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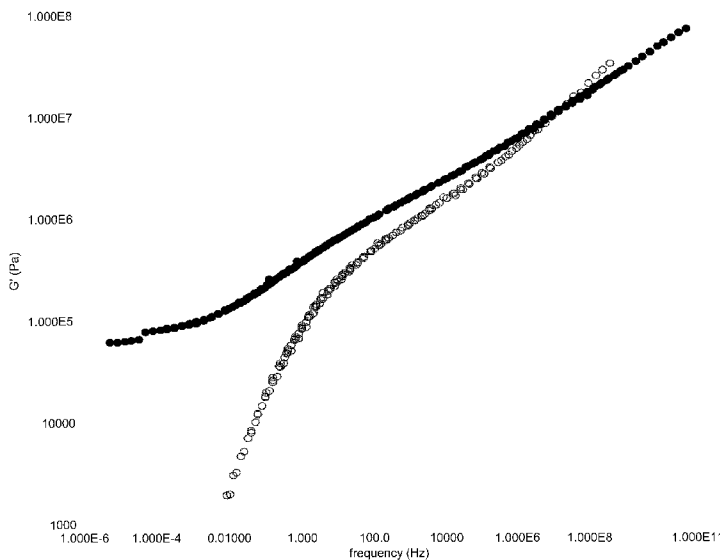
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(54) **Title:** ELECTRO-OPTIC DISPLAYS, AND MATERIALS FOR USE THEREIN



(57) **Abstract:** A flexible electro-optic display comprises a layer of solid electro-optic material; a backplane comprising at least one electrode arranged to apply an electric field to the layer of electro-optic material; and an adhesive layer disposed between the layer of electro-optic material and the backplane and adhesively securing the layer of electro-optic material to the backplane, the adhesive layer having a storage modulus (G') of at least about 10⁴ Pascals at 10-3 Hz and 70°C.

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For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

ELECTRO-OPTIC DISPLAYS, AND MATERIALS FOR USE THEREIN

[Para 1] This application is related to:

- (a) U.S. Patent No. 6,831,769, issued December 14, 2004;
- (b) U.S. Patent Publication No. 2005/0122563;
- (c) U.S. Patent No. 7,012,735, issued March 14, 2006;
- (d) U.S. Patent No. 7,110,164, issued September 19, 2006.

[Para 2] The reader is referred to these patents and publication for background information useful in understanding the present invention.

[Para 3] The present invention relates to electro-optic displays and to materials for use therein. More specifically, this invention relates to adhesive compositions for use in electro-optic displays, and to electro-optic displays incorporating such adhesive compositions. The present invention is especially, though not exclusively, intended for use in displays containing encapsulated electrophoretic media.

[Para 4] Electro-optic displays comprise a layer of electro-optic material, a term which 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 5] Some electro-optic materials are solid in the sense that the materials have solid external surfaces, although the materials may, and often do, have internal liquid- or gas-filled spaces. Such displays using solid electro-optic materials may hereinafter for convenience be referred to as "solid electro-optic displays". Thus, the term "solid electro-optic displays" includes rotating bichromal member displays, encapsulated

electrophoretic displays, microcell electrophoretic displays and encapsulated liquid crystal displays.

[Para 6] 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 7] Several types of electro-optic displays are known. One type of electro-optic display is a rotating bichromal member type as described, for example, in U.S. Patents Nos. 5,808,783; 5,777,782; 5,760,761; 6,054,071 6,055,091; 6,097,531; 6,128,124; 6,137,467; and 6,147,791 (although this type of display is often referred to as a "rotating bichromal ball" display, the term "rotating bichromal member" is preferred as more accurate since in some of the patents mentioned above the rotating members are not spherical). Such a display uses a large number of small bodies (typically spherical or cylindrical) which have two or more sections with differing optical characteristics, and an internal dipole. These bodies are suspended within liquid-filled vacuoles within a matrix, the vacuoles being filled with liquid so that the bodies are free to rotate. The appearance of the display is changed by applying an electric field thereto, thus rotating the bodies to various positions and varying which of the sections of the bodies is seen through a viewing surface. This type of electro-optic medium is typically bistable.

[Para 8] Another 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 9] 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 copending Application Serial No. 10/711,802, filed October 6, 2004 (Publication No. 2005/0151709), that such electro-wetting displays can be made bistable.

[Para 10] 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. Nevertheless, problems with the long-term image quality of these displays have prevented their widespread usage. For example, particles that make up electrophoretic displays tend to settle, resulting in inadequate service-life for these displays.

[Para 11] As noted above, electrophoretic media require the presence of a fluid. In most prior art electrophoretic media, this fluid is a liquid, but electrophoretic media can be produced using gaseous fluids; see, for example, Kitamura, T., et al., "Electrical toner movement for electronic paper-like display", IDW Japan, 2001, Paper HCS1-1, and Yamaguchi, Y., et al., "Toner display using insulative particles charged

triboelectrically", IDW Japan, 2001, Paper AMD4-4). See also U.S. Patent Publication No. 2005/0001810; European Patent Applications 1,462,847; 1,482,354; 1,484,635; 1,500,971; 1,501,194; 1,536,271; 1,542,067; 1,577,702; 1,577,703; and 1,598,694; and International Applications WO 2004/090626; WO 2004/079442; and WO 2004/001498. Such gas-based electrophoretic media appear to be susceptible to the same types of problems due to particle settling as liquid-based electrophoretic media, when the media are used in an orientation which permits such settling, for example in a sign where the medium is disposed in a vertical plane. Indeed, particle settling appears to be a more serious problem in gas-based electrophoretic media than in liquid-based ones, since the lower viscosity of gaseous suspending fluids as compared with liquid ones allows more rapid settling of the electrophoretic particles.

[Para 12] Numerous patents and applications assigned to or in the names of the Massachusetts Institute of Technology (MIT) and E Ink Corporation have recently been published describing encapsulated electrophoretic media. Such encapsulated media comprise numerous small capsules, each of which itself comprises an internal phase containing electrophoretically-mobile particles suspended in a liquid suspending 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. Encapsulated media of this type are described, for example, in U.S. Patents Nos. 5,930,026; 5,961,804; 6,017,584; 6,067,185; 6,118,426; 6,120,588; 6,120,839; 6,124,851; 6,130,773; 6,130,774; 6,172,798; 6,177,921; 6,232,950; 6,249,271; 6,252,564; 6,262,706; 6,262,833; 6,300,932; 6,312,304; 6,312,971; 6,323,989; 6,327,072; 6,376,828; 6,377,387; 6,392,785; 6,392,786; 6,413,790; 6,422,687; 6,445,374; 6,445,489; 6,459,418; 6,473,072; 6,480,182; 6,498,114; 6,504,524; 6,506,438; 6,512,354; 6,515,649; 6,518,949; 6,521,489; 6,531,997; 6,535,197; 6,538,801; 6,545,291; 6,580,545; 6,639,578; 6,652,075; 6,657,772; 6,664,944; 6,680,725; 6,683,333; 6,704,133; 6,710,540; 6,721,083; 6,724,519; 6,727,881; 6,738,050; 6,750,473; 6,753,999; 6,816,147; 6,819,471; 6,822,782; 6,825,068; 6,825,829; 6,825,970; 6,831,769; 6,839,158; 6,842,167; 6,842,279;

6,842,657; 6,864,875; 6,865,010; 6,866,760; 6,870,661; 6,900,851; 6,922,276; 6,950,200; 6,958,848; 6,967,640; 6,982,178; 6,987,603; 6,995,550; 7,002,728; 7,012,600; 7,012,735; 7,023,420; 7,030,412; 7,030,854; 7,034,783; 7,038,655; 7,061,663; 7,071,913; 7,075,502; 7,075,703; 7,079,305; 7,106,296; 7,109,968; 7,110,163; 7,110,164; 7,116,318; 7,116,466; 7,119,759; 7,119,772; 7,148,128; 7,167,155; 7,170,670; 7,173,752; 7,176,880; and 7,180,649; and U.S. Patent Applications Publication Nos. 2002/0060321; 2002/0090980; 2003/0011560; 2003/0102858; 2003/0151702; 2003/0222315; 2004/0014265; 2004/0075634; 2004/0094422; 2004/0105036; 2004/0112750; 2004/0119681; 2004/0136048; 2004/0155857; 2004/0180476; 2004/0190114; 2004/0196215; 2004/0226820; 2004/0257635; 2004/0263947; 2005/0000813; 2005/0007336; 2005/0012980; 2005/0017944; 2005/0018273; 2005/0024353; 2005/0062714; 2005/0067656; 2005/0078099; 2005/0099672; 2005/0122284; 2005/0122306; 2005/0122563; 2005/0134554; 2005/0146774; 2005/0151709; 2005/0152018; 2005/0152022; 2005/0156340; 2005/0168799; 2005/0179642; 2005/0190137; 2005/0212747; 2005/0213191; 2005/0219184; 2005/0253777; 2005/0270261; 2005/0280626; 2006/0007527; 2006/0024437; 2006/0038772; 2006/0139308; 2006/0139310; 2006/0139311; 2006/0176267; 2006/0181492; 2006/0181504; 2006/0194619; 2006/0197736; 2006/0197737; 2006/0197738; 2006/0198014; 2006/0202949; and 2006/0209388; and International Applications Publication Nos. WO 00/38000; WO 00/36560; WO 00/67110; and WO 01/07961; and European Patents Nos. 1,099,207 B1; and 1,145,072 B1.

[Para 13] 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 14] A related type of electrophoretic display is a so-called "microcell electrophoretic display". 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 assigned to Sipix Imaging, Inc.

[Para 15] 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, the aforementioned U.S. Patents Nos. 6,130,774 and 6,172,798, and U.S. Patents Nos. 5,872,552; 6,144,361; 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.

[Para 16] 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 US Patent Publication Number 2004/0226820); 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 17] Other types of electro-optic media may also be used in the displays of the present invention.

[Para 18] An electro-optic display normally comprises a layer of electro-optic material and at least two other layers disposed on opposed sides of the electro-optic 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 electro-optic 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 electro-optic layer comprises an electrode, the layer on the opposed side of the electro-optic layer typically being a protective layer intended to prevent the movable electrode damaging the electro-optic layer.

[Para 19] The manufacture of a three-layer electro-optic display normally involves at least one lamination operation. For example, in several of the aforementioned MIT and E Ink patents and applications, there is described a process for manufacturing an encapsulated electrophoretic display in which an encapsulated electrophoretic medium comprising capsules in a binder is coated on to a flexible substrate comprising indium-tin-oxide (ITO) or a similar conductive coating (which acts as one electrode of the final

display) on a plastic film, the capsules/binder coating being dried to form a coherent layer of the electrophoretic medium firmly adhered to the substrate. Separately, a backplane, containing an array of pixel electrodes and an appropriate arrangement of conductors to connect the pixel electrodes to drive circuitry, is prepared. To form the final display, the substrate having the capsule/binder layer thereon is laminated to the backplane using a lamination adhesive. (A very similar process can be used to prepare an electrophoretic display usable with a stylus or similar movable electrode by replacing the backplane with a simple protective layer, such as a plastic film, over which the stylus or other movable electrode can slide.) In one preferred form of such a process, the backplane is itself flexible and is prepared by printing the pixel electrodes and conductors on a plastic film or other flexible substrate. The obvious lamination technique for mass production of displays by this process is roll lamination using a lamination adhesive.

[Para 20] In the processes described above, the lamination of the substrate carrying the electro-optic layer to the backplane may advantageously be carried out by vacuum lamination. Vacuum lamination is effective in expelling air from between the two materials being laminated, thus avoiding unwanted air bubbles in the final display; such air bubbles may introduce undesirable artifacts in the images produced on the display. However, vacuum lamination of the two parts of an electro-optic display in this manner imposes stringent requirements upon the lamination adhesive used, especially in the case of a display using an encapsulated electrophoretic medium. The lamination adhesive should have sufficient adhesive strength to bind the electro-optic layer to the layer (typically an electrode layer) to which it is to be laminated, and in the case of an encapsulated electrophoretic medium, the adhesive should also have sufficient adhesive strength to mechanically hold the capsules together. If the electro-optic display is to be of a flexible type (and one of the important advantages of rotating bichromal member and encapsulated electrophoretic displays is that they can be made flexible), the adhesive should have sufficient flexibility not to introduce defects into the display when the display is flexed. The lamination adhesive should have adequate flow properties at

the lamination temperature to ensure high quality lamination, and in this regard, the demands of laminating encapsulated electrophoretic and some other types of electro-optic media are unusually difficult; the lamination has to be conducted at a temperature of not more than about 130°C since the medium cannot be exposed to substantially higher temperatures without damage, but the flow of the adhesive must cope with the relatively uneven surface of the capsule-containing layer, the surface of which is rendered irregular by the underlying capsules. The lamination temperature should indeed be kept as low as possible, and room temperature lamination would be ideal, but no commercial adhesive has been found which permits such room temperature lamination. The lamination adhesive should be chemically compatible with all the other materials in the display.

[Para 21] As discussed in detail in the aforementioned U.S. Patent No. 6,831,769, a lamination adhesive used in an electro-optic display should meet certain electrical criteria, and this introduces considerable problems in the selection of the lamination adhesive. Commercial manufacturers of lamination adhesives naturally devote considerable effort to ensuring that properties, such as strength of adhesion and lamination temperatures, of such adhesives are adjusted so that the adhesives perform well in their major applications, which typically involve laminating polymeric and similar films. However, in such applications, the electrical properties of the lamination adhesive are not relevant, and consequently the commercial manufacturers pay no heed to such electrical properties. Indeed, substantial variations (of up to several fold) in certain electrical properties may exist between different batches of the same commercial lamination adhesive, presumably because the manufacturer was attempting to optimize non-electrical properties of the lamination adhesive (for example, resistance to bacterial growth) and was not at all concerned about resulting changes in electrical properties.

[Para 22] However, in electro-optic displays, in which the lamination adhesive is normally located between the electrodes, which apply the electric field needed to change the electrical state of the electro-optic medium, the electrical properties of the adhesive may become crucial. As will be apparent to electrical engineers, the volume

resistivity of the lamination adhesive becomes important, since the voltage drop across the electro-optic medium is essentially equal to the voltage drop across the electrodes, minus the voltage drop across the lamination adhesive. If the resistivity of the adhesive layer is too high, a substantial voltage drop will occur within the adhesive layer, requiring an increase in voltage across the electrodes. Increasing the voltage across the electrodes in this manner is undesirable, since it increases the power consumption of the display, and may require the use of more complex and expensive control circuitry to handle the increased voltage involved. On the other hand, if the adhesive layer, which extends continuously across the display, is in contact with a matrix of electrodes, as in an active matrix display, the volume resistivity of the adhesive layer should not be too low, or lateral conduction of electric current through the continuous adhesive layer may cause undesirable cross-talk between adjacent electrodes. Also, since the volume resistivity of most materials decreases rapidly with increasing temperature, if the volume resistivity of the adhesive is too low, the performance of the display at temperatures substantially above room temperature is adversely affected. For these reasons, there is an optimum range of lamination adhesive resistivity values for use with any given electro-optic medium, this range varying with the resistivity of the electro-optic medium. The volume resistivities of encapsulated electrophoretic media are typically around 10^{10} ohm cm, and the resistivities of other electro-optic media are usually of the same order of magnitude. Accordingly, the volume resistivity of the lamination adhesive should normally be around 10^8 to 10^{12} ohm cm, or about 10^9 to 10^{11} ohm cm, at the operating temperature of the display, typically around 20°C . The lamination adhesive should also have a variation of volume resistivity with temperature which is similar to that of the electro-optic medium itself.

[Para 23] The number of commercial materials which can meet most of the previously discussed, rather disparate requirements for a lamination adhesive for use in an electro-optic display is small, and in practice a small number of water-dispersed urethane emulsions have been used for this purpose. A similar group of materials have been used as the binder for an encapsulated electrophoretic medium.

[Para 24] Furthermore, in considering the choice of a lamination adhesive for use in an electro-optic display, attention must be paid to the process by which the display is to be assembled. Most prior art methods for final lamination of solid electro-optic displays are essentially batch methods in which (typically) the electro-optic medium, a lamination adhesive and a backplane are brought together immediately prior to final assembly, and it is desirable to provide methods better adapted for mass production. 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. 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 25] The aforementioned U.S. Patent No. 6,982,178 also describes a method for testing the electro-optic medium in a front plane laminate prior to incorporation of the front plane laminate into a display. In this testing method, the release sheet is provided with an electrically conductive layer, and a voltage sufficient to change the optical state of the electro-optic medium is applied between this electrically conductive layer and the electrically conductive layer on the opposed side of the electro-optic medium. Observation of the electro-optic medium will then reveal any faults in the medium, thus avoiding laminating faulty electro-optic medium into a display, with the resultant cost of scrapping the entire display, not merely the faulty front plane laminate.

[Para 26] The aforementioned U.S. Patent No. 6,982,178 also describes a second method for testing the electro-optic medium in a front plane laminate by placing an electrostatic charge on the release sheet, thus forming an image on the electro-optic medium. This image is then observed in the same way as before to detect any faults in the electro-optic medium.

[Para 27] 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, electro-optic medium layer 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 28] The aforementioned 2004/0155857 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 29] Copending Application Serial No. 11/550,114, filed October 17, 2006 describes a so-called "inverted front plane laminate", which is a variant of the front plane laminate described in the aforementioned U.S. Patent No. 6,982,178. This inverted front plane laminate comprises, in order, at least one of a light-transmissive protective layer and a light-transmissive electrically-conductive layer; an adhesive layer; a layer of a solid electro-optic medium; and a release sheet. This inverted front plane laminate is used to form an electro-optic display having a layer of lamination adhesive between the electro-optic layer and the front electrode or front substrate; a second, typically thin layer of adhesive may or may not be present between the electro-optic layer and a backplane. Such electro-optic displays can combine good resolution with good low temperature performance. This Application also describes various methods designed for high volume manufacture of electro-optic displays using inverted front plane laminates; preferred forms of these methods are "multi-up" methods designed to allow lamination of components for a plurality of electro-optic displays at one time.

[Para 30] In view of the advantages of the assembly methods using a front plane laminate, inverted front plane laminate or double release film described in the aforementioned patents and applications, it is desirable that a lamination adhesive be capable of being incorporated into such a front plane laminate, inverted front plane laminate or double release film.

[Para 31] U.S. Patent Publication No. 2003/0025855 describes (see especially Paragraphs [0162] to [0191] thereof) certain polyurethane dispersions especially formulated for use in electro-optic displays.

[Para 32] The aforementioned U.S. Patent No. 7,012,735 describes an electro-optic display comprising first and second substrates, and an adhesive layer and a layer of electro-optic material disposed between the first and second substrates, the adhesive layer comprising a mixture of a polymeric adhesive material and a salt or other polyelectrolyte. The salt may be for example a tetraalkylammonium salt such as tetrabutylammonium chloride, or potassium acetate. (It has also been found that tetrabutylammonium hexafluorophosphate may advantageously be substituted for the chloride on a mole-for-mole basis.) The polyelectrolyte may be a polymeric material, for example the sodium salt of polyacrylic acid. The salt or polyelectrolyte serves to vary the volume resistivity of the adhesive material but typically does not substantially affect the mechanical properties of this material.

[Para 33] This patent also describes an electrophoretic medium comprising a plurality of capsules, each of the capsules comprising a capsule wall, a suspending fluid encapsulated within the capsule wall and a plurality of electrically charged particles suspended in the suspending fluid and capable of moving therethrough on application of an electric field to the medium, the medium further comprising a binder surrounding the capsules, the binder comprising a mixture of a polymeric adhesive material and a salt or other polyelectrolyte. The salt or polyelectrolyte may be any of those previously described.

[Para 34] The displays and media described in the aforementioned U.S. Patent No. 7,012,735 give good results. However, in at least some cases, there are concerns that addition of ionic species to adhesives and/or binders used in electro-optic displays might possibly cause corrosion problems in certain materials used in electro-optic displays, in particular the backplanes thereof which are typically in direct contact with

the lamination adhesive. This invention relates to modification of polyurethane adhesives to render them more suitable for use in electro-optic displays.

[Para 35] As already mentioned, the lamination processes used to manufacture electro-optic displays impose stringent requirements upon both the mechanical and electrical properties of the lamination adhesive. In addition, there are other constraints which the lamination adhesive must satisfy. Void growth may be encountered in various types of solid electro-optic displays, and to ensure a high quality display, it is essential that the final display be free from voids, since such voids produce visible defects in images written on the display, as illustrated below. To ensure that the final display is free from voids, it is essential that both the lamination to form the front plane laminate (when effected) and the final lamination to the backplane be carried out without the formation of voids. It is also necessary that the final display be able to withstand substantial temperature changes (such as may occur, for example, when a portable computer or personal digital assistant is removed from an air-conditioned car to outdoor sun on a hot day) without inducing or aggravating the formation of voids, since it has been found that some displays, which initially appear essentially free from voids, can develop objectionable voids when exposed to such temperature changes. This phenomenon may be termed "void re-growth".

[Para 36] The aforementioned U.S. Patent No. 7,173,752 describes an electro-optic display generally similar to those described in the aforementioned U.S. Patent No. 7,012,735, but in which the adhesive layer is doped with a hydroxyl containing polymer having a number average molecular weight not greater than about 5000, a preferred dopant being poly(ethylene glycol). The aforementioned U.S. Patent No. 7,012,735 also describes a polymer dispersed electro-optic medium in which the continuous phase comprises a mixture of a polymeric adhesive material and a hydroxyl containing polymer having a number average molecular weight not greater than about 5000. The hydroxyl containing polymer serves the same functions as the dopants described in the aforementioned U.S. Patent No. 7,012,735 but is less likely to lead to corrosion problems and improves the operating temperature range of the display. The hydroxyl

containing polymer doped adhesive may be incorporated into a front plane laminate or double release film as described above.

[Para 37] The aforementioned U.S. Patent No. 7,173,752 also describes an electro-optic display generally similar to those described in the aforementioned 2005/0007653, but in which the adhesive layer is cross-linked with a thermal cross-linking agent. Such cross-linking helps to avoid void regrowth problems. The cross-linking affects the volume resistivity of the adhesive, but this resistivity can be readjusted to a desirable level by controlling the level of dopant.

[Para 38] In flexible electro-optic displays, there is one further problem which must be taken into account when choosing a lamination adhesive, namely the problem known as "creep". When a flexible display is repeatedly flexed or rolled up, there is a tendency for the lamination adhesive layer to undergo plastic flow, such that when the display is restored to a notionally flat state, the display does not in fact lie flat but is warped. Creep is a particular problem in color displays where a color filter array is present on the opposed side of the electro-optic layer from the backplane, since creep in the adhesive layer can lead to relative movement between the backplane and the color filter array, thus leading to mis-alignment between elements of the color filter array and electrodes on the backplane with which these elements are intended to be aligned, and thus adversely affecting the color of an image written on the display.

[Para 39] It has now been found that creep in flexible displays can be reduced or eliminated by using a lamination adhesive having a storage modulus at least as great as a predetermined value, and this invention relates to electro-optic displays having such a lamination adhesive. The necessary storage modulus can be provided by cross-linking of the lamination adhesive or in other ways.

[Para 40] This invention provides a flexible electro-optic display comprising:
a layer of solid electro-optic material capable of changing at least one optical characteristic upon application of an electric field thereto;

a backplane comprising at least one electrode arranged to apply an electric field to the layer of electro-optic material; and

an adhesive layer disposed between the layer of electro-optic material and the backplane and adhesively securing the layer of electro-optic material to the backplane,

the adhesive layer having a storage modulus (G') of at least about 10^4 Pascals at 10^{-3} Hz and 70°C .

[Para 41] The term "flexible" is used herein consistently with its normal meaning in the display art to refer to a display which is capable of being bent repeatedly without macroscopic damage to the display. (For example, a flexible medium could be incorporated into a wrist band which is bent perhaps several times a day as the wrist band is put on and removed from a user's wrist.) Such displays typically have front substrates and backplanes formed from polymeric films having thicknesses not greater than about $300\ \mu\text{m}$, and preferably not greater than about $100\ \mu\text{m}$. It is of course necessary to ensure that the conductors, electrodes and any non-linear devices, especially transistors, present on the backplane can withstand repeated flexing, and it may be desirable to use non-linear devices comprising conductive polymers.

[Para 42] The displays of the present invention may be "rollable", a term which is used herein to denote a display which can be moved repeatedly between a rolled position (in which the display is wrapped around a mandrel or, for example, the casing of a cellular telephone), and an operating position, in which an image can be viewed on the display. Such a rollable display requires that the display be moved from its rolled position to its operating position each time the display is to be used, and hence the display must be able to withstand a large number of unrolling/re-rolling cycles during its operating lifetime. The number of such cycles required in a rollable display is much greater than for a flexible display; for example, a flexible display used on a wrist strap may typically undergo only two (or perhaps four) flexures per day as the wrist strap is placed on and removed from the user's wrist, whereas a rollable display used to provide a large display screen for the display of electronic mail received by a cellular telephone

might undergo 20 or more unrolling/re-rolling cycles per day as the user receives numerous electronic mail messages.

[Para 43] The adhesive layer used in the display of the present invention must have a storage modulus of at least about 10^4 Pascals at 10^{-3} Hz and 70°C . The storage modulus is desirably at least about 3×10^4 Pascals, and preferably at least about 6×10^4 Pascals at 10^{-3} Hz and 70°C . The modulus is measured at 70°C , which is typically the upper end of the operating range of electrophoretic and many other electro-optic displays. The modulus of an adhesive layer normally decreases as its temperature rises. Accordingly, creep is more likely to occur at high temperatures. Hence it is appropriate to measure creep at the upper end of the operating range. Should the display be intended to operate above 70°C , it will of course be appropriate to use an adhesive layer having the necessary modulus at the upper end of the actual operating temperature range of the display. In displays having two or more adhesive layers, for example displays using the double release films and inverted front plane laminates described above, desirably both or all of the adhesive layers should meet the storage modulus criterion set out above.

[Para 44] As is well known to those skilled in electro-optic display technology, most known processes for producing adhesive layers in such displays require that the coating of the adhesive layer be effected with the adhesive in a fluid form having a viscosity not greater than about 10000, and desirably not greater than about 5000, cP, although these limits are subject to some variation depending upon the exact coating technique employed. Accordingly, in the production of the displays of the present invention, it will typically be necessary to coat the adhesive layer in a fluid form and thereafter to dry, polymerize, cross-link or otherwise treat the adhesive material to provide the necessary storage modulus in the completed display. Typically, the curing of the adhesive material is effected by including a cross-linking agent therein. The cross-linking agent may be an ultra-violet activated cross-linking agent. Alternatively, the cross-linking agent may be a thermally-activated cross-linking agent and may comprise an epoxy group, which may be in the form of a glycidyl grouping (i.e., an epoxymethyl grouping). The cross-linking agent may also comprise a tertiary amine.

For example, the cross-linking agent may comprise N,N-diglycidylaniline, which may be present in the adhesive layer in a concentration of at least about 5,000, and preferably at least about 10,000, parts per million by weight. Other useful types of cross-linking agents include epoxy ethers of alkyl or cycloalkyl polyols having at least two hydroxyl groups, and polymers having a main chain and a plurality of epoxy groups depending from the main chain. Specific useful cross-linking agents include 1,4-cyclohexanedimethanol diglycidyl ether, neopentyl glycol diglycidyl ether, O,O,O-triglycidylglycerol, and homopolymers and copolymers of glycidyl methacrylate.

[Para 45] It should be noted that, although in most cases it will be desirable to carry out curing of the adhesive in the final display, in some cases, as for example when the adhesive layer is incorporated into a front plane laminate or similar structure, it may be possible to partially or completely cure the adhesive before the final assembly of the display.

[Para 46] The adhesive layer may comprise a polyurethane as described in the aforementioned U.S. Patent No. 7,173,752, or a polyacrylate. Other types of adhesive material may also be used.

[Para 47] This invention also provides a process for producing a flexible electro-optic display, the process comprising:

providing an assembly comprising a layer of solid electro-optic material capable of changing at least one optical characteristic upon application of an electric field thereto; a flexible backplane comprising at least one electrode arranged to apply an electric field to the layer of electro-optic material; and an adhesive layer disposed between the layer of electro-optic material and the backplane and adhesively securing the layer of electro-optic material to the backplane, the adhesive layer comprising a cross-linking agent capable of cross-linking the adhesive layer, and

exposing the adhesive layer to conditions effective to activate the cross-linking agent, thereby cross-linking the adhesive layer to produce an adhesive layer having a storage modulus (G') of at least about 10^4 Pascals at 10^{-3} Hz and 70°C .

[Para 48] This invention also provides an electro-optic display comprising:
a layer of solid electro-optic material capable of changing at least one optical characteristic upon application of an electric field thereto;
a flexible backplane comprising at least one electrode arranged to apply an electric field to the layer of electro-optic material; and
an adhesive layer disposed between the layer of electro-optic material and the backplane and adhesively securing the layer of electro-optic material to the backplane, the adhesive layer having a storage modulus (G') of at least about 10^4 Pascals at 10^{-3} Hz and 70°C .

[Para 49] Figure 1 of the accompanying drawings is a graph showing the variation with frequency of the storage modulus of cross-linked and non-cross-linked adhesive materials, as measured at 50°C , in the experiments described in the Example below.

[Para 50] Figure 2 is a graph showing the variation with frequency of the loss modulus of cross-linked and non-cross-linked adhesive materials, calculated for 50°C , in the experiments described in the Example below.

[Para 51] Figure 3 is a graph showing the variation with frequency of $\tan \Delta$ as calculated from the data shown in Figures 1 and 2.

[Para 52] Figures 4 to 6 are graphs similar to Figures 1 to 3 respectively but showing data calculated for 70°C in the experiments described in the Example below.

[Para 53] As already indicated, the present invention relates to the use, in a flexible electro-optic display, of an adhesive layer having a high storage modulus. This invention also provides a process for producing an electro-optic display with such an adhesive layer, an electro-optic display produced by such a process, and components (namely a front plane laminate, inverted front plane and double release film) useful for forming such displays. The use of such an adhesive layer has been found effective in reducing or eliminating creep in flexible electro-optic displays.

[Para 54] When a thermal cross-linking agent is used in the present display, the cross-linking agent may be any of those described in the aforementioned U.S. Patent

No. 7,173,752; other cross-linking agents may also be used. The adhesive used may be a polyurethane adhesive such as those described in the aforementioned U.S. Patent No. 7,173,752, or a polyacrylate. Any of the dopants previously mentioned may be used to adjust the volume resistivity of the adhesive to the desired range.

[Para 55] The displays of the present invention may incorporate any of the types of electro-optic media previously described. Thus, for example, the present displays may include an encapsulated electrophoretic, microcell electrophoretic or polymer dispersed electrophoretic medium. The present displays may be manufactured using front plane laminates or double release films as described above.

[Para 56] The following Example is now given, though by way of illustration only, to show how cross-linking affects adhesives used in electro-optic displays.

[Para 57] Example

[Para 58] Tests were conducted on cross-linked and non-cross-linked forms of a custom polyurethane lamination adhesive of the type described in U.S. Patent Publication No. 2005/0107564 (this material had a molar mass of 52,000, with M_w/M_n equal to 2.7-3.2 and was in the form of an aqueous latex). The cross-linking agent used was diglycidyl aniline at a concentration of 20,000 ppm. To ensure complete cross-linking, the cross-linked samples were stored at 60°C for 120 hours before testing.

[Para 59] Cross-linked and non-cross-linked samples of the adhesive were coated at a thickness of about 17 μm on to 2 inch (51 mm) squares of metal foil. For the dynamic mechanical analysis (DMA) measurements described below, the layer of lamination adhesive was removed from the metal foil, and folded to produce a layer of adhesive approximately 600 μm thick, which was then subjected to DMA testing using a TA Instruments DMA tester. The adhesive samples were subjected to DMA testing at a constant stress of 1000 Pa, and a small deformation (0.01 per cent of strain) was applied over a wide frequency range, and at varying temperatures, to determine the storage (elastic) modulus (G') of the samples; the loss (viscous) modulus (G'') and the ratio ($\tan \Delta$) of the two moduli. Figures 1 to 3 of the accompanying drawings show the

storage moduli, loss moduli and $\tan \Delta$ respectively of the two samples as a function of frequency at 50°C, while Figures 4 to 6 show the same parameters at 70°C; in each case, the measured moduli and temperatures were adjusted to 50°C and 70°C by standard techniques well known to those skilled in effecting DMA measurements. In each Figure, the open circles represent the non-cross-linked samples and the closed circles the cross-linked samples.

[Para 60] It will be seen from the Figures that the storage modulus of the non-cross-linked samples drops dramatically below about 1 Hz, at both 50°C and 70°C, whereas the storage modulus of the cross-linked samples does not undergo this dramatic decline. Consequently, at 10^{-2} Hz, the storage modulus of the cross-linked sample is approximately two orders of magnitude greater than that of the non-cross-linked sample. The variation of the loss modulus with frequency is similar but less dramatic. Accordingly, the value of $\tan \Delta$ of the non-cross-linked samples shows an abrupt increase below about 1 Hz, whereas there is no such increase in the cross-linked samples.

[Para 61] Based upon the data in the Figures, the cross-linked polyurethane adhesive is useful in flexible displays and exhibits low creep whereas the non-cross-linked polyurethane would be subject to excessive creep. These results have been confirmed empirically.

CLAIMS

1. A flexible electro-optic display comprising:
a layer of solid electro-optic material capable of changing at least one optical characteristic upon application of an electric field thereto;
a backplane comprising at least one electrode arranged to apply an electric field to the layer of electro-optic material; and
an adhesive layer disposed between the layer of electro-optic material and the backplane and adhesively securing the layer of electro-optic material to the backplane,
the adhesive layer having a storage modulus (G') of at least about 10^4 Pascals at 10^{-3} Hz and 70°C .
2. An electro-optic display according to claim 1 wherein the backplane comprises a polymeric film having a thickness not greater than about $300\ \mu\text{m}$, the display further comprising a front plane lying on the opposed side of the electro-optic material from the backplane and comprising a polymeric film having a thickness not greater than about $300\ \mu\text{m}$.
3. A display according to claim 2 wherein the backplane and front plane each comprise a polymeric film having a thickness not greater than about $100\ \mu\text{m}$.
4. A display according to claim 1 wherein the backplane comprises a plurality of non-linear devices comprising conductive polymers.
5. A display according to claim 1 wherein the adhesive layer has a storage modulus (G') of at least about 3×10^4 Pascals at 10^{-3} Hz and 70°C .
6. A display according to claim 5 wherein the adhesive layer has a storage modulus (G') of at least about 6×10^4 Pascals at 10^{-3} Hz and 70°C .
7. A display according to claim 1 having a second layer of adhesive material disposed on the opposed side of the electro-optic material from the backplane, the second layer of adhesive having a storage modulus (G') of at least about 10^4 Pascals at 10^{-3} Hz and 70°C .

8. A display according to claim 1 wherein the adhesive layer comprises a polyurethane.

9. A display according to claim 1 wherein the adhesive layer comprises a cross-linked polymer.

10. A display according to claim 1 wherein the adhesive layer has been cross-linked with a thermally-activated cross-linking agent.

11. A display according to claim 10 wherein the adhesive layer has been cross-linked with a thermally-activated cross-linking agent containing an epoxy group.

12. A display according to claim 10 wherein the adhesive layer has been cross-linked with N, N-diglycidylaniline.

13. A display according to claim 9 wherein the adhesive layer has been cross-linked with an ultra-violet activated cross-linking agent.

14. A process for producing a flexible electro-optic display, the process comprising:

providing an assembly comprising a layer of solid electro-optic material capable of changing at least one optical characteristic upon application of an electric field thereto; a flexible backplane comprising at least one electrode arranged to apply an electric field to the layer of electro-optic material; and an adhesive layer disposed between the layer of electro-optic material and the backplane and adhesively securing the layer of electro-optic material to the backplane, the adhesive layer comprising a cross-linking agent capable of cross-linking the adhesive layer, and

exposing the adhesive layer to conditions effective to activate the cross-linking agent, thereby cross-linking the adhesive layer to produce an adhesive layer having a storage modulus (G') of at least about 10^4 Pascals at 10^{-3} Hz and 70°C .

15. A process according to claim 14 wherein the adhesive layer produced by cross-linking has a storage modulus (G') of at least about 3×10^4 Pascals at 10^{-3} Hz and 70°C .

16. A process according to claim 15 wherein the adhesive layer produced by cross-linking has a storage modulus (G') of at least about 6×10^4 Pascals at 10^{-3} Hz and 70°C .
17. A process according to claim 14 wherein the adhesive layer comprises a polyurethane.
18. A process according to claim 14 wherein the adhesive layer is cross-linked with a thermally-activated cross-linking agent.
19. A process according to claim 18 wherein the adhesive layer is cross-linked with a thermally-activated cross-linking agent containing an epoxy group.
20. A process according to claim 19 wherein the adhesive layer is cross-linked with N, N-diglycidylaniline.

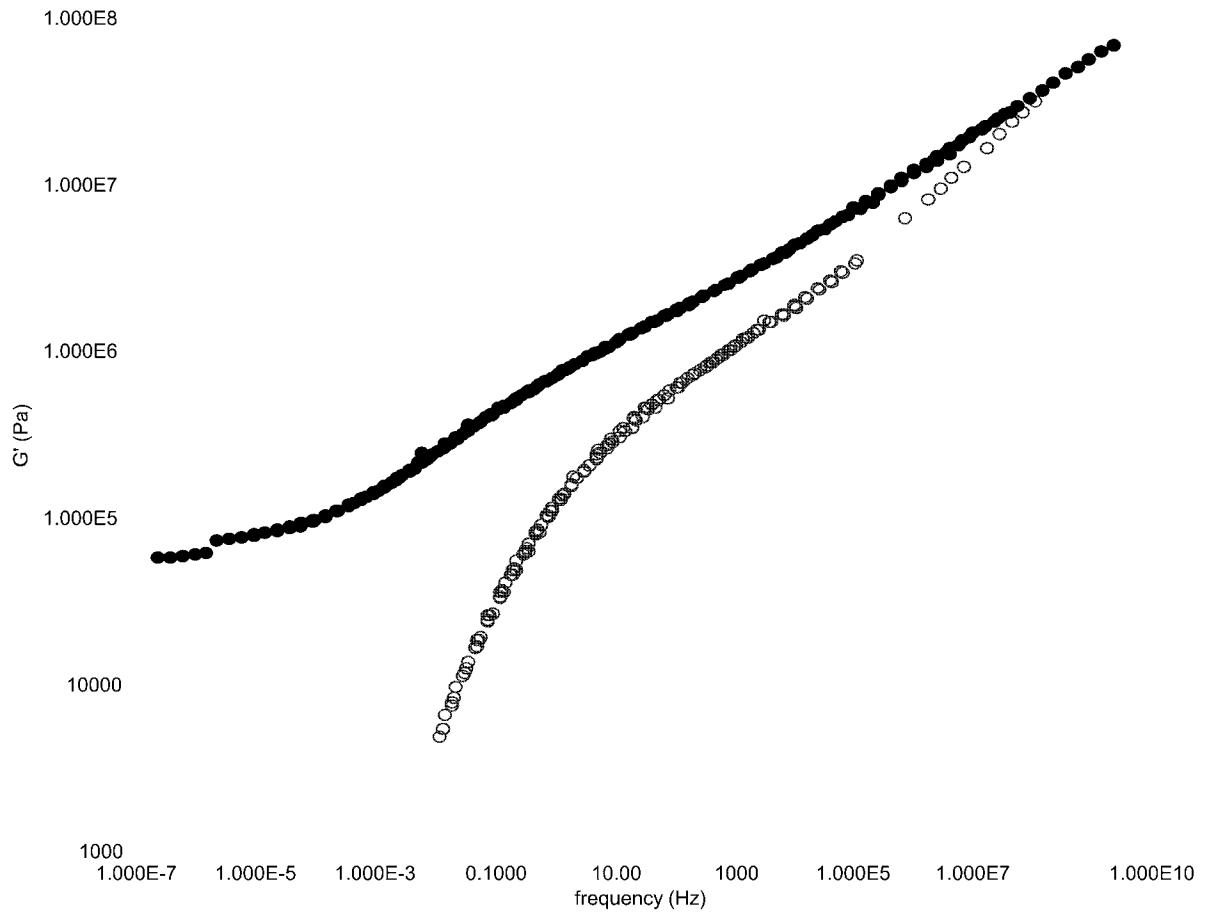


Fig. 1

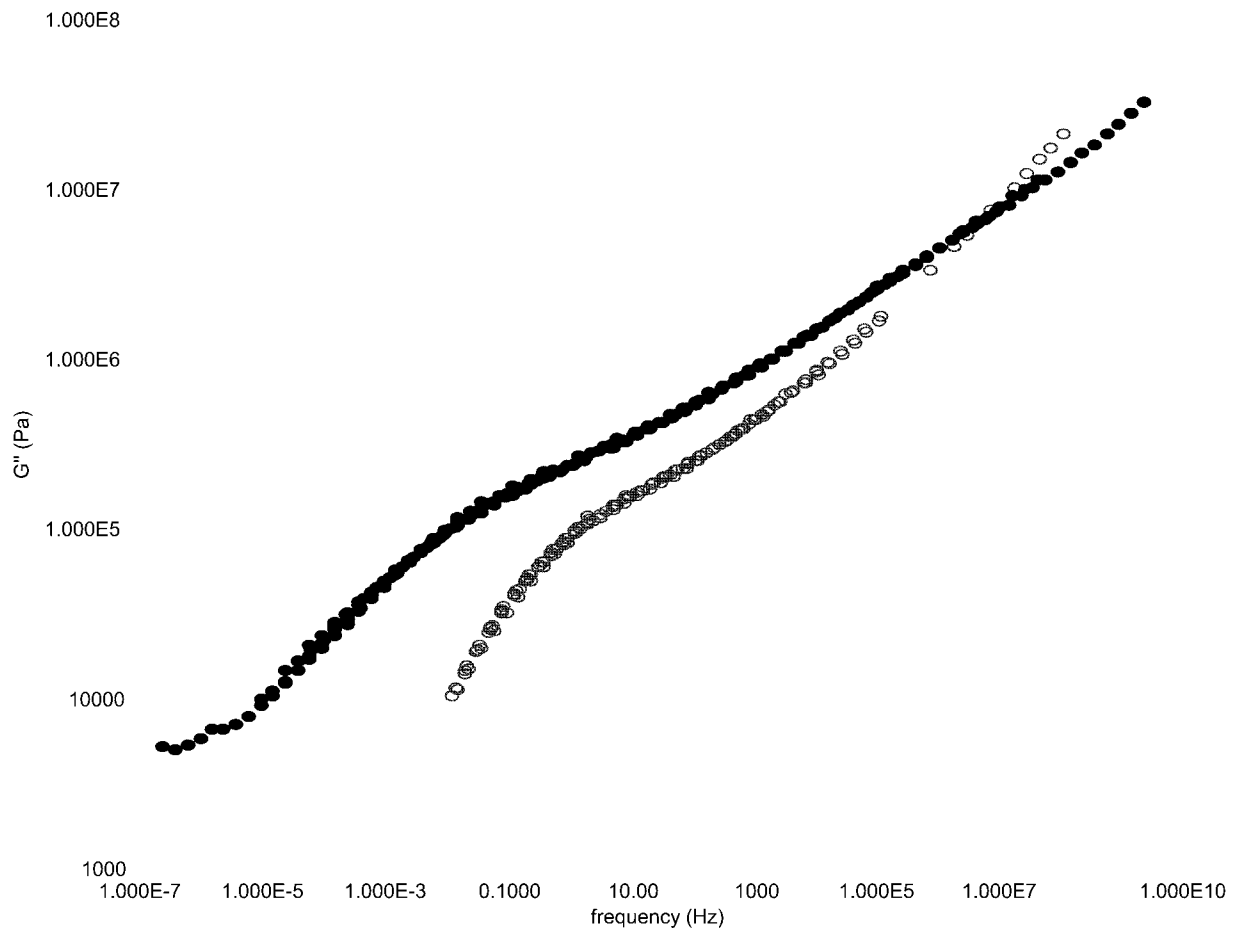


Fig. 2

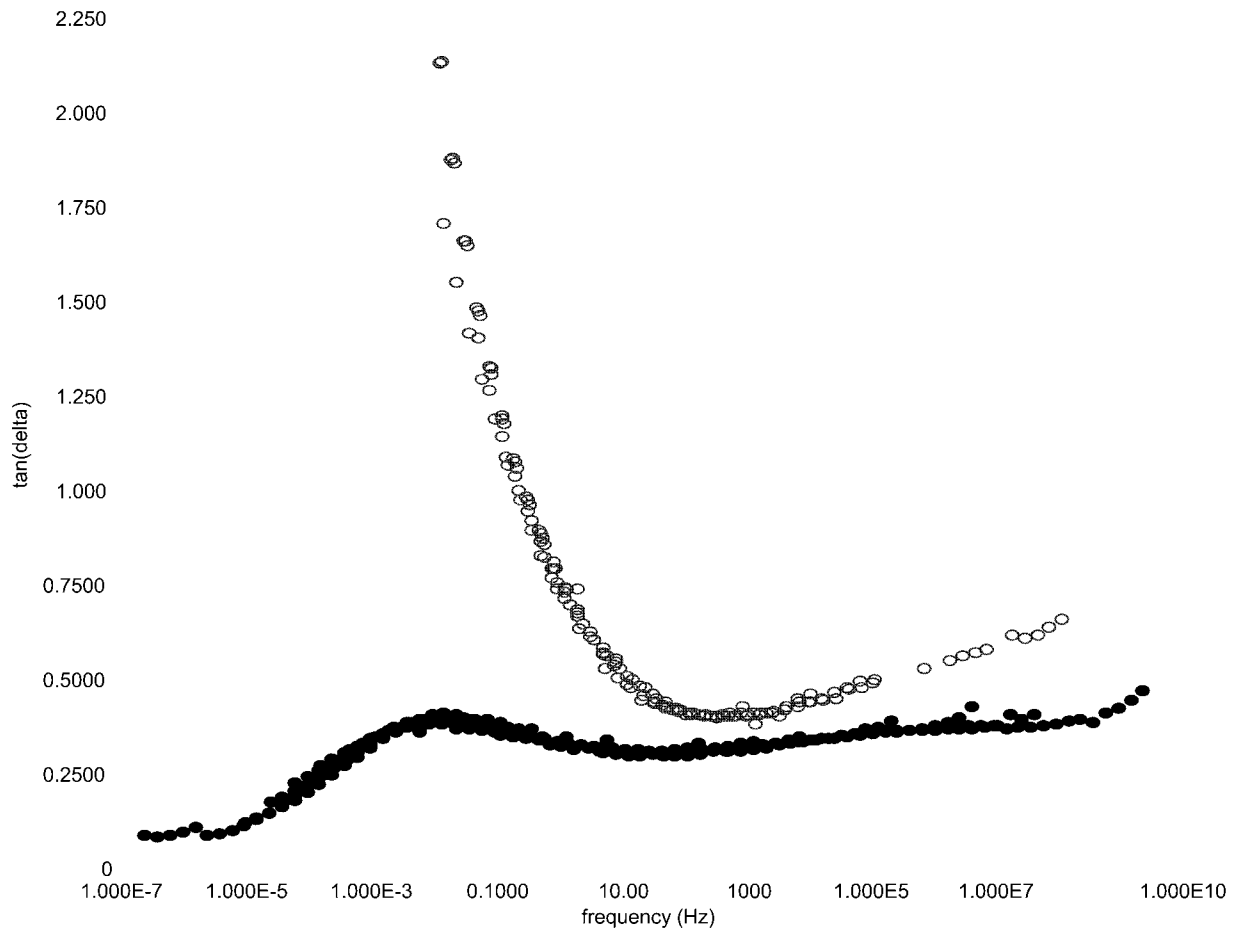


Fig. 3

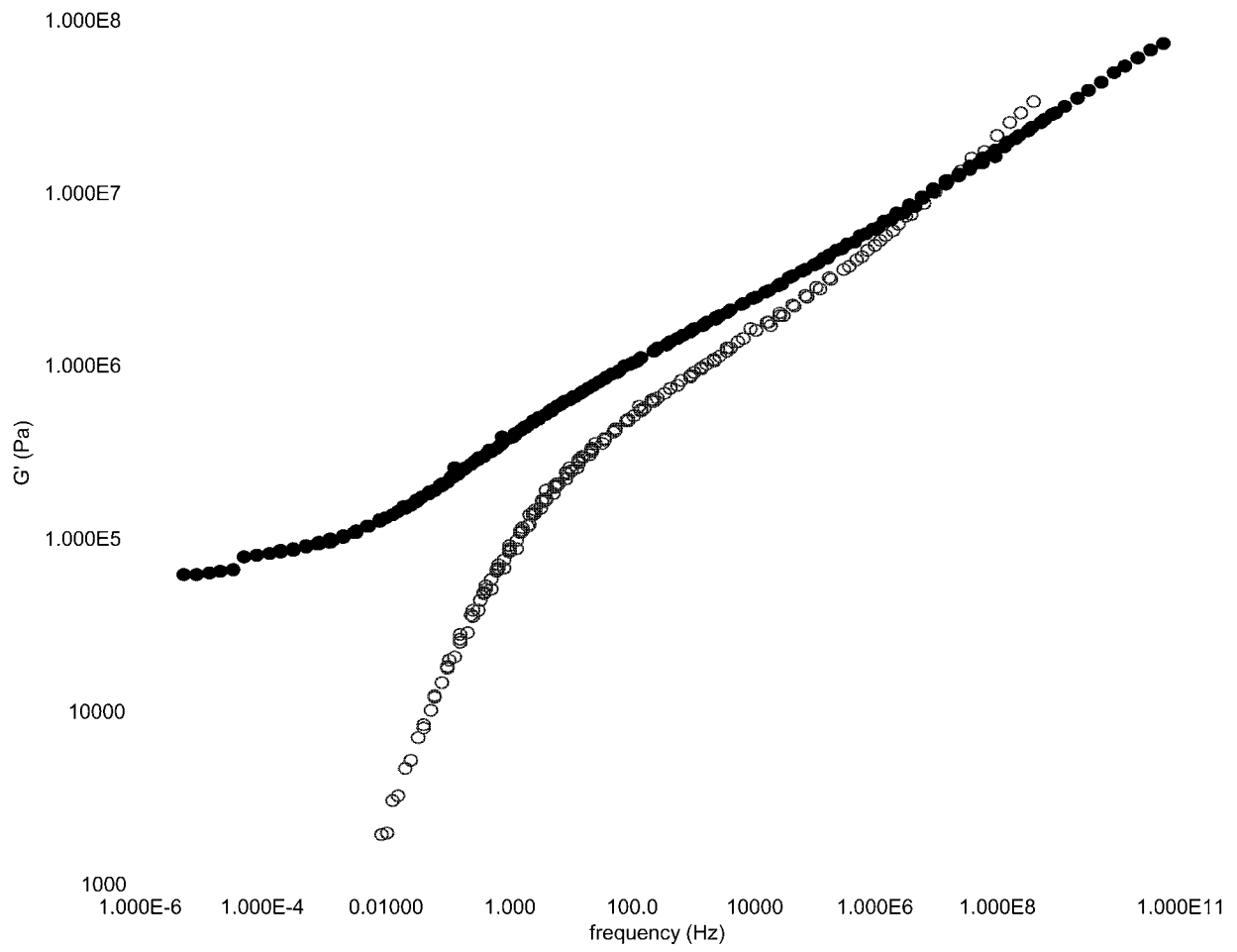


Fig. 4

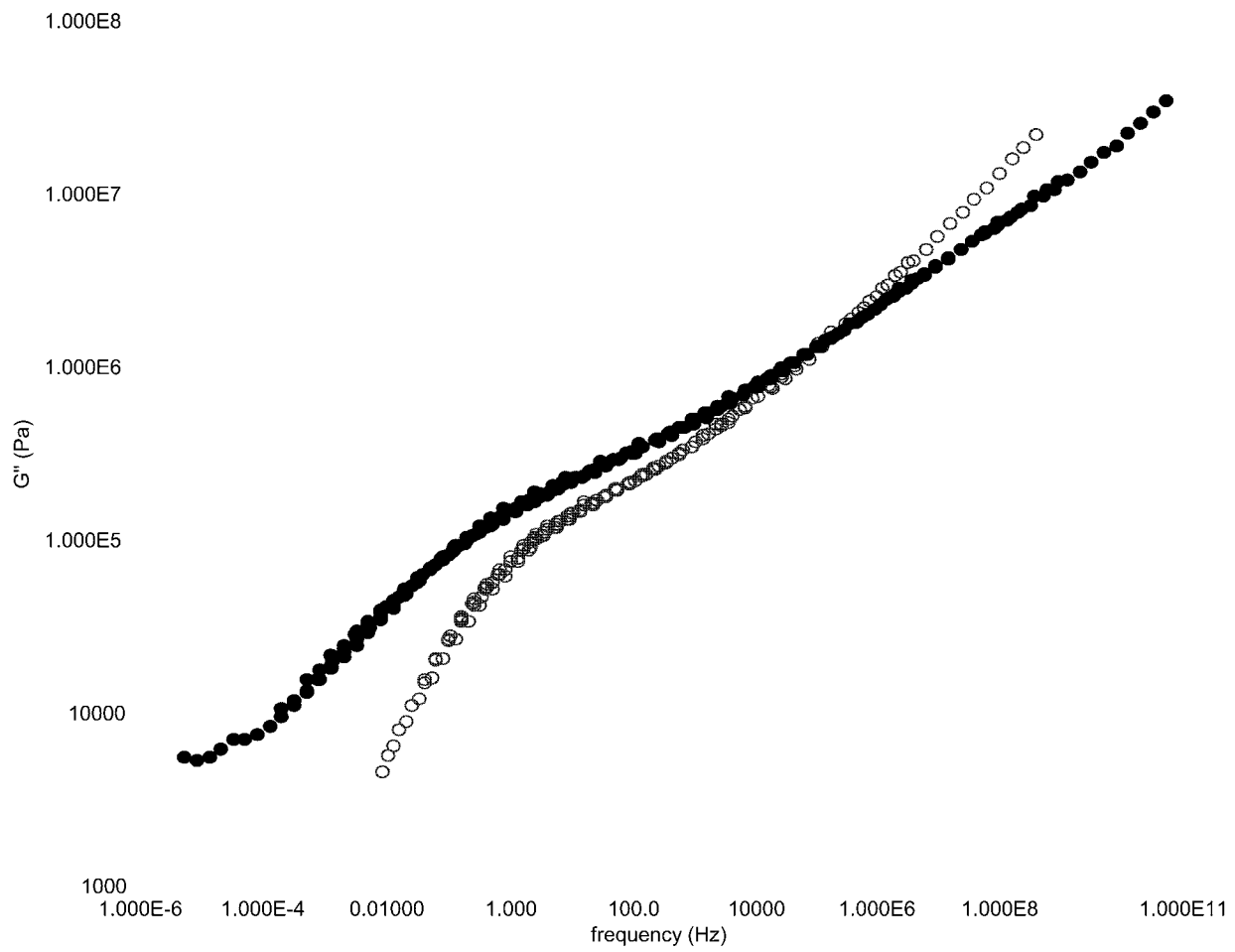


Fig. 5

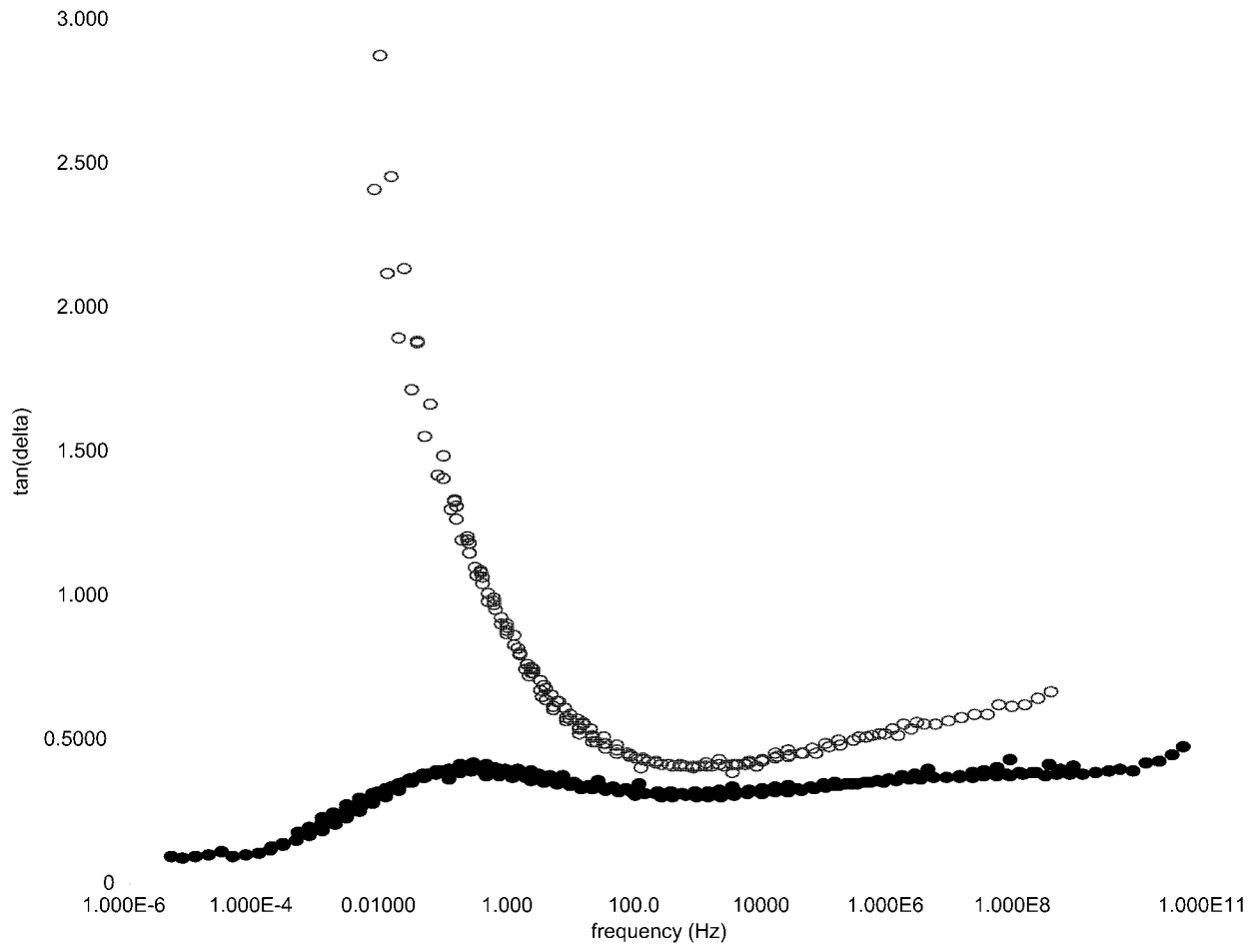


Fig. 6