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(54) Title: PIEZO-ELECTROPHORETIC FILM INCLUDING PATTERNED PIEZO POLARITIES FOR CREATING IMAGES VIA ELECTROPHORETIC MEDIA

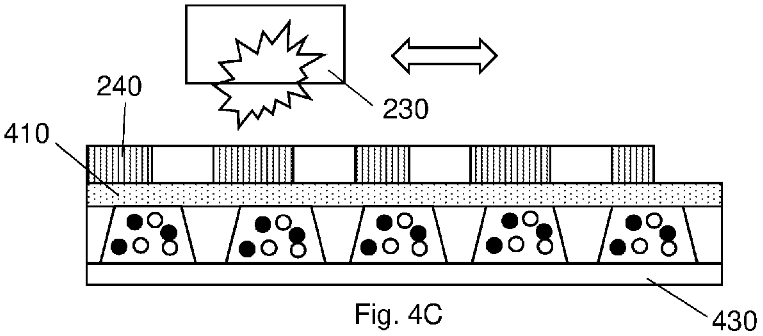

Fig. 4C

(57) Abstract: Low-profile piezo-electrophoretic films and display films including low profile piezoelectrophoretic films. In some embodiments, the piezoelectric material of the piezoelectrophoretic films can be patterned with high-voltage electric fields after fabrication of the piezo-electrophoretic films. Such films are useful as security markers, authentication films, or sensors. The films are generally flexible. Some films are less than 100 μm in thickness. Displays formed from the films do not require an external power source.

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PIEZO-ELECTROPHORETIC FILM INCLUDING PATTERNED PIEZO POLARITIES FOR CREATING IMAGES VIA ELECTROPHORETIC MEDIA

REFERENCE TO RELATED APPLICATIONS

[Para 1] This application claims priority to U.S. Patent Application No. 63/314,584, filed February 28, 2022. All patents and publications disclosed herein are incorporated by reference in their entireties.

BACKGROUND

[Para 2] 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. The electrophoretic fluid typically includes a non-polar solvent and one or more sets of charged particles. The particles may have different optical properties (colors), different charges (positive or negative), different charge magnitudes (zeta potentials), and/or different absorptive properties (broadly light-absorbing, 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 particle of one set to appear at the viewing surface while the other particle is driven away from the viewing surface.

[Para 3] Many electrophoretic displays are bi-stable: their optical state persists even after the activating electric field is removed. Bistability is mostly due to 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.

[Para 4] Driving an electrophoretic display requires a power source to provide the electric field between the electrodes. The power source is typically a battery, which provides power to the electrodes via driving circuitry. One or more electrodes may be incorporated into an active matrix backplane. The power supply could also be, e.g., a photovoltaic cell, a fuel cell, or a power supply that operates from wall current. The power supply could also be a piezo-electric element which creates charge through physical motion or thermal expansion, as described in US Patent No. 5,930,026, which is incorporated by reference in its entirety. In all of these examples, some type of driving circuitry is required to provide an electrical pathway between the power source and the electrodes and typically, the circuitry includes control elements such as switches, transistors, etc. In most instances, the circuitry is fairly routine, however it typically adds bulk and structural limitations (i.e., not flexible or twistable) to the final display. There is a need for very simple, flexible, durable, and thin electrophoretic displays for applications such as security markers, sensors, and indicators.

SUMMARY

[Para 5] 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.

[Para 6] In a first aspect, the invention includes an electrophoretic display film, less than 100 μm 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 sealing layer. In some embodiments, the film is less than 50 μm 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.

[Para 7] 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 μm 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.

[Para 8] 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 µm 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 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 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 µm 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.

[Para 9] In a fourth aspect, an electrophoretic display film, less than 100 µm 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 sealing layer is conductive. In some embodiments, the film is less than 50 μm 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.

[Para 10] 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.

[Para 11] In a sixth aspect, the invention includes an electrophoretic display film, less than 100 µm 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 µm 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.

[Para 12] In a seventh aspect, the invention includes an electrophoretic display film, less than 100 µm 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.

BRIEF DESCRIPTION OF DRAWINGS

[Para 13] 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 100µm, e.g., less than 50µm, e.g., less than 25µm.

[Para 14] 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.

[Para 15] Fig. 2A shows an exemplary thin layer of piezoelectric material on a substrate.

[Para 16] 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.

[Para 17] 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.

[Para 18] Fig. 2D illustrates a polarization (poling) pattern that can be achieved with the methods of Fig. 2B and Fig. 2C.

[Para 19] Fig. 3A illustrates a side view of a piezo-electric film poled in the A direction.

[Para 20] Fig. 3B illustrates a top view of a piezo-electric film poled in the A direction.

[Para 21] Fig. 3C illustrates a side view of a piezo-electric film poled in the G direction using a conductive mask.

[Para 22] Fig. 3D illustrates a top view of a piezo-electric film poled in the G direction using a conductive mask.

[Para 23] Fig. 4A shows an exemplary thin layer of a piezo-microcell precursor film on a substrate.

[Para 24] 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.

[Para 25] 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.

[Para 26] 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.

[Para 27] Fig. 5A is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[Para 28] Fig. 5B is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[Para 29] Fig. 5C is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[Para 30] Fig. 5D is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[Para 31] Fig. 6A is a schematic cross section of an embodiment of a piezo-electrophoretic display.

[Para 32] Fig. 6B is a schematic cross section of an embodiment of a piezo-electrophoretic display.

[Para 33] Fig. 7 details a method for creating a piezo-electrophoretic film or (optionally) display.

[Para 34] Fig. 8A is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[Para 35] Fig. 8B is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[Para 36] Fig. 9A is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[Para 37] Fig. 9B is a schematic cross section of an embodiment of a piezo-electrophoretic film.

[Para 38] Fig. 10A is a schematic cross section of an embodiment of a piezo-electrophoretic display.

[Para 39] Fig. 10B is a schematic cross section of an embodiment of a piezo-electrophoretic display.

[Para 40] Fig. 10C is a schematic cross section of an embodiment of a piezo-electrophoretic display.

[Para 41] Fig. 11 details a method for creating a low-profile piezo-electrophoretic film.

[Para 42] Fig. 12A is a schematic cross section of a piezo-electrophoretic film created with the method shown in Fig. 11.

[Para 43] Fig. 12B is a schematic cross section of a piezo-electrophoretic display created with the method shown in Fig. 11.

[Para 44] Fig. 13A is a schematic cross section of an alternative piezo-electrophoretic film created with the method shown in Fig. 11.

[Para 45] Fig. 13B is a schematic cross section of an alternative piezo-electrophoretic display created with the method shown in Fig. 11.

DETAILED DESCRIPTION

[Para 46] Low-profile piezo-electrophoretic films and display films including low profile piezo-electrophoretic films 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. Such films are useful as security markers, authentication films, or sensors. The films are generally flexible. Some films are less than 100 μm in thickness. In some embodiments, the piezo-electrophoretic films are less than 50 μm and foldable without breaking. Displays formed with the films do not require an external power source.

[Para 47] 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.

[Para 48] 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. Patent 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.

[Para 49] 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.

[Para 50] 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.

[Para 51] 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. Patents Nos. 6,301,038; 6,870,657; and 6,950,220. This type of medium is also typically bistable.

[Para 52] 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. Patent No. 7,420,549 that such electro-wetting displays can be made bistable.

[Para 53] 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 move 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.

[Para 54] 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.

[Para 55] 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:

(a) Electrophoretic particles, fluids and fluid additives; see for example U.S. Patents Nos. 7,002,728 and 7,679,814;

(b) Capsules, binders and encapsulation processes; see for example U.S. Patents Nos. 6,922,276 and 7,411,719;

(c) Films and sub-assemblies containing electro-optic materials; see for example U.S. Patents Nos. 6,982,178 and 7,839,564;

(d) Backplanes, adhesive layers and other auxiliary layers and methods used in displays; see for example U.S. Patents Nos. 7,116,318 and 7,535,624;

(e) Color formation and color adjustment; see for example U.S. Patents Nos. 7,075,502 and 7,839,564;

(f) Methods for driving displays; see for example U.S. Patents Nos. 7,012,600 and 7,453,445;

(g) Applications of displays; see for example U.S. Patents Nos. 7,312,784 and 8,009,348;

(h) Non-electrophoretic displays, as described in U.S. Patents Nos. 6,241,921; 6,950,220; 7,420,549 and 8,319,759; and U.S. Patent Application Publication No. 2012/0293858;

(i) Microcell structures, wall materials, and methods of forming microcells; see for example United States Patents Nos. 7,072,095 and 9,279,906; and

(j) Methods for filling and sealing microcells; see for example United States Patents Nos. 7,144,942 and 7,715,088.

[Para 56] 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. Patent 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.

[Para 57] A related type of electrophoretic display is a 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. Patents Nos. 6,672,921 and 6,788,449, both of which are incorporated by reference in their entireties.

[Para 58] 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. Patents 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. Patent 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.

[Para 59] 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. Patent 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.

[Para 60] The aforementioned U.S. Patent 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 μm), preferably about 2 to about 10 mil (51 to 254 μm). 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 Trade Mark) 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.

[Para 61] 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.

[Para 62] U.S. Patent 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. Patent 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.

[Para 63] The subject matter presented herein, in particular, relates to piezo-electrophoretic display structural designs which do not need a power supply (e.g., battery or wired power supply, photovoltaic source, etc.,) in order for the electrophoretic display to operate. The assembly of such an electrophoretic display is therefore simplified. In some embodiments, the piezoelectric material and the electrophoretic media are directly laminated together. The electrophoretic medium may be contained in microcells, microcapsules, or the electrophoretic medium may be dispersed in a polymer matrix, as described above. In some embodiments the piezoelectric material is polarized (i.e., written) with a high-voltage electric field after the piezo-electrophoretic film or piezo-electrophoretic display has been created, as discussed below.

[Para 64] 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 (SiO_2), berlinite ($AlPO_4$), gallium orthophosphate ($GaPO_4$), tourmaline, barium titanate ($BaTiO_3$), lead zirconate titanate (PZT), zinc oxide (ZnO), aluminum nitride (AlN), lithium tantalite, lanthanum gallium silicate, potassium sodium tartrate and any other known piezo materials. In piezoelectric materials,

[Para 65] Piezo-electrophoretic films and piezo-electrophoretic displays described herein use piezoelectricity to drive the charged pigments of an electrophoretic medium Thus, when the piezoelectric material coupled to an electrophoretic media layer is manipulated, the color of the electrophoretic material at the viewing surface changes. For example, by bending or introduce stress to a piece of piezo material, voltage may be generated and this voltage can be utilized to cause movement of the color pigments of the electrophoretic material. If segments of piezoelectric material with different polarizations are used, or if areas of differential polarization are created in a piezoelectric film, 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" (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, or CR, is a desired aspect of a display.

[Para 66] Figures 1A and 1B illustrates 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.

[Para 67] 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^2. 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 aligned these regions is known as poling.

[Para 68] 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. Patent 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 μm, e.g., less than 10 μm, less than 5 μm, 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.

[Para 69] 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 spun-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 μm, e.g., less than 10 μm, less than 5 μm) 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.

[Para 70] As shown in Fig. 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 μm of the piezo material that will be poled. The spatial focus can be accomplished with 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.

[Para 71] 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 $\mathbf{P}_1$ and $\mathbf{P}_2$, 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 techniques 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 are 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.

[Para 72] Figs. 2A-3D illustrate the various techniques that can be used to create areas of differential polarization in an 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 spun-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 a acrylates, vinyl ethers, or epoxides, as described in detail in, for example, U.S. Patent 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. Patent 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 US Patent 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 sealing layer is in contact with the thin film of piezoelectric material 410.

[Para 73] 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 $\mathbf{P}_1$ and $\mathbf{P}_2$, 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.

[Para 74] The techniques described above can be used to achieve a great variety of thin piezo-electrophoretic films as described in the following figures.

[Para 75] 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 μm thick or less, e.g., 70 μm thick or less, e.g., 50 μm thick or less, e.g., 35 μm thick or less, e.g., 20 μm thick or less, e.g., 10 μm 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.

[Para 76] 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. 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 10^7 to 10^8 Ohm-m, and the piezoelectric material has a resistance of 10^{11} to 10^{14} Ohm-m.)

[Para 77] 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 10^6 Ohm*cm to 10^8 Ohm*cm, preferably less than 10^{12} Ohm*cm.

[Para 78] 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.

[Para 79] In many embodiments, a piezo-electrophoretic film or a piezo-electrophoretic display will often include a release sheet. The release may be use 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.

[Para 80] 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).

[Para 81] 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.

[Para 82] 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 Irostic® polymers.

[Para 83] 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.

[Para 84] 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 Fig. 6A and 6B, it is typical to add a release 510 to the completed piezo-electrophoretic displays (601, 602) to improve handling, and the 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

[Para 85] A series of piezo-electrophoretic films of the type exemplified in Figs. 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 1: Prototype piezo-electrophoretic films

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

[Para 86] 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 Fig. 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 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 μm thick) is exceedingly thin (i.e., less than 25 μm 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 μm. 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).

[Para 87] 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 are 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 μm 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).

[Para 88] 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 μm thick or less, e.g., 20 μm 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 substrate1255 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. Patent 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 μm thick or less, e.g., 20 μm 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.

[Para 89] 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 displays 1202 does not require for 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.

[Para 90] 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.

[Para 91] 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.

[Para 92] 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. An electrophoretic display film, less than 100 µm thick (top to bottom), comprising, in order:

a first adhesive layer (520);

an electrophoretic medium layer (405, 530);

a patterned piezo electric layer (410, 560, 960) comprising zones of differential polarization; and

a flexible, light-transmissive electrode layer (580).

2. The electrophoretic display film of claim 1, wherein the electrophoretic medium layer (405, 530) comprises a plurality of microcapsules (990) containing a non-polar fluid (425) and charged pigment particles (423, 427) that move toward or away from the patterned piezo electric layer (410, 560, 960) when the patterned piezo electric layer (410, 560, 960) is flexed, wherein the microcapsules (990) are coupled to each other with a polymer binder (995), or wherein the electrophoretic medium layer (405, 530) comprises a plurality of microcells (420, 530) containing a non-polar fluid (425) and charged pigment particles (423, 427) that move toward or away from the patterned piezo electric layer (410, 560, 960) when the patterned piezo electric layer (410, 560, 960) is flexed, wherein the non-polar fluid (425) and charged pigment particles (423, 427) are sealed in the microcells (420, 530) with a sealing layer (430).

3. The electrophoretic display film of claim 1, wherein the electrophoretic display film is less than 50 µm thick.

4. The electrophoretic display film of claim 1, wherein the patterned piezo electric layer (410, 560, 960) comprises polyvinylidene fluoride (PVDF), which is optionally poled to create the zones of differential polarization (460, 470).

5. The electrophoretic display film of claim 1, wherein the flexible, light-transmissive electrode layer (580) comprises a metal oxide comprising tin or zinc, or poly(3,4-ethylenedioxythiophene) (PEDOT).

6. An electrophoretic display film assembly comprising a release sheet (510) coupled to an electrophoretic display film of claim 1, wherein the release sheet (510) is coupled to the first adhesive layer (520).

7. The electrophoretic display film assembly of claim 6, further comprising a second adhesive layer coupled to the flexible, light-transmissive electrode layer (580), and a second release sheet coupled to the second adhesive layer.

8. A method of making an electrophoretic display film comprising:

coupling a film (1260) of polyvinylidene fluoride (PVDF) to a polymer film (1230) 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 (1280);

coupling the flexible, light-transmissive electrode layer (1280) to a first release film (1255) with a first adhesive layer (1250);

embossing the piezo-microcell precursor film to create an array of microcells (1230), wherein the microcells have a bottom, walls, and a top opening;

filling the microcells with an electrophoretic medium (405, 530) through the top opening; and

sealing off the top opening of the filled microcells with a water-soluble polymer (1240) to make an electrophoretic medium layer (405, 530).

9. The method of claim 8, further comprising applying a primer (1235) to the polymer film (1230) comprising acrylates, vinyl ethers, or epoxides before coupling the polymer film (1230) to the film (1260) of polyvinylidene fluoride (PVDF).

10. The method of claim 8, further comprising coupling the water-soluble polymer (1240) to a second release film (1210) with a second adhesive layer (1220).

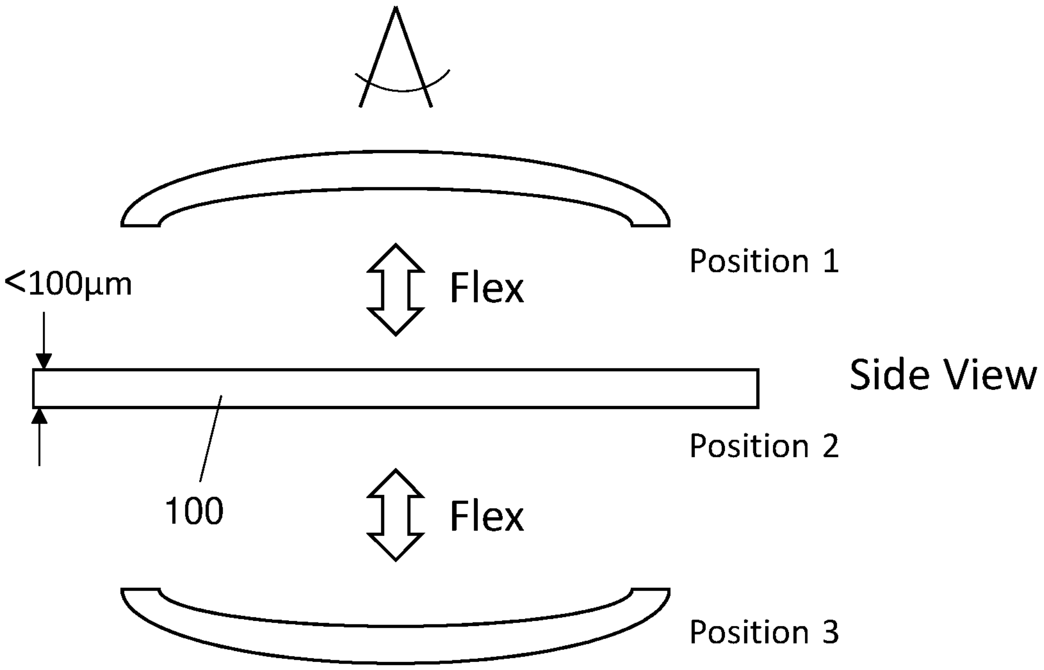
11. The method of claim 8, further comprising removing the first release film (1255) to produce an electrophoretic display film that is less than 100 µm thick.

12. The method of claim 8, wherein the electrophoretic medium layer (405, 530) comprises a non-polar fluid (425) and charged pigment particles (423, 427) that move toward or away from the film (1260) of polyvinylidene fluoride (PVDF) when film (1260) of polyvinylidene fluoride (PVDF) is flexed.

13. The method of claim 8, wherein the film (1260) of polyvinylidene fluoride (PVDF) is poled to create differential zones of polarization (460, 470).

14. The method of claim 8, wherein the flexible, light-transmissive electrode layer (1280) comprises a metal oxide comprising tin or zinc, or poly(3,4-ethylenedioxythiophene) (PEDOT).

15. The method of claim 8, wherein the film (1260) of polyvinylidene fluoride (PVDF) is patterned with an electric field to create areas of differing polarization (460, 470).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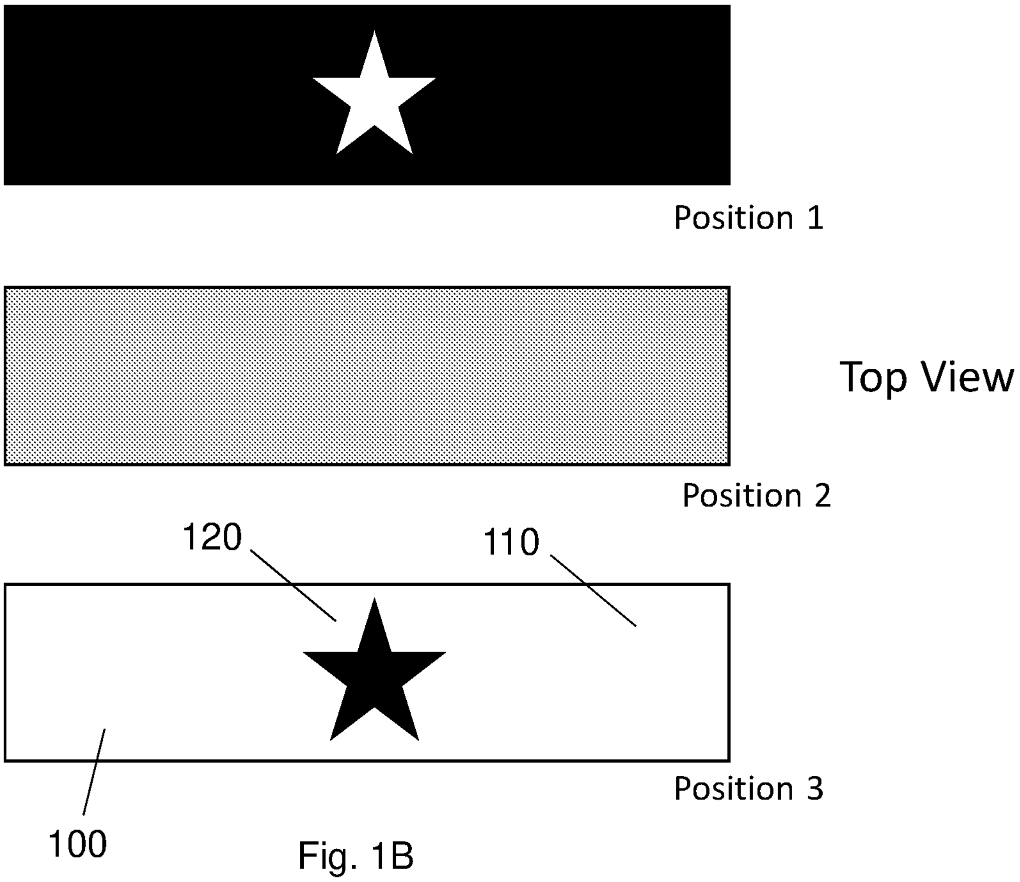
Flex
Position 1
Side View
Position 2
100
Flex
Position 3

Fig. 1A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Top View
Position 2
120
110
Position 3
100

Fig. 1B

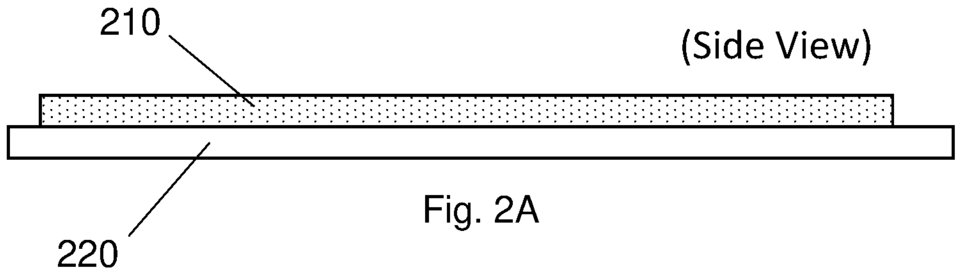

Fig. 2A

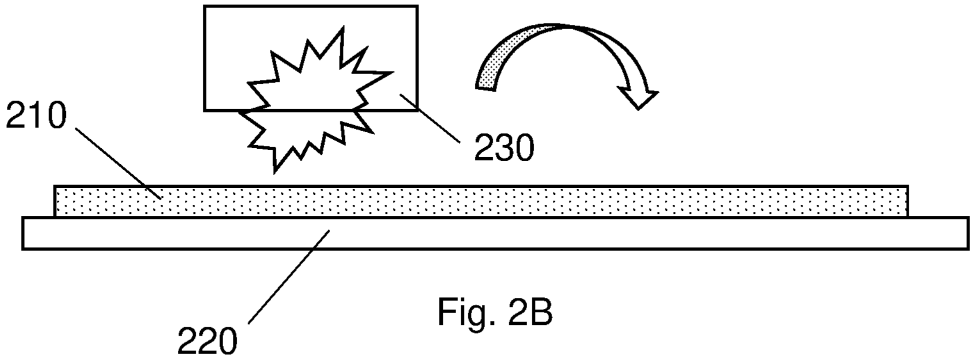

Fig. 2B

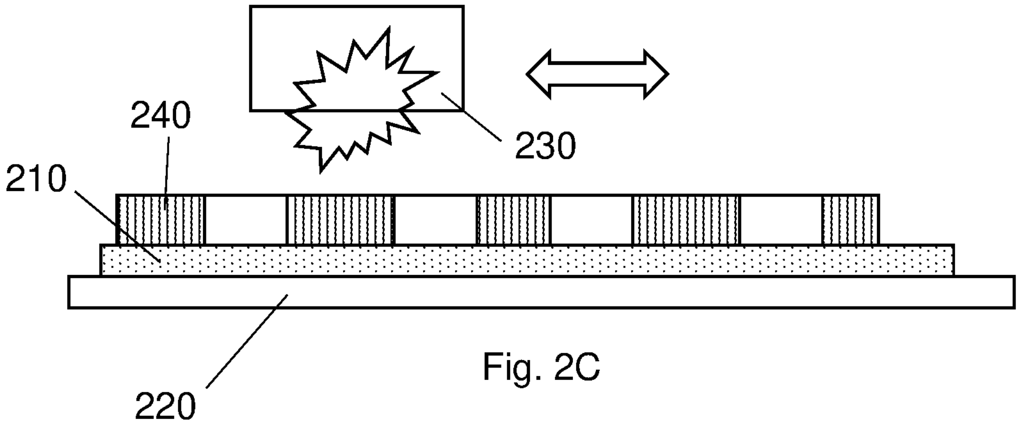

Fig. 2C

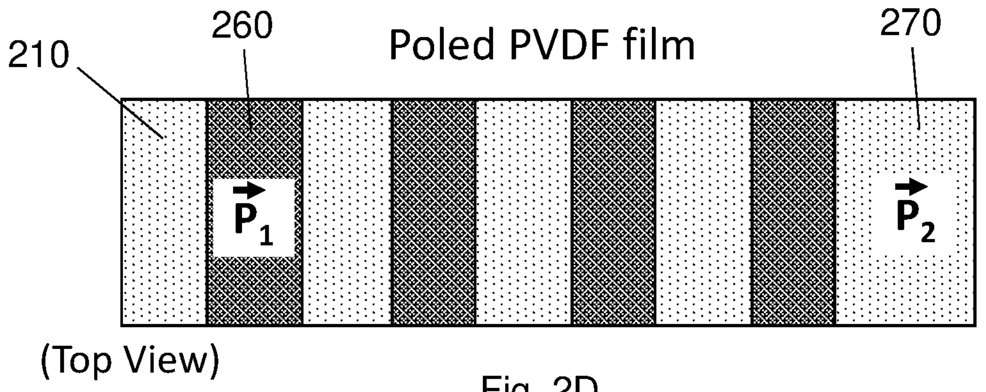

Fig. 2D

Poling into A direction

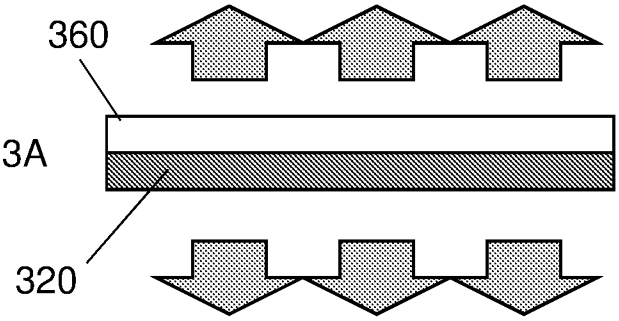

Fig. 3A

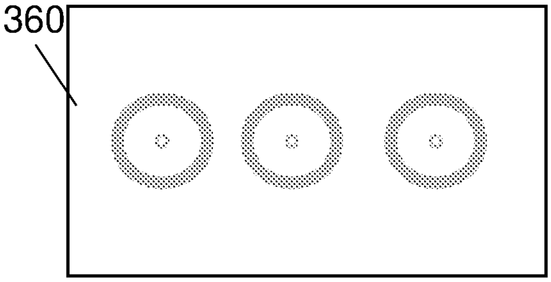

Fig. 3B

Poling into G direction

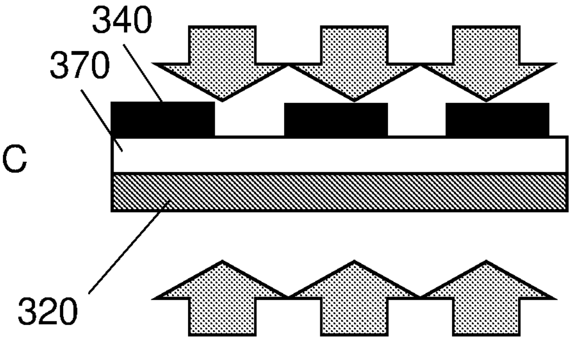

Fig. 3C

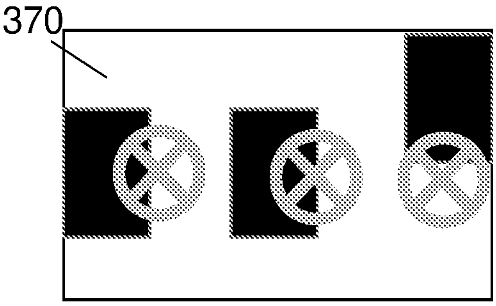

Fig. 3D

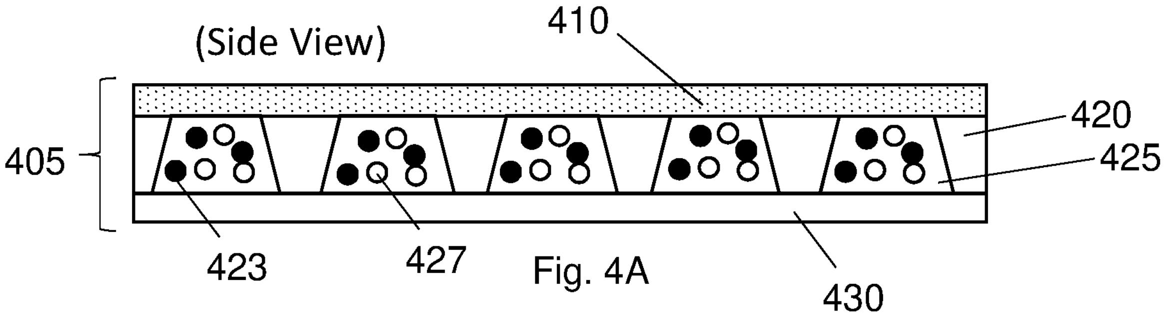

Fig. 4A

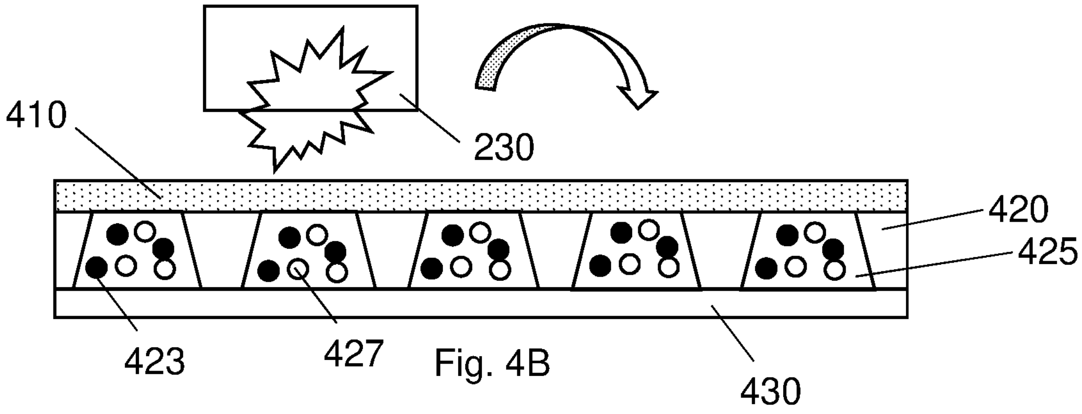

Fig. 4B

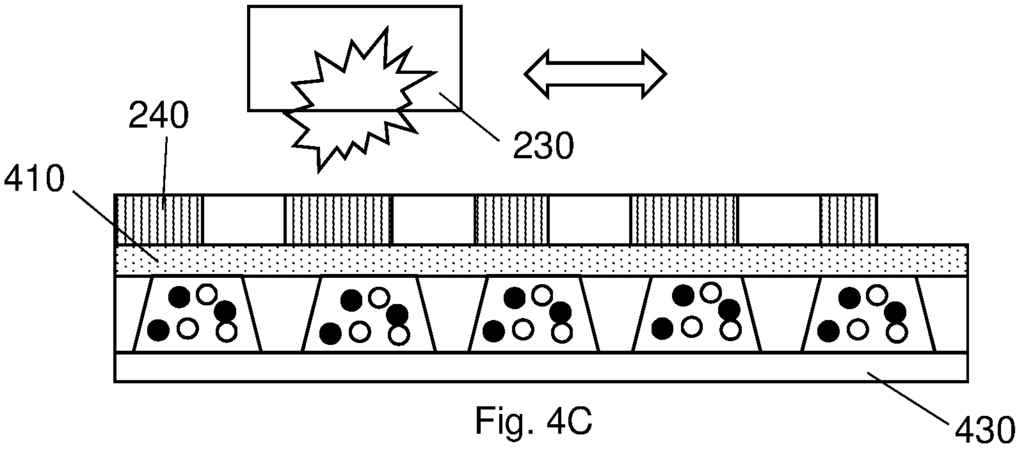

Fig. 4C

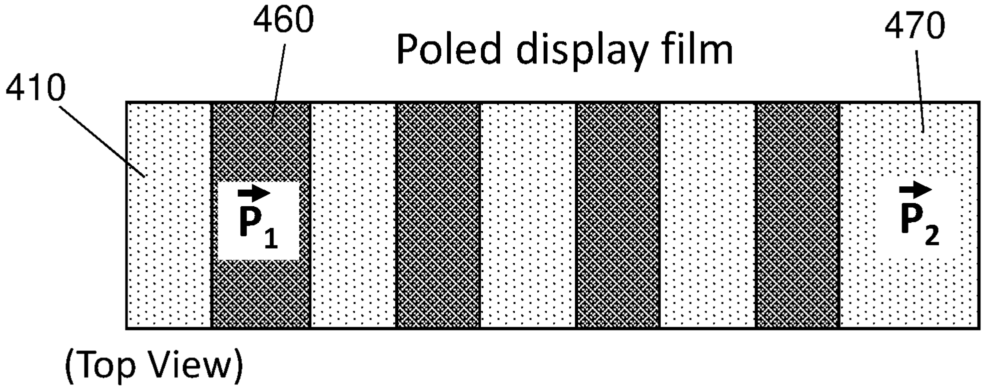

Fig. 4D

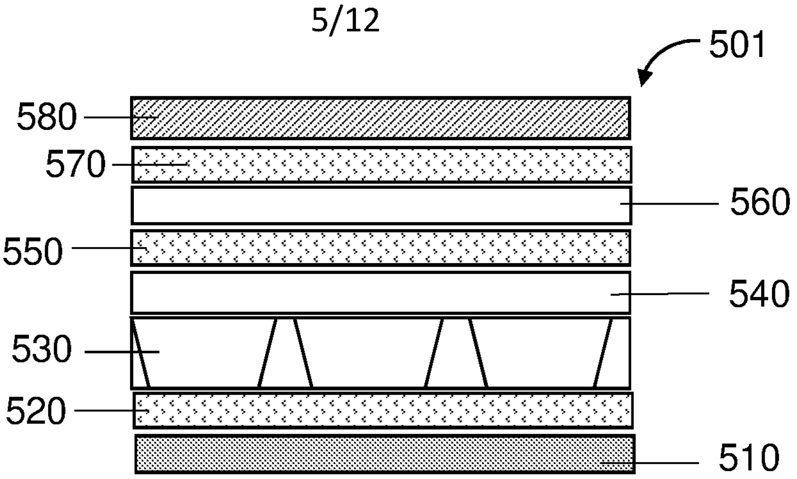

Fig. 5A

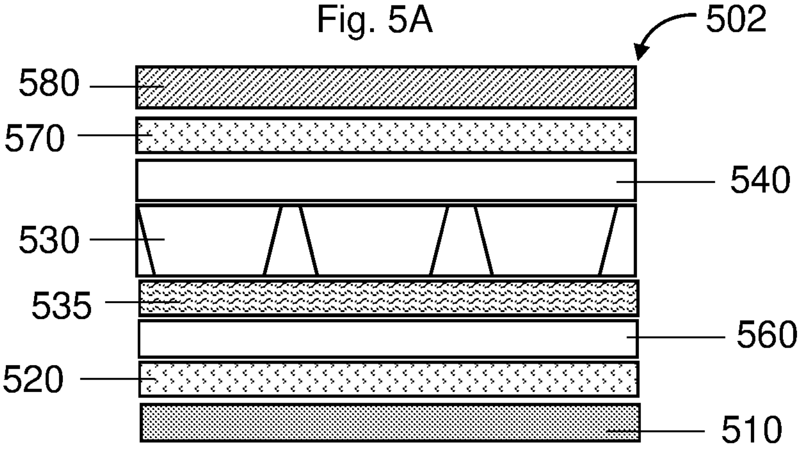

Fig. 5B

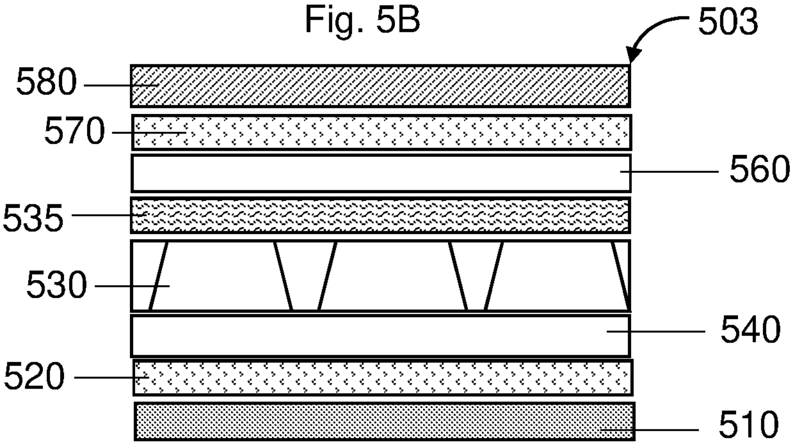

Fig. 5C

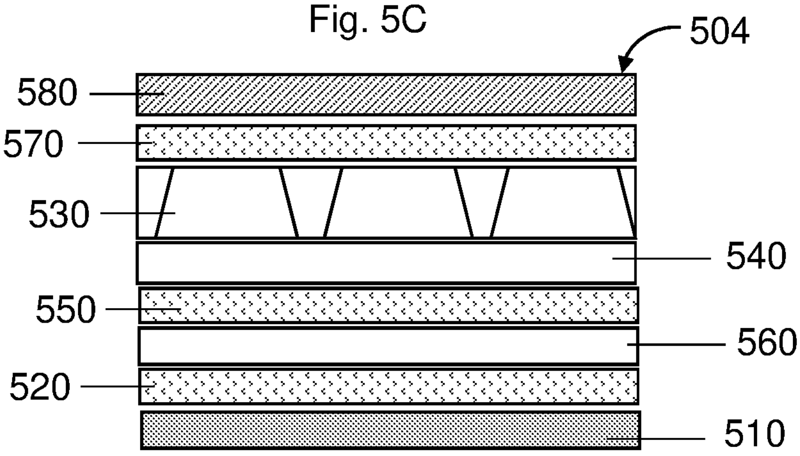

Fig. 5D

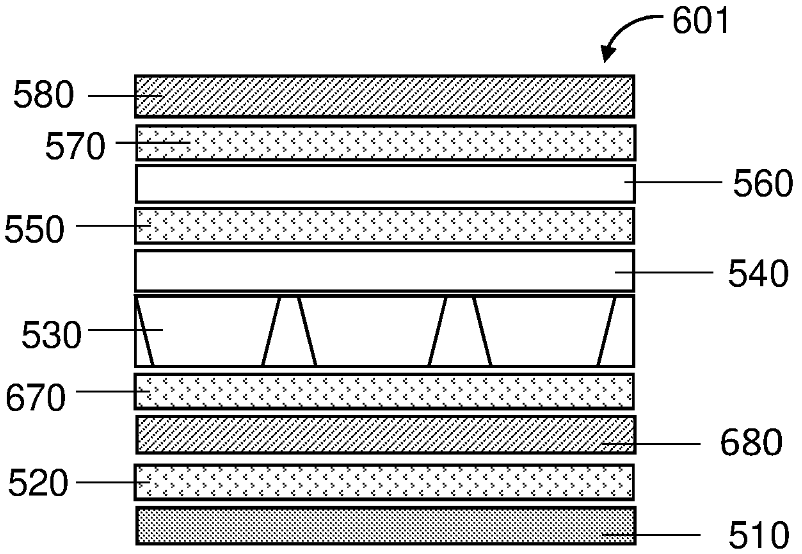
601
580
570
560
550
540
530
670
680
520
510

Fig. 6A

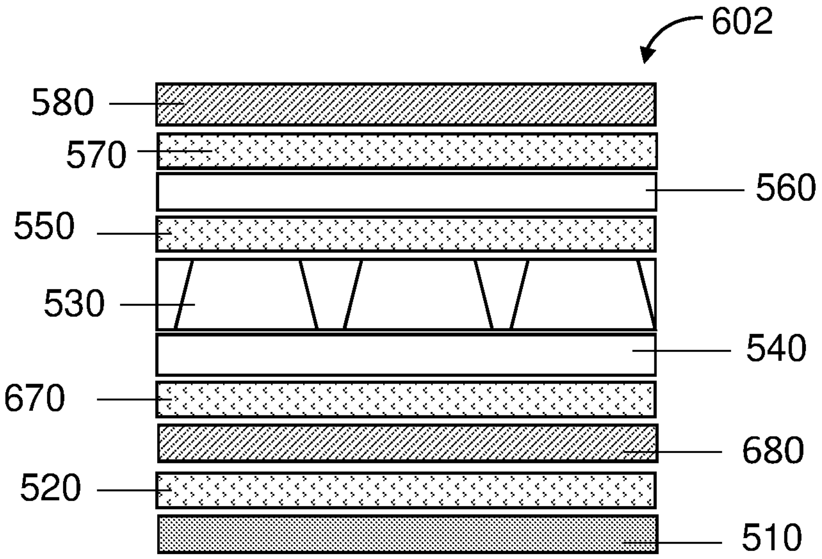
602
580
570
560
550
530
540
670
680
520
510

Fig. 6B

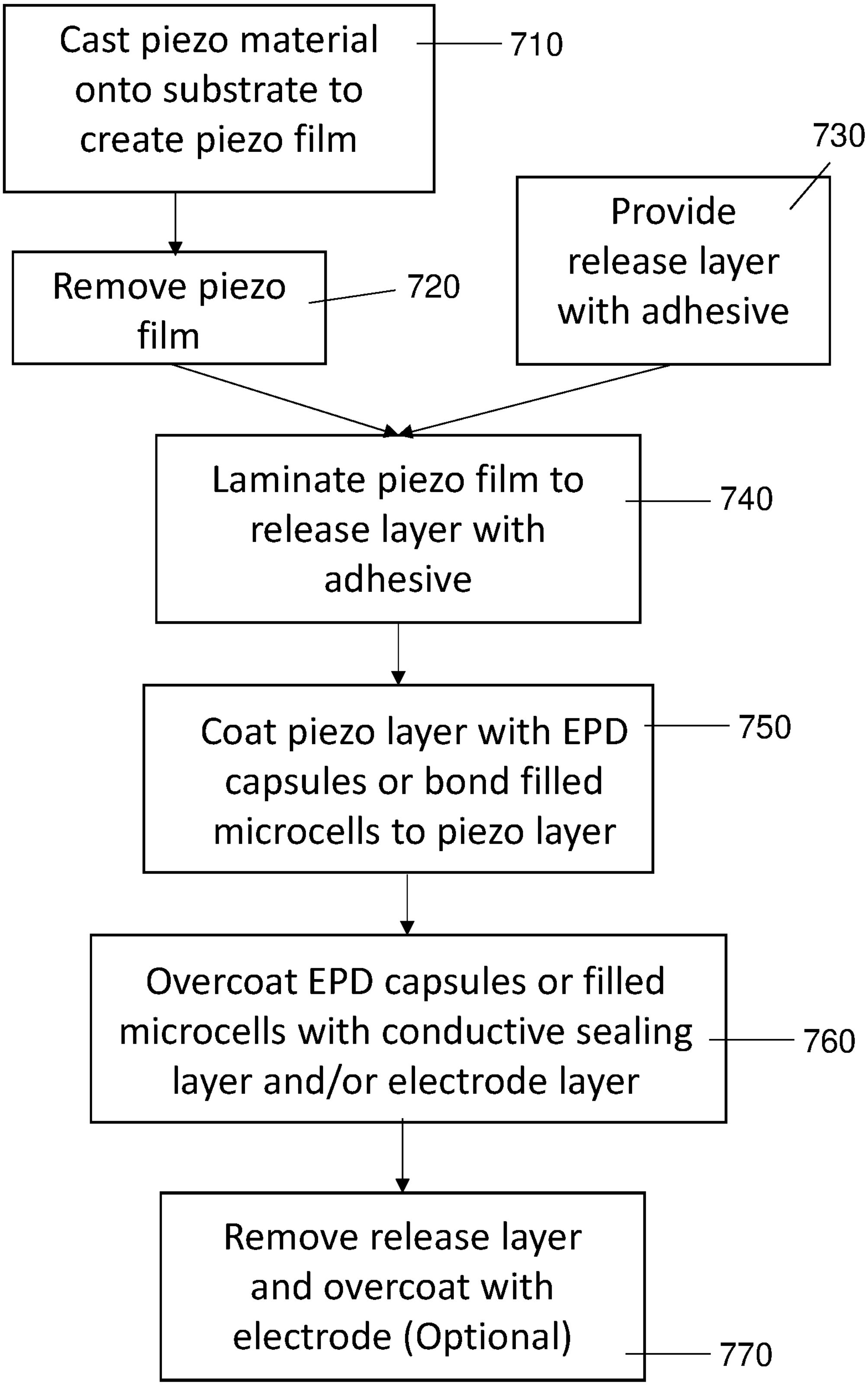
Cast piezo material onto substrate to create piezo film
710
Remove piezo film
720
Provide release layer with adhesive
730
Laminate piezo film to release layer with adhesive
740
Coat piezo layer with EPD capsules or bond filled microcells to piezo layer
750
Overcoat EPD capsules or filled microcells with conductive sealing layer and/or electrode layer
760
Remove release layer and overcoat with electrode (Optional)
770

Fig. 7

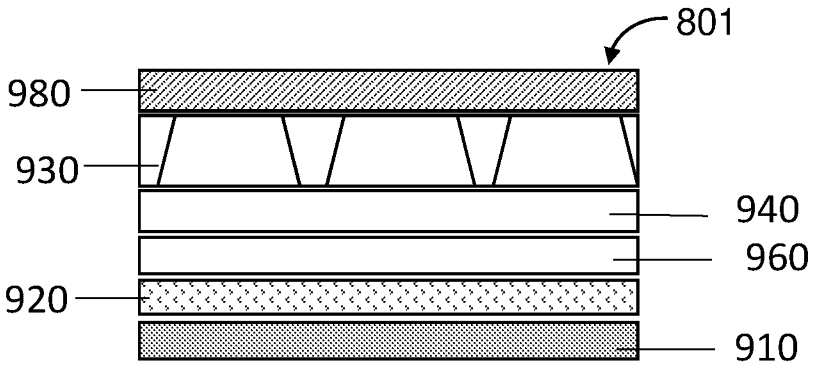

Fig. 8A

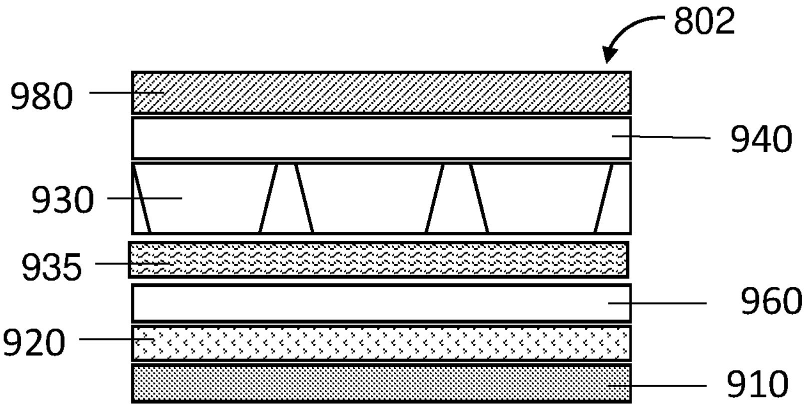

Fig. 8B

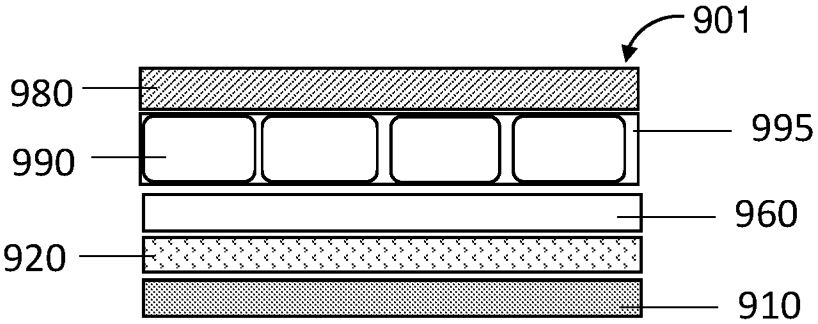

Fig. 9A

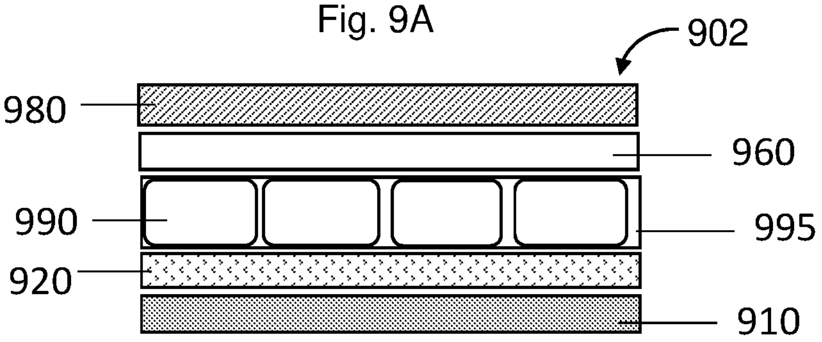

Fig. 9B

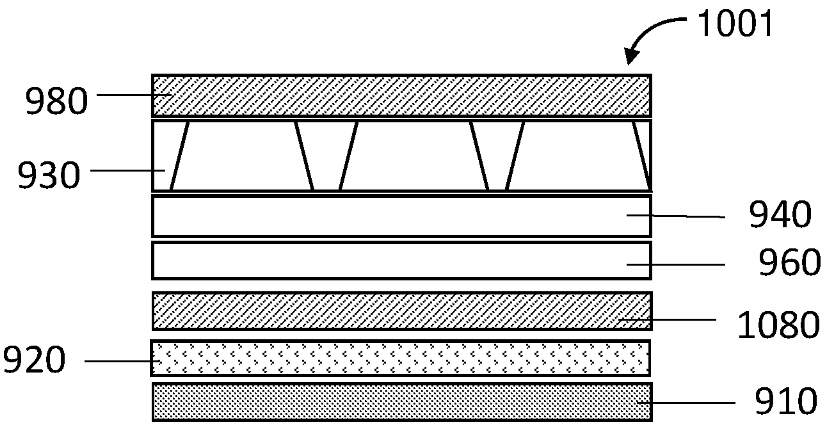

Fig. 10A

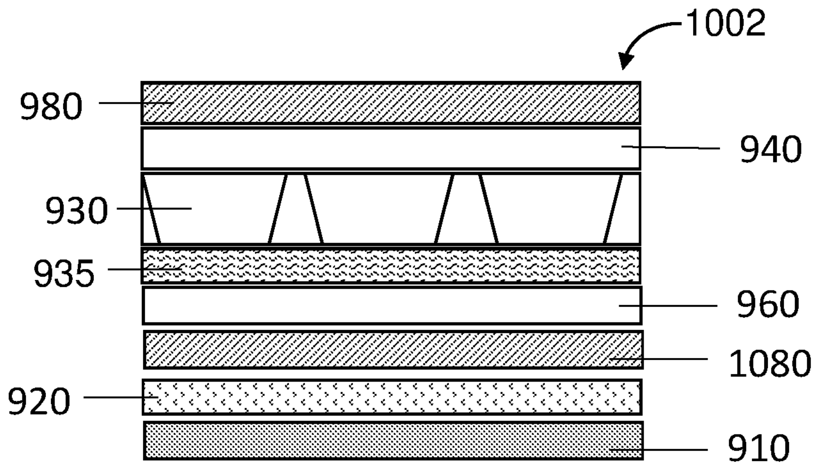

Fig. 10B

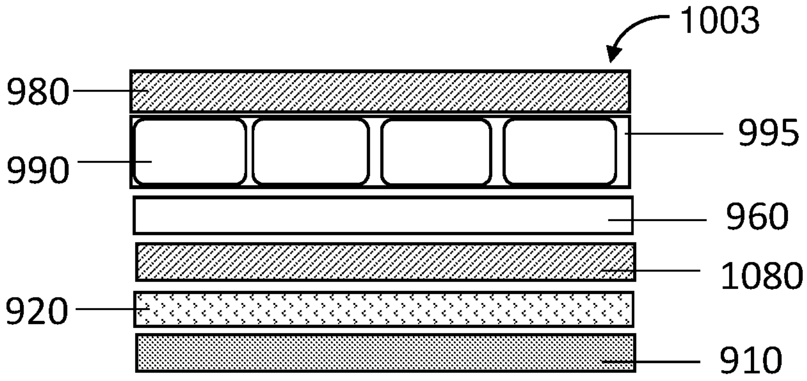

Fig. 10C

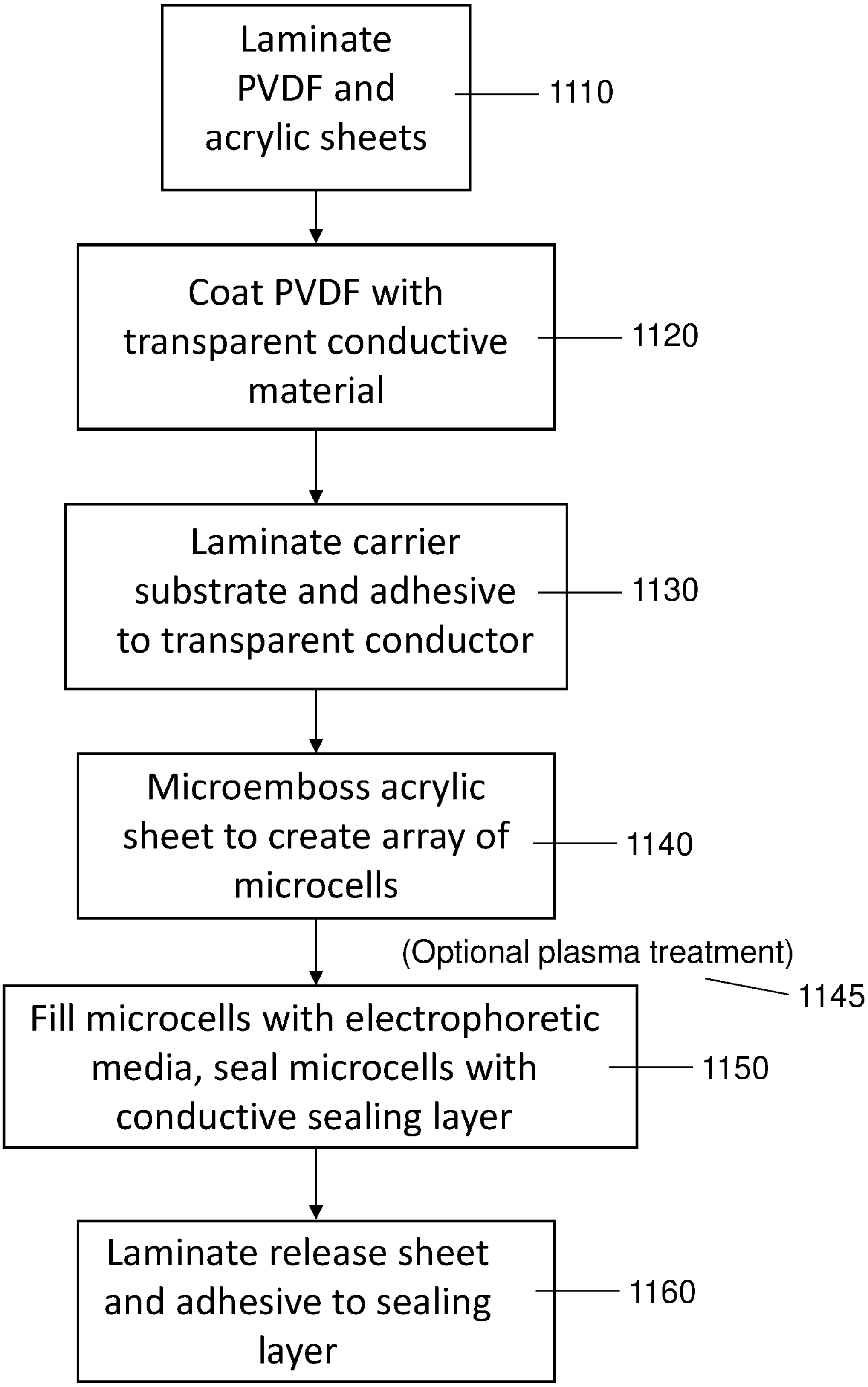
Laminate PVDF and acrylic sheets
1110
Coat PVDF with transparent conductive material
1120
Laminate carrier substrate and adhesive to transparent conductor
1130
Microemboss acrylic sheet to create array of microcells
1140
(Optional plasma treatment)
1145
Fill microcells with electrophoretic media, seal microcells with conductive sealing layer
1150
Laminate release sheet and adhesive to sealing layer
1160

Fig. 11

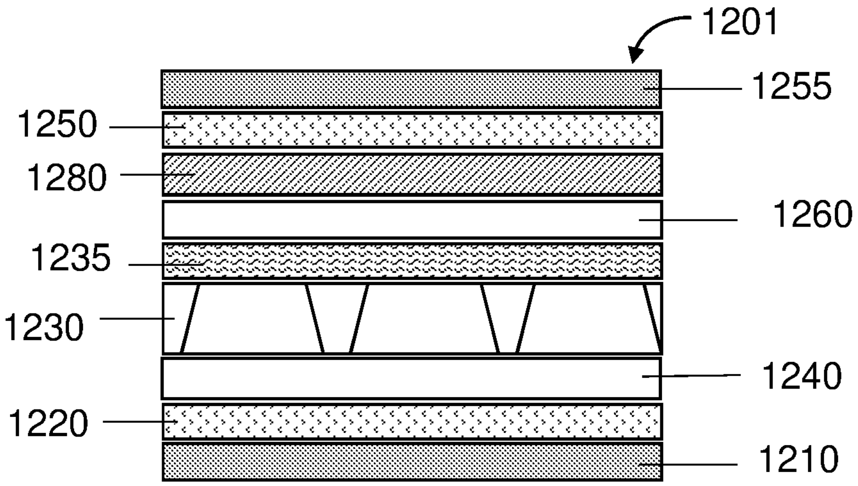

Fig. 12A

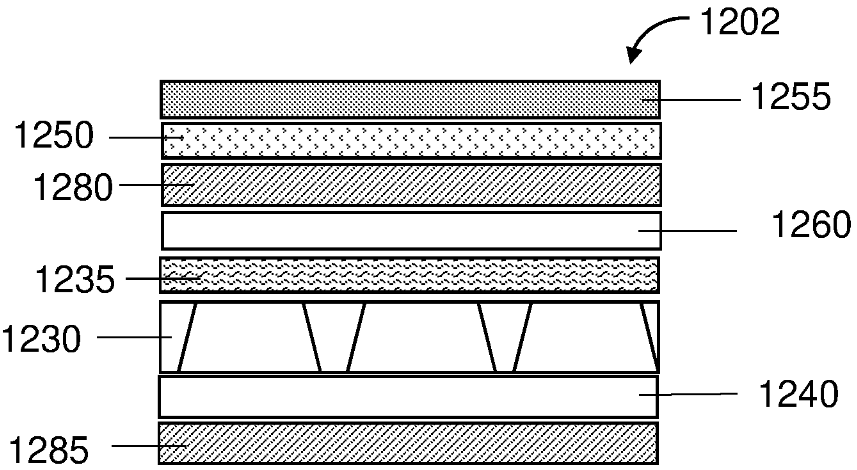

Fig. 12B

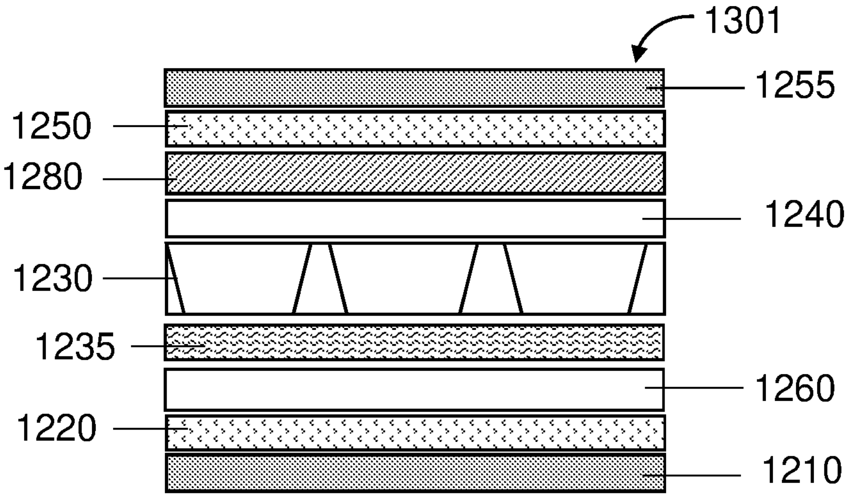

Fig. 13A

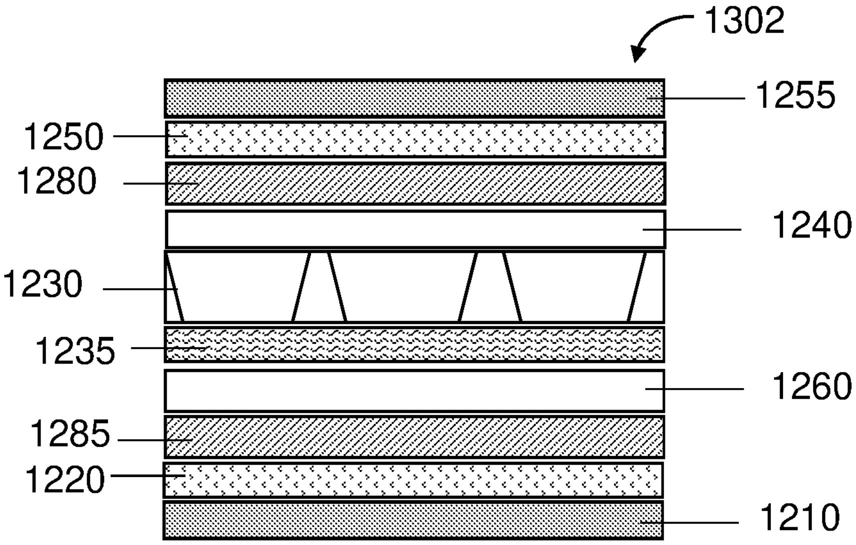

Fig. 13B

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2023/062969

A. CLASSIFICATION OF SUBJECT MATTER
INV. G02F1/00 G02F1/01 G02F1/167
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
G02F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	US 2022/035219 A1 (GU HAIYAN [US] ET AL) 3 February 2022 (2022-02-03) the whole document -----	1-7 8-15
A	US 2004/263483 A1 (AUFDERHEIDE BRIAN E [US]) 30 December 2004 (2004-12-30) the whole document -----	1-15

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

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"P" document published prior to the international filing date but later than the priority date claimed

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"&" document member of the same patent family

Date of the actual completion of the international search 2 June 2023	Date of mailing of the international search report 13/06/2023
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Brumbarov, Jassen

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/US2023/062969

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