretic and other electro-optic media. Such encapsulated media comprise numerous small capsules, each of which itself comprises an internal phase containing electrophoretically-mobile particles in a fluid medium, and a capsule wall surrounding the internal phase. Typically, the capsules are themselves held within a polymeric binder to form a coherent layer positioned between two electrodes. The technologies described in these patents and applications include: [0102] (a) Electrophoretic particles, fluids and fluid additives; see for example U.S. Pat. Nos. 7,002,728 and 7,679,814; [0103] (b) Capsules, binders and encapsulation processes; see for example U.S. Pat. Nos. 6,922,276 and 7,411,719; [0104] (c) Films and sub-assemblies containing electro-optic materials; see for example U.S. Pat. Nos. 6,982,178 and 7,839,564; [0105] (d) Backplanes, adhesive layers and other auxiliary layers and methods used in displays; see for example U.S. Pat. Nos. 7,116,318 and 7,535,624; [0106] (e) Color formation and color adjustment; see for example U.S. Pat. Nos. 7,075,502 and 7,839,564; [0107] (f) Methods for driving displays; see for example U.S. Pat. Nos. 7,012,600 and 7,453,445; [0108] (g) Applications of displays; see for example U.S. Pat. Nos. 7,312,784 and 8,009,348; [0109] (h) Non-electrophoretic displays, as described in U.S. Pat. Nos. 6,241,921; 6,950,220; 7,420,549 and 8,319,759; and U.S. Patent Application Publication No. 2012/0293858; [0110] (i) Microcell structures, wall materials, and methods of forming microcells; see for example U.S. Pat. Nos. 7,072,095 and 9,279,906; and [0111] (j) Methods for filling and sealing microcells; see for example

U.S. Pat. Nos. 7,144,942 and 7,715,088.

[0112] Many of the aforementioned patents and applications recognize that the walls surrounding the discrete microcapsules in an encapsulated electrophoretic medium could be replaced by a continuous phase, thus producing a so-called polymer-dispersed electrophoretic display, in which the electrophoretic medium comprises a plurality of discrete droplets of an electrophoretic fluid and a continuous phase of a polymeric material, and that the discrete droplets of electrophoretic fluid within such a polymer-dispersed electrophoretic display may be regarded as capsules or microcapsules even though no discrete capsule membrane is associated with each individual droplet; see for example, the aforementioned U.S. Pat. No. 6,866,760. Accordingly, for purposes of the present application, such polymer-dispersed electrophoretic media are regarded as sub-species of encapsulated electrophoretic media.

[0113] A related type of electrophoretic display is a so-called “microcell electrophoretic display,” also known as MICROCUP®. In a microcell electrophoretic display, the charged particles and the fluid are not encapsulated within microcapsules but instead are retained within a plurality of cavities formed within a carrier medium, typically a polymeric film. See, for example, U.S. Pat. Nos. 6,672,921 and 6,788,449, both of which are incorporated herein by reference in their entireties.

[0114] Although electrophoretic media are often opaque (since, for example, in many electrophoretic media, the particles substantially block transmission of visible light through the display) and operate in a reflective mode, many electrophoretic displays can be made to operate in a so-called “shutter mode” in which one display state is substantially opaque and one is light-transmissive. See, for example, U.S. Pat. Nos. 5,872,552; 6,130,774; 6,144,361; 6,172,798; 6,271,823; 6,225,971; and 6,184,856. Dielectrophoretic displays, which are similar to electrophoretic displays but rely upon variations in electric field strength, can operate in a similar mode; see U.S. Pat. No. 4,418,346. Other types of electro-optic displays may also be capable of operating in shutter mode. Electro-optic media operating in shutter mode may be useful in multi-layer structures for full color displays; in such structures, at least one layer adjacent the viewing surface of the display operates in shutter mode to expose or conceal a second layer more distant from the viewing surface.

[0115] An encapsulated electrophoretic display typically does not suffer from the clustering and settling failure mode of traditional electrophoretic devices and provides further advantages, such as the ability to print or coat the display on a wide variety of flexible and rigid substrates. (Use of the word “printing” is intended to include all forms of printing and coating, including, but without limitation: pre-metered coatings such as patch die coating, slot or extrusion coating, slide or cascade coating, curtain coating; roll coating such as knife over roll coating, forward and reverse roll coating; gravure coating; dip coating; spray coating; meniscus coating; spin coating; brush coating; air knife coating; silk screen printing processes; electrostatic printing processes; thermal printing processes; ink jet printing processes; electrophoretic deposition (See U.S. Pat. No. 7,339,715); and other similar techniques.) Thus, the resulting display can be flexible. Further, because the display medium can be printed, using a variety of methods, the display itself can be made inexpensively.

[0116] The aforementioned U.S. Pat. No. 6,982,178 describes a method of assembling a solid electro-optic display (including an encapsulated electrophoretic display) which is well adapted for mass production. Essentially, this patent describes a so-called “front plane laminate” (“FPL”) which comprises, in order, a light-transmissive electrically-conductive layer; a layer of a solid electro-optic medium in electrical contact with the electrically-conductive layer; an adhesive layer; and a release sheet. Typically, the light-transmissive electrically-conductive layer will be carried on a light-transmissive substrate, which is preferably flexible, in the sense that the substrate can be manually wrapped around a drum (say) 10 inches (254 mm) in diameter without permanent deformation. The term “light-transmissive” is used in this patent and herein to mean that the layer

thus designated transmits sufficient light to enable an observer, looking through that layer, to observe the change in display states of the electro-optic medium, which will normally be viewed through the electrically-conductive layer and adjacent substrate (if present); in cases where the electro-optic medium displays a change in reflectivity at non-visible wavelengths, the term “light-transmissive” should of course be interpreted to refer to transmission of the relevant non-visible wavelengths. The substrate will typically be a polymeric film, and will normally have a thickness in the range of about 1 to about 25 mil (25 to 634  $\mu\text{m}$ ), preferably about 2 to about 10 mil (51 to 254  $\mu\text{m}$ ). The electrically-conductive layer is conveniently a thin metal or metal oxide layer of, for example, aluminum or ITO, or may be a conductive polymer. Poly(ethylene terephthalate) (PET) films coated with aluminum or ITO are available commercially, for example as “aluminized Mylar” (“Mylar” is a Registered Trademark) from E.I. du Pont de Nemours & Company, Wilmington DE, and such commercial materials may be used with good results in the front plane laminate.

[0117] Assembly of an electro-optic display using such a front plane laminate may be effected by removing the release sheet from the front plane laminate and contacting the adhesive layer with the backplane under conditions effective to cause the adhesive layer to adhere to the backplane, thereby securing the adhesive layer, layer of electro-optic medium and electrically-conductive layer to the backplane. This process is well-adapted to mass production since the front plane laminate may be mass produced, typically using roll-to-roll coating techniques, and then cut into pieces of any size needed for use with specific backplanes.

[0118] U.S. Pat. No. 7,561,324 describes a so-called “double release sheet” which is essentially a simplified version of the front plane laminate of the aforementioned U.S. Pat. No. 6,982,178. One form of the double release sheet comprises a layer of a solid electro-optic medium sandwiched between two adhesive layers, one or both of the adhesive layers being covered by a release sheet. Another form of the double release sheet comprises a layer of a solid electro-optic medium sandwiched between two release sheets. Both forms of the double release film are intended for use in a process generally similar to the process for assembling an electro-optic display from a front plane laminate already described, but involving two separate laminations. Typically, in a first lamination the double release sheet is laminated to a front electrode to form a front sub-assembly, and then in a second lamination the front sub-assembly is laminated to a backplane to form the final display, although the order of these two laminations could be reversed if desired.

[0119] The subject matter presented herein relates to structural designs and manufacturing processes for piezo-electrophoretic films and displays that do not need a power supply (e.g., battery, wired power supply, photovoltaic source, etc.) in order for the display to operate. The assembly process is therefore simplified, and the thickness of such displays is substantially less than that of conventional piezo-electrophoretic displays.

[0120] Piezoelectricity is the charge which accumulates in a solid material in response to applied mechanical stress. Suitable materials for the subject matter disclosed herein may include polyvinylidene fluoride (PVDF), quartz ( $\text{SiO}_2$ ), berlinite ( $\text{AlPO}_4$ ), gallium orthophosphate ( $\text{GaPO}$ ), tourmaline, barium titanate ( $\text{BaTiO}_3$ ), lead zirconate titanate (PZT), zinc oxide ( $\text{ZnO}$ ), aluminum nitride ( $\text{AlN}$ ), lithium tantalite, lanthanum gallium silicate, potassium sodium tartrate and any other known piezoelectric materials.

[0121] Piezo-electrophoretic films and piezo-electrophoretic displays described herein use the piezoelectricity to drive the charged pigment particles of an electrophoretic medium toward one of the display electrodes. Thus, manipulating or physically straining the piezoelectric material when coupled to an electrophoretic media layer can cause the color of the electrophoretic material at the viewing surface to change. For example, by bending or introducing other mechanical stress to a piece of piezoelectric material, voltage may be generated across the electrophoretic medium and this voltage can be utilized to cause movement of the color pigment particles of the electrophoretic medium. If only portions of an electrophoretic media layer are coupled to a piezoelectric material, or if areas of differential polarization are created in the piezoelectric material, an electrophoretic

medium having two types of oppositely-charged pigments can be used to create patterns with high contrast ratios, as shown in FIGS. 1A and 1B. As used herein, the term “contrast ratio” or “CR” for an electro-optic display (e.g., an electrophoretic display) is defined as the ratio of the luminance of the brightest color (white) to that of the darkest color (black) that the display is capable of producing. Normally a high contrast ratio is a desired aspect of an electro-optic display.

[0122] FIGS. 1A and 1B illustrate side and top views of an exemplary piezo-electrophoretic display **100** in accordance with the subject matter disclosed herein. In this embodiment, a piezoelectric material is laminated to an electrophoretic medium layer (discussed below), and one or more electrodes are included to provide a suitable electric field to cause the electrophoretic particles to travel toward (or away from the viewing surface). In the embodiment shown in FIGS. 1A and 1B, a second area **120** of the piezoelectric material of the piezo-electrophoretic display **100** has been polarized in a direction opposite the first area **110**, thus when the piezo-electrophoretic display **100** is manipulated from a neutral state (position 2) to either a first (position 1) or a second (position 3) optical state, the first and second areas (**110**, **120**) will achieve different colors in the two areas. In the instance of an electrophoretic medium having oppositely charged particle sets of black and white, a high contrast image will be formed, e.g., as shown in FIG. 1B. Because the first and second areas (**110**, **120**) of the piezoelectric material can be polarized with good resolution (as discussed below), a variety of images/information can be encoded to “appear” when the piezo-electrophoretic display **100** is manipulated. For example, a security ribbon may be created that exists in a neutral state as a gray strip, but when the security ribbon is flexed, the ribbon will display a security seal, such as the star shape shown in FIG. 1B. Of course, the security seal may alternatively include a bar code, a number, a word, a phone number, and internet address, a QR code, a photograph, a half-tone image, or a logo.

[0123] In principle, a piezoelectric material (optionally adjacent an electrophoretic material) can be polarized with a localized strong electric field, as shown in FIGS. 2A-3D. It is known that piezoelectric material (especially films) can be stimulated to move between polarization states with a variety of external stresses, such as mechanical stretching, heat, electromagnetic fields, and applied force. The piezoelectric effect is closely related to the occurrence of electric dipole moments in solids. The dipole density or polarization ( $P$ ) corresponds to the dipole moments per volume of the crystallographic unit cell, typically measured in C/m.<sup>sup.2</sup>. The resulting dipole density,  $P$ , is a vector field, specific for a particular region of the material (i.e., differential polarization). Similar to magnets, dipoles near each other tend to be aligned in regions (Weiss domains). When first created, the domains are usually randomly oriented. However, using a variety of multi-step processes, the domains can be aligned producing localized areas of differential polarization. The process of aligning these regions is known as poling.

[0124] While many piezoelectric materials are crystalline, a number of flexible piezo-active polymers are known, such as polyvinylidene fluoride (PVDF) and its copolymers, polyamides, and parylene-C. Non-crystalline polymers, such as polyimide and polyvinylidene chloride (PVDC), fall under amorphous bulk polymers. The standard procedure to make piezo active films, such as polyvinylidene fluoride (PVDF), is to create the polymer film and stretch it to create stress and align the dipoles. Stretching transforms unpolarized alpha phase regions of PVDF to polarized beta phase. A subsequent stimulus is added to pole regions of beta phase, for example, using strong electric fields. Other methods of aligning beta phases have been described in the literature, such as laser irradiation and intense magnetic fields. See, e.g., U.S. Pat. No. 9,831,417. If the stimulus is can be done with sufficiently high resolution, the poles can be used to create visible patterns, e.g., as illustrated in FIGS. 1A and 1B. In some embodiments, the electric field is applied at elevated temperatures, however it is not always necessary. In particular, for very thin piezoelectric films, e.g., less than 20  $\mu\text{m}$ , e.g., less than 10  $\mu\text{m}$ , less than 5  $\mu\text{m}$ , it is feasible to pole the film without elevated temperature provided that the electric field is sufficiently strong. In the instance of PVDF, an additional benefit is that such films are also optically transparent, thus they can be coupled to an

electrophoretic medium either between the viewing surface and the electrophoretic medium or the electrophoretic medium can be layered between the piezoelectric film and the viewing surface. [0125] An exemplary method for poling a thin film of piezoelectric material is illustrated in FIGS. 2A-2D. A thin film of piezoelectric material **210**, such as PVDF can be melted and spin-coated on a substrate **220** to form a thin film. The thin film may optionally be thermally-conditioned or stretched prior to poling. Suitable bulk PVDF is available from, e.g., Sigma-Aldrich as a bulk powder or as a film. Pre-stretched piezoactive PVDF films are also available from, e.g., PolyK Technologies (State College, PA). Such films may also be procured with metalized electrode coatings on one side, which may also be used for piezo-electrophoretic films and displays, however poling piezo-electrophoretic with backing metal layers using electric fields is difficult. Co-polymers of PVDF, such as polyvinylidene fluoride-trifluoroethylene (PVDF-TrFE) are also available from both Sigma-Aldrich and PolyK. In some embodiments, thin films of PVDF and PVDF co-polymers can be produced by preparing a concentrated solution of bulk PVDF in a compatible, volatile solvent, such as dimethylformamide (DMF) and slot-coating the concentrated solution on a suitable transfer substrate or release, e.g., using a roll-to-roll process. The PVDF-coated substrate is then heated to drive off the DMF, resulting in a thin film (e.g., less than 20  $\mu\text{m}$ , e.g., less than 10  $\mu\text{m}$ , less than 5  $\mu\text{m}$ ) of PVDF. By carefully controlling the thermal cycles, the resulting film can be pre-conditioned to have larger numbers of beta phase domains, suitable for poling.

[0126] As shown in FIGS. 2B and 2C, the thin film of piezoelectric material **210** can be poled with a high voltage corona discharge **230** with spatial focus. Suitable corona discharge equipment is available from, e.g., Simco-Ion (Alameda, CA). Such devices can create localized 10-50 kV fields, e.g., 30 kV fields, e.g., 20 kV fields that can be brought within a few  $\mu\text{m}$  of the piezo material that will be poled. The spatial focus can be accomplished by steering electric fields and/or gas flow which focus/steer the flow of ions emanating from the corona discharge. As shown in FIG. 2B, the high-voltage corona discharge **230** can be moved in three dimensions to create areas of differential polarization, i.e., to pattern the piezoelectric material **210**. Alternatively, the piezoelectric material **210** can be mounted on an XYZ stage allowing the film work piece to approach the high voltage corona discharge **230** in a controlled fashion. In an alternative embodiment, a conductive mask **240** can be used to protect areas of the piezoelectric material **210** from the high voltage corona discharge **230**, as shown in FIG. 2C. A conductive mask may be fabricated from, e.g., conductive stainless steel or another conductive material that can withstand proximity to the corona discharge. Alternative masks, made from charge-absorbing or charge-blocking materials, such as glass, plastic, or rubber will also work. When the high voltage corona discharge **230** is moved over the thin film of piezoelectric material **210**, the thin film of piezoelectric material **210** is poled only in the areas where the conductive mask **240** is not covering the thin film of piezoelectric material **210**. Additionally, the polarity of the high voltage corona discharge **230** can be reversed, so that some areas can be polarized in a first direction, some areas are polarized in a second direction, and some areas are randomly polarized or unpolarized. See also FIGS. 3A-3D.

[0127] Using the techniques shown in FIGS. 2B and 2C, it is straightforward to create a thin film of piezoelectric material **210** with areas of differential polarization P.sub.1 and P.sub.2, shown as **260** and **270** in FIG. 2D. The areas of differential polarization **260** and **270** do not necessarily have opposite polarities of equal magnitude, however such an arrangement is common to provide better contrast ratios when a two-particle electrophoretic medium is used in conjunction with thin film of piezoelectric material **210**. For example, as shown in 2D, the first area **260** may be polarized toward the viewer, while the second area **270** may be polarized away from the viewer. This technique is further illustrated in FIGS. 3A-3D, which show how a single area **360** of a thin film of piezoelectric material deposited on a substrate **320** can be poled to have a polarization vector coming out of the page, as shown in FIG. 3B. Accordingly, when the thin film of piezoelectric material is manipulated (flexed) it will preferentially drive one polarity of electrophoretic particles



toward a viewing surface. As shown in FIG. 3C, a second area **370** of the thin film of piezoelectric material can be polarized in a different direction, with or without the addition of a conductive mask **340**, resulting in some patterned combination of polarity and magnitude, as needed for the application. As shown in FIG. 3D some portions of area **370** are polarized into the viewing surface, but with shadows created by the conductive mask **340**. Accordingly, when the piezoelectric material is manipulated (flexed) it will preferentially drive one polarity of electrophoretic particles toward a viewing surface, except in the areas where the polarization has been masked, which will remain in a neutral color stage, thereby giving rise to a pattern, e.g., a security seal.

[0128] FIGS. 2A-3D illustrate the various techniques that can be used to create areas of differential polarization in a thin film of piezoelectric material **210**. As illustrated in FIGS. 4A-4D, these same techniques can be used to create areas of differential polarization in a thin piezo-electrophoretic medium film **405** as well. As shown in FIG. 4A, a thin film of piezoelectric material **410** can be coupled to a layer of electrophoretic microcells **420** to create a piezo-electrophoretic medium film **405**. The thin film of piezoelectric material **410** can be coupled to a layer of electrophoretic microcells **420** with an adhesive layer (not shown) or the thin film of piezoelectric material **410** can be spin-coated directly to the layer of electrophoretic microcells **420**, i.e., as discussed above with respect to FIG. 2A. The electrophoretic microcells **420** are typically formed from a polymer, such as from acrylates, vinyl ethers, or epoxides, as described in detail in, for example, U.S. Pat. Nos. 6,930,818, 7,052,571, 7,616,374, 8,361,356, and 8,830,561, all of which are incorporated by reference in their entireties. In some embodiments, the layer of electrophoretic microcells **420** may be filled with an electrophoretic medium **425** including two or more electrophoretic particles **423** and **427**, which typically have different electrophoretic mobilities and optical properties. The electrophoretic medium **425** may be sealed with a sealing layer **430**, preferably a water-soluble sealing layer as described in U.S. Pat. Nos. 7,560,004, 7,572,491, 9,759,978, or 10,087,344, all of which are incorporated by reference in their entireties. In some embodiments, the layer of electrophoretic microcells **420** is created on a release, filled with electrophoretic medium **425** and sealed with sealing layer **430**, and then the filled and sealed electrophoretic microcells **420** are used as the substrate for the creation of the thin film of piezoelectric material **410**. The resulting structure is a thin piezo-electrophoretic medium film **405**. In other embodiments the thin film of piezoelectric material **410** is laminated to an acrylate, vinyl ether, or epoxide film that is a precursor to a layer of electrophoretic microcells **420**. The combined thin film of piezoelectric material **410** and precursor material is then embossed on the precursor side (discussed below), and subsequently filled with electrophoretic medium **425** and sealed with sealing layer **430** in order to produce a thin piezo-electrophoretic medium film **405**. In yet another embodiment (not shown in FIGS. 4A-4D), a complete microcell front plane laminate, of the type described in U.S. Pat. No. 7,158,282 and available commercially from E Ink Corporation, can be used as the substrate for a thin film of piezoelectric material **410**, which can be poled as described below. Notably, when a front plane laminate material is used, the final structure additionally includes a conductive layer, which is typically light-transmissive. The front plane laminate can be oriented so that the light-transmissive electrode layer is in contact with the thin film of piezoelectric material **410**, or the front plane laminate can be flipped over so that the scaling layer is in contact with the thin film of piezoelectric material **410**.

[0129] Once the thin piezo-electrophoretic medium film **405** has been created, thin film of piezoelectric material **410** can be addressed as described above with respect to FIGS. 2A-3D. That is the thin film of piezoelectric material **410** can be poled with a high voltage corona discharge **230** with spatial focus, as shown in FIG. 4B, e.g., by mounting the thin piezo-electrophoretic medium film **405** on an XYZ stage allowing the film work piece to approach the high voltage corona discharge **230** in a controlled fashion. In an alternative embodiment, a conductive mask **240** can be used to protect areas of thin piezo-electrophoretic medium film **405** from the high voltage corona discharge **230**, as shown in FIG. 4C. As discussed with respect to FIG. 2A-3D, the polarity of the

high voltage corona discharge **230** can be reversed, so that some areas can be polarized in a first direction, some areas are polarized in a second direction, and some areas are randomly polarized or unpolarized. Like FIG. **2D** above, poling the thin film of piezoelectric material **410** in the thin piezo-electrophoretic medium film **405** results in areas of differential polarization P.sub.1 and P.sub.2, shown as **460** and **470** in FIG. **4D**. Importantly, because the thin piezo-electrophoretic medium film **405** can be fabricated before poling, it is feasible for an end-customer to control the final step of creating the desired poling design in the thin piezo-electrophoretic medium film **405**. Thus, if the final product will include a security seal or serial number, the security seal or serial number can be placed after the final product has been completed and verified, etc. For example, a United States \$100 bill may be printed at the United States Treasury with a serial number in metallic ink at the same time that a security ribbon comprising a thin piezo-electrophoretic medium film **405** is poled to create a verification code corresponding to the serial number. This feature eliminates many logistical problems, and associated costs, because it is not necessary to, for example, match a pre-fabricated security marker with a specific product further downstream in the supply chain.

[0130] The techniques described above can be used to achieve a great variety of thin piezo-electrophoretic films as described in the following figures.

[0131] As shown in FIGS. **5A-6B**, **8A-10C**, and **12A-13B**, a piezo-electrophoretic film or a piezo-electrophoretic display includes a layered stack of some number of components including a thin piezo-electric film and a layer of electrophoretic media. The piezoelectric material can be any of the materials listed above, however polymers, such as PVDF and its copolymers are preferred because they can be fabricated into very thin films. The electrophoretic media typically includes one or more sets of charged particles that move through a non-polar solvent in the presence of an electric field. The electrophoretic media is typically contained, i.e., in microcapsules, microcells, or dispersed droplets. The electrophoretic media can also be contained in open troughs or wells which are sealed in a larger flexible container. The piezo-electrophoretic films and piezo-electrophoretic displays exemplified herein can be made quite thin, e.g., 100  $\mu\text{m}$  thick or less, e.g., 70  $\mu\text{m}$  thick or less, e.g., 50  $\mu\text{m}$  thick or less, e.g., 35  $\mu\text{m}$  thick or less, e.g., 20  $\mu\text{m}$  thick or less, e.g., 10  $\mu\text{m}$  thick or less. Such thin materials are able to flex without breaking or leaking and are also not noticeable when incorporated into final products, such as paper or a bank note. Additionally, many of the piezo-electrophoretic film or a piezo-electrophoretic displays include layers that are all light-transmissive and/or sufficiently thin to be light transmissive thus allowing the piezo-electrophoretic response to be viewed from above and below. In such piezo-electrophoretic film or a piezo-electrophoretic displays, when a first image is viewable from the top surface, e.g., Position 1 of FIG. **1B**, the bottom surface will typically show the negative, e.g., Position 3 of FIG. **1B**. However, when incorporating electrophoretic media with more than two types of particles, the top and bottom may not show reversed images due to mixed particle states at one of the two surfaces.

[0132] A piezo-electrophoretic film or a piezo-electrophoretic display will often include at least one electrode layer, which may be light-transmissive, and which may be flexible. Suitable materials include commercial ITO-coated PET, which may be used as substrate for manufacturing. In some other embodiments, flexible and transparent conductive coatings including other transparent conductive oxides (TCOs) may be used, such as, zinc oxide, zinc tin oxide, indium zinc oxide, aluminum zinc oxide, indium tin zirconium oxide, indium gallium oxide, indium gallium zinc oxide, or fluorinated variants of these oxides such as fluorine-doped tin oxide. In many of the embodiments described herein, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) is used because it has excellent bending properties and is optically transparent. While the overall conductivity is not as high as, e.g., PET/ITO, PEDOT:PSS is sufficient to provide the necessary electric field to drive the electrophoretic particles in the electrophoretic medium. Unless otherwise noted, the term PEDOT is used interchangeably with PEDOT:PSS herein. Other materials include polymers, typically light-transmissive polymers, that are doped with conductive

materials such as carbon black, metal flakes, metal whiskers, carbon nanotubes, silicon nitride nanotubes, or graphene. In some instances, the electrode layer is a metal film, such as a copper, silver, gold, or aluminum film or foil. Metal-coated polymer films may also be suitable for use as an electrode layer. The resistance of the electrode layer may be at 500 ohm-m or less, e.g., 100 ohm-m or less, e.g., 1 ohm-m or less, e.g., 0.1 ohm-m or less, e.g., 0.01 ohm-m or less. (For comparison the electrophoretic medium layer typically has a resistance of approximately 10.sup.7 to 10.sup.8 ohm-m, and the piezoelectric material has a resistance of 10.sup.11 to 10.sup.14 ohm-m.)

[0133] A piezo-electrophoretic film or a piezo-electrophoretic display will often include at least one adhesive layer, formed from a polymer such as an acrylic or a polyurethane, polyurethanes, polyureas, polycarbonates, polyamides, polyesters, polycaprolactones, polyvinyl alcohol, polyethers, polyvinyl acetate derivatives such as poly(ethylene-co-vinylacetate), polyvinyl fluoride, polyvinylidene fluoride, polyvinyl butyral, polyvinylpyrrolidone, poly(2-ethyl-2-oxazoline), acrylic or methacrylic copolymers, maleic anhydride copolymers, vinyl ether copolymers, styrene copolymers, diene copolymers, siloxane copolymers, cellulose derivatives, gum Arabic, alginate, lecithin, polymers derived from amino acids, and the like. The adhesives may additionally include one or more low dielectric polymers or oligomers, ionic liquids, or conductive fillers such as carbon black, metal flakes, metal whiskers, carbon nanotubes, silicon nitride nanotubes, or graphene. Adhesives including such charged and/or conducting materials are conductive adhesives. The polymers and oligomers used in the adhesive layer may have functional group(s) for chain extension or crosslinking during or after lamination. The adhesive layer may have a resistivity value of roughly 10.sup.6 ohm\*cm to 10.sup.8 ohm\*cm, preferably less than 10.sup.12 ohm\*cm.

[0134] Among the polymers and oligomers mentioned above, polyurethanes, polyureas, polycarbonates, polyesters and polyamides, especially those comprising a functional group, are particularly preferred because of their superior adhesion and optical properties and high environmental resistance. Examples for the functional groups may include, but are not limited to, —OH, —SH, —NCO, —NCS, —NHR, —NRCONHR, —NRCSNHR, vinyl or epoxide and derivatives thereof, including cyclic derivatives. The “R” in the functional groups mentioned above may be hydrogen or alkyl, aryl, alkylaryl or arylalkyl of up to 20 carbon atoms which alkyl, aryl, alkylaryl or arylalkyl may be optionally substituted or interrupted by N, S, O or a halogen. The “R” preferably is hydrogen, methyl, ethyl, phenyl, hydroxymethyl, hydroxyethyl, hydroxybutyl or the like. Functionalized polyurethanes, such as hydroxyl terminated polyester polyurethanes or polyether polyurethanes, isocyanate terminated polyester polyurethanes or polyether polyurethanes or acrylate terminated polyester polyurethanes or polyether polyurethanes are particularly preferred.

[0135] In many embodiments, a piezo-electrophoretic film or a piezo-electrophoretic display will often include a release sheet. The release may be used temporarily to facilitate processing piezo-electrophoretic film or a piezo-electrophoretic display, e.g., when embossing, filling, cutting, etc. In other embodiments the release may be used to deliver a final piezo-electrophoretic film or a piezo-electrophoretic display that will be adhered to a final product. In some instances the release will protect a functional adhesive layer that will be used to manipulate the piezo-electrophoretic film or a piezo-electrophoretic display prior to the piezo-electrophoretic film or a piezo-electrophoretic display being disposed in a final product. The release may be formed from a material selected from the group consisting of polyethylene terephthalate (PET), polycarbonate, polyethylene (PE), polypropylene (PP), paper and a laminated or cladding film thereof. The release may also be metalized to facilitate quality control measurements and/or to control static electricity during handling, shipping, and downstream incorporation into products. In some embodiments, a silicone release coating may be applied onto the release to improve the release properties.

[0136] While not shown in FIGS. 5A-6B, 8A-10C, and 12A-13B, a piezo-electrophoretic film or a piezo-electrophoretic display may also include an additional edge seal and/or barrier material to

allow the a piezo-electrophoretic film or a piezo-electrophoretic display to maintain the desired humidity level and to prevent leakage of e.g., non-polar solvent or adhesive, and to prevent ingress of water, dirt, or gasses. The barrier materials can be any flexible material, typically a polymer with low to negligible WVTR (water vapor transmission rate). Suitable materials include polyethylene terephthalate, polyethylene naphthalate, polycarbonate, polyimides, cyclic olefins, and combinations thereof. If the piezo-electrophoretic film or a piezo-electrophoretic display will be exposed to particularly harsh conditions, a flexible glass such as WILLOW® glass (Corning, Inc.) may be used for the barrier layer. The edge seal can be a metallized foil or other barrier foil adhered over the edge of the piezo-electrophoretic film or a piezo-electrophoretic display. The edge seal may also be formed from dispensed sealants (thermal, chemical, and/or radiation cured), polyisobutylene or acrylate-based sealants, which may be cross-linked. In some embodiments, the edge seal may be a sputtered ceramic, such as alumina or indium tin oxide, or advanced ceramics such as available from Vitex Systems, Inc. (San Jose, CA).

[0137] In general, the layers of a piezo-electrophoretic film **501-504** can be arranged/laminated in the order that produces the best performance for an end application. For example, as shown in FIG. 5A, a piezo-electrophoretic film **501**, may be prepared by disposing a microcell precursor material on a release **510**, including a release adhesive **520**. The microcell precursor can then be embossed or photolithographed to create an array of microcells **530**. The microcells **530** may be thermally cured or cured with electromagnetic radiation, such as U.V. light. The microcells **530** can then be filled with electrophoretic media and sealed with a sealing layer **540**, as discussed above with respect to FIG. 4A. (It is to be understood that microcells **530** adjacent a sealing layer **540** are filled with an electrophoretic medium including charged particles in a non-polar solvent even though the electrophoretic media are not shown in the subsequent figures.) A piezoelectric layer **560** can be laminated to the sealing layer **540** using an adhesive **550**, which will typically be an optically-clear adhesive formed from one of the materials listed above. Finally, a flexible electrode **580** will be coupled to the piezo-electrophoretic film with a conductive adhesive **570**. Such a piezo-electrophoretic film **501** may be subsequently manipulated by handling release **510** until such a time as the stack, minus release **510**, is affixed to a final product. In the piezo-electrophoretic film **501** the piezoelectric layer **560** is typically poled to create areas of differential polarization before the flexible electrode **580** is coupled to the piezo-electrophoretic film. In some embodiments the flexible electrode **580** and the conductive adhesive **570** can be replaced with a thin layer of a transparent conductive oxide, such as ITO. The ITO can be sputtered directly onto the piezoelectric layer **560**.

[0138] Closely-related, but alternative stacks are shown in FIGS. 5B-5D. In FIG. 5B, a piezo-electrophoretic film **502** is created in which a piezoelectric layer **560** is prepared prior to fabrication on a separate release **510**. For example, the piezoelectric layer **560** may be a pre-stretched PVDF film that has already been poled to create a security pattern. The piezoelectric layer **560** is then coupled to a sealed microcell layer **530**, which has been coupled to a flexible electrode **580**. Notably, in piezo-electrophoretic film **502**, the openings of the microcell layer **530** face away from the piezoelectric layer **560**, which can facilitate a good bond between the microcell layer **530** and the piezoelectric layer **560**. This bond may be improved with the introduction of a primer **535** to improve adhesion of the piezoelectric layer **560** to the microcell material, typically a polymer comprising acrylates, vinyl ethers, or epoxides. The primer **535** may be a polar oligomeric or polymeric material, such as polyhydroxy functionalized polyester acrylates (e.g., BOMAR® BDE 1025 from Dymax) or alkoxylated acrylates, such as ethoxylated nonyl phenol acrylate (e.g., SR504 from Sartomer), ethoxylated trimethylolpropane triacrylate (e.g., SR9035 from Sartomer) or ethoxylated pentaerythritol tetraacrylate (e.g., SR494 from Sartomer). Examples of polar polymers suitable for use a primer **535** include solvent urethane polymers, such as Irostatic® polymers.

[0139] Of course, it is also possible to build the stack such that the openings of the microcell layer **530** face toward the piezoelectric layer **560**, as in piezo-electrophoretic film **504** illustrated in FIG.

5D. As a further alternative, shown in FIG. 5C, piezo-electrophoretic film 503 is arranged such that the openings of the microcell layer 530 face away from the piezoelectric layer 560, however the piezoelectric layer 560 is coupled directly to the flexible electrode 580.

[0140] The piezo-electrophoretic films (501, 502, 503, 504) shown in FIGS. 5A-5D can be transformed to piezo-electrophoretic displays (601, 602) with the addition of a second flexible electrode 680 in place of the release layer in FIGS. 5A-5D. Piezo-electrophoretic displays (601, 602) typically will also include a second conductive adhesive 670, however it should be noted that in some instances the conductive adhesive 670, alone, may be sufficient to provide the necessary electric field to switch the electrophoretic material. Additionally, it is possible to directly coat the bottom of the microcell layer 530 (FIG. 6A) or the sealing layer 540 (FIG. 6B) with a thin layer of a transparent conductive oxide to create a second electrode. Also, if it is not necessary to see through both the top and bottom of the piezo-electrophoretic displays (601, 602), a conductive metal foil can be used as the second flexible electrode 680. As shown in FIGS. 6A and 6B, it is typical to add a release 510 to the completed piezo-electrophoretic displays (601, 602) to improve handling, and to provide a ready-to-use adhesive to affix the piezo-electrophoretic displays (601, 602). In some embodiments, a piezo-electrophoretic display 601 can be formed by simply bonding a piezoelectric layer 560 to a commercial front plane laminate including the second flexible electrode 680 and a sealed microcell layer 530 including an electrophoretic medium. In such instances, the piezoelectric layer 560 is typically poled to create areas of differential polarization before the front plane laminate is coupled to the piezoelectric layer 560. While piezo-electrophoretic displays (601, 602) of FIGS. 6A and 6B are shown with the piezoelectric layer 560 above the sealed microcell layer 530, it is to be understood that the piezoelectric layer 560 can also be placed below the sealed microcell layer 530 to create piezo-electrophoretic displays analogous to FIGS. 5B and 5D.

#### Prototype Performance

[0141] A series of piezo-electrophoretic films of the type exemplified in FIG. 5A were created using PEDOT:PSS film as the flexible electrode 580. The piezoelectric layer 560 was varied as shown in Table 1 (composition and thickness). The piezoelectric films were sourced from TE Connectivity (Norwood, MA), Fishman (Andover, MA), or casted in-house and cured using PVDF powder from Sigma-Aldrich. Using the described poling techniques the polarization direction was altered to create patterns. The electrophoretic media included low-voltage formulations of black and white particles, or black and red particles, or red and black particles, designed to switch color states with +/-3V. As shown in Table 1, all of the variations provided suitable switching.

TABLE-US-00001 TABLE 1 Prototype piezo-electrophoretic films

| Experiment | current | Thickness       | contact | to Performance  | number              | collector       | Piezoelement                | in um | EPD |
|------------|---------|-----------------|---------|-----------------|---------------------|-----------------|-----------------------------|-------|-----|
| 1          | PEDOT   | TE              | PVDF    | 10 G            | Pre-fab             | good            | extruded, pulled, polled    | 2     |     |
| 2          | PEDOT   | TE              | PVDF    | 10 G            | Pre-fab             | good            | extruded, pulled, polled    | 3     |     |
| 3          | PEDOT   | TE              | PVDF    | 10 A            | Pre-fab             | good            | extruded, pulled, polled    | 4     |     |
| 4          | PEDOT   | TE              | PVDF    | 10 A            | Pre-fab             | good            | extruded, pulled, polled    | 5     |     |
| 5          | PEDOT   | Fishman         | casted  | 5 A             | Pre-fab             | good            | copolymer 80/20             | 6     |     |
| 6          | PEDOT   | Fishman         | casted  | 5 A             | Pre-fab             | good            | copolymer 80/20             | 7     |     |
| 7          | PEDOT   | Casted in-house | 3 A     | Poled w/E Field | good                | copolymer 80/20 | 8                           |       |     |
| 8          | PEDOT   | TE              | PVDF    | 10 G and A      | in Corona discharge | good            | extruded, pulled one Polled |       |     |

[0142] Table 1 suggests that multiple types of electrophoretic media will respond suitably to the small electric fields produced by flexing thin piezo films. In particular, it was found that a spin-coated polyvinylidene fluoride-trifluoroethylene (PVDF-TrFE) film of less than 3 μm had sufficient charge injection to cause DV electrophoretic media to switch. See Experiment number 7. Such a piezo-electrophoretic film 801 (see FIG. 8A) can be formed using the method described in FIG. 7. First, a thin film of piezoelectric material 940 is created by casting (slot-dye coating) a concentrated PVDF/DMF solution on a suitable substrate and heating to drive off the solvent, as in step 710 of FIG. 7. In step 720, the piezo film 960 is removed from the substrate. The cast piezo film 960 may be 10 μm thick or less, e.g., 5 μm thick or less, e.g., 3 μm thick or less. The piezo

film **960** may also be stretched to increase the number of beta phase domains and/or poled with suitable electric fields as discussed above. In step **730**, a release **910** is provided along with an adhesive **920** and the release **910** and adhesive **920** are subsequently laminated to the cast piezo film **960** in step **740**. The piezo film **960** is then coated with/bonded to an electrophoretic layer in step **750**. The electrophoretic layer can be a sealed microcell layer, including filled microcells **930** and a sealing layer **940**, or alternatively, the electrophoretic layer can include encapsulated electrophoretic media **990** in a polymer binder **995**, as shown in FIGS. **9A** and **9B**. Bonding the piezo film **960** to an electrophoretic layer may be facilitated with an intervening primer layer **935**, e.g., using one of the primer materials discussed above. If the electrophoretic layer is a sealed microcell layer, the microcells **930** can be disposed such that the sealing layer **940** is adjacent the piezo film **960** as in FIG. **8A**, or the microcells **930** can be disposed such that the sealing layer **940** is disposed on the side opposite the piezo film **960**, i.e., as in FIG. **8B**. As a final step, **760** an electrode layer **980** is created and bonded to/deposited on either the microcells **930** as in FIG. **8A**, or bonded to/deposited on the sealing layer **940** as in FIG. **8B**. As described above, the electrode layer **980** can include a flexible conductive material such as PEDOT:PSS or it may include a directly-deposited (e.g., sputtered or vapor deposited) transparent conductive oxide (TCO). In some embodiments, the electrode **980** may include a pre-fabricated film of ITO on a polymer substrate, such as PET. A piezo-electrophoretic film **801** including directly-deposited TCO electrode layer **980**, a thin piezo layer **960**, and a thin layer of microcells **930** (approximately 10  $\mu\text{m}$  thick) is exceedingly thin (i.e., less than 25  $\mu\text{m}$  thick excluding the release **910**), which allows the piezo-electrophoretic film **801** to be bent without failure and is not noticeable when affixed to an object such as a bank note. A corresponding piezo-electrophoretic film **901**, including microcapsules, can also be fabricated with a total thickness less than 25  $\mu\text{m}$ . Of course, alternative constructions using the thin piezo film **960** are also possible, such as locating the piezo film **960** between the electrode **980** and the electrophoretic layer, i.e., the layer of microcapsules **990**, as shown in FIG. **9B**. As an alternative, the electrode **980** in FIGS. **8A-9B** may be replaced with a conductive adhesive (not shown) or a conductive adhesive in conjunction with an additional release layer (not shown).

[0143] Similar to FIGS. **6A** and **6B**, the piezo-electrophoretic films of FIGS. **8A-9B** can include a second electrode layer to form corresponding displays (**1001**, **1002**, **1003**) as shown in FIGS. **10A-10C**. The electrode layer **980** and the second electrode layer **1080** can both comprise a flexible conductive material such as PEDOT:PSS, or the electrode layer **980** and the second electrode layer **1080** may both comprise a directly-deposited (e.g., sputtered or vapor deposited) transparent conductive oxide (TCO), or some combination thereof. Again, in the instance where both the electrode layer **980** and the second electrode layer **1080** use thin TCO films, the resulting piezo-electrophoretic displays (**1001**, **1002**, **1003**) can be made very thin, i.e., less than 25  $\mu\text{m}$  thick excluding the release **910**. In some embodiments, the electrode layer **980** is created bonded to/deposited on the microcells **930** as in FIG. **10A**. In other embodiments, the electrode layer **980** is bonded to/deposited on the sealing layer **940** as in FIG. **10B**. The assemblies of piezo-electrophoretic displays **1001** and **1002** can also be used with microcapsules **990** containing electrophoretic media held together with a binder **995**, thus creating a piezo-electrophoretic display **1003**, as shown in FIG. **10C**. As an alternative, the electrodes **980/1080** in FIGS. **10A-10C** may be replaced with conductive adhesives (not shown) or conductive adhesive in conjunction with additional release layers (not shown).

[0144] An alternative method of constructing piezo-electrophoretic films and piezo-electrophoretic displays is described with respect to the flow chart of FIG. **11**. A piezoelectric film **1260** is procured, which may be a commercial film, or a cast film as described above. The piezoelectric film **1260** is laminated to a microcell precursor material in step **1110**. The piezoelectric film **1260** may be stretched and/or poled prior to laminating. The precursor material is typically an acrylate polymer, however any suitable embossable material, such as vinyl ether polymers, or epoxide polymers film can be used. Typically, the precursor film is 30  $\mu\text{m}$  thick or less, e.g., 20  $\mu\text{m}$  thick or

less. The precursor film may be treated with a primer **1235** prior to the lamination step **1110**. Once the piezoelectric film **1260** and the microcell precursor materials have been joined, the side of the piezoelectric film **1260** opposite the microcell precursor material is coated with a transparent conductive material, e.g., selected from those described above, typically indium tin oxide. (Alternatively, depending upon the application, the side of the piezoelectric film **1260** opposite the microcell precursor material can be coated with a conductive adhesive, which may be carried by a release layer.) This coating step creates the electrode **1280**, shown in the piezo-electrophoretic film **1201** and the piezo-electrophoretic display **1202**, shown in FIGS. **12A** and **12B**, respectively. (While it is not shown in FIG. **11**, an alternative construction is to obtain a piezoelectric film **1260** that is pre-coated with transparent conductive materials and subsequently laminate the pre-coated piezoelectric film **1260** and the microcell precursor material together, including the optional use of a primer **1235**.) After a stack of electrode **1280**, piezoelectric film **1260**, and microcell precursor is created, the stack is laminated to a carrier substrate **1255** using an adhesive layer **1250**, as shown in step **1130**. The carrier substrate **1255** may be any of the materials described above for use as a release, and the adhesive **1250** may be any of the adhesives described above. In practice the carrier substrate **1255** is typically PET because PET sheets are easy to handle during the embossing step **1140**. In step **1140**, the stack comprising carrier substrate **1255**, adhesive **1250**, piezoelectric film **1260**, and microcell precursor is microembossed using the techniques described above with respect to U.S. Pat. Nos. 6,930,818, 7,052,571, 7,616,374, 8,361,356, and 8,830,561. When this procedure is completed with a thin piezoelectric film and a thin microcell precursor, the final stack thickness (not including the carrier substrate) can be 30  $\mu\text{m}$  thick or less, e.g., 20  $\mu\text{m}$  thick or less. This results in an open microcell structure that is subsequently filled with the desired electrophoretic medium and sealed with a water-soluble sealing layer **1240** at step **1150**. The sealing layer **1240** can be made conductive with the inclusion of conductive species. The sealing layer **1240** is typically light-transmissive or transparent. The open microcells may be cleaned/activated with a vapor plasma treatment **1145** before the microcells are filled with the desired electrophoretic medium. Finally, a release sheet **1210** is coupled to the sealing layer **1240** with an adhesive **1220** in step **1160**, to make transportation of the piezo-electrophoretic film **1201** easier and to facilitate placement of the electrophoretic film **1201** on the final product. The adhesive **1220** may also be conductive. The resulting structure is shown in FIG. **12A**. Importantly, it is possible to complete the steps of FIG. **11** without poling the piezoelectric film **1260**, thereby allowing the final customer to pattern the piezo-electrophoretic film **1201** at the location of final assembly, e.g., by creating areas of differential polarity with a corona discharge as described above.

[0145] As shown in FIG. **12B**, the method of FIG. **11** can be extended to creating piezo-electrophoretic displays **1202** with the addition of a second electrode **1285**. The second electrode **1285** may also include a transparent conductive material that is added directly to the sealing layer **1240** in lieu of the release **1210** and adhesive **1220**. However, in other embodiments the release **1210** will be removed and a second electrode **1285** will be laminated to the sealing layer **1240** with the adhesive **1220**. If the piezo-electrophoretic display **1202** does not require the electrophoretic medium to be visible from both sides, the second electrode **1285** can be a metal film. Alternatively, the second electrode **1285** may be a conductive polymer such as PEDOT:PSS. In some other embodiments, the adhesive **1220** may be a conductive adhesive that provides sufficient conductivity to act as the second electrode **1285**.

[0146] Finally, it is to be appreciated that an electrode need not be coupled to the piezoelectric film **1260** prior to embossing the stack comprising the piezoelectric film **1260** and the microcell precursor material. Rather a stack including release **1210**, adhesive **1220**, piezoelectric film **1260**, and microcell precursor can be prepared and the microcell precursor subsequently embossed, filled, and sealed as described above. Alternatively, a stack including release **1210**, adhesive **1220**, electrode **1285**, piezoelectric film **1260**, and microcell precursor can also be prepared and the microcell precursor subsequently embossed, filled, and sealed as described above, as shown in FIG.

**13B.** The resulting piezo-electrophoretic film **1301** and piezo-electrophoretic display **1302** are shown in FIGS. **13A** and **13B**, respectively. The piezo-electrophoretic film **1301** and piezo-electrophoretic display **1302** may be favored for applications where it is desired to have the piezoelectric film **1260** as close as possible to the attachment surface on the final product, i.e., if the piezo-electrophoretic film **1301** is used as a strain sensor and it is important the intervening electrophoretic media layers do not dissipate the forces from the surface.

[0147] FIG. **14** is a flow chart detailing the steps of a method **1400** for creating high contrast piezo-electrophoretic films and piezo-electrophoretic displays. Method **1400** has been optimized for creating a piezo-electrophoretic film using a roll-to-roll manufacturing process.

[0148] Method **1400** is described with reference to FIGS. **15A-15F**. Method **1400** begins at step **1410**, at which a first electrode, electrode **1550**, is formed on a first substrate, substrate **1555**, by depositing an electrically-conductive material, e.g., selected from those described above, onto a substrate. For example, a thin layer of electrically-conductive material can be directly-deposited (e.g., sputtered, vapor deposited) onto a suitable substrate, such as a polymer substrate (e.g., PET). In some embodiments, substrate **1555** may be any of the materials described above for use as a release sheet that is used temporarily to facilitate fabrication of the piezo-electrophoretic film. In some embodiments, the electrically-conductive material used to form electrode **1550** is an adhesive or tie layer comprising a transparent conductive material (e.g., a first electrically-conductive adhesive) including a conductive metal oxide, conductive polymer, and/or other suitable conductive agent that is coated onto substrate **1555**. In some embodiments, to form electrode **1550**, an adhesive or tie layer is deposited on the substrate **1555**, and a conductive polymer such as PEDOT is deposited over the tie layer. In some instances, the electrode **1550** is a metal film, such as a copper, silver, gold, or aluminum film or foil that is bonded to a flexible, light-transmissive substrate such as a polymeric film. In some embodiments, the electrode **1550** is less than 5  $\mu\text{m}$  in thickness. In some embodiments, the electrode **1550** is between 1 and 3  $\mu\text{m}$  in thickness.

[0149] Next, at step **1420**, a piezoelectric layer **1560** is formed on the electrode **1550** by depositing a piezoelectric material onto the electrically-conductive material (e.g., electrode **1550**). For example, the electrode **1550** can be coated with a thin film of piezoelectric material, e.g., selected from those described above, such as PVDF, using a spin-coating process or casting (e.g., slot-dye coating) as described above. In some embodiments, a film deposition process such as printing, spraying, or gravure coating is used to form the piezoelectric layer **1560** on the electrode **1550**. In some embodiments, the resulting piezoelectric layer **1560** is less than 10  $\mu\text{m}$  in thickness. In some embodiments, the resulting piezoelectric layer **1560** is about 3  $\mu\text{m}$  in thickness.

[0150] As discussed above, the properties of PEDOT can make it a desirable material for the electrode onto which the layer of piezoelectric material is formed. For example, PEDOT is conductive, optically transparent, and exhibits greater flexibility (e.g., has a smaller bend radius) than many other conductive polymers. Further, polymerization of hydrophobic PEDOT in the presence of the polyanion PSS renders it water dispersible, thereby enabling PEDOT to be deposited by roll-to-roll processes that are commonly used to manufacture solution-processable organic electronics.

[0151] However, PEDOT:PSS can potentially become unstable under an electrical potential leading to oxidation/reduction, and can adhere poorly to secondary layers that may be applied onto the PEDOT:PSS, such as PVDF. For example, the hygroscopic nature of PSS can lead to moisture absorption by the electrode layer which can disrupt the film morphology. In addition, the presence of water soluble PSS can make it difficult to subsequently deposit a water-based secondary material (e.g., an aqueous solution of piezoelectric material, a water-based microcell primer, etc.) onto an electrode film formed from PEDOT. The water-based secondary layer can wet and disrupt the PEDOT electrode film, especially when the secondary material is applied using mechanical force (e.g., via Mayer rods). As a result, conventional processes are limited to using non-aqueous based solutions when applying a secondary layer of material onto PEDOT:PSS-based electrode films.



[0152] The inventive subject matter described herein includes methods for overcoming the above-identified deficiencies of conventional processes that incorporate electrodes formed from PEDOT. FIG. 20 shows a flow chart detailing the steps of a method 2000 for integrating a thin film of piezoelectric material with an electrode. The steps of method 2000 can be performed in lieu of steps 1410 and 1420 of method 1400 to integrate the film of piezoelectric material with an electrode formed from a material such as PEDOT.

[0153] Method 2000 includes providing (2010) a first substrate (e.g., substrate 1555). For example, a suitable substrate, such as a polymer substrate (e.g., PET) can be provided as the first substrate. In some embodiments, the first substrate is any of the materials described above for use as a release sheet that is used temporarily to facilitate fabrication of the piezo-electrophoretic film. In some embodiments, the first substrate is a release sheet having a thickness of approximately 1 mil or 25  $\mu\text{m}$ .

[0154] Next, method 2000 includes depositing (2020) an electrically-conductive material onto the first substrate. For example, an aqueous solution including PEDOT can be deposited onto the first substrate using a roll-to-roll process that incorporates a Mayer bar/rod (also commonly referred to as a Meyer bar/rod) or other coating rod, a doctor blade, a slot die, or gravure/microgravure coating, or a combination thereof (e.g., a doctor blade used in conjunction with a gravure/microgravure roll).

[0155] Method 2000 also includes forming (2030) a first electrode from the electrically-conductive material. For example, the solution including PEDOT that is deposited onto the first substrate can be dried to form an electrically-conductive film that serves as a first electrode (e.g., electrode 1550) for the piezo-electrophoretic display.

[0156] In some embodiments, the solution including PEDOT includes a crosslinker that causes crosslinks to form between the PEDOT polymer chains as the solution dries. The resulting film of crosslinked PEDOT can increase the moisture stability of the first electrode. In some embodiments, the crosslinker is a molecule having long flexible chains, and the film produced by crosslinking the PEDOT using the crosslinker improves the flexibility of the first electrode.

[0157] Next, method 2000 includes depositing (2040) an adhesive material onto the first electrode. For example, a tie layer (also referred to as a hard coat layer) of material can be deposited onto the first electrode using one of the roll-to-roll processes discussed above in connection with step 2020. The tie layer/hard coat layer is made from a conductive material that can be applied thinly and also has adhesive properties while being transparent. In some embodiments, the tie layer or hard coat layer includes a mixture of acrylates (e.g., acrylic monomers), polyurethane (e.g., thermoplastic polyurethane or “TPU”), and a solvent based on methyl ethyl ketone (“MEK”).

[0158] Method 2000 also includes forming (2050) a tie layer on the first electrode. For example, the adhesive tie layer or hard coat material that is deposited onto the first electrode can be thermally cured, or cured with electromagnetic radiation, such as UV light. Although the tie layer is not shown in FIGS. 15A-15F, in view of the disclosure herein, a person of ordinary skill in the art would understand that the tie layer is located between electrode 1550 and the piezoelectric layer 1560 for piezo-electrophoretic films and displays formed using the steps of method 2000.

[0159] In some embodiments, the tie layer or hard coat material is made from polyurethane and acrylic monomers crosslinked via photoinitiators and UV light. In some embodiments, the cured tie layer has a thickness less than 1  $\mu\text{m}$ . In some embodiments, the cured tie layer has a thickness between 0.25  $\mu\text{m}$  and up to 3  $\mu\text{m}$ .

[0160] Finally, method 2000 includes forming (2060) a layer of piezoelectric material (e.g., piezoelectric layer 1560) on the tie layer. For example, the tie layer can be coated with a thin film of piezoelectric material, e.g., one of the materials selected from those described above, such as PVDF, using a spin-coating process or casting (e.g., slot-dye coating) as described above. In some embodiments, a film deposition process such as printing, spraying, or gravure coating is used to form the piezoelectric layer 1560 on the tie layer. One of ordinary skill in the art will appreciate

that other processes can be used to deposit the layer of piezoelectric material onto the tie layer, such as those processes described above in connection with step **2020** of method **2000**.

[0161] In some embodiments, the resulting piezoelectric layer **1560** is less than 10  $\mu\text{m}$  in thickness. In some embodiments, the resulting piezoelectric layer **1560** is about 3  $\mu\text{m}$  in thickness.

[0162] Method **2000** provides several advantages over conventional processes for integrating a thin film of piezoelectric material with an electrode formed from PEDOT. For example, formation of the adhesive tie layer onto the PEDOT-based electrode improves the mechanical stability of the PEDOT. This improved durability enables the PEDOT to better withstand subsequent processing stages that would otherwise cause cracks or breaks in the PEDOT using conventional processes. Further, crosslinking the PEDOT to itself improves the moisture stability and has better structural properties than electrodes formed using conventional processes.

[0163] The tie layer also improves the adhesion of the electrode to the layer of piezoelectric material, and enables the electrode layer to withstand more flexing without cracking or breaking in adverse environments.

[0164] These features are particularly beneficial for applications in which piezo-electrophoretic films and displays are bonded to a thin target object such as a bank note or currency bill as a verification code. For example, using method **2000**, aqueous solutions of materials and common roll-to-roll processes can be used throughout the manufacturing process, allowing the finished piezo-electrophoretic film or display to be significantly thinner than those made using conventional processes. Using this process, layers such as the piezoelectric layer can be made much thinner (e.g., about 3  $\mu\text{m}$  in thickness) as compared to the same layer formed using conventional methods.

[0165] In contrast, conventional piezo-electrophoretic films and displays typically use an extruded film of PVDF that can be 10  $\mu\text{m}$  or more in thickness. The extruded sheet of piezoelectric material must then be pulled/stretched to a size of three to four times its original size to make it thin enough to integrate into a piezo-electrophoretic film or display for use on a bank note, and the stretching process is required to be tightly controlled to avoid tears or holes in the piezoelectric material.

[0166] Accordingly, method **2000** provides several advantages over conventional processes for integrating a thin film of piezoelectric material with an electrode formed from PEDOT.

[0167] Returning to method **1400** shown in FIG. **14**, after the piezoelectric layer **1560** is formed on the electrode **1550**, a mask **1540** is applied to the piezoelectric material of piezoelectric layer **1560**, as shown in step **1430**. As in the embodiments described above, the mask **1540** can be used to shield or insulate a first plurality of areas of the piezoelectric layer **1560** from the high voltage corona discharge **1533** that is used for poling the piezoelectric layer **1560**. This allows the piezoelectric layer **1560** to be patterned with images, text, and other information (e.g., machine-readable codes). The mask **1540** can be fabricated from a material having a dielectric strength sufficient to withstand at least a localized 5 kV field. In some embodiments, the mask **1540** is formed from a disposable material that can be applied in a roll-to-roll process. For example, the mask **1540** can be formed from paper such as an electrical insulation paper having a pressure-sensitive adhesive applied to one surface for bonding to the piezoelectric layer **1560**. Alternatively, the mask **1540** can be a reusable fixture made from charge-absorbing or charge-blocking materials that is integrated into a roll-to-roll translation stage similar to a screen printing process or a rotational roller.

[0168] FIG. **15A** is a schematic cross section **1501** of a piezo-electrophoretic film at step **1440** of the method shown in FIG. **14**. As shown in FIG. **15A**, the mask **1540** includes masking portions **1542** that shield or insulate corresponding areas of the piezoelectric layer **1560** from the high voltage corona discharge **1533**, and non-masking portions **1544** that allow corresponding areas of the piezoelectric layer **1560** to be poled in the areas where the mask **1540** is not covering the piezoelectric layer **1560**. The piezoelectric material is then polarized at step **1440**. For example, a high voltage corona discharge (e.g., high voltage corona discharge **1533**) or other suitable electric field discussed above can be used to polarize the unmasked portions of piezoelectric layer **1560** in a

first direction while leaving the masked portions unpolarized. If a disposable material such as paper was used for the mask, the mask is then removed from piezoelectric layer **1560**.

[0169] In some embodiments, the process of patterning of the piezoelectric material can start from a sheet or a roll of poled PVDF film. The film can be patterned by laser cutting, laser ablation (e.g., laser photoablation), die cutting, or other cutting methods, and then laminated to an electrode layer, or to a microcapsule- or microcell-based front plane laminate or FPL as described above.

[0170] FIG. **15B** is a schematic cross section **1502** of a piezo-electrophoretic film after completion of step **1440** of the method shown in FIG. **14**. As shown in FIG. **15B**, the piezoelectric layer **1560** now includes unpolarized portions **1562** corresponding to the locations of the masking portions **1542** of the mask **1540**, and polarized portions **1564** corresponding to the locations of the non-masking portions **1544** of the mask **1540**.

[0171] The piezoelectric material is then bonded to an electrophoretic material in step **1450**. For example, the piezoelectric layer **1560** can be coated with an electrophoretic medium layer including a plurality of microcapsules containing a non-polar fluid and charged pigment particles (not shown in FIG. **15B**). Alternatively, an electrophoretic medium layer comprising microcells **1530** can be formed on the piezoelectric layer **1560** using a similar process to the one shown in the flow chart of FIG. **11**. For example, an embossable microcell precursor material can be laminated to the piezoelectric layer **1560**. Prior to lamination, the precursor material may be treated or coated with a microcell primer e.g., using one of the primer materials discussed above. In some examples, the microcell primer includes a thermoplastic or thermoset material or a precursor thereof, such as polyurethane, a multifunctional acrylate or methacrylate such as lauryl methacrylate, a vinylbenzene, a vinyl ether, an epoxide or an oligomers or polymer thereof.

[0172] The microcell precursor is microembossed using the techniques described above resulting in an open microcell structure that is subsequently filled with the desired electrophoretic medium and sealed with a sealing layer **1535** as shown in FIG. **15C**, which is schematic cross section **1503** of a piezo-electrophoretic film after completion of step **1450** of the method shown in FIG. **14**. The open microcells may optionally be cleaned/activated with a vapor plasma treatment before the microcells **1530** are filled with the desired electrophoretic medium.

[0173] In some embodiments, the layer of microcells **1530** is between 8 and 20  $\mu\text{m}$  in thickness, and the sealing layer **1535** is between 3 and 10  $\mu\text{m}$  in thickness. In some embodiments, the layer of microcells **1530** is about 10  $\mu\text{m}$  in thickness, and the sealing layer **1535** is about 5  $\mu\text{m}$  in thickness.

[0174] In an alternate embodiment, instead of forming the microcells on the piezoelectric layer **1560**, a piezoelectric layer **1560** such as the one shown in FIG. **15B** is bonded or laminated to a microcapsule- or microcell-based front plane laminate or FPL as described above.

[0175] At step **1460**, a second electrode, electrode **1585**, is formed on a second substrate, substrate **1586**. The electrode **1585** can be formed on the substrate **1586** using one of the processes described above with respect to the electrode **1550** and the substrate **1555**. As one example, the substrate **1586** may be a release sheet that is used temporarily to facilitate fabrication of the piezo-electrophoretic film, and electrode **1585** can be formed from an adhesive or tie layer comprising a transparent conductive material that is deposited onto substrate **1586**. In some embodiments, a process similar to method **2000** is used to form electrode **1585** and adhesive tie layer on substrate **1586**. In some embodiments, the electrode **1585** is less than 5  $\mu\text{m}$  in thickness. In some embodiments, the electrode **1585** is between 1 and 3  $\mu\text{m}$  in thickness.

[0176] The electrically-conductive material deposited on the second substrate is then bonded with the electrophoretic material at step **1470**. For example, the substrate **1586** and the electrode **1585** can be laminated to the sealing layer **1535** of the microcells **1530** to form the structure shown in FIG. **15D**, which is schematic cross section **1504** of a piezo-electrophoretic film after completion of step **1470** of the method shown in FIG. **14**. The addition of the electrode **1585** (and substrate **1586**) to the piezo-electrophoretic film forms a piezo-electrophoretic display that can be bonded to a target object. For example, the structure shown in FIG. **15D** can be a piezo-electrophoretic

display sandwiched between two release sheets (e.g., substrates **1555** and **1586**).

[0177] In an alternative embodiment not shown in FIG. **14**, steps **1460-1470** are replaced by the following process. The sealing layer **1535** of the microcells **1530** can be coated with an electrically-conductive material such as PEDOT or one of the materials discussed above to form the electrode **1585**. Subsequently, the electrode **1585** can be coated with an adhesive material (e.g., a heat seal adhesive (“HSA”), or one of the materials discussed above) for bonding the piezo-electrophoretic display to a target object in step **1480** described below.

[0178] Returning to FIG. **14**, at step **1480**, the piezo-electrophoretic display can be bonded with a target object. For example, a piezo-electrophoretic display such as the one shown in FIG. **15D** can be processed and affixed to a target object **1588** such as paper or a bank note, as shown in FIG. **15E**, which is a cross section **1505** of a piezo-electrophoretic film bonded with a target object after completion of step **1480** of the method shown in FIG. **14**.

[0179] In some embodiments, the substrate **1586** is a release sheet that is peeled or removed from the electrode **1585**, and electrode **1585** is bonded to the surface of the target object **1588** using a hot stamping process. For example, heat and pressure can be applied to the piezo-electrophoretic display and/or the target object **1588** in a roll-to-roll hot stamping process that presses the piezo-electrophoretic display against the target object **1588**. An adhesive (not shown) left behind on the electrode **1585** after peeling off the substrate **1586** is activated by the heat and pressure and bonds the piezo-electrophoretic display to the target object **1588**.

[0180] In some embodiments, the electrode **1585** is formed from an adhesive or tie layer that is activated during the bonding process to bond the piezo-electrophoretic display to the target object **1588**. In some embodiments, a roll-to-roll lamination process is used to bond the piezo-electrophoretic display to the target object **1588**.

[0181] In some embodiments, the substrate **1555** and the substrate **1586** are release sheets and the force required to remove each release sheet is individually tuned to ensure the substrate **1586** is removed prior to the substrate **1555** during the bonding process. For example, the adhesives used to temporarily adhere the substrate **1555** and the substrate **1586** to the electrode **1550** the electrode **1585**, respectively, can be formulated or chosen such that the force required to peel the substrate **1586** from the piezo-electrophoretic display is less than the force required to peel the substrate **1555** from the piezo-electrophoretic display.

[0182] FIG. **15F** is a cross section **1506** of a piezo-electrophoretic film bonded with a target object and coated with a protective coating after completion of step **1480** of the method shown in FIG. **14**. As exemplified in FIG. **15F**, subsequent to bonding the piezo-electrophoretic display to the target object **1588**, the substrate **1555** can be peeled away from the electrode **1550**, and a protective coating **1589** can be applied to the piezo-electrophoretic display and the surface of the target object **1588** to which the piezo-electrophoretic display is bonded. In some embodiments, the protective coating **1589** is a layer of lacquer that is applied to the piezo-electrophoretic display and the surface of the target object **1588** using a printing process. Suitable materials for the protective coating **1589** can include UV-curable polyester acrylates, polyurethane acrylates, UV-curable epoxides, and thermal curable epoxides, or any material sufficient to seal the piezo-electrophoretic display and the target object **1588** to repel dirt and avoid excessive absorption of moisture.

[0183] Accordingly, the process described in connection with FIG. **14** can be used to produce a piezo-electrophoretic display having a thickness far less than conventional displays and can therefore be bonded to a target object such as a bank note or currency bill without substantially increasing the overall thickness or being substantially noticeable. In some embodiments, the total thickness of the piezo-electrophoretic display can be between 50  $\mu\text{m}$  and 100  $\mu\text{m}$ . In some embodiments, the total thickness of the piezo-electrophoretic display can be between 25  $\mu\text{m}$  and 50  $\mu\text{m}$ . In some embodiments, the total thickness of the piezo-electrophoretic display can be less than 25  $\mu\text{m}$ .

[0184] FIG. **16** shows an enlarged view **1600** of a partial cross section of a piezo-electrophoretic

display **1601** fabricated in accordance with the subject matter disclosed herein. For the purpose of describing the operation of the display, only a subset of the layers of the piezo-electrophoretic display **1601** are shown: the electrode **1550**, the piezoelectric layer **1560** including unpolarized portions **1562** and polarized portions **1564**, the microcells **1530**, the sealing layer **1535**, and the second electrode **1585**. In the enlarged view **1600**, the microcells **1530** and the sealing layer **1535** are represented by electrophoretic layer **1631**.

[0185] The enlarged view **1600** includes one of the unpolarized portions **1562** and one of the polarized portions **1564**. A first portion **1632** of the electrophoretic layer **1631** is positioned above the unpolarized portion **1562**, and a second portion **1634** is positioned above polarized portion **1564**, as delineated by dashed line **1602**. The first portion **1632** and second portion **1634** each have an electrical resistance that is based on the volume of the electrophoretic layer **1631** they encompass. Further, the unpolarized portion **1562** also has an electrical resistance that is based on the volume of the piezoelectric layer **1560** it encompasses. As denoted by the “+” and “-” symbols, a voltage has been generated in polarized portion **1564** of the piezoelectric layer **1560**, for example, in response to bending or mechanical stress to the piezoelectric material.

[0186] FIG. **17** illustrates an exemplary equivalent circuit **1700** of the enlarged cross section shown in FIG. **16**. The three nodes or points, ‘A’ at polarized portion **1564**, ‘B’ at the electrode **1585**, and ‘C’ at the electrode **1550**, shown in FIG. **16** correspond to the same points shown in equivalent circuit **1700** of FIG. **17**. Resistance R.sub.1 corresponds to the sum of the electrical resistances of the first portion **1632** of the electrophoretic layer **1631** and the unpolarized portion **1562** of the piezoelectric layer **1560**. Resistance R.sub.2 corresponds to the electrical resistances of the second portion **1634** of the electrophoretic layer **1631**.

[0187] The polarized portion **1564** of the piezoelectric layer **1560** is represented as a battery, and voltage V.sub.PZ is the voltage generated by the polarized portion **1564** across points A and C. Resistance R.sub.1 and Resistance R.sub.2 are represented in series because the presence of a voltage source beneath a portion of the electrophoretic layer **1631** effectively divides the layer into separate sections (as delineated by dashed line **1602**) having different electrical properties. For example, when voltage V.sub.PZ has been generated, the voltage potential at point A is higher than at points B or C. Using a conventional current flow paradigm, current **1701** flows from point A through resistance R.sub.1 to point B, and from point B through resistance R.sub.2 to point C. It follows that the voltage generated across resistance R.sub.1 is opposite in polarity to the voltage generated across resistance R.sub.2. In effect, two opposite voltages are created in series across separate portions of the electrophoretic layer.

[0188] Accordingly, fabricating the piezo-electrophoretic display as described with respect to method **1400** provides advantages over conventional piezo-electrophoretic displays. For example, selectively poling the piezoelectric layer advantageously provides an improved means to drive the oppositely-charged pigment particles in the electrophoretic medium in opposite directions from one another in the absence of a matrix of individually-addressable pixel electrodes. Therefore, piezo-electrophoretic displays produced according to the method **1400** can be made thin enough for use in applications requiring them to be durable and substantially unnoticeable when incorporated into thin, low-profile final products such as paper or bank notes while still providing a high contrast ratio between the polarized and unpolarized regions due to the effects described above. Furthermore, in this embodiment, only a single poling operation is required for the piezoelectric layer.

[0189] FIG. **18** is a flow chart detailing the steps of a method **1800** for creating high contrast piezo-electrophoretic films and piezo-electrophoretic displays. Method **1800** has been optimized for creating a piezo-electrophoretic film using a roll-to-roll manufacturing process.

[0190] Method **1800** is described with reference to FIGS. **19A-19G**. To facilitate understanding, where possible the same or similar reference numerals and names have been used in FIGS. **19A-19G** to refer to elements that correspond to, or are similar in function to, the elements shown in

FIGS. 15A-15F. However, one of ordinary skill in the art will appreciate based on the description herein that the elements of each embodiment need not necessarily be identical in composition and structure to one another, and that elements disclosed with reference to one embodiment may be beneficially utilized for other embodiments without specific recitation.

[0191] Method **1800** begins at step **1810**, at which a piezoelectric layer **1960** is formed on a temporary substrate **1965** by depositing a piezoelectric material onto the temporary substrate **1965**. For example, the temporary substrate **1965** can be coated with a thin film of piezoelectric material, e.g., selected from those described above, such as PVDF, using a spin-coating process or casting (e.g., slot-dye coating) as described above. The temporary substrate **1965** may be a release sheet that is used temporarily to facilitate fabrication of the piezo-electrophoretic film. In some embodiments, the temporary substrate **1965** is a release sheet that may be formed from a material selected from the group consisting of polyethylene terephthalate (PET), polycarbonate, polyethylene (PE), polypropylene (PP), paper and a laminated or cladding film thereof. A silicone release coating may also be applied onto the temporary substrate **1965** to improve the release properties. FIG. **19A** is a schematic cross section **1901** of a piezo-electrophoretic film at step **1810** of method **1800**.

[0192] In some embodiments, a film deposition process such as printing, spraying, or gravure coating is used to form the piezoelectric layer **1960** on the temporary substrate **1965**. In some embodiments, the resulting piezoelectric layer **1960** is less than 10  $\mu\text{m}$  in thickness. In some embodiments, the resulting piezoelectric layer **1960** is about 3  $\mu\text{m}$  in thickness.

[0193] At step **1820**, the piezoelectric material is bonded with an electrically-conductive material that is coated on a first substrate. For example, the temporary substrate **1965** can be peeled from the piezoelectric layer **1960**, and the piezoelectric layer **1960** can be laminated onto a first electrode, electrode **1950**, that is formed on a first substrate, substrate **1955**. The electrode **1950** can be formed on the substrate **1955** using one of the processes described above with respect to the electrode **1550** and the substrate **1555**. As one example, the substrate **1955** may be a release sheet that is used temporarily to facilitate fabrication of the piezo-electrophoretic film, and electrode **1950** can be formed from an adhesive or tie layer comprising a transparent conductive material that is deposited onto substrate **1955**. In some embodiments, the electrode **1950** is less than 5  $\mu\text{m}$  in thickness. In some embodiments, the electrode **1950** is between 1 and 3  $\mu\text{m}$  in thickness.

[0194] In an alternative embodiment not shown in FIG. **18**, steps **1810** and **1820** are replaced by the following process. In lieu of using the temporary substrate **1965**, a substrate **1955** can be coated with an electrically-conductive material (e.g., one of the materials discussed above) to form the electrode **1950**. In some embodiments, to form electrode **1950**, an adhesive or tie layer is deposited on the substrate **1955**, and a conductive polymer such as PEDOT is deposited over the tie layer. In some embodiments, a process similar to method **2000** is used to form electrode **1950** and adhesive tie layer on substrate **1955**. Subsequent to forming the electrode **1950**, the piezoelectric material is bonded with the electrode **1950** to form the piezoelectric layer **1960**. For example, the piezoelectric material can be coated onto electrode **1950** as described above.

[0195] Returning to FIG. **18**, after the piezoelectric layer **1960** is bonded to the electrode **1950**, a mask **1940** is applied to the piezoelectric layer **1960**, as shown in step **1830**. As in the embodiments described above, the mask **1940** can be used to shield or insulate areas of the piezoelectric layer **1960** from the high voltage corona discharge **1933** that is used for poling the piezoelectric layer **1960**. This allows the piezoelectric layer **1960** to be patterned with images, text, and other information (e.g., machine-readable codes). The mask **1940** can be fabricated from a material having a dielectric strength sufficient to withstand at least a localized 5 kV field. In some embodiments, the mask **1940** is formed from a disposable material that can be applied in a roll-to-roll process. For example, the mask **1940** can be formed from paper such as an electrical insulation paper having a pressure-sensitive adhesive applied to one surface for bonding to the piezoelectric layer **1960**. Alternatively, the mask **1940** can be a reusable fixture made from charge-absorbing or

charge-blocking materials that is integrated into a roll-to-roll translation stage similar to a screen printing process or a rotational roller.

[0196] FIG. **19B** is a schematic cross section **1902** of a piezo-electrophoretic film at step **1830** of the method shown in FIG. **18**. As shown in FIG. **19B**, the mask **1940** includes masking portions **1942** that shield or insulate corresponding areas of the piezoelectric layer **1960** from the high voltage corona discharge **1933**, and non-masking portions **1944** that allow corresponding areas of the piezoelectric layer **1960** to be poled in the areas where the mask **1940** is not covering the piezoelectric layer **1960**.

[0197] The piezoelectric material is then polarized or poled at step **1840**. For example, a high voltage corona discharge (e.g., high voltage corona discharge **1933**) or other suitable electric field discussed above can be used to polarize the unmasked portions of piezoelectric layer **1960** in a first direction while leaving the masked portions unpolarized. If a disposable material such as paper was used for the mask, the mask is then removed from piezoelectric layer **1960**.

[0198] FIG. **19C** is a schematic cross section **1903** of a piezo-electrophoretic film after completion of step **1840** of the method shown in FIG. **18**. As shown in FIG. **19C**, the piezoelectric layer **1960** now includes unpolarized portions **1962** corresponding to the locations of the masking portions **1942** of the mask **1940**, and polarized portions **1964** corresponding to the locations of the non-masking portions **1944** of the mask **1940**.

[0199] At step **1850**, a second electrode, electrode **1985**, is formed on a second substrate, substrate **1986**, by depositing an electrically-conductive material, e.g., selected from those described above, onto a substrate. The electrode **1985** can be formed on the substrate **1986** using one of the processes described above with respect to the electrode **1550** and the substrate **1555**. In one example, to form electrode **1985**, an adhesive or tie layer is deposited on the substrate **1986**, and a conductive polymer such as PEDOT is deposited over the tie layer. In some embodiments, a process similar to method **2000** is used to form electrode **1985** and adhesive tie layer on substrate **1986**. In some embodiments, the electrode **1985** is less than 5  $\mu\text{m}$  in thickness. In some embodiments, the electrode **1985** is between 1 and 3  $\mu\text{m}$  in thickness.

[0200] The electrophoretic material is then bonded to the electrically-conductive material at step **1860**. For example, the electrode **1985** can be coated with an electrophoretic medium layer including a plurality of microcapsules containing a non-polar fluid and charged pigment particles (not shown in FIG. **19D**). Alternatively, an electrophoretic medium layer comprising microcells **1930** can be formed on the electrode **1985** using a similar process to the one shown in the flow chart of FIG. **11**. For example, an embossable microcell precursor material can be laminated to the electrode **1985**. Prior to lamination, the precursor material may be treated with a primer e.g., using one of the primer materials discussed above. The microcell precursor is microembossed using the techniques described above resulting in an open microcell structure that is subsequently filled with the desired electrophoretic medium and sealed with a sealing layer **1935** as shown in FIG. **19D**, which is schematic cross section **1904** of a piezo-electrophoretic film after completion of steps **1850** and **1860** of the method shown in FIG. **18**. The open microcells may optionally be cleaned/activated with a vapor plasma treatment before the microcells **1930** are filled with the desired electrophoretic medium.

[0201] In some embodiments, the layer of microcells **1930** is between 8 and 20  $\mu\text{m}$  in thickness, and the sealing layer **1935** is between 3 and 10  $\mu\text{m}$  in thickness. In some embodiments, the layer of microcells **1930** is about 10  $\mu\text{m}$  in thickness, and the sealing layer **1535** is about 5  $\mu\text{m}$  in thickness.

[0202] In an alternate embodiment, instead of forming the microcells on the electrode **1985**, an electrode **1985** such as the one shown in FIG. **19D** is bonded or laminated to a microcapsule- or microcell-based front plane laminate or "FPL" as described above.

[0203] The piezoelectric material deposited on the first electrode is then bonded with the electrophoretic material at step **1870**. For example, piezoelectric layer **1960** can be laminated to the sealing layer **1935** of the microcells **1930** to form the structure shown in FIG. **19E**, which is

schematic cross section **1905** of a piezo-electrophoretic film after completion of step **1870** of the method shown in FIG. **18**. In some embodiments, the piezoelectric layer **1960** can be coupled to the sealing layer **1935** with an adhesive layer (not shown in FIG. **19E**).

[0204] The addition of the piezoelectric layer **1960** with the electrode **1550** (and substrate **1555**) to the piezo-electrophoretic film forms a piezo-electrophoretic display that can be bonded to a target object. For example, the structure shown in FIG. **19E** can be a piezo-electrophoretic display sandwiched between two release sheets (e.g., substrates **1955** and **1986**).

[0205] At step **1880**, the piezo-electrophoretic display can be bonded with a target object. For example, a piezo-electrophoretic display such as the one shown in FIG. **19E** can be processed and affixed to a target object **1988** such as paper or a bank note, as shown in FIG. **19F**, which is a cross section **1906** of a piezo-electrophoretic film bonded with a target object according to the method shown in FIG. **18**.

[0206] In some embodiments, the substrate **1986** is a release sheet that is peeled or removed from the electrode **1985**, and electrode **1985** is bonded to the surface of the target object **1988** using a hot stamping process as described above.

[0207] In some embodiments, the electrode **1985** is formed from an adhesive or tie layer that is activated during the bonding process to bond the piezo-electrophoretic display to the target object **1988**. In some embodiments, a roll-to-roll lamination process is used to bond the piezo-electrophoretic display to the target object **1988**.

[0208] In some embodiments, the substrate **1955** and the substrate **1986** are release sheets and the force required to remove each release sheet is individually tuned to ensure the substrate **1986** is removed prior to the substrate **1955** during the bonding process. For example, the adhesives used to temporarily adhere the substrate **1955** and the substrate **1986** to the electrode **1950** the electrode **1985**, respectively, can be formulated or chosen such that the force required to peel the substrate **1986** from the piezo-electrophoretic display is less than the force required to peel the substrate **1955** from the piezo-electrophoretic display.

[0209] FIG. **19G** is a cross section **1907** of a piezo-electrophoretic film bonded with a target object and coated with a protective coating after completion of step **1880** of the method shown in FIG. **18**. As exemplified in FIG. **19G**, subsequent to bonding the piezo-electrophoretic display to the target object **1988**, the substrate **1955** can be peeled away from the electrode **1950**, and a protective coating **1989** can be applied to the piezo-electrophoretic display and the surface of the target object **1988** to which the piezo-electrophoretic display is bonded, as described above.

[0210] Accordingly, the process described in connection with FIG. **18** can be used to produce a piezo-electrophoretic display having a thickness far less than conventional displays and can therefore be bonded to a target object such as a bank note or currency bill without substantially increasing the overall thickness or being substantially noticeable. In some embodiments, the total thickness of the piezo-electrophoretic display can be between 50  $\mu\text{m}$  and 100  $\mu\text{m}$ . In some embodiments, the total thickness of the piezo-electrophoretic display can be between 25  $\mu\text{m}$  and 50  $\mu\text{m}$ . In some embodiments, the total thickness of the piezo-electrophoretic display can be less than 25  $\mu\text{m}$ . The resulting piezo-electrophoretic display produced using method **1800** therefore provides substantially similar benefits and advantages to a piezo-electrophoretic display produced using method **1400** described above.

[0211] One of ordinary skill in the art will appreciate that the steps of method **1400** and method **1800** need not be carried out in the precise order in which the steps were presented herein. As one example, steps **1810-1840** of method **1800** need not necessarily occur prior to steps **1850** and **1860**.

[0212] It is to be appreciated that piezo-electrophoretic films and piezo-electrophoretic displays described herein can be combined with other known techniques for creating security markers or authenticity labels. For example, a piezo-electrophoretic film or piezo electrophoretic display may additionally include a semi-transparent overlay that does not change optical properties when the piezoelectric film is manipulated. For example, a smiley-face overlay may include eyes constructed



from piezo-electrophoretic displays such that when the layered material is bent, the eyes appear to blink. In some embodiments, images or shapes may be printed or laminated onto a solid-colored (e.g., white) background, and must be viewed through the piezo-electrophoretic films to see a pre-arranged pattern. Thus, when not in use, a viewer only sees the solid color, i.e., the printed image or shape will be hidden. However, the printed image or shape will be displayed when the device is manipulated. It is also feasible to adhere a piezo-electrophoretic film or piezo-electrophoretic display to a separate light-transmissive polymer film included in the target product (e.g., bank note) such that the pattern in the piezoelectric layer is only viewable when the target product is held up to a light source and manipulated.

[0213] It will be apparent to those skilled in the art that numerous changes and modifications can be made to the specific embodiments of the invention described above without departing from the scope of the invention. Accordingly, the whole of the foregoing description is to be interpreted in an illustrative and not in a limitative sense.

## Claims

1. A method for integrating a layer of piezoelectric material with an electrode, the method comprising: providing a first substrate; depositing an electrically-conductive material onto the first substrate; forming a first electrode on the first substrate from the electrically-conductive material; depositing an adhesive material onto the first electrode; forming a tie layer on the first electrode from the adhesive material; and depositing a piezoelectric material comprising polyvinylidene fluoride (PVDF) solution on the tie layer to produce a piezoelectric layer less than 5  $\mu\text{m}$  in thickness.
2. The method of claim 1 wherein the first substrate is a release film.
3. The method of claim 1 wherein the electrically-conductive material comprises a light-transmissive conductive polymer.
4. The method of claim 3 wherein the light-transmissive conductive polymer comprises poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS).
5. The method of claim 4 wherein the light-transmissive conductive polymer further comprises a crosslinker.
6. The method of claim 4 wherein the light-transmissive conductive polymer comprises an aqueous solution comprising PEDOT:PSS.
7. The method of claim 1 wherein depositing the electrically-conductive material onto the first substrate comprises using a Mayer rod, a doctor blade, a slot die, or gravure coating, or a combination thereof.
8. The method of claim 1 wherein the polyvinylidene fluoride (PVDF) solution is an aqueous solution.
9. The method of claim 1 wherein the adhesive material comprises a mixture of acrylates, polyurethane, and a solvent based on methyl ethyl ketone.
10. The method of claim 1 wherein forming the tie layer on the first electrode comprises curing the adhesive material with electromagnetic radiation.
11. A method for integrating a layer of microcells with an electrode, the method comprising: providing a first substrate; depositing an electrically-conductive material onto the first substrate; forming a first electrode on the first substrate from the electrically-conductive material; depositing an adhesive material onto the first electrode; forming a tie layer on the first electrode from the adhesive material; bonding a microcell precursor material to the tie layer; embossing the microcell precursor material to create a layer of microcells, wherein the microcells have a bottom, walls, and a top opening; filling the microcells with an electrophoretic medium through the top opening; and sealing off the top opening of the filled microcells with a water-soluble polymer to create a sealing layer.

- 12.** The method of claim 11 wherein the first substrate is a release film.
  - 13.** The method of claim 11 wherein the electrically-conductive material comprises a light-transmissive conductive polymer.
  - 14.** The method of claim 13 wherein the light-transmissive conductive polymer comprises poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS).
  - 15.** The method of claim 14 wherein the light-transmissive conductive polymer further comprises a crosslinker.
  - 16.** The method of claim 14 wherein the light-transmissive conductive polymer comprises an aqueous solution comprising PEDOT:PSS.
  - 17.** The method of claim 11 wherein depositing the electrically-conductive material onto the first substrate comprises using a Mayer rod, a doctor blade, a slot die, or gravure coating, or a combination thereof.
  - 18.** The method of claim 11 further comprising applying a primer to the microcell precursor material before bonding the microcell precursor material to the tie layer.
  - 19.** The method of claim 18 further comprising activating the microcells with a vapor plasma treatment before filling the microcells with the electrophoretic medium.
  - 20.** The method of claim 18 wherein the primer is in an aqueous solution.
  - 21.** The method of claim 11 wherein the adhesive material comprises a mixture of acrylates, polyurethane, and a solvent based on methyl ethyl ketone.
  - 22.** The method of claim 11 wherein forming the tie layer on the first electrode comprises curing the adhesive material with electromagnetic radiation.
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