



- (51) International Patent Classification:  
*B32B 17/10* (2006.01) *B32B 27/30* (2006.01)
- (21) International Application Number:  
PCT/US2017/016898
- (22) International Filing Date:  
8 February 2017 (08.02.2017)
- (25) Filing Language: English
- (26) Publication Language: English
- (30) Priority Data:  
15/051,183 23 February 2016 (23.02.2016) US
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(81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

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(54) Title: INTERLAYERS HAVING ENHANCED OPTICAL PROPERTIES

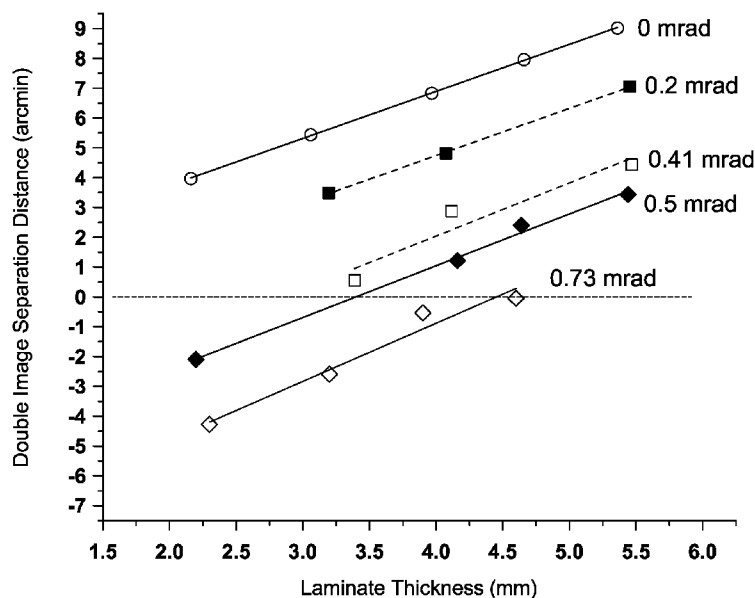


FIG. 12

(57) Abstract: A tapered interlayer comprising at least one polymer layer and a having tapered zone with an overall wedge angle of not more than 0.85 mrad is provided. The refractive index of the polymer layer is 1.480 or higher. Multiple layer panels formed from such interlayers may exhibit desirable optical properties, including, for example, less image "ghosting," when used as part of a heads-up-display (HUD) display panel for use in automotive and aircraft applications.

**Published:**

— *with international search report (Art. 21(3))*

## **INTERLAYERS HAVING ENHANCED OPTICAL PROPERTIES**

### **CROSS-REFERENCE TO RELATED APPLICATIONS**

[0001] This application is a Continuation-in-Part of U.S. Patent Application Serial No. 14/563,011 filed on December 8, 2014, which is currently pending, the disclosure of which is incorporated herein by reference in its entirety.

### **BACKGROUND**

#### **1. Field of the Invention**

[0002] This disclosure relates to polymer resins and, in particular, to polymer resins suitable for use in polymer interlayers, including those utilized in multiple layer panels.

#### **2. Description of Related Art**

[0003] Poly(vinyl butyral) (PVB) is often used in the manufacture of polymer sheets that can be used as interlayers in multiple layer panels, including, for example, light-transmitting laminates such as safety glass or polymeric laminates. PVB is also used in photovoltaic solar panels to encapsulate the panels which are used to generate and supply electricity for commercial and residential applications.

[0004] Safety glass generally refers to a transparent laminate that includes at least one polymer sheet, or interlayer, disposed between two sheets of glass. Safety glass is often used as a transparent barrier in architectural and automotive applications, and one of its primary functions is to absorb energy resulting from impact or a blow without allowing penetration of the object through the glass and to keep the glass bonded even when the applied force is sufficient to break the glass. This prevents dispersion of sharp glass shards, which minimizes injury and damage to people or objects within an enclosed area. Safety glass may also provide other benefits, such as a reduction in ultraviolet (UV) and/or infrared (IR) radiation, and it may also enhance the aesthetic appearance of window openings through addition of

color, texture, and the like. Additionally, safety glass with desirable acoustic properties has also been produced, which results in quieter internal spaces.

[0005] Laminated safety glass has been used in vehicles equipped with heads-up display (HUD) systems, which project an image of an instrument cluster or other important information to a location on the windshield at the eye level of the vehicle operator. Such a display allows the driver to stay focused on the upcoming path of travel while visually accessing dash board information. When projected onto a flat windshield having uniform and consistent thickness, an interfering double, or reflected “ghost,” image is created due to the differences in the position of the projected image as it is reflected off the inside and outside surfaces of the glass.

[0006] One method of addressing these ghost images has been to include a coating, such as a dielectric coating, on one of the surfaces of the windshield between the glass and the polymeric interlayer. The coating is designed to produce a third ghost image at a location very close to the primary image, while significantly reducing the brightness of the secondary image, so that the secondary image appears to blend into the background. Unfortunately, at times, the effectiveness of such a coating can be limited and the coating itself may interfere with the adhesion of the polymeric interlayer to the glass substrates, resulting in optical distortion and other issues.

[0007] At the same time, due in part to desire for increased fuel efficiency for vehicles, demand has grown for multiple layer panels that are lighter in weight than traditional panels. In general, the multiple layer panels utilized in a vehicle, which may include, for example, its windshield, side windows, and other panels, make up a notable portion of the total weight of the vehicle – at times up to 5 percent. As a result, reduction in the total weight of these panels would generally result in a decrease in the weight of the vehicle and a corresponding increase in fuel efficiency. Most of the weight in these panels is not in the interlayer, but is instead attributable to the substrates, which typically include glass.

[0008] Therefore, a need exists for polymeric interlayers suitable for use in HUD applications that can also be utilized in lightweight multiple layer panels. Such

interlayers should exhibit desirable optical, acoustic, and visual properties, while reducing double image separation and permitting usage of thinner substrates.

## SUMMARY

[0009] One embodiment of the present invention relates a tapered interlayer comprising at least one polymer layer comprising a poly(vinyl acetal) resin and at least one plasticizer. The polymer layer has a refractive index of at least about 1.480, and the interlayer comprises a tapered zone having an overall wedge angle of not more than 0.85 mrad.

[0010] Another embodiment of the present invention relates to a multiple layer panel comprising a pair of rigid substrates; and an interlayer disposed between the substrates. The interlayer comprises a tapered zone having an overall wedge angle of less than 0.85 mrad and the equivalent refractive index of the multiple layer panel is at least 0.010 higher than the refractive index of each of the rigid substrates.

[0011] Still another embodiment of the present invention relates to a multiple layer panel comprising a pair of rigid substrates and a tapered interlayer disposed between the substrates. The tapered interlayer comprises at least one polymeric resin. The refractive index of the interlayer is at least 5 percent higher than the refractive index of each of the rigid substrates.

## BRIEF DESCRIPTION OF THE DRAWINGS

[0012] Various embodiments of the present invention are described in detail below with reference to the attached drawing Figures, wherein:

[0013] FIG. 1 is a cross-sectional view of a tapered interlayer configured in accordance with one embodiment of the present invention, where various features of the tapered interlayer are labeled for ease of reference;

[0014] FIG. 2 is a cross-sectional view of a tapered interlayer having a tapered zone that extends over the entire width of the interlayer, where the entire tapered zone has a constant wedge angle and a linear thickness profile;

[0015] FIG. 3 is a cross-sectional view of a tapered interlayer having a tapered zone that extends over part of the width of the interlayer and a flat edge zone that extends over part of the width of the interlayer, where the tapered zone includes a constant angle zone and a variable angle zone;

[0016] FIG. 4 is a cross-sectional view of a tapered interlayer having a tapered zone that extends over part of the width of the interlayer and two flat edge zones that extend over part of the width of the interlayer, where the tapered zone includes a constant angle zone and two variable angle zones;

[0017] FIG. 5 is a cross-sectional view of a tapered interlayer having a tapered zone that extends over part of the width of the interlayer and two flat edge zones that extend over part of the width of the interlayer, where the tapered zone is formed entirely of a variable angle zone having a curved thickness profile;

[0018] FIG. 6 is a cross-sectional view of a tapered interlayer having a tapered zone that extends over the entire width of the interlayer, where the tapered includes three constant angle zones spaced apart from one another by two variable angle zones;

[0019] FIG. 7 is a cross-sectional view of a tapered interlayer having a tapered zone that extends over part of the width of the interlayer and two flat edge zones that extend over part of the width of the interlayer, where the tapered zone includes three constant angle zones and four variable angle zones;

[0020] FIG. 8a is a plan view of a tapered interlayer configured for use in a vehicle windshield, where the thickness profile of the interlayer is similar to the thickness profile of the interlayer depicted in FIG. 2;

[0021] FIG. 8b is a cross-sectional view of the interlayer of FIG. 8a, showing the thickness profile of the interlayer;

[0022] FIG. 9 is a schematic diagram of an apparatus used to measure the double image separation distance of a multiple layer panel;

[0023] FIG. 10 is an example of a projection image formed when measuring the double image separation distance of a multiple layer panel using the apparatus of FIG. 9;

[0024] FIG. 11 is an example of a profile formed by analyzing the projection image shown in FIG. 10 by plotting the number of pixels as a function of intensity;

[0025] FIG. 12 is a graphical depiction of the results of the double image separation distance analyses performed on several Comparative and Disclosed samples as described in Example 11, particularly showing the double image separation distance of several samples of varying wedge angle as a function of total thickness;

[0026] FIG. 13 is graphical depiction of the results of the double image separation distance analyses performed on several Comparative and Disclosed samples as described in Example 11, particularly showing the double image separation of several samples of varying glass configuration as a function of wedge angle; and

[0027] FIG. 14 is a graphical depiction of the results of the double image separation distance analysis performed in Example 13, particularly showing the double image separation distance of several panels utilizing panels having varying glass thickness as a function of the equivalent refractive index.

## **DETAILED DESCRIPTION**

[0028] Resin compositions, layers, and interlayers according to various embodiments of the present invention can comprise one or more thermoplastic polymers and a refractive index (RI) balancing agent. As used herein, the term “refractive index balancing agent” or “RI balancing agent” refers to any component or additive included in the composition, layer, or interlayer for adjusting the refractive index of at least one of the resins or layers. The RI balancing agent may increase or reduce the refractive index of at least one of the resins or layers within an interlayer, which may improve the optical properties of the interlayer, including mottle, haze, and/or clarity, as compared to an identical interlayer formed without an RI balancing agent.

[0029] As used herein, the terms “polymer resin composition” and “resin composition” refer to compositions including one or more polymer resins. Polymer compositions may optionally include other components, such as plasticizers and/or other additives. As used herein, the terms “polymer resin layer,” “polymer layer,” and “resin layer” refer to one or more polymer resins, optionally combined with one or more plasticizers, that have been formed into a polymeric sheet. Again, polymer layers can include additional additives, although these are not required. As used herein, the term “interlayer” refers to a single or multiple layer polymer sheet suitable for use with at least one rigid substrate to form a multiple layer panel. The terms “single-sheet” interlayer and “monolithic” interlayer refer to interlayers formed of one single resin sheet, while the terms “multiple layer” and “multilayer” interlayer refer to interlayers having two or more resin sheets coextruded, laminated, or otherwise coupled to one another.

[0030] The resin compositions, layers, and interlayers described herein may include one or more thermoplastic polymers. Examples of suitable thermoplastic polymers can include, but are not limited to, poly(vinyl acetal) resins, polyurethanes (PU), poly(ethylene-co-vinyl) acetates (EVA), polyvinyl chlorides (PVC), poly(vinylchloride-co-methacrylate), polyethylenes, polyolefins, ethylene acrylate ester copolymers, poly(ethylene-co-butyl acrylate), silicone elastomers, epoxy resins, and acid copolymers such as ethylene/carboxylic acid copolymers and ionomers thereof, derived from any of the previously-listed polymers, and combinations thereof. In some embodiments, the thermoplastic polymer can be selected from the group consisting of poly(vinyl acetal) resins, polyvinyl chloride, and polyurethanes, or the resin can comprise one or more poly(vinyl acetal) resins. Although described herein with respect to poly(vinyl acetal) resins, it should be understood that one or more of the above polymer resins could be included with, or in the place of, the poly(vinyl acetal) resins described below in accordance with various embodiments of the present invention.



[0031] When the resin compositions, layers, and interlayers described herein include poly(vinyl acetal) resins, the poly(vinyl acetal) resins can be formed according to any suitable method. Poly(vinyl acetal) resins can be formed by acetalization of polyvinyl alcohol with one or more aldehydes in the presence of an acid catalyst. The resulting resin can then be separated, stabilized, and dried according to known methods such as, for example, those described in U.S. Patent Nos. 2,282,057 and 2,282,026, as well as "Vinyl Acetal Polymers," in the *Encyclopedia of Polymer Science & Technology*, 3<sup>rd</sup> ed., Volume 8, pages 381-399, by B.E. Wade (2003). The resulting poly(vinyl acetal) resins may have a total percent acetalization of at least about 50, at least about 60, at least about 70, at least about 75, at least about 80, at least about 85 weight percent, measured according to ASTM D1396, unless otherwise noted. The total amount of aldehyde residues in a poly(vinyl acetal) resin can be collectively referred to as the acetal component, with the balance of the poly(vinyl acetal) resin being residual hydroxyl and residual acetate groups, which will be discussed in further detail below.

[0032] According to some embodiments, the resin composition, layer, or interlayer may include at least one poly(vinyl acetal) resin, which may be present in the composition, layer, or interlayer in an amount of at least about 0.5, at least about 1, at least about 2, at least about 3, at least about 5, at least about 10, at least about 15, at least about 20, at least about 30, at least about 40, or at least about 45 weight percent, based on the combined weight of all resins in the composition, layer, or interlayer. Together, the at least one poly(vinyl acetal) resins can make up at least about 10, at least about 20, at least about 30, at least about 40, at least about 50, at least about 60, at least about 70, or at least about 80 weight percent of composition, layer, or interlayer, based on the combined weight of all resins. In some embodiments, the amount of resins other than the at least one poly(vinyl acetal) resin can be not more than about 20, not more than about 15, not more than about 10, not more than about 5, not more than about 2, or not more than about 1 weight percent, based on the combined weight of all resins.

[0033] In some embodiments, the resin composition, layer, or interlayer can include at least a first poly(vinyl acetal) resin and a second poly(vinyl acetal) resin, each of which may be present in the composition, layer, or interlayer in an amount of at least about 0.5, at least about 1, at least about 2, at least about 3, at least about 5, at least about 10, at least about 15, at least about 20, at least about 30, at least about 40, or at least about 45 weight percent, based on the combined weight of all resins in the composition, layer, or interlayer. Together, the first and second poly(vinyl acetal) resins can make up at least about 10, at least about 20, at least about 30, at least about 40, at least about 50, at least about 60, at least about 70, or at least about 80 weight percent of composition, layer, or interlayer, based on the combined weight of all resins. In some embodiments, the amount of resins other than the first and second poly(vinyl acetal) resins can be not more than about 20, not more than about 15, not more than about 10, not more than about 5, not more than about 2, or not more than about 1 weight percent, based on the combined weight of all resins.

[0034] In some embodiments, one of the first and the second poly(vinyl acetal) resins can be present in the composition, layer, or interlayer in an amount of less than 12 weight percent, based on the combined weight of the first and second poly(vinyl acetal) resins. For example, the first or the second poly(vinyl acetal) resin can be present in the composition, layer, or interlayer in an amount of at least about 0.5, at least about 1, at least about 1.5, at least about 2, at least about 2.5, at least about 3, at least about 3.5, at least about 4, at least about 4.5, at least about 5, at least about 5.5, at least about 6, at least about 6.5, at least about 7 weight percent and/or not more than about 12, not more than about 11.5, not more than about 11, not more than about 10.5, not more than about 10, not more than about 9.5, not more than about 9, not more than about 8.5, not more than about 8, not more than about 7.5 weight percent, based on the combined weight of the first and second poly(vinyl acetal) resins. In some embodiments, one of the first and second poly(vinyl acetal) resins can be present in the composition, layer, or interlayer in an amount in the range of from about 0.5 to about 12, about 1.5 to about 11.5, about 2 to about 11, about 2.5

to about 10 weight percent, based on the combined weight of the first and second poly(vinyl acetal) resins.

[0035] The first and second poly(vinyl acetal) resins can include residues of any suitable aldehyde and, in some embodiments, can include residues of at least one C<sub>1</sub> to C<sub>10</sub> aldehyde, or at least one C<sub>4</sub> to C<sub>8</sub> aldehyde. Examples of suitable C<sub>4</sub> to C<sub>8</sub> aldehydes can include, but are not limited to, n-butyraldehyde, iso-butyraldehyde, 2-methylvaleraldehyde, n-hexyl aldehyde, 2-ethylhexyl aldehyde, n-octyl aldehyde, and combinations thereof. At least one of the first and second poly(vinyl acetal) resins can include at least about 20, at least about 30, at least about 40, at least about 50, at least about 60, or at least about 70 weight percent of residues of at least one C<sub>4</sub> to C<sub>8</sub> aldehyde, based on the total weight of aldehyde residues of the resin, and/or can include not more than about 90, not more than about 85, not more than about 80, not more than about 75, not more than about 70, or not more than about 65 weight percent of at least one C<sub>4</sub> to C<sub>8</sub> aldehyde, or in the range of from about 20 to about 90, about 30 to about 80, or about 40 to about 70 weight percent of at least one C<sub>4</sub> to C<sub>8</sub> aldehyde. The C<sub>4</sub> to C<sub>8</sub> aldehyde may be selected from the group listed above, or it can be selected from the group consisting of n-butyraldehyde, iso-butyraldehyde, 2-ethylhexyl aldehyde, and combinations thereof.

[0036] In various embodiments, the first and/or second poly(vinyl acetal) resin may be a polyvinyl n-butyral (PVB) resin. In other embodiments, the first and/or second poly(vinyl acetal) resin can be a poly(vinyl n-butyral) resin that mainly comprises residues of n-butyraldehyde, and may, for example, include not more than about 50, not more than about 40, not more than about 30, not more than about 20, not more than about 10, not more than about 5, or not more than about 2 weight percent of residues of an aldehyde other than n-butyraldehyde, based on the total weight of all aldehyde residues of the resin. When the first and/or second poly(vinyl acetal) resins are PVB resins, the molecular weight of the resins can be at least about 50,000, at least about 70,000, at least about 100,000 Daltons and/or not more than about 600,000, not more than about 550,000, not more than about 500,000, not more

than about 450,000, or not more than 425,000 Daltons, measured by size exclusion chromatography using low angle laser light scattering (SEC/LALLS) method of Cotts and Ouano. As used herein, the term “molecular weight” refers to weight average molecular weight ( $M_w$ ). The molecular weight of the first and/or second poly(vinyl acetal) resin can be in the range of from about 50,000 to about 600,000, about 70,000 to about 450,000, or about 100,000 to about 425,000 Daltons.

[0037] Although generally described herein with respect to first and second poly(vinyl acetal) resins, it should be understood that, in some embodiments, an equivalent single poly(vinyl acetal) resin including first and second acetal moieties may be substituted for the first and second poly(vinyl acetal) resins with similar results. As used herein, the term “poly(vinyl acetal) resin component” can refer to an individual poly(vinyl acetal) resin present in a blend of resins or to an acetal moiety present on a single poly(vinyl acetal) resin. In various embodiments, the ratio, by weight, of the amount of the first poly(vinyl acetal) resin component to the second poly(vinyl acetal) resin component in a layer, interlayer, or blend can be in the range of from about 0.5:99.5 to about 99.5:0.5, about 1:99 to 99:1, about 10:90 to about 90:10, about 25:75 to about 75:25, or about 40:60 to about 60:40.

[0038] In some embodiments, at least one resin composition, layer, or interlayer can include at least a first poly(vinyl acetal) resin component and a second poly(vinyl acetal) resin component. In some embodiments, the first and second resin components may comprise first and second poly(vinyl acetal) resins that can be physically mixed to form a resin blend, which may be combined with one or more plasticizers or other additives to provide a blended polymer layer or interlayer. In other embodiments, the first and second poly(vinyl acetal) resin components may be present as respective first and second acetal moieties in a single poly(vinyl acetal) resin. As with the resin blend, this single “hybrid” poly(vinyl acetal) resin can be optionally blended with a plasticizer and utilized in polymer layers and interlayers.

[0039] In some embodiments, when the resin components include poly(vinyl acetal) resins, the first and second poly(vinyl acetal) resins may be blended such that

one of the first and second resins is dispersed within the other of the first and second resins, which can form domains of one of the first and second poly(vinyl acetal) resins within the other of the first and second poly(vinyl acetal) resins. Such a blended resin may be used as a single layer interlayer or it may be combined with one or more adjacent layers to form a multilayer interlayer. In other embodiments, the first and second poly(vinyl acetal) resins can be present in adjacent layers of a multilayer interlayer, such that one of the layers of the interlayer includes the first poly(vinyl acetal) resin and another layer of the interlayer includes the second poly(vinyl acetal) resin. Additional layers can also be present adjacent to at least one of the layers.

[0040] The resin compositions, layers, and interlayers according to various embodiments of the present invention can further include at least one plasticizer. Depending on the specific composition of the resin or resins in a composition, layer, or interlayer, the plasticizer may be present in an amount of at least about 5, at least about 10, at least about 15, at least about 20, at least about 25, at least about 30, at least about 35, at least about 40, at least about 45, at least about 50, at least about 55, at least about 60 parts per hundred parts of resin (phr) and/or not more than about 120, not more than about 110, not more than about 105, not more than about 100, not more than about 95, not more than about 90, not more than about 85, not more than about 75, not more than about 70, not more than about 65, not more than about 60, not more than about 55, not more than about 50, not more than about 45, or not more than about 40 phr, or in the range of from about 5 to about 120, about 10 to about 110, about 20 to about 90, or about 25 to about 75 phr. Specific embodiments are discussed in detail shortly.

[0041] As used herein, the term “parts per hundred parts of resin” or “phr” refers to the amount of plasticizer present as compared to one hundred parts of resin, on a weight basis. For example, if 30 grams of plasticizer were added to 100 grams of a resin, the plasticizer would be present in an amount of 30 phr. If the resin composition, layer, or interlayer includes two or more resins, the weight of plasticizer is compared to the combined amount of all resins present to determine the parts per

hundred resin. Further, when the plasticizer content of a layer or interlayer is provided herein, it is provided with reference to the amount of plasticizer in the mix or melt that was used to produce the layer or interlayer.

[0042] Examples of suitable plasticizers can include, but are not limited to, triethylene glycol di-(2-ethylhexanoate) ("3GEH"), triethylene glycol di-(2-ethylbutyrate), triethylene glycol diheptanoate, tetraethylene glycol diheptanoate, tetraethylene glycol di-(2-ethylhexanoate) ("4GEH"), dihexyl adipate, dioctyl adipate, hexyl cyclohexyladipate, diisononyl adipate, heptylnonyl adipate, di(butoxyethyl) adipate, and bis(2-(2-butoxyethoxy)ethyl) adipate, dibutyl sebacate, dioctyl sebacate, and mixtures thereof. The plasticizer may be selected from the group consisting of triethylene glycol di-(2-ethylhexanoate) and tetraethylene glycol di-(2-ethylhexanoate), or the plasticizer can comprise triethylene glycol di-(2-ethylhexanoate).

[0043] According to some embodiments, the first and second poly(vinyl acetal) resins in the compositions, layers, and interlayers described herein can have different compositions. For example, in some embodiments, the first poly(vinyl acetal) resin can have a residual hydroxyl content and/or residual acetate content that is at least about 2, at least about 3, at least about 4, at least about 5, at least about 6, or at least about 8 weight percent higher or lower than the residual hydroxyl content and/or residual acetate content of the second poly(vinyl acetal) resin. As used herein, the terms "residual hydroxyl content" and "residual acetate content" refer to the amount of hydroxyl and acetate groups, respectively, that remain on a resin after processing is complete. For example, polyvinyl n-butyral can be produced by hydrolyzing polyvinyl acetate to polyvinyl alcohol, and then acetalizing the polyvinyl alcohol with n-butyraldehyde to form polyvinyl n-butyral. In the process of hydrolyzing the polyvinyl acetate, not all of the acetate groups are converted to hydroxyl groups, and residual acetate groups remain on the resin. Similarly, in the process of acetalizing the polyvinyl alcohol, not all of the hydroxyl groups are converted to acetal groups, which also leaves residual hydroxyl groups on the resin.

As a result, most poly(vinyl acetal) resins include both residual hydroxyl groups (as vinyl hydroxyl groups) and residual acetate groups (as vinyl acetate groups) as part of the polymer chain. The residual hydroxyl content and residual acetate content are expressed in weight percent, based on the weight of the polymer resin, and are measured according to ASTM D1396, unless otherwise noted.

[0044] The difference between the residual hydroxyl content of the first and second poly(vinyl acetal) resins could also be at least about 2, at least about 5, at least about 10, at least about 12, at least about 15, at least about 20, or at least about 30 weight percent. As used herein, the term “weight percent different” or “the difference is at least weight percent” refers to a difference between two given weight percentages, calculated by subtracting the one number from the other. For example, a poly(vinyl acetal) resin having a residual hydroxyl content of 12 weight percent has a residual hydroxyl content that is 2 weight percent lower than a poly(vinyl acetal) resin having a residual hydroxyl content of 14 weight percent (14 weight percent – 12 weight percent = 2 weight percent). As used herein, the term “different” can refer to a value that is higher than or lower than another value.

[0045] At least one of the first and second poly(vinyl acetal) resins can have a residual hydroxyl content of at least about 14, at least about 14.5, at least about 15, at least about 15.5, at least about 16, at least about 16.5, at least about 17, at least about 17.5, at least about 18, at least about 18.5, at least about 19, at least about 19.5 and/or not more than about 45, not more than about 40, not more than about 35, not more than about 33, not more than about 30, not more than about 27, not more than about 25, not more than about 22, not more than about 21.5, not more than about 21, not more than about 20.5, or not more than about 20 weight percent, or in the range of from about 14 to about 45, about 16 to about 30, about 18 to about 25, about 18.5 to about 24, or about 19.5 to about 21 weight percent.

[0046] The other poly(vinyl acetal) resin can have a residual hydroxyl content of at least about 8, at least about 9, at least about 10, at least about 11 weight percent and/or not more than about 16, not more than about 15, not more than about 14.5,

not more than about 13, not more than about 11.5, not more than about 11, not more than about 10.5, not more than about 10, not more than about 9.5, or not more than about 9 weight percent, or in the range of from about 8 to about 16, about 9 to about 15, or about 9.5 to about 14.5 weight percent, and can be selected such that the difference between the residual hydroxyl content of the first and second poly(vinyl acetal) resin is at least about 2 weight percent, as mentioned previously. One or more other poly(vinyl acetal) resins may also be present in the resin composition, layer, or interlayer and can have a residual hydroxyl within the ranges provided above. Additionally, the residual hydroxyl content of the one or more other poly(vinyl acetal) resins can be the same as or different than the residual hydroxyl content of the first and/or second poly(vinyl acetal) resins.

[0047] In some embodiments, at least one of the first and second poly(vinyl acetal) resins can have a residual acetate content different than the other. For example, in some embodiments, the difference between the residual acetate content of the first and second poly(vinyl acetal) resins can be at least about 2, at least about 3, at least about 4, at least about 5, at least about 8, at least about 10 weight percent. One of the poly(vinyl acetal) resins may have a residual acetate content of not more than about 4, not more than about 3, not more than about 2, or not more than about 1 weight percent, measured as described above. In some embodiments, at least one of the first and second poly(vinyl acetal) resins can have a residual acetate content of at least about 5, at least about 8, at least about 10, at least about 12, at least about 14, at least about 16, at least about 18, at least about 20, or at least about 30 weight percent. The difference in the residual acetate content between the first and second poly(vinyl acetal) resins can be within the ranges provided above, or the difference can be less than about 3, not more than about 2, not more than about 1, or not more than about 0.5 weight percent. Additional poly(vinyl acetal) resins present in the resin composition or interlayer can have a residual acetate content the same as or different than the residual acetate content of the first and/or second poly(vinyl acetal) resin.



[0048] In some embodiments, the difference between the residual hydroxyl content of the first and second poly(vinyl acetal) resins can be less than about 2, not more than about 1, not more than about 0.5 weight percent and the difference in the residual acetate content between the first and second poly(vinyl acetal) resins can be at least about 3, at least about 5, at least about 8, at least about 15, at least about 20, or at least about 30 weight percent. In other embodiments, the difference in the residual acetate content of the first and second poly(vinyl acetal) resins can be less than about 3, not more than about 2, not more than about 1, or not more than about 0.5 weight percent and the difference in the residual hydroxyl content of the first and second poly(vinyl acetal) resins can be at least about 2, at least about 5, at least about 10, at least about 12, at least about 15, at least about 20, or at least about 30 weight percent.

[0049] In various embodiments, the differences in residual hydroxyl and/or residual acetate content of the first and second poly(vinyl acetal) resins can be selected to control or provide certain performance properties, such as strength, impact resistance, penetration resistance, processability, or acoustic performance to the final composition, layer, or interlayer. For example, poly(vinyl acetal) resins having a higher residual hydroxyl content, usually greater than about 16 weight percent, can facilitate high impact resistance, penetration resistance, and strength to a resin composition or layer, while lower hydroxyl content resins, usually having a residual hydroxyl content of less than 16 weight percent, can improve the acoustic performance of the interlayer or blend.

[0050] Poly(vinyl acetal) resins having higher or lower residual hydroxyl contents and/or residual acetate contents may also, when combined with at least one plasticizer, ultimately include different amounts of plasticizer. As a result, layers or domains formed of first and second poly(vinyl acetal) resins having different compositions may also have different properties within a single polymer layer or interlayer. Although not wishing to be bound by theory, it is assumed that the compatibility of a given plasticizer with a poly(vinyl acetal) resin can depend, at least

in part, on the composition of the polymer, and, in particular, on its residual hydroxyl content. Overall, poly(vinyl acetal) resins with higher residual hydroxyl contents tend to exhibit a lower compatibility (or capacity) for a given plasticizer as compared to similar resins having a lower residual hydroxyl content. As a result, poly(vinyl acetal) resins with higher residual hydroxyl contents tend to be less plasticized and exhibit higher stiffness than similar resins having lower residual hydroxyl contents. Conversely, poly(vinyl acetal) resins having lower residual hydroxyl contents may tend to, when plasticized with a given plasticizer, incorporate higher amounts of plasticizer, which may result in a softer polymer layer that exhibits a lower glass transition temperature than a polymer layer including a similar resin having a higher residual hydroxyl content. Depending on the specific resin and plasticizer, these trends could be reversed.

[0051] When two poly(vinyl acetal) resins having different levels of residual hydroxyl content are blended with a plasticizer, the plasticizer may partition between the polymer layers or domains, such that more plasticizer can be present in the layer or domain having the lower residual hydroxyl content and less plasticizer may be present in the layer or domain having the higher residual hydroxyl content. Ultimately, a state of equilibrium is achieved between the two resins. The correlation between the residual hydroxyl content of a poly(vinyl acetal) resin and plasticizer compatibility/capacity can facilitate addition of a proper amount of plasticizer to the polymer resin. Such a correlation also helps to stably maintain the difference in plasticizer content between two or more resins when the plasticizer would otherwise migrate between the resins.

[0052] In some embodiments, a polymer layer or interlayer can include at least a first polymer layer comprising a first poly(vinyl acetal) resin and a first plasticizer, and a second polymer layer, adjacent to the first polymer layer, comprising a second poly(vinyl acetal) resin and a second plasticizer. The first and second plasticizer can be the same type of plasticizer, or the first and second plasticizers may be different. In some embodiments, at least one of the first and second plasticizers may also be a

blend of two or more plasticizers, which can be the same as or different than one or more other plasticizers. When one of the first and second poly(vinyl acetal) resins has a residual hydroxyl content that is at least 2 weight percent higher or lower than the residual hydroxyl content of the other, the difference in plasticizer content between the polymer layers can be at least about 2, at least about 5, at least about 8, at least about 10, at least about 12, or at least about 15 phr. In most embodiments, the polymer layer that includes the resin having a lower hydroxyl content can have the higher plasticizer content. In order to control or retain other properties of the polymer layer or interlayer, the difference in plasticizer content between the first and second polymer layers may be not more than about 40, not more than about 30, not more than about 25, not more than about 20, or not more than about 17 phr. In other embodiments, the difference in plasticizer content between the first and second polymer layers can be at least about 40, at least about 50, at least about 60, or at least about 70 phr.

[0053] As a result, in some embodiments, wherein the first and second poly(vinyl acetal) resins are present in adjacent layers of a multilayer interlayer, the first and second polymer layers can exhibit different glass transition temperatures. Similarly, when the first and second poly(vinyl acetal) resins are present in a blend, the domains of one of the first and second poly(vinyl acetal) resins can exhibit a different glass transition temperature than the other of the first and second poly(vinyl acetal) resins. Glass transition temperature, or  $T_g$ , is the temperature that marks the transition from the glass state of the polymer to the rubbery state. The glass transition temperatures of the resins and layers described herein were determined by dynamic mechanical thermal analysis (DMTA). The DMTA measures the storage (elastic) modulus ( $G'$ ) in Pascals, loss (viscous) modulus ( $G''$ ) in Pascals, and the tan delta ( $G''/G'$ ) of the specimen as a function of temperature at a given oscillation frequency and temperature sweep rate. The glass transition temperature is then determined by the position of the tan delta peak on the temperature scale. Glass transition

temperatures provided herein were determined at an oscillation frequency of 1 Hz under shear mode and a temperature sweep rate of 3°C/min.

[0054] The difference in the glass transition temperature of the first polymer layer and the second polymer layer, or between various regions of a blended resin or polymer layer, can be at least about 3, at least about 5, at least about 8, at least about 10, at least about 12, at least about 15, at least about 18, at least about 20, at least about 22, or at least about 25°C. One of the first and second polymer layers can have a glass transition temperature of at least about 26, at least about 28, at least about 30, at least about 33, at least about 35°C and/or not more than about 70, not more than about 65, not more than about 60, not more than about 55, not more than about 50°C, or in the range of from about 26 to about 70, about 30 to about 60, about 35 to about 50°C. The other of the first and second poly(vinyl acetal) resins can have a glass transition temperature of not more than 25, not more than about 20, not more than about 15, not more than about 10, not more than about 5, not more than about 0, not more than about -5, or not more than about -10°C.

[0055] When the first and second poly(vinyl acetal) resins are blended with one another such that domains of one resin are dispersed within the other, such differences in plasticizer content and/or glass transition temperature may also exist between domains of the first and second resins. For example, in some embodiments, a polymer layer or interlayer may include various domains of higher or lower plasticizer content and/or domains having higher or lower glass transition temperatures, as described previously. In some embodiments, at least a portion of the polymer layer or interlayer can have a glass transition temperature of at least about 26, at least about 28, at least about 30, at least about 33, at least about 35°C and/or not more than about 70, not more than about 65, not more than about 60, not more than about 55, not more than about 50°C, or in the range of from about 26 to about 70, about 28 to about 60, about 35 to about 50°C and/or at least a portion of the polymer layer or interlayer can have a glass transition temperature of not more than 25, not more than about 20, not more than about 15, not more than about 10,

not more than about 5, not more than about 0°C, not more than about -5°C, or not more than about -10°C.

[0056] One or more resin blends, layers, and interlayers described herein may include various other additives to impart particular properties or features to the interlayer. Such additives can include, but are not limited to, dyes, pigments, stabilizers such as ultraviolet stabilizers, antioxidants, anti-blocking agents, flame retardants, IR absorbers or blockers such as indium tin oxide, antimony tin oxide, lanthanum hexaboride (LaB<sub>6</sub>) and cesium tungsten oxide, processing aides, flow enhancing additives, lubricants, impact modifiers, nucleating agents, thermal stabilizers, UV absorbers, dispersants, surfactants, chelating agents, coupling agents, adhesives, primers, reinforcement additives, and fillers.

[0057] Additionally, various adhesion control agents (“ACAs”) can be used in the interlayers of the present disclosure to control the adhesion of the sheet to glass. In various embodiments, the amount of ACAs present in a resin composition, layer, or interlayer can be at least about 0.003, at least about 0.01, at least about 0.025 and/or not more than about 0.15, not more than about 0.10, or not more than about 0.04 phr, or in the range of from about 0.003 to about 0.15, about 0.01 to about 0.10, or about 0.025 to about 0.04 phr. Suitable ACAs can include, but are not limited to, sodium acetate, potassium acetate, magnesium bis(2-ethyl butyrate), magnesium bis(2-ethylhexanoate), and combinations thereof, as well as the ACAs disclosed in U.S. Patent No. 5,728,472.

[0058] Resins having different compositions and plasticized polymer layers having different properties also tend to exhibit different refractive indices, which can reduce the optical quality of the resulting layer or blend. Although not wishing to be bound by theory, it is believed that such differences in refractive index may cause light that passes through the different polymer layers or domains to be refracted in different directions, which may cause haze in the final product. At times, the absolute value of the difference between the refractive index of the first poly(vinyl acetal) resin or layer and the refractive index of the second poly(vinyl acetal) resin or layer,

measured according to ASTM D542 at a wavelength of 589 nm and 25°C, can exceed 0.010. As a result, these compositions, layers, or interlayers can have a haze value greater than 5 percent and/or a mottle value greater than 3.

[0059] However, in various embodiments of the present invention, compositions, layers, and interlayers comprising a poly(vinyl acetal) resin may further include at least one refractive index (RI) balancing agent for adjusting the refractive index of the composition, layer, or interlayer. In some embodiments, the composition, layer, or interlayer may include at least a first poly(vinyl acetal) resin and a second poly(vinyl acetal) resin along with at least one RI balancing agent. In other embodiments, the composition, layer, or interlayer may include a single poly(vinyl acetal) resin along with at least one RI balancing agent. As discussed above, the RI balancing agent can be any suitable agent present in a resin or a resin blend, layer, or interlayer, or portion thereof, that increases or reduces the refractive index of at least one resin or layer, which may improve the optical properties of the interlayer as compared to an identical interlayer formed without an RI balancing agent. In some embodiments, the resin blend, layer, or interlayer may have a haze value of at least 5 percent when formed in the absence of the RI balancing agent.

[0060] The RI balancing agent can be in any suitable form and may be physically blended with one or more resins or it can be chemically bonded, or reacted, with at least one resin so that the RI balancing agent is incorporated into the polymer chain. Examples of RI balancing agents can include, but are not limited to, liquid RI additives, solid RI additives, and residues of at least one aldehyde present in one or more of the poly(vinyl acetal) resins. Various embodiments of RI balancing agents, as well as resin compositions, layers, and interlayers including the same, will now be discussed in detail below.

[0061] The RI balancing agent may be present in the resin, polymer layer, or interlayer in an amount sufficient to modify the refractive index of poly(vinyl acetal) resin, polymer layer, or interlayer. The RI balancing agent may also be present in the composition, layer, or interlayer in an amount sufficient to modify the refractive index

of at least one of the two poly(vinyl acetal) resins, thereby minimizing the difference between the refractive indices of two poly(vinyl acetal) polymer layers having different refractive indices. The RI balancing agent may also minimize the difference between the refractive index of one or more poly(vinyl acetal) resins and one or more plasticizers within a resin composition, layer, or interlayer. In some embodiments, the RI balancing agent may be present in an amount sufficient to reduce the absolute value of the difference between the refractive index the first poly(vinyl acetal) polymer layer and the refractive index of the second poly(vinyl acetal) polymer layer to not more than 0.010, not more than about 0.009, not more than about 0.008, not more than about 0.007, not more than about 0.006, not more than about 0.005, not more than about 0.004, or not more than about 0.003. When a multilayer interlayer includes two or more polymer layers, the RI balancing agent may be present in one or both layers and can, in some embodiments, be present in one of the layers in a higher amount than in one or more of the other layers.

[0062] In some embodiments, the RI balancing agent can comprise one or more residues of an aldehyde having a refractive index of at least 1.421, as measured by ASTM D542 at a wavelength of 589 nm and a temperature of 25°C. The RI balancing aldehyde, which may also be referred to herein as a “high refractive index aldehyde” or “high RI aldehyde,” can have a refractive index of at least about 1.425, at least about 1.450, at least about 1.475, at least about 1.500, at least about 1.510, or at least about 1.515 and/or not more than about 1.675, not more than about 1.650, or not more than about 1.625, or in the range of from about 1.425 to about 1.675, about 1.475 to about 1.650, or about 1.515 to about 1.625. The high RI aldehyde may be an aromatic aldehyde that includes at least one aromatic ring or group. Examples of aromatic aldehydes can include, but are not limited to, C<sub>7</sub> to C<sub>30</sub> aromatic aldehydes, C<sub>8</sub> to C<sub>25</sub> aromatic aldehydes, or C<sub>9</sub> to C<sub>20</sub> aromatic aldehydes. Specific examples of high RI aldehydes that can be used as RI balancing agents in various embodiments of the present invention are listed in Table 1, below.

**Table 1: Exemplary High RI Aldehydes**

<b>Aldehyde</b>	<b>Refractive Index</b>
Hexylcinnamaldehyde	1.517
Benzaldehyde	1.545
Cinnamaldehyde	1.620
4-Chlorobenzaldehyde	1.585
4-t-butylphenylacetaldehyde	1.530
2-phenylpropionaldehyde	1.517
Hydrocinnamaldehyde	1.523

[0063] When the RI balancing agent includes residues of at least one high RI aldehyde, at least one of the first and second poly(vinyl acetal) resins can include residues of at least one high RI aldehyde in an amount of at least about 0.5, at least about 1, at least about 5, at least about 10, at least about 15, at least about 20, at least about 25, at least about 30, at least about 35, at least about 40, at least about 45, at least about 50, at least about 55, at least about 60, at least about 65, at least about 70, at least about 75, at least about 80, at least about 85, at least about 90, at least about 95 percent and/or not more than about 99.5, not more than about 99, not more than about 97, not more than about 95, not more than about 90, not more than about 85, not more than about 80, not more than about 75, not more than about 70, not more than about 65, or not more than about 60 weight percent, based on the total weight of aldehyde residues of the first or second poly(vinyl acetal) resin. At least one of the first and second poly(vinyl acetal) resins can include residues of at least one high RI aldehyde in an amount in the range of from about 0.5 to about 99.5, about 10 to about 90, about 25 to about 75, or about 40 to about 60 weight percent, based on the total weight of aldehyde residues of the first or second poly(vinyl acetal) resin.

[0064] The amount of high RI aldehyde residues can be determined using a combination of Fourier Transform Infrared Spectroscopy (FT-IR) and Size Exclusion Chromatography (SEC) with UV detection. In particular, FT-IR is used to measure residual hydroxyl content of the resin and SEC is used to determine the amount of high RI aldehyde residues, with the amount of any other aldehyde residues being determined by the difference. The FT-IR analysis is performed using a Perkin Elmer



Spectrum 100 FT-IR Spectrometer (commercially available from Perkin Elmer, Waltham, Massachusetts) with an ATR sampling attachment. The analysis is performed using 8 scans at a 4 cm<sup>-1</sup> resolution. Prior to testing, a calibration is generated from several poly(vinyl n-butyral) samples of varying residual hydroxyl content which have been dried in a desiccator with silica overnight at room temperature to remove excess moisture. The peak maximum wave number of the hydroxyl stretching band is correlated with the molar vinyl alcohol content of each sample, which was previously determined by ASTM D1396, and the resulting linear curve fits are used to predict molar residual hydroxyl content of the samples being analyzed. These values can be converted to weight percent by calculation after determination of the composition of the poly(vinyl acetal) resin using SEC analysis has been completed, as described below.

[0065] The SEC analysis is performed using a Waters 2695 Alliance pump and autosampler with a Waters 410 inline differential refractive index detector and a Waters 2998 PDA inline UV detector (commercially available from Waters Corporation, Milford, Massachusetts) with Dionex Chromeleon v. 6.8 data acquisition software with an extension pack (commercially available from Thermo Fischer Scientific, Sunnyvale, CA). The analysis is performed with a PL Gel Mixed C (5 micron) column and Mixed E (3 micron) columns with an injection volume of 50 microliters at a flow rate of 1.0 mL/minute. Samples are prepared by dissolving between 0.03 and 0.09 grams of resin in 10-15 mL of stabilized tetrahydrofuran and then filtering each through a 0.22 micron PTFE filter. Initial calibrations of the chromatograph are performed using a narrow molecular weight polystyrene standard and a poly(vinyl acetal) resin including only residues of the high RI aldehyde, and subsequent samples were calibrated with a broad molecular weight polystyrene (commercially available as PSBR250K from American Polymer Standard Corporation, Mentor, Ohio).

[0066] In some embodiments, only one of the first and second poly(vinyl acetal) resins includes residues of the high RI aldehyde, while, in other embodiments, both of the resins may include such residues. The refractive index of a resin

comprising residues of a high RI aldehyde can be at least about 1.492, at least about 1.495, at least about 1.500, at least about 1.505, at least about 1.510, or at least about 1.515.

[0067] In various embodiments, at least one of the first and second poly(vinyl acetal) resins may also include residues of at least one aldehyde having a refractive index of less than 1.421. Examples of these aldehydes can include, for example, aliphatic aldehydes such as the C<sub>4</sub> to C<sub>8</sub> aldehydes discussed above. The aldehydes having a refractive index of less than 1.421 can be selected from the group consisting of n-butyraldehyde, iso-butyraldehyde, and 2-ethylhexyl aldehyde.

[0068] When these residues are present, the first and/or second poly(vinyl acetal) resin can include at least about 10, at least about 15, at least about 20, at least about 25, at least about 30, at least about 35, at least about 40, at least about 45, at least about 50, at least about 55, at least about 60, at least about 65, at least about 70, at least about 75, at least about 80, at least about 85, at least about 90, at least about 95 percent and/or not more than about 99, not more than about 97, not more than about 95, not more than about 90, not more than about 85, not more than about 80, not more than about 75, not more than about 70, not more than about 65, or not more than about 60 weight percent, of these aldehydes, based on the total weight of aldehyde residues of the first or second poly(vinyl acetal) resin.

[0069] The amount of residues of an aldehyde having a refractive index of less than 1.421 are determined using the FT-IR/SEC method described above and then by calculation according to the following formula: 100 weight percent – weight percent residual hydroxyl (from FT-IR) – weight percent of high RI aldehyde residues (from SEC) – weight percent residual acetate (from FT-IR) = weight percent of residues of aldehyde having refractive index less than 1.421. The first and/or second poly(vinyl acetal) resin can include residues of an aldehyde having a refractive index of less than 1.421 in an amount in the range of from about 10 to about 99, about 25 to about 75, or about 40 to about 60 weight percent, based on the total weight of aldehyde residues of the first or second poly(vinyl acetal) resin. The refractive index of one of

these resins can be less than about 1.492, less than about 1.491, or less than about 1.490, measured as described previously.

[0070] According to some embodiments, one of the first and second poly(vinyl acetal) resins primarily includes residues of a high RI aldehyde, while the other of the first and second poly(vinyl acetal) resins primarily includes residues of at least one aldehyde having a refractive index of less than 1.421. As used herein, the term “primarily” means at least 75 weight percent, so that a poly(vinyl acetal) resin primarily including residues of a specified aldehyde includes at least 75 weight percent of residues of the specified aldehyde, based on the total weight of aldehyde residues of that resin. The poly(vinyl acetal) resin primarily including residues of a high RI aldehyde can include not more than about 25, not more than about 20, not more than about 15, not more than about 10, not more than about 5, not more than about 2, or not more than about 1 weight percent of residues of other aldehydes having a refractive index less than 1.421, based on the total weight of aldehyde residues of the resin.

[0071] Similarly, the other poly(vinyl acetal) resin, which can primarily include residues of an aldehyde having a refractive index of less than 1.421, may comprise not more than about 25, not more than about 20, not more than about 15, not more than about 10, not more than about 5, not more than about 2, or not more than about 1 weight percent of residues of a high RI aldehyde, based on the total weight of aldehyde residues of the resin, and may include at least about 75, at least about 80, at least about 85, at least about 90, at least about 95, at least about 97, or at least about 99 percent of residues of one or more aldehydes having a refractive index less than 1.421. In some embodiments, the ratio of the resin primarily including residues of the high RI aldehyde to the other resin or resins in the composition can be at least about 1:99, at least about 5:95, at least about 10:90, at least about 20:80, at least about 25:75, at least about 30:70, at least about 40:60 and/or not more than about 99:1, not more than about 95:5, not more than about 90:10, not more than about 85:15, not more than about 75:25, not more than about 70:30, or not more than about

60:40, or in the range of from about 1:99 to 99:1, about 10:90 to about 90:10, about 25:75 to 75:25, or about 40:60 to 60:40.

[0072] In other embodiments, at least one of the first and second poly(vinyl acetal) resins includes residues of a high RI aldehyde and at least one aldehyde having a refractive index of less than 1.421, thereby forming a “hybrid” resin that includes residues of both high and low RI aldehydes. According to these embodiments, the amounts of the high RI aldehyde residues and the residues of aldehydes having a refractive index of less than 1.421, as well as the weight ratios of one to the other, in the hybrid resin can be within the same ranges provided above with respect to the resin blends. When the first or second poly(vinyl acetal) resin includes residues of both high RI and lower RI aldehydes, the other of the two poly(vinyl acetal) resins may also include residues of at least one high RI aldehyde. Alternatively, the other of the two resins may include little or no high RI aldehyde residues, such that it includes less than about 10, less than about 5, less than about 2, or less than about 1 weight percent of residues of a high RI aldehyde, with the balance being residues of an aldehyde having a refractive index of less than 1.421, including, for example, aldehydes selected from the group consisting of n-butyraldehyde, iso-butyraldehyde, 2-ethylhexyl aldehyde, and combinations thereof.

[0073] When the interlayer is a multilayer interlayer, it can include at least one polymer layer having at least a first poly(vinyl acetal) resin and another polymer layer comprising at least a second poly(vinyl acetal) resin, wherein the difference between the residual hydroxyl content of the first poly(vinyl acetal) resin and the second poly(vinyl acetal) resin is at least 2 weight percent. One or both of the poly(vinyl acetal) resins can include residues of a high RI aldehyde and one of the polymer layers may have a refractive index that is higher or lower than the other by at least about 0.002, at least about 0.003, at least about 0.004 and/or not more than about 0.010, not more than about 0.009, not more than about 0.008, or not more than about 0.007, or by an amount in the range of from about 0.002 to about 0.010, about 0.003 to about 0.009, or about 0.004 to about 0.007. In some embodiments when the interlayer

includes at least three polymer layers, the innermost polymer layer can have the higher refractive index, while in other embodiments, the refractive index of one or both of the outer polymer layers may be higher. In some embodiments, only one of the first and second poly(vinyl acetal) resins may include residues of the high RI aldehyde. In other embodiments, both of the poly(vinyl acetal) resins may include residues of at least one high RI aldehyde, but the resins can still exhibit a difference in refractive index within the ranges provided above.

[0074] One or both of the poly(vinyl acetal) resins can include residues of at least one high RI aldehyde. In some embodiments, when the poly(vinyl acetal) resin including such residues has a residual hydroxyl content of not more than, for example, 15 weight percent, the polymer layer including such a resin may have a glass transition temperature of less than 20, not more than about 15, not more than about 10, not more than about 5, not more than about 0, not more than about -5, or not more than about -10°C and a refractive index of at least about 1.465, at least about 1.470, at least about 1.475, at least about 1.480, at least about 1.485, at least about 1.490, at least about 1.495, at least about 1.500, at least about 1.510, at least about 1.520, at least about 1.525, at least about 1.540, at least about 1.550, at least about 1.575, at least about 1.590, at least about 1.600, at least about 1.615, at least about 1.625, or at least about 1.650, each measured as described previously. The plasticizer content of the layer, according to various embodiments, can be at least about 50, at least about 55, at least about 60, at least about 65 phr and/or not more than about 120, not more than about 110, not more than about 90, not more than about 85, not more than about 80, or not more than about 75 phr, or in the range of from about 50 to about 120, about 55 to about 110, about 60 to about 90, or about 65 to about 75 phr.

[0075] When the resin having the residues of a high RI aldehyde in the multilayer interlayer discussed above has a residual hydroxyl content greater than, for example 16 weight percent, the polymer layer including that resin may have a glass transition temperature of at least about 26, at least about 30, at least about 33, or at least about 35°C, and a refractive index of at least about 1.470, at least about 1.475,

at least about 1.480, at least about 1.485, at least about 1.490, at least about 1.495, at least about 1.500, at least about 1.510, at least about 1.520, at least about 1.525, at least about 1.540, at least about 1.550, at least about 1.575, at least about 1.590, at least about 1.600, at least about 1.615, at least about 1.625, or at least about 1.650, each measured as described previously. The plasticizer content of the layer, according to some embodiments, can be less than 50 phr, not more than about 45 phr, not more than about 40 phr, not more than about 30, not more than about 20 phr.

[0076] The refractive index of the entire interlayer can be at least about 1.477, at least about 1.478, at least about 1.480, at least about 1.485, at least about 1.490, at least about at least about 1.495, at least about 1.500, at least about 1.505, at least about 1.510, at least about 1.515, at least about 1.520, at least about 1.540, at least about 1.550, at least about 1.575, at least about 1.580, at least about 1.590, at least about 1.600, at least about 1.610, at least about 1.620, at least about 1.630, at least about 1.640, or at least about 1.650, measured as described previously.

[0077] According to various embodiments of the present invention, the RI balancing agent can comprise a liquid RI balancing agent. As used herein, the term “liquid RI balancing agent” refers to an RI balancing agent that is liquid at standard conditions of 25°C and 1 atm. In some embodiments, the liquid RI balancing agent can be, for example, a high RI plasticizer. As used herein, the term “high RI plasticizer,” refers to a plasticizer having a refractive index of at least 1.460, measured as described previously. The high RI plasticizers suitable for use as RI balancing agents can have a refractive index of at least about 1.470, at least about 1.480, at least about 1.490, at least about 1.500, at least about 1.510, at least about 1.520 and/or not more than about 1.600, not more than about 1.575, or not more than about 1.550, measured as discussed above. The refractive index of the high RI plasticizers may be in the range of from about 1.460 to about 1.600, about 1.470 to about 1.575, about 1.480 to about 1.550, about 1.490 to about 1.525.

[0078] Examples of types or classes of high RI plasticizers can include, but are not limited to, polyadipates (RI of about 1.460 to about 1.485); epoxides such as

epoxidized soybean oils (RI of about 1.460 to about 1.480); phthalates and terephthalates (RI of about 1.480 to about 1.540); benzoates and toluates (RI of about 1.480 to about 1.550); and other specialty plasticizers (RI of about 1.490 to about 1.520). Specific examples of suitable RI plasticizers can include, but are not limited to, dipropylene glycol dibenzoate, tripropylene glycol dibenzoate, polypropylene glycol dibenzoate, isodecyl benzoate, 2-ethylhexyl benzoate, diethylene glycol benzoate, butoxyethyl benzoate, butoxyethoxyethyl benzoate, butoxyethoxyethoxyethyl benzoate, propylene glycol dibenzoate, 2,2,4-trimethyl-1,3-pentanediol dibenzoate, 2,2,4-trimethyl-1,3-pentanediol benzoate isobutyrate, 1,3-butanediol dibenzoate, diethylene glycol di-o-toluate, triethylene glycol di-o-toluate, dipropylene glycol di-o-toluate, 1,2-octyl dibenzoate, tri-2-ethylhexyl trimellitate, di-2-ethylhexyl terephthalate, bis-phenol A bis(2-ethylhexanoate), di-(butoxyethyl) terephthalate, di-(butoxyethoxyethyl) terephthalate, and mixtures thereof. The high RI plasticizer may be selected from dipropylene glycol dibenzoate and tripropylene glycol dibenzoate, and/or 2,2,4-trimethyl-1,3-pentanediol dibenzoate.

[0079] When the polymer layer or interlayer includes a high RI plasticizer, the plasticizer can be present in the layer alone or it can be blended with one or more additional plasticizers. The other plasticizer or plasticizers may also comprise high RI plasticizers, or one or more may be a lower RI plasticizer having a refractive index of less than 1.460. In some embodiments, the lower RI plasticizer may have a refractive index of less than about 1.450, less than about 1.445, or less than about 1.442 and can be selected from the group listed previously. When a mixture of two or more plasticizers are employed as a RI balancing agent, the mixture can have a refractive index within one or more of the above ranges.

[0080] When used as an RI balancing agent in a multilayer interlayer, a high RI plasticizer may be present in different amounts in two or more of the polymer layers. Similarly, when used as an RI balancing agent in a resin composition or blended polymer layer, the high RI plasticizer may be partitioned as described previously, such that polymer layers or domains having a lower residual hydroxyl content may have

higher amounts of the high RI plasticizer. In some embodiments, at least one layer or portion of a polymer layer or interlayer may comprise a high RI plasticizer as an RI balancing agent in an amount of at least about 5, at least about 10, at least about 15, at least about 20, at least about 25, at least about 30, at least about 35 and/or not more than about 50, not more than about 45, or not more than about 40 phr. The high RI plasticizer can be present in the polymer layer or interlayer in an amount in the range of from about 5 to about 50, about 10 to about 45, about 20 to about 40 phr. In some embodiments, one or more other layers or portions can include the high RI plasticizer in an amount of at least about 50, at least about 55, at least about 60, at least about 65, at least about 70 and/or not more than about 120, not more than about 110, not more than about 100, not more than about 90, or not more than about 75 phr, or in the range of from about 50 to about 120, about 55 to about 110, about 60 to about 90, about 65 to about 75 phr. These amounts may include any other plasticizers present in the composition, including those having a refractive index less than 1.460, or may include only the high RI plasticizer.

[0081] When a high RI plasticizer is used as an RI balancing agent in a multilayer interlayer, the interlayer can include at least one polymer layer having a first poly(vinyl acetal) resin and another polymer layer comprising a second poly(vinyl acetal) resin, wherein the difference between the residual hydroxyl content of the first poly(vinyl acetal) resin and the second poly(vinyl acetal) resin is at least 2 weight percent. At least one of the polymer layers can include a high RI plasticizer in an amount sufficient so that the absolute value of the difference between the refractive index of the polymer layer and the refractive index of the another polymer layer is not more than 0.010. In some embodiments when the interlayer includes at least three polymer layers, the innermost polymer layer can have a higher refractive index, while in other embodiments, the refractive index of one or both of the outer polymer layers may be higher.

[0082] When a high RI plasticizer is included in a polymer layer that includes at least one poly(vinyl acetal) resin having the lower residual hydroxyl content, at least



a portion of the polymer layer can have a glass transition temperature of not more than 25, not more than about 20, not more than about 15, not more than about 10, not more than about 5, not more than about 0, not more than about -5, or not more than about -10°C, and the layer can have a refractive index of at least about 1.465, at least about 1.470, at least about 1.475, at least about 1.480, at least about 1.485, at least about 1.490, at least about 1.495, at least about 1.500, at least about 1.510, at least about 1.520, at least about 1.525, at least about 1.540, at least about 1.550, at least about 1.575, at least about 1.590, at least about 1.600, at least about 1.615, at least about 1.625, or at least about 1.650, measured as described previously. The plasticizer content of this layer, in some embodiments, can be at least about 50, at least about 55, at least about 60 phr and/or not more than about 120, not more than about 110, not more than about 90, not more than about 85, not more than about 80, or not more than about 75 phr, or in the range of from about 50 to about 120, about 55 to about 110, about 60 to about 90, or about 60 to about 75 phr.

[0083] When the high RI plasticizer is present in the polymer layer that comprises the poly(vinyl acetal) resin having a higher residual hydroxyl content, at least a portion of the layer can have a glass transition temperature of at least about 26, at least about 30, at least about 33, or at least about 35°C, and the layer can have a refractive index of at least about 1.470, at least about 1.475, at least about 1.480, at least about 1.485, at least about 1.490, at least about 1.495, at least about 1.500, at least about 1.510, at least about 1.520, at least about 1.525, at least about 1.540, at least about 1.550, at least about 1.575, at least about 1.590, at least about 1.600, at least about 1.615, at least about 1.625, or at least about 1.650, measured as described previously. The plasticizer content of this layer, according to some embodiments, can be less than 50 phr, not more than about 45 phr, not more than about 40 phr, not more than about 30, or not more than about 20 phr.

[0084] According to various embodiments of the present invention, the RI balancing agent may be a solid RI additive present in one or more layers or portions of a layer or interlayer. As used herein, the term “solid RI additive” refers to an

additive used to adjust the refractive index of a poly(vinyl acetal) resin, polymer layer, or interlayer and which is solid at ambient conditions of 25°C and 1 atm. In various embodiments, the solid RI additive may have a melting point of at least about 27, at least about 30, at least about 35, at least about 40, at least about 45, at least about 50, at least about 55, at least about 60, at least about 75, at least about 80, at least about 85, at least about 90, at least about 95, or at least about 100°C. When employed in a resin blend, layer, or interlayer, the solid RI additive can be present in an amount sufficient such that the absolute value of the difference between the refractive indices of the first and second polymer layers is not more than about 0.010. The difference between the refractive index of the first and second polymer layers may be greater than 0.010, when formulated into identical polymer layers in the absence of the solid RI additive.

[0085] In some embodiments, the solid RI additive can be a high RI solid additive for increasing the refractive index of at least one polymer layer or interlayer. The refractive index of the high RI solid additive can be at least about 1.460, at least about 1.465, at least about 1.470, at least about 1.475, at least about 1.480, at least about 1.485, at least about 1.490, at least about 1.495, at least about 1.500, at least about 1.505, at least about 1.510, at least about 1.525, at least about 1.550, at least about 1.575, or at least about 1.600, measured as described previously. In other embodiments, the solid RI additive may be a RI lowering solid additive for reducing the refractive index of at least one resin or polymer layer. The RI lowering solid additive can have a refractive index of less than 1.460, not more than about 1.455, not more than about 1.450, not more than about 1.445, or not more than about 1.440, measured as described previously. Whether higher or lower, the solid RI additive can have a refractive index that is at least about 0.005, at least about 0.010, at least about 0.050, at least about 0.10 and/or not more than about 0.50, not more than about 0.35, or not more than about 0.20 different than the refractive index of the poly(vinyl acetal) resin. The difference in refractive index between the solid RI additive and the

poly(vinyl acetal) resin can be in the range of from about 0.005 to about 0.50, about 0.010 to about 0.35, or about 0.050 to about 0.35.

[0086] In various embodiments, the solid RI additive can be present in a resin composition or interlayer in an amount of at least about 0.5, at least about 1, at least about 1.5, at least about 2, or at least about 5 phr, depending on the specific type of additive and layer or interlayer. The solid RI additive, whether a high RI additive or an RI lowering additive, may comprise a physical solid RI additive capable of being physically mixed or blended with at least one poly(vinyl acetal) resin in a resin composition or layer, or it can be a reactive solid RI additive, which may react with and become incorporated into the backbone of one or more poly(vinyl acetal) resins.

[0087] The solid RI additive can be used in combination with one or more low RI plasticizers. Examples of low RI plasticizers can include, but are not limited to, triethylene glycol di-(2-ethylhexanoate) ("3GEH"), triethylene glycol di-(2-ethylbutyrate), triethylene glycol diheptanoate, tetraethylene glycol diheptanoate, tetraethylene glycol di-(2-ethylhexanoate) ("4GEH"), dihexyl adipate, dioctyl adipate, hexyl cyclohexyladipate, diisononyl adipate, heptylnonyl adipate, di(butoxyethyl) adipate, and bis(2-(2-butoxyethoxy)ethyl) adipate, dibutyl sebacate, dioctyl sebacate, and mixtures thereof. The plasticizer may be selected from the group consisting of triethylene glycol di-(2-ethylhexanoate) and tetraethylene glycol di-(2-ethylhexanoate), or the plasticizer can comprise triethylene glycol di-(2-ethylhexanoate). The solid RI additive can also be used in combination with one or more high RI plasticizers mentioned previously.

[0088] When the solid RI additive is a physical solid RI additive, it may be combined with one or more poly(vinyl acetal) resins or layers in an interlayer. In some embodiments, the physical solid RI additive may be present in at least one layer or in an interlayer in an amount of at least about 1, at least about 2, at least about 3, at least about 5, at least about 8, at least about 10, at least about 12, at least about 15, at least about 20 and/or not more than about 60, not more than about 55, not more than about 50, not more than about 45, not more than about 40, not more than about

35, not more than about 30, not more than about 25, not more than about 20, or not more than about 15 phr, or in an amount in the range of from about 1 to about 60, about 5 to about 50, or about 10 to about 45 phr. Examples of suitable physical solid high RI additives can include, but are not limited to, polyadipates, polystyrene having a molecular weight of less than 2500, epoxides, phthalic acid esters, benzoic acid esters, inorganic oxides such as, for example, zirconium oxide, and combinations thereof. Physical solid RI lowering additives can be selected from the group consisting of halogenated additives, and silicon-containing additives.

[0089] When utilized in a multilayer interlayer, the physical solid RI additive may be present in one of the polymer layers in a higher amount than one or more other layers. The difference between the amount of the physical solid RI additive present in one of the polymer layers and the amount of physical solid RI additive present in another layer, including, for example, an adjacent layer, can be at least about 2, at least about 5, at least about 8, at least about 10 phr and/or not more than about 30, not more than about 25, or not more than about 20 phr, or it can be in the range of from about 2 to about 30, about 5 to about 25, or about 10 to about 20 phr. According to some embodiments, at least one of the layers can include at least about 1, at least about 5, at least about 10, at least about 15 phr and/or not more than about 60, not more than about 55, not more than about 50, not more than about 45 phr of the physical solid RI additive, or the physical solid RI additive can be present in an amount in the range of from about 1 to about 60, about 10 to about 50, or about 15 to about 45 phr. In some embodiments, the physical solid RI additive can be present in one or more layers in an amount of at least about 5, at least about 10, at least about 15, at least about 20 phr and/or not more than about 60, not more than about 55, not more than about 50 phr, or in an amount in the range of from about 5 to about 60, about 15 to about 55, or about 20 to about 50 phr.

[0090] When the multilayer interlayer comprises three or more polymer layers and the solid RI additive is a solid high RI additive, the inner, or core, layer or layers can include higher amounts of the physical solid RI additive than the outer, or skin,

layers. However, if the solid RI additive is a solid RI lowering additive, the outer skin layers may include higher amounts of the solid RI additive than the inner core layer. The core layer can include at least about 30, at least about 40, at least about 50, at least about 60, at least about 70, or at least about 80 percent of the total amount of physical solid RI additive present in the interlayer.

[0091] When the solid RI additive is a reactive solid RI additive, it may be reacted with at least one poly(vinyl acetal) resin such that at least a portion of the additive is incorporated into the polymer chain. The reactive RI additive can be an aromatic additive and can comprise, in some embodiments, phthalic anhydrides and phenylalkoxysilanes including, for example, diphenyldimethoxysilane.

[0092] In some embodiments, the reactive RI additive may be present in one layer of a multilayer interlayer in a higher amount than it is present in one or more other layers of the interlayer. In some embodiments, it may be absent, or substantially absent, from one or more polymer layers. For example, when the interlayer is a multilayer interlayer including at least three polymer layers, the inner core layer may include at least about 0.5, at least about 1, at least about 1.5, at least about 2, at least about 2.5, at least about 3 and/or not more than about 50, not more than about 30, not more than about 20, not more than about 15, not more than about 12, not more than about 10, or not more than about 8 phr of one or more reactive solid RI additives, or an amount in the range of from about 0.5 to about 20, about 1 to about 12, or about 2 to about 8 phr. The outer skin layer or layers may include not more than about 10, not more than about 5, not more than about 2, not more than about 1, or not more than about 0.5 phr of the reactive solid RI additive. The core layer can include at least about 65, at least about 75, at least about 85, at least about 95, or at least about 99 percent of the total amount of the reactive RI additive present in the interlayer.

[0093] When a solid RI additive is used as an RI balancing agent in a multilayer interlayer, the interlayer can include at least one polymer layer having a first poly(vinyl acetal) resin and another polymer layer comprising a second poly(vinyl acetal) resin, wherein the difference between the residual hydroxyl content of the first poly(vinyl

acetal) resin and the second poly(vinyl acetal) resin is at least 2 weight percent. At least one of the polymer layers can include the high RI additive in an amount sufficient so that the absolute value of the difference between the refractive index of the first polymer layer and the refractive index of the second polymer layer is not more than 0.010. In some embodiments when the interlayer includes at least three polymer layers, the innermost polymer layer can have a higher refractive index, while in other embodiments, the refractive index of one or both of the outer polymer layers may be higher.

[0094] When a solid RI additive is included in a polymer layer comprising the poly(vinyl acetal) resin having the lower residual hydroxyl content, the polymer layer can have a glass transition temperature of not more than 25, not more than about 20, not more than about 15, not more than about 10, not more than about 5, not more than about 0, not more than about -5, or not more than about -10°C, and a refractive index of at least about 1.465, at least about 1.470, at least about 1.475, at least about 1.480, at least about 1.485, at least about 1.495, at least about 1.500, at least about 1.510, at least about 1.520, at least about 1.525, at least about 1.540, at least about 1.550, at least about 1.575, at least about 1.590, at least about 1.600, at least about 1.615, at least about 1.625, or at least about 1.650, each measured as described previously. The plasticizer content of this layer, in some embodiments, can be at least about 50, at least about 55, at least about 60, at least about 65 phr and/or not more than about 120, not more than about 110, not more than about 90, not more than about 85, not more than about 80, or not more than about 75 phr, or in the range of from about 50 to about 120, about 55 to about 110, about 60 to about 90, about 65 to about 75 phr.

[0095] When the solid RI additive is present in the polymer layer that comprises the poly(vinyl acetal) resin having a higher residual hydroxyl content, the layer can have a glass transition temperature of at least about 26, at least about 30, at least about 33, or at least about 35°C. In some embodiments, the layer can have a refractive index of at least about 1.470, at least about 1.475, at least about 1.480, at

least about 1.485, at least about 1.490, at least about 1.500, at least about 1.510, at least about 1.520, at least about 1.525, at least about 1.540, at least about 1.550, at least about 1.575, at least about 1.590, at least about 1.600, at least about 1.615, at least about 1.625, or at least about 1.650. The plasticizer content of this layer, according to some embodiments, can be less than 50 phr, not more than about 45 phr, not more than about 40 phr, not more than about 30, or not more than about 20 phr.

[0096] According to some embodiments, the refractive index of the interlayer can be greater than 1.475, at least about 1.480, at least about 1.490, at least about 1.500, at least about 1.510, at least about 1.520, at least about 1.530, at least about 1.540, at least about 1.550, at least about 1.560, at least about 1.570, at least about 1.580, at least about 1.590, at least about 1.600, at least about 1.610, at least about 1.620, at least about 1.630, at least about 1.640, at least about 1.650, at least about 1.660, or at least about 1.670, measured as described above. The interlayer can include one or more of the RI balancing agents described herein, or one or more other RI balancing agents not specifically mentioned. The interlayer may be a single layer (or monolithic interlayer), or it may comprise two or more layers adjacent to one another, as discussed herein.

[0097] Resin compositions, layers, and interlayers formulated according to various embodiments of the present invention to include at least two poly(vinyl acetal) resins and an RI balancing agent can exhibit enhanced optical properties without sacrificing other properties such as impact resistance and acoustic performance. As discussed above, due to differences in properties or composition of the resins, such as, for example, residual hydroxyl content, residual acetate content, or aldehyde residues, identical blends of the same resins formulated without the RI balancing agent may provide compositions, layers, and interlayers with reduced optical performance.

[0098] Clarity is one parameter used to describe the optical performance of compositions, layers, and interlayers described herein and may be determined by

measuring haze value or percent. Haze value represents the quantification of light scattered by a sample in contrast to the incident light. In some embodiments, the resin blends, layers, and interlayers described herein may have a haze value of less than 5 percent, less than about 4 percent, less than about 3 percent, less than about 2 percent, less than about 1, or less than about 0.5 percent, as measured in accordance with ASTM D1003-13– Procedure B using Illuminant C, at an observer angle of 2 degrees. The test is performed with a spectrophotometer, such as a Hunterlab UltraScan XE instrument (commercially available from Hunter Associates, Reston, VA), on a polymer sample having a thickness of 0.76 mm, which has been laminated between two sheets of clear glass each having a thickness of 2.3 mm (commercially available from Pittsburgh Glass Works of Pennsylvania).

[0099] Additionally, the polymer layers and interlayers described herein may have a mottle value of not more than 3, not more than 2, or not more than 1. Mottle is another measure of optical quality, which is detected as a texture or graininess. Mottle is a visual defect if the level is too high or too severe, thereby causing objectionable visual appearance. Mottle is assessed and categorized by a side-by-side qualitative comparison of shadowgraph projections for a test laminate with a set of standard laminate shadowgraphs that represent a series, or scale, of mottle values ranging from 1 to 4, with 1 representing a standard of low mottle (*i.e.*, a low number of disruptions) and 4 representing a standard of high mottle (*i.e.*, a high number of disruptions). High mottle is generally considered objectionable, particularly in automotive and architectural applications. Optionally, a model laminate having a single layer interlayer with zero mottle (no mottle) is used to facilitate the evaluation in a test laminate that has a mottle rating lower than the scale of the standard set, such as lower than a rating of 1. A test laminate that shows a shadowgraph projection similar to that of a zero-mottle laminate is assessed to have a mottle rating of zero. The test laminate is prepared with two sheets of clear glass each having a thickness of 2.3 mm (commercially available from Pittsburgh Glass Works of Pennsylvania) and an



interlayer. The interlayer typically has a random rough surface  $R_z$  of about 35 to 40 microns and thickness of 0.76 to 0.86 mm.

[0100] The mottle values provided herein were determined using a Clear Mottle Analyzer (CMA) that includes a xenon arc lamp, a sample holder, a projection screen, and a digital camera. The xenon arc lamp is used to project a shadowgraph of a laminated sample onto the screen and the camera is configured to capture an image of the resulting shadowgraph. The image is then digitally analyzed using computer imaging software and compared to images of previously-captured standard samples to determine the mottle of the sample. A method of measuring mottle using a CMA is described in detail in U.S. Patent Application Publication No. US 2012-0133764.

[0101] Another parameter used to determine the optical performance is transparency, or percent visual transmittance ( $\%T_{vis}$ ), which is measured using a spectrophotometer, such as a HunterLab UltraScan EX, in accordance with ASTM D1003, Procedure B using Illuminant C at an observer angle of 2 degrees. The values provided herein were obtained by analyzing a glass laminate samples having an interlayer thickness of about 0.76 mm and a clear glass thickness of 2.3 mm (commercially available from Pittsburgh Glass Works of Pennsylvania). In some embodiments, the polymer layers and interlayers of the present invention can have a percent visual transmittance of at least about 70, at least about 75, at least about 80, at least about 81, at least about 82, at least about 83, at least about 84, at least about 85, at least about 85.5, at least about 86, at least about 86.5, at least about 87, at least about 87.5, at least about 88, or at least about 88.5 percent. More specifically, the polymer layers and interlayers of the present invention have a  $\%T_{vis}$  of greater than 85 for the interlayers containing only additives of ACAs, UV stabilizers, and antioxidant, or greater than 80% for the interlayers containing additional additives such as pigments, IR absorbers or blockers as mentioned above. Polymer interlayers containing high levels of pigments and/or dyes may have lower  $\%T_{vis}$  values as desired, such as in mass pigmented or colored polymer interlayers.

[0102] In addition to exhibiting one or more optical properties within the ranges above, the polymer layers and interlayers described herein may also exhibit acoustic properties within a desirable range. In some embodiments, as discussed above, at least a portion of a polymer layer or interlayer may have a glass transition temperature of not more than 25, not more than about 20, not more than about 15, not more than about 10, not more than about 5, not more than about 0, not more than about -5, or not more than about -10°C, which may facilitate acoustic performance of the layer or interlayer. At the same time, at least a portion of the layer or interlayer may have a glass transition temperature of at least about 26, at least about 30, at least about 35°C, which may facilitate impact resistance properties and strength.

[0103] In some embodiments, a polymer layer or interlayer according to the present invention may have a tan delta value of at least about 0.70. Tan delta is the ratio of the loss modulus ( $G''$ ) in Pascals to the storage modulus ( $G'$ ) in Pascals of a specimen measured by Dynamic Mechanical Thermal Analysis (DMTA). The DMTA is performed with an oscillation frequency of 1 Hz under shear mode and a temperature sweep rate of 3°C/min. The peak value of the  $G''/G'$  curve at the glass transition temperature is the tan delta value. Polymer layers or interlayers as described according to various embodiment herein can have a tan delta of at least about 1.0, at least about 1.05, at least about 1.10, at least about 1.25, at least about 1.50, at least about 1.75, at least about 2.0, or at least about 2.25.

[0104] Additionally, the polymer layers and interlayers can have a damping loss factor, or loss factor, of at least about 0.10, at least about 0.15, at least about 0.17, at least about 0.20, at least about 0.25, at least about 0.27, at least about 0.30, at least about 0.33, or at least about 0.35. Loss factor is measured by Mechanical Impedance Measurement as described in ISO Standard 16940. A polymer sample is laminated between two sheets of clear glass, each having a thickness of 2.3 mm, and is prepared to have a width of 25 mm and a length of 300 mm. The laminated sample is then excited at the center point using a vibration shaker, commercially available

from Brüel and Kjær (Nærum, Netherlands) and an impedance head (Brüel and Kjær) is used to measure the force required to excite the bar to vibrate and the velocity of the vibration. The resultant transfer function is recorded on a National Instrument data acquisition and analysis system and the loss factor at the first vibration mode is calculated using the half power method. In some embodiments, when the RI balancing agent is a high RI plasticizer, the layer or interlayer may have a loss factor greater than 0.25, greater than 0.27, greater than 0.30, or greater than 0.35 at 20°C, while, in other embodiments, when the RI balancing agent is a solid RI additive or residues of at least one high RI aldehyde, the layer or interlayer may have a loss factor of at least about 0.10, at least about 0.15, at least about 0.20, at least about 0.25, or at least about 0.30°C at 20°C.

[0105] Similar to the resin blend of two distinct resins of the first poly(vinyl acetal) resin and the second poly(vinyl acetal) resin, blending two or more distinct polymer layers or interlayers can often result in the new polymer layer(s) or interlayer(s) having unexpected properties and performance attributes. For example, a polymer layer or interlayer having lower residual hydroxyl content and lower glass transition temperature may be blended with another polymer layer or interlayer having higher residual hydroxyl content and higher glass transition temperature, resulting a new polymer layer or interlayer having soft domains of lower glass transition temperature, which enhances its acoustic performance, and stiff domains of higher glass transition temperature, which imparts enhanced processability, strength, and impact resistance to the polymer layer or interlayer. Other example includes blending a single sheet interlayer and multilayer interlayer, blending two multilayer interlayers, or blending one multilayer interlayer into a polymer layer of another multilayer interlayer. In essence, the effect arising from blending two materials can also be achieved from blending two or more resins, plasticizers, and other additives according to the contents of the materials. As used herein, a “blend resin material” or “blend material” refers to the resin composition, polymer layer or interlayer to be blended into another resin composition, polymer layer or interlayer.

In blending two polymer layers or two interlayers, at least one of the two materials to be blended can include the polymer layer or interlayer of the present invention. In other embodiments, both materials can include the polymer layers or interlayers of the present invention.

[0106] According to some embodiments, at least a portion of the resin compositions, layers, or interlayers described herein may comprise another resins, layers, or interlayers. In some embodiments, at least about 0.5, at least about 1, at least about 5, at least about 10, at least about 15, at least about 20, at least about 25, at least about 30, or at least about 50 percent of the total amount of resin in a composition, layer, or interlayer can originate from a blend resin material.

[0107] Often, when the type and/or amount of resins and plasticizers in the blend resin material differ substantially from the type and/or amount of the resin or plasticizer being produced, and into which the blend resin material is being added, the optical performance, as determined by the clarity or haze, of the resulting resin composition, layer, or interlayer that includes the blend resin material may be adversely impacted. According to embodiments of the present invention, polymer layers and interlayers that include higher levels of blend resin material can be produced by utilizing one or more of the RI balancing agents discussed above.

[0108] When the RI balancing agent includes a high RI plasticizer, higher amounts of blend resin materials can be added to a process for producing a resin composition, layer, or interlayer described herein without reducing the clarity or increasing the haze of the final composition, layer, or interlayer. In some embodiments, the composition that includes blend materials can include a first poly(vinyl acetal) resin and a second poly(vinyl acetal) resin, wherein one of the resins has a residual hydroxyl content that can be at least 2 weight percent lower than the residual hydroxyl content of the other resin. Such a composition may further include at least one high RI plasticizer having a refractive index of 1.460, and, in some embodiments, more than 3 percent of the combined amount of the first and second poly(vinyl acetal) resins present in the composition, layer, or interlayer may have

originated from a blend composition, layer, or interlayer. Despite the difference in residual hydroxyl contents of the first and second poly(vinyl acetal) resins, the composition that includes more than 0.5 weight percent of blend resin materials may have a haze value of not more than about 5, not more than about 4, not more than about 3, not more than about 2, or not more than about 1, or not more than about 0.5.

[0109] The high RI plasticizer used as an RI balancing agent with blend resin compositions can have a refractive index within one or more of the ranges described previously. The high RI plasticizer may be added during production of the composition, layer, or interlayer along with the blend material and/or at least a portion of the high RI plasticizer may be present in the blend resin material added to the process. Additionally, one or more other plasticizers may also be present in the resin material being blended and/or in the resin composition, layer, or interlayer being produced, including, for example, those having a refractive index less than about 1.450, less than about 1.445, or less than about 1.442, measured as described previously. In some embodiments, one or more additional high RI plasticizers may also be present in the blend material and/or in the resin composition, layer, or interlayer into which the materials are being blended.

[0110] The resin composition that includes blended resin material as described above can be used to form layers and interlayers according to various embodiments of the present invention. For example, the resin composition including a blend resin material can be used to form a single monolithic interlayer, or it may be used to form one or more layers of a multilayer interlayer. When used in various layers and interlayers, additional plasticizer may be added such that the total amount of plasticizer present in the polymer layer or interlayer can be within the range described previously. Similarly, the glass transition temperature and refractive indices of polymer layers and interlayers formed from a composition that includes a blend resin material may also be within the ranges provided above. Additionally, polymer layers and interlayers formed from a composition that includes blended materials may also

exhibit acoustic properties as described previously and may be included in any of the applications described below.

[0111] According to some embodiments, at least a portion of the resin compositions, layers, or interlayers described herein may comprise one or more recycled resin materials, including, for example, recycled layers or interlayers. As used herein, the term “recycled” means removed from and subsequently returned to a production line. Often, utilizing recycled materials may adversely affect the optical performance of the final composition, layer, or interlayer, as determined by clarity or haze, because of the different compositions and properties of the materials being blended or combined. However, in some embodiments, layers or interlayers as described herein may include at least one recycled resin material, while still exhibiting the same optical and/or acoustic properties as described herein. The type and/or amount of recycle resin material may fall within one or more of the ranges described previously and the layer or interlayer may further include at least one RI balancing agent. Additionally, the polymer layers and interlayers including recycled resin material may also have optical and/or acoustic performance within one or more of the ranges described below.

[0112] The resin compositions, layers, and interlayers described above may be produced according to any suitable method. In various embodiments, the method for producing these compositions, layers, and interlayers can include providing two or more poly(vinyl acetal) resins, blending at least one resin with an RI balancing agent and, optionally, at least one plasticizer or other additive, to form a blended composition, and forming a layer from the blended composition.

[0113] In some embodiments, the resins provided in the initial steps of the method can be in the form of one or more poly(vinyl acetal) resins, while, in other embodiments, one or more resin precursors can also be provided. In some embodiments, when two or more poly(vinyl acetal) resins are physically blended, the blending of the two resins can comprise melt blending and may be performed at a temperature of at least about 140, at least about 150, at least about 180, at least about

200, at least about 250°C. In other embodiments, when the poly(vinyl acetal) resin components provided include resin precursors, the blending step may include reacting two or more aldehydes with a polyvinyl alcohol to provide a single poly(vinyl acetal) resin having two or more aldehyde moieties. Additionally, a portion of the blending step can include blending one or more of the resins with at least one plasticizer and/or with one or more of the RI balancing agents described previously.

[0114] The resulting blended resins can then be formed into one or more polymer layers according to any suitable method. Exemplary methods of forming polymer layers and interlayers can include, but are not limited to, solution casting, compression molding, injection molding, melt extrusion, melt blowing, and combinations thereof. Multilayer interlayers including two or more polymer layers may also be produced according to any suitable method such as, for example, co-extrusion, blown film, melt blowing, dip coating, solution coating, blade, paddle, air-knife, printing, powder coating, spray coating, and combinations thereof. In various embodiments of the present invention, the layers or interlayers may be formed by extrusion or co-extrusion. In an extrusion process, one or more thermoplastic polymers, plasticizers, and, optionally, at least one additive, including one or more RI balancing agents as described previously, can be pre-mixed and fed into an extrusion device. Other additives, such as ACAs, colorants, and UV inhibitors, which can be in liquid, powder, or pellet form, may also be used and may be mixed into the thermoplastic polymers or plasticizers prior to entering the extrusion device. These additives can be incorporated into the polymer resin and, by extension, the resultant polymer sheet, thereby enhancing certain properties of the polymer layer or interlayer and its performance in the final multiple layer glass panel or other end product.

[0115] In various embodiments, the thickness, or gauge, of the layers or interlayers can be at least about 10, at least about 15, at least about 20 mils and/or not more than about 100, not more than about 90, not more than about 60, not more than about 50, or not more than about 35 mils, or it can be in the range of from about 10 to about 100, about 15 to about 60, or about 20 to about 35 mils. In millimeters,

the thickness of the polymer layers or interlayers can be at least about 0.25, at least about 0.38, at least about 0.51 mm and/or not more than about 2.54, not more than about 2.29, not more than about 1.52, or not more than about 0.89 mm, or in the range of from about 0.25 to about 2.54 mm, about 0.38 to about 1.52 mm, or about 0.51 to about 0.89 mm. In some embodiments, the polymer layers or interlayers can comprise flat polymer layers having substantially the same thickness along the length, or longest dimension, and/or width, or second longest dimension, of the sheet, while, in other embodiments, one or more layers of a multilayer interlayer, for example, can be tapered or can have a wedge-shaped profile, such that the thickness of the interlayer changes along the length and/or width of the sheet, such that one edge of the layer or interlayer has a thickness greater than the other. When the interlayer is a multilayer interlayer, at least one, at least two, or at least three of the layers of the interlayer can comprise at least one tapered zone. When the interlayer is a monolithic interlayer, the polymer sheet can be flat or may include at least one tapered zone. Tapered interlayers may be useful in, for example, heads-up-display (HUD) panels in automotive and aircraft applications.

[0116] Turning now to FIGS. 1 through 8, several embodiments of tapered interlayers according to the present invention are provided. FIG. 1 is a cross-sectional view of an exemplary tapered interlayer that includes a tapered zone of varying thickness. As shown in FIG. 1, the tapered zone has a minimum thickness,  $T_{\min}$ , measured at a first boundary of the tapered zone and a maximum thickness,  $T_{\max}$ , measured at a second boundary of the tapered zone. In certain embodiments,  $T_{\min}$  can be at least about 0.25, at least about 0.40, or at least about 0.60 millimeters (mm) and/or not more than 1.2, not more than about 1.1, or not more than about 1.0 mm. Further,  $T_{\min}$  can be in the range of 0.25 to 1.2 mm, 0.4 to 1.1 mm, or 0.60 to 1.0 mm. In certain embodiments,  $T_{\max}$  can be at least about 0.38, at least about 0.53, or at least about 0.76 mm and/or not more than 2.2, not more than about 2.1, or not more than about 2.0 mm. Further,  $T_{\max}$  can be in the range of 0.38 to 2.2 mm, 0.53 to 2.1 mm, or 0.76 to 2.0 mm. In certain embodiments, the difference between  $T_{\max}$  and  $T_{\min}$  can



be at least about 0.13, at least about 0.15, at least about 0.2, at least about 0.25, at least about 0.3, at least about 0.35, at least about 0.4 mm and/or not more than 1.2, not more than about 0.9, not more than about 0.85, not more than about 0.8, not more than about 0.75, not more than about 0.7, not more than about 0.65, or not more than about 0.6 mm. Further, the difference between  $T_{\max}$  and  $T_{\min}$  can be in the range of 0.13 to 1.2 mm, 0.25 to 0.75 mm, or 0.4 to 0.6 mm. In certain embodiments, the distance between the first and second boundaries of the tapered zone (i.e. the “tapered zone width”) can be at least about 5, at least about 10, at least about 15, at least about 20, or at least about 30 centimeters (cm) and/or not more than about 200, not more than about 150, not more than about 125, not more than about 100 or not more than about 75 cm. Further, the tapered zone width can be in the range of 5 to 200 cm, 15 to 125 cm, or 30 to 75 cm.

[0117] As shown in FIG. 1, the tapered interlayer includes opposite first and second outer terminal edges. In certain embodiments, the distance between the first and second outer terminal edges (i.e., the “interlayer width”) can be at least about 20, at least about 40, or at least about 60 cm and/or not more than about 400, not more than about 200, or not more than about 100 cm. Further the interlayer width can be in the range of 20 to 400 cm, 40 to 200 cm, or 60 to 100 cm. In the embodiment depicted in FIG. 1, the first and second boundaries of the tapered zone are spaced inwardly from the first and second outer terminal edges of the interlayer. In such embodiments, only a portion of the interlayer is tapered. When the tapered zone forms only a portion of the interlayer, the ratio of the interlayer width to the tapered zone width can be at least about 0.05:1, at least about 0.1:1, at least about 0.2:1, at least about 0.3:1, at least about 0.4:1 at least about 0.5:1, at least about 0.6:1, or at least about 0.7:1 and/or not more than about 1:1, not more than about 0.95:1, not more than about 0.9:1, not more than about 0.8:1, or not more than about 0.7:1. Further, the ratio of interlayer width to the tapered zone width can be in the range of 0.05:1 to 1:1 or 0.3:1 to 0.9:1. In an alternative embodiment, discussed below, the entire interlayer is tapered. When the entire interlayer is tapered, the tapered zone

width is equal to the interlayer width and the first and second boundaries of the tapered zone are located at the first and second terminal edges, respectively.

[0118] As illustrated in FIG. 1, the tapered zone of the interlayer has a wedge angle, which is defined as the angle formed between a first reference line extending through two points of the interlayer where the first and second tapered zone boundaries intersect a first (upper) surface of the interlayer and a second reference line extending through two points where the first and second tapered zone boundaries intersect a second (lower) surface of the interlayer. In certain embodiments, the wedge angle of the tapered zone can be at least about 0.13, at least about 0.15, at least about 0.2, at least about 0.25, at least about 0.3, at least about 0.35, at least about 0.4 milliradians (mrad) and/or not more than about 1.2, not more than about 1.0, not more than about 0.9, not more than about 0.85, not more than about 0.8, not more than about 0.75, not more than about 0.7, not more than about 0.65, not more than about 0.6, not more than about 0.55, not more than about 0.5, or not more than about 0.45 mrad. Further, the wedge angle of the tapered zone can be in the range of 0.13 to 1.2 mrad, 0.2 to 0.8 mrad, 0.25 to 0.75 mrad, 0.3 to 0.6, or 0.4 to 0.55 mrad.

[0119] When the first and second surfaces of the tapered zone are each planar, the wedge angle of the tapered zone is simply the angle between the first (upper) and second (lower) surfaces. However, as discussed in further detail below, in certain embodiments, the tapered zone can include at least one variable angle zone having a curved thickness profile and a continuously varying wedge angle. Further, in certain embodiments, the tapered zone can include two or more constant angle zones, where the constant angle zones each have a linear thickness profile, but at least two of the constant angle zones have different wedge angles.

[0120] FIGS. 2-7 illustrate various tapered interlayers configured according to embodiments of the present invention. FIG. 2 depicts an interlayer 20 that includes a tapered zone 22 extending entirely from a first terminal edge 24a of the interlayer 20 to a second terminal edge 24b of the interlayer 20. In this configuration, the first

and second boundaries of the tapered zone are located at the first and second terminal edges 24a,b of the interlayer. The entire tapered zone 22 of the interlayer 20 depicted in FIG. 2 has a constant wedge angle  $\Theta$  that is simply the angle formed between the first (upper) planar and second (lower) planar surfaces of the interlayer 20.

[0121] FIG. 3 illustrates an interlayer 30 that includes a tapered zone 32 and a flat edge zone 33. The first boundary 35a of the tapered zone 32 is located at the first terminal edge 34a of the interlayer 30, while the second boundary 35b of the tapered zone 32 is located where the tapered zone 32 and the flat edge zone 33 meet. The tapered zone 32 includes a constant angle zone 36 and a variable angle zone 37. The constant angle zone 36 has a linear thickness profile and a constant wedge angle,  $\Theta_c$ , while the variable angle zone 37 has a curved thickness profile and a continuously varying wedge angle. The starting wedge angle of the variable angle zone 37 is equal to the constant wedge angle  $\Theta_c$  and the ending wedge angle of the variable angle zone 37 is zero. The interlayer 30 depicted in FIG. 3 has a constant wedge angle  $\Theta_c$  that is greater than the overall wedge angle of the entire tapered zone 32.

[0122] FIG. 4 illustrates an interlayer 40 that includes a tapered zone 42 located between first and second flat edge zones 43a,b. The first boundary 45a of the tapered zone 42 is located where the tapered zone 42 and the first flat edge zone 43a meet, while the second boundary 45b of the tapered zone 42 is located where the tapered zone 42 and the second flat edge zone 43b meet. The tapered zone 42 includes a constant angle zone 46 located between first and second variable angle zones 47a,b. The first variable angle zone 47a forms a transition zone between the first flat edge zone 43a and the constant angle zone 46. The second variable angle zone 47b forms a transition zone between the second flat edge zone 43b and the constant angle zone 46. The constant angle zone 46 has a linear thickness profile and a constant wedge angle,  $\Theta_c$ , while the first and second variable angle zones 47a,b have curved thickness profiles and continuously varying wedge angles. The starting wedge angle of the first variable angle zone 47a is equal to zero and the ending wedge angle

of the first variable angle zone 47b is equal to the constant wedge angle  $\Theta_c$ . The starting wedge angle of the second variable angle zone 47b is equal to the constant wedge angle  $\Theta_c$  and the ending wedge angle of the second variable angle zone 47b is zero. The interlayer 40 depicted in FIG 4 has a constant wedge angle  $\Theta_c$  that is greater than the overall wedge angle of the entire tapered zone 42.

[0123] FIG. 5 illustrates an interlayer 50 that includes a tapered zone 52 located between first and second flat edge zones 53a,b. The tapered zone 52 of the interlayer 50 does not include a constant angle zone. Rather, the entire tapered zone 52 of the interlayer 50 is a variable angle zone having a curved thickness profile and a continuously varying wedge angle. As described above, the overall wedge angle,  $\Theta$ , of the tapered zone 52 is measured as the angle between a first reference line “A” extending through the two points where the first and second boundaries 55a,b of the tapered zone 52 meet the first (upper) surface of the interlayer 50 and a second reference line “B” extending through the two points where the first and second boundaries 55a,b of the tapered zone 52 meet the second (lower) surface of the interlayer 50. However, within the tapered zone 52, the curved thickness profile provides an infinite number of wedge angles, which can be greater than, less than, or equal to the overall wedge angle  $\Theta$  of the entire tapered zone 52.

[0124] FIG. 6 illustrates an interlayer 60 that does not include any flat end portions. Rather, the tapered zone 62 of the interlayer 60 forms the entire interlayer 60. Thus, the first and second boundaries 65a,b of the tapered zone 60 are located at the first and second terminal edges 64a,b of the interlayer 60. The tapered zone 62 of the interlayer 60 includes first, second, and third constant angle zones 46a,b,c separated by first and second variable angle zones 47a,b. The first, second, and third constant angle zones 46a,b,c each have a linear thickness profile and each have unique first, second, and third constant wedge angles,  $\Theta_{c1}, \Theta_{c2}, \Theta_{c3}$ , respectively. The first variable angle zone 47a acts as a transition zone between the first and second constant angle zones 46a,b. The second variable angle zone 47b acts as a transition zone between the second and third constant angle zones 46b,c. As discussed above,

the overall wedge angle,  $\Theta$ , of the tapered zone 62 is measured as the angle between a first reference line "A" and a second reference line "B." The first constant wedge angle  $\Theta_{c1}$  is less than the overall wedge angle  $\Theta$  of the tapered zone 62. The second constant wedge angle  $\Theta_{c2}$  is greater the overall wedge angle  $\Theta$  of the tapered zone 62. The third constant wedge angle  $\Theta_{c3}$  is less than the overall wedge angle  $\Theta$  of the tapered zone 62. The wedge angle of the first variable angle zone 47a continuously increases from the first constant wedge angle  $\Theta_{c1}$  to the second constant wedge angle,  $\Theta_{c2}$ . The wedge angle of the second variable angle zone 47b continuously decreases from the second constant wedge angle  $\Theta_{c2}$  to the third wedge angle  $\Theta_{c3}$ .

[0125] FIG. 7 illustrates an interlayer 70 that includes a tapered zone 72 located between first and second flat edge zones 73a,b. The first and second boundaries 75a,b of the tapered zone 72 are spaced inwardly from the first and second outer edges 74a,b of the interlayer 70. The tapered zone 72 of the interlayer 70 includes first, second, third, and fourth variable angle zones 77a,b,c,d and first, second, and third constant angle zones 76a,b,c. The first variable angle zone 77a acts as a transition zone between the first flat edge zone 73a and the first constant angle zone 76a. The second variable angle zone 77b acts as a transition zone between the first constant angle zone 76a and the second constant angle zone 76b. The third variable angle zone 77c acts as a transition zone between the second constant angle zone 76b and the third constant angle zone 76c. The fourth variable angle zone 77d acts as a transition zone between the third constant angle zone 76c and the second flat edge zone 73b. The first, second, and third constant angle zones 76a,b,c each have a linear thickness profile and each have unique first, second, and third constant wedge angles,  $\Theta_{c1}, \Theta_{c2}, \Theta_{c3}$ , respectively. As discussed above, the first, second, third, and fourth variable angle zones 77a,b,c,d have wedge angles that continuously transition from the wedge angle of the constant angle zone on one side of the variable angle zone 77 to the wedge angle of the constant angle zone on the other side of the variable angle zone 77.

[0126] As discussed above, the tapered interlayer can include one or more constant angle tapered zones, each having a width that is less than the overall width of the entire tapered zone and each having a wedge angle that is the same as or different than the overall wedge angle of the entire tapered zone. For example, the tapered zone can include one, two, three, four, five, or more constant angle tapered zones. When multiple constant angle tapered zones are employed, the constant angle tapered zones can be separated from one another by variable angle tapered zones that serve to transition between adjacent constant angle tapered zones.

[0127] In certain embodiments, the width of each constant angle tapered zone can be at least about 2, at least about 5, at least about 10, at least about 15, or at least about 20 cm and/or not more than about 150, not more than about 100, or not more than about 50 cm. In certain embodiments, the ratio of the width of each constant angle tapered zone to the overall width of the entire tapered zone can be at least about 0.1:1, at least about 0.2:1, at least about 0.3:1 or at least about 0.4:1 and/or not more than about 0.9:1, not more than about 0.8:1, not more than about 0.7:1, not more than about 0.6:1, or not more than about 0.5:1.

[0128] In certain embodiments, the wedge angle of each constant angle tapered zone can be at least about 0.13, at least about 0.15, at least about 0.2, at least about 0.25, at least about 0.3, at least about 0.35, at least about 0.4 mrad and/or not more than about 1.2, not more than about 1.0, not more than about 0.9, not more than about 0.85, not more than about 0.8, not more than about 0.75, not more than about 0.7, not more than about 0.65, or not more than about 0.6 mrad. Further, the wedge angle of each constant angle tapered zone can be in the range of 0.13 to 1.2 mrad, 0.25 to 0.75 mrad, or 0.4 to 0.6 mrad. In certain embodiments, the wedge angle of at least one constant angle tapered zone is at least about 0.01, at least about 0.05, at least about 0.1, at least about 0.2, at least about 0.3, or at least about 0.4 mrad greater than the overall wedge angle of the entire tapered zone. In certain embodiments, the wedge angle of at least one constant angle tapered zone is at least about 0.01, at least about 0.05, at least about 0.1, at least about 0.2, at least about

0.3, or at least about 0.4 mrad less than the overall wedge angle of the entire tapered zone. In certain embodiments, the wedge angle of at least one constant angle tapered zone is not more than about 0.4, not more than about 0.3, not more than about 0.2, not more than about 0.1, not more than about 0.05, or not more than about 0.01 mrad greater than the overall wedge angle of the entire tapered zone. In certain embodiments, the wedge angle of at least one constant angle tapered zone is not more than about 0.4, not more than about 0.3, not more than about 0.2, not more than about 0.1, not more than about 0.05, or not more than about 0.01 mrad less than the overall wedge angle of the entire tapered zone.

[0129] FIGS. 8a and 8b illustrate an interlayer 80 that is similar in thickness profile to the interlayer 30 of FIG. 3. The interlayer 80 of FIGS. 8a and 8b is configured for use in a vehicle windshield by fixing the interlayer between two sheets of glass. As depicted in FIG. 8a, the first terminal edge 84a of the interlayer 80 can be located at the bottom of the windshield, while the second terminal edge 84b of the interlayer 80 can be located at the top of the windshield. The tapered zone 82 of the interlayer 80 is positioned in an area of the windshield where a heads-up display is to be located. The tapered zone 82 of interlayer 80 includes a constant angle zone 86 and a variable angle zone 87. As depicted in FIG. 8a, in certain embodiments, the tapered zone 82 extends entirely across the interlayer 80 between a first side edge 88a and a second side edge 88b of the interlayer 80. FIG. 8b, which is similar to FIG. 3, shows the thickness profile of the interlayer 80 between the bottom of the windshield and the top of the windshield.

[0130] Although not illustrated in the drawings, it should be understood that in certain embodiments, the tapered interlayer can be a multilayered interlayer. When the tapered interlayer comprises multiple individual layers, all of the individual layers can be tapered, some of the individual layers can be tapered, or only one of the individual layers can be tapered. Further, in certain embodiments the glass transition temperatures of the individual layers can be different than another. For example, in one embodiment, the interlayer includes a tapered middle layer having a lower glass

transition temperature than two tapered outer layers of the interlayer, where the glass transition temperature of one or both of the outer layers exceeds the glass transition temperature of the middle layer by at least about 10, at least about 20, at least about 30, at least about 40, or at least about 50°C.

[0131] In some embodiments, the polymer layers or interlayers can comprise flat polymer layers having substantially the same thickness along the length, or longest dimension, and/or width, or second longest dimension, of the sheet, while, in other embodiments, one or more layers of a multilayer interlayer, for example, can be wedge-shaped or can have a wedge-shaped profile, such that the thickness of the interlayer changes along the length and/or width of the sheet, such that one edge of the layer or interlayer has a thickness greater than the other. When the interlayer is a multilayer interlayer, at least one, at least two, or at least three of the layers of the interlayer can be wedge-shaped. When the interlayer is a monolithic interlayer, the polymer sheet can be flat or wedge shaped. Wedge-shaped interlayers may be useful in, for example, heads-up-display (HUD) panels in automotive and aircraft applications.

[0132] The resin compositions, layers, and interlayers according to embodiments of the present invention may be utilized in a multiple layer panel that comprises a polymer layer or interlayer and at least one rigid substrate. In some embodiments, the multilayer panels include a pair of rigid substrates with the resin interlayer disposed therebetween. The total thickness of the panel, measured as the combined thickness of the substrates and the interlayer, can be at least about 2, at least about 3, at least about 3.5, at least about 4, at least about 4.5, at least about 4.65, at least about 4.75, or at least about 5 mm and/or not more than about 6.5, not more than about 6.25, not more than about 6.0, not more than about 5.75, not more than about 5.5, or not more than about 5 mm. The thickness of one or both substrates may be substantially less than would be expected for a conventional multiple layer panel. For example, in some embodiments, the combined thickness of each of the rigid substrates can be not more than about 4.0, not more than about 3.9, not more



than about 3.8, not more than about 3.7, not more than about 3.6, not more than about 3.5, not more than about 3.4 mm.

[0133] Any suitable rigid substrate may be used to form a multiple layer panel, and in some embodiments, the substrates may comprise a material selected from the group consisting of glass, polycarbonate, biaxially oriented PET, copolyesters, acrylic, and combinations thereof. When the rigid substrate includes a polymeric material, the polymeric material may or may not include a hard coat surface layer. In certain embodiments, the rigid substrates may have a refractive index less than the refractive index of the interlayer used to form the multiple layer panel. For example, the refractive index of at least one of the rigid substrates can be not more than about 1.550, not more than about 1.545, not more than about 1.540, not more than about 1.535, not more than about 1.530, or not more than about 1.525, measured as described above. In some embodiments, the refractive index of at least one of the rigid substrates can be at least about 5, at least about 10, at least about 15, at least about 20 percent lower than the refractive index of the interlayer used to form the multiple layer panel. In certain embodiments, the refractive index of one or both substrates can be at least about 0.25 units, at least about 0.50 units, at least about 0.75 units, at least about 1.0 units, at least about 1.1 units, or at least about 1.5 units lower than the refractive index of the interlayer. Alternatively, it can be said that the refractive index of the interlayer is higher than the refractive index of at least one of the substrates by the amounts or percentages described herein.

[0134] Multiple layer panels, configured according to certain embodiments of the present invention, may have an equivalent refractive index that is higher than the refractive index of one or more of the rigid substrates. As used herein, the term “equivalent refractive index” refers to the overall refractive index of a composite material, such as, for example, a multiple layer panel. The equivalent refractive index ( $n_{eq}$ ) of a particular multiple layer panel is calculated according the following equations (1) and (2):

$$\tan \theta_{eq} = \frac{x_T}{t_T}$$

(1)

$$n_A \sin \theta_1 = n_{eq} \sin \theta_{eq}$$

(2),

wherein  $x_T$  and  $t_T$  are defined as follows:

$$t_T = \sum_{i=1}^m t_i$$

(3)

$$x_T = \sum_{i=1}^m x_i = \sum_{i=1}^m t_i \tan \theta_i$$

(4)

[0135] In the above equations,  $\theta_{eq}$  is the equivalent incident angle of the composite,  $x_T$  is the distance light travels through the entire composite,  $t_T$  is the total thickness of the composite having  $m$  layers,  $n_A$  is the refractive index of air,  $\theta_1$  is the incident angle of light passing through the air-layer 1 interface,  $n_{eq}$  is the equivalent refractive index of the composite,  $t_i$  is the thickness of layer  $i$ , and  $\theta_i$  is the incident angle of light passing through layer  $i$ . The equivalent refractive index of a multiple layer panel as described herein can be at least about 1.490, at least about 1.495, at least about 1.500, at least about 1.505, at least about 1.510, at least about 1.515, at least about 1.520, at least about 1.525, at least about 1.530, at least about 1.535, at least about 1.540, or at least about 1.545.

[0136] In some embodiments, the multiple layer panel can have an equivalent refractive index that is at least about 0.25, at least about 0.5, at least about 0.75, at least about 1, or at least about 1.5 percent higher than the refractive index of at least

one of the substrates used to form the multiple layer interlayer. For example, the panel can have an equivalent refractive index that is at least 0.010, at least about 0.015, at least about 0.020, or at least about 0.025 higher than the refractive index of one or more rigid substrates used to form the panel. In certain embodiments, the equivalent refractive index of a multiple layer panel may be less than the refractive index of the interlayer used to form the panel and can be, for example, at least about 0.5, at least about 1, at least about 1.5, at least about 2, at least about 2.5, at least about 3, at least about 5, or at least about 7 percent lower than the refractive index of the interlayer used to form the multiple layer panel. The difference between the refractive index of the interlayer and the equivalent refractive index of the panel can be at least about 0.010, at least about 0.025, at least about 0.050, at least about 0.10, at least about 0.125, at least about 0.50, or at least about 0.75.

[0137] Multiple layer panels as described herein can be used for a variety of end use applications, including, for example, for automotive windshields and windows, aircraft windshields and windows, panels for various transportation applications such as marine applications, rail applications, etc., structural architectural panels such as windows, doors, stairs, walkways, balusters, decorative architectural panels, weather-resistant panels, such as hurricane glass or tornado glass, ballistic panels, and other similar applications.

[0138] In certain embodiments, multiple layer panels as described herein may exhibit a reduction in interfering double or reflected “ghost” images when, for example, used for projecting a heads-up display (HUD) image onto the windshield of an automobile or aircraft. Typically, ghost images are most problematic when the windshield has a generally uniform thickness profile, due to the differences in position of the projected image when it is reflected off the inside and outside surfaces of the glass. In some embodiments, however, multiple layer panels as described herein can minimize projection of ghost images such that, for example, the panel has a double image separation distance significantly lower than would be exhibited by a conventional panel.

[0139] The method of determining double image separation includes providing a multiple layer panel that includes at least a pair of rigid substrates and an interlayer disposed therebetween. The interlayer can include any properties of, or may be, any of the interlayers described herein. In some embodiments, the interlayer may be a tapered interlayer comprising, for example, at least one tapered zone having a wedge angle of at least 0.05 mrad. The substrates may also include one or more properties of the substrates described herein and, in certain embodiments, may comprise glass.

[0140] To measure the double image separation of a given panel, a projection image can be generated by passing light through at least a portion of the panel. In some embodiments, the light passing through the panel includes an image such as, for example, a grid, a line, a shape, or a picture. In some embodiments, the image may be generated by reflecting a thin film transistor display off of a substantially flat mirrored surface, although other suitable methods of generating images may be used. If the multiple layer panel is formed using a tapered interlayer, at least a portion of the light passing through the panel may be passed through at least a portion of the tapered zone. Alternatively, or in addition, the light may pass through one or more other zones of interest along the panel in order to determine the double image separation formed as light projects through those zones.

[0141] Once light has passed through and is reflected off the surfaces of the panel, the projection image can be projected onto a surface and then captured to form a captured image. In some embodiments, the projected image displayed on the surface may include a primary image and a secondary "ghost" image, off-set and slightly overlapping the primary image, as shown in FIG. 10. The projected image may be captured using a digital camera or other suitable device, and the capture may include digitizing the projected image to form a digital projection image comprising a plurality of pixels.

[0142] Once digitized, the captured image can be quantitatively analyzed to form a profile that includes at least one primary image indicator and at least one

secondary image indicator. The analyzing may be performed by converting at least a portion of the digital projection image to a vertical image matrix that includes a numerical value representing the intensity of pixels in that portion of the image. A column of the matrix can then be extracted and graphed against pixel number, as shown in FIG. 11, to provide the profile. The primary image indicator of the profile can then be compared with the secondary image indicator of the profile to determine a difference. In some embodiments, the primary image indicator may comprise the higher intensity peaks of the graph, while the secondary image indicator may be the lower intensity peaks. Any suitable difference between the two indicators can be determined and, in some embodiments, can be the difference in position between the two indicators in the profile graph. Based on the difference, the double image separation distance for the portion of the panel being tested can be calculated. A specific use of the above method for determining double image separation distance is described in Example 11.

[0143] Multiple layer panels configured according to embodiments of the present invention may have a significantly lower double image separation distance than conventionally-formed panels. For example, in some embodiments, the double image separation distance of panels as described herein may be at least about 10, at least about 20, at least about 30, at least about 40 percent less than the double image separation distance of panels formed from conventional interlayers such as, for example, those having a refractive index less than 1.480, all other things being equal. The double image separation distance for a given panel may depend, in part, on its specific configuration, including the type and dimensions of its interlayer and substrates. Specific examples of double image separation distances for several inventive and comparative panels are provided in the Examples.

[0144] When laminating the polymer layers or interlayers between two rigid substrates, such as glass, the process can include at least the following steps: (1) assembly of the two substrates and the interlayer; (2) heating the assembly via an IR radiant or convective device for a first, short period of time; (3) passing the assembly

into a pressure nip roll for the first de-airing; (4) heating the assembly for a short period of time to about 60°C to about 120°C to give the assembly enough temporary adhesion to seal the edge of the interlayer; (5) passing the assembly into a second pressure nip roll to further seal the edge of the interlayer and allow further handling; and (6) autoclaving the assembly at temperature between 135°C and 150°C and pressures between 150 psig and 200 psig for about 30 to 90 minutes. Other methods for de-airing the interlayer-glass interface, as described according to some embodiments in steps (2) through (5) above include vacuum bag and vacuum ring processes, and both may also be used to form interlayers of the present invention as described herein.

[0145] In some embodiments, the multiple layer panel may include at least one polymer film disposed on the layer or interlayer, forming a multiple layer panel referred to as a “bilayer.” In some embodiments, the interlayer utilized in a bilayer may include a multilayer interlayer, while, in other embodiments, a monolithic interlayer may be used. The use of a polymer film in multiple layer panels as described herein may enhance the optical character of the final panel, while also providing other performance improvements, such as infrared absorption. Polymer films differ from polymer layers or interlayers in that the films alone do not provide the necessary penetration resistance and glass retention properties. The polymer film can also be thinner than the sheet, and may have a thickness in the range of from 0.001 to 0.25 mm. Poly(ethylene terephthalate) (“PET”) is one example of a material used to form the polymer film.

[0146] The following examples are intended to be illustrative of the present invention in order to teach one of ordinary skill in the art to make and use the invention and are not intended to limit the scope of the invention in any way.

## EXAMPLES

[0147] The following Examples describe the preparation of several resin compositions, layers, and interlayers that include various poly(vinyl acetal) reins. As

described below, several tests performed on many of the compositions, layers, and interlayers were used to evaluate the acoustic and optical properties of several comparative and inventive materials.

**Example 1: Preparation of High Refractive Index Poly(vinyl acetal) Resins**

[0148] Several comparative poly(vinyl acetal) resins, referred to as Comparative Resins CR-1 through CR-12 in Table 2 below, were prepared by acetalizing polyvinyl alcohol with one or more aldehydes including n-butyraldehyde (nBuCHO; RI = 1.377), iso-butyraldehyde (iBuCHO; RI = 1.374), and 2-ethylhexyl aldehyde (2EHCHO; RI = 1.414). The composition of the resulting resins were measured using either the ASTM D1396 or FT-IR/SEC method described in detail previously. The refractive index and glass transition temperature ( $T_g$ ) of several of the resins was also measured according to the methods described previously, and the results are provided in Table 2, below.

**Table 2: Properties of Several Comparative Poly(vinyl acetal) Resins**

Resin	Residual Hydroxyl (wt%)	Residual Acetate (wt%)	Residual Polyvinyl nBuCHO (wt%)	Residual Polyvinyl iBuCHO (wt%)	Residual Polyvinyl 2EHCHO (wt%)	Resin Refractive Index (nD25)
CR-1	21	1	78	-	-	1.491
CR-2	19	1	80	-	-	1.490
CR-3	19	1	46	-	34	1.489
CR-4	16	1	83	-	-	1.488
CR-5	15	1	39	-	45	1.487
CR-6	13	1	86	-	-	1.486
CR-7	12	18	70	-	-	1.481
CR-8	12	12	76	-	-	nd
CR-9	12	10	78	-	-	1.483
CR-10	11	1	88	-	-	1.485
CR-11	12	1	-	88	-	nd
CR-12	13	1	-	86	-	nd

nd = not determined

[0149] Additionally, several poly(vinyl acetal) resins according to embodiments of the present invention were also prepared in a similar manner. These inventive resins, referred to as Disclosed Resins DR-1 through DR-21 in Table 3, were formed by acetalizing polyvinyl alcohol with mixtures of n-butyraldehyde and various high refractive index aldehydes, including benzaldehyde (BzCHO RI = 1.545), cinnamaldehyde (CCHO; RI = 1.620), 4-chlorobenzaldehyde (4-ClBzCHO; RI = 1.5850), 2-phenylpropionaldehyde (2PHPrCHO; RI = 1.517), and hydrocinnamaldehyde (HCCHO; RI = 1.523). The refractive index of several of the Disclosed Resins was also determined and the results are summarized in Table 3, below.



Table 3: Properties of Disclosed Resins

Resin	Residual Hydroxyl (wt%)	Residual Acetate (wt%)	Aldehyde Component (wt%)	HCCHO/ BuCHO (mol/mol)	CCHO/ BuCHO (mol/mol)	2PhPrCHO/ BuCHO (mol/mol)	BzCHO/Bu CHO (mol/mol)	4ClBzCHO/ BuCHO (mol/mol)	HCCHO/ iBuCHO	Resin Refractive Index (nD25)
DR-1	11	1	88	5/95	-	-	-	-	-	1.496
DR-2	11	1	88	10/90	-	-	-	-	-	1.500
DR-3	11	1	88	15/85	-	-	-	-	-	1.504
DR-4	11	1	88	20/80	-	-	-	-	-	1.510
DR-5	11	1	88	25/75	-	-	-	-	-	1.511
DR-6	11	1	88	50/50	-	-	-	-	-	1.528
DR-7	11	1	88	100/0	-	-	-	-	-	1.552
DR-8	19	1	80	10/90	-	-	-	-	-	nd
DR-9	19	1	80	25/75	-	-	-	-	-	nd
DR-10	19	1	80	50/50	-	-	-	-	-	nd
DR-11	19	1	80	100/0	-	-	-	-	-	nd
DR-12	12	18	70	25/75	-	-	-	-	-	nd
DR-13	12	10	78	25/75	-	-	-	-	-	nd
DR-14	17	1	82	-	25/75	-	-	-	-	1.507
DR-15	15	1	84	-	-	25/75	-	-	-	1.505
DR-16	14	1	85	-	-	25/75	-	-	-	1.503
DR-17	18	1	81	-	-	-	25/75	-	-	1.507
DR-18	14	1	85	-	-	-	25/75	-	-	1.501
DR-19	20	1	79	-	-	-	-	25/75	-	1.507
DR-20	14	1	85	-	-	-	-	25/75	-	1.509
DR-21	11	1	88	-	-	-	-	-	25/75	nd

nd = not determined

[0150] As shown in Tables 2 and 3, above, polyvinyl acetal resins comprising residues of at least one high refractive index aldehyde, including those listed above, tend to exhibit higher refractive indices than those comprising residues of aldehydes such as n-butyraldehyde, iso-butyraldehyde, and 2-ethylhexyl aldehyde.

#### **Example 2: Preparation of High Refractive Index Resin Interlayers**

[0151] Several Comparative and Disclosed Interlayers were formed by mixing and melt blending several of the Comparative Resins listed in Table 2 and several of the Disclosed Resins listed in Table 3 above, with varying amounts of the plasticizer triethylene glycol bis(2-ethylhexanoate) (3GEH). The composition, refractive index, and glass transition temperature of each of the resulting interlayers, referred to as Comparative Interlayers CL-1 through CL-14 and Disclosed Interlayers DL-1 through DL-26, were measured as described previously, and the results are respectively summarized in Tables 4 and 5, below.

**Table 4: Properties of Several Comparative Interlayers**

Interlayer	Resin	Plasticizer (phr)	Refractive Index (nD25)	T <sub>g</sub> (°C)
CL-1	CR-1	34	1.478	36
CL-2	CR-2	38	1.477	30
CL-3	CR-3	38	1.475	28
CL-4	CR-4	50	1.472	17
CL-5	CR-4	30	1.477	30
CL-6	CR-5	75	1.468	6
CL-7	CR-6	75	1.467	2
CL-8	CR-7	75	1.464	-4
CL-9	CR-8	75	1.466	-3
CL-10	CR-9	75	1.465	-3
CL-11	CR-10	75	1.466	-2
CL-12	CR-10	50	1.47	12
CL-13	CR-11	75	nd	nd
CL-14	CR-12	75	nd	nd

nd = not determined

**Table 5: Properties of Several Disclosed Interlayers**

Interlayer	Resin	Plasticizer content (phr)	Sheet Refractive Index (nD25)	T <sub>g</sub> (°C)
DL-1	DR-1	75	1.473	1
DL-2	DR-1	50	1.479	12
DL-3	DR-2	75	1.475	0
DL-4	DR-3	75	1.477	0
DL-5	DR-4	75	1.480	1
DL-6	DR-5	75	1.482	1
DL-7	DR-6	75	1.490	2
DL-8	DR-7	75	1.506	3
DL-9	DR-14	75	1.480	12
DL-10	DR-15	75	1.478	11
DL-11	DR-16	75	1.477	7
DL-12	DR-17	75	1.480	22
DL-13	DR-18	75	1.476	12
DL-14	DR-19	75	1.480	17
DL-15	DR-20	75	1.479	12
DL-16	DR-12	75	nd	nd
DL-17	DR-13	75	nd	nd
DL-18	DR-21	75	nd	nd
DL-19	DR-8	38	1.477	30
DL-20	DR-9	38	1.487	nd
DL-21	DR-10	38	1.501	nd
DL-22	DR-11	38	1.525	nd
DL-23	DR-5	38	nd	nd
DL-24	DR-5	50	1.488	nd
DL-25	DR-5	60	1.484	2
DL-26	DR-7	38	nd	nd

nd = not determined

[0152] As shown in Tables 4 and 5, above, Disclosed Interlayers utilizing higher refractive index Disclosed Resins from Table 3 exhibit higher refractive indices than Comparative Interlayers formulated with lower refractive index resins, such as the

Comparative Resins listed in Table 2. Additionally, as shown by comparison of Disclosed Interlayers DL-1 and DL-2 and DL-6 and DL-23 through DL-25, the amount of plasticizer used to form an interlayer affects both the glass transition temperature and refractive index of the layer, although not necessarily to the same extent for all resins. For example, as shown by the comparison of DL-1 (75 phr 3GEH; DR-1) and DL-2 (50 phr 3GEH; DR-1), reducing the amount of plasticizer by about 33 percent increases the glass transition temperature of the interlayer by 11°C (from 1°C to 12°C), but only increases the refractive index by 0.006 (from 1.473 to 1.479). However, as shown by the comparison of DL-25 (60 phr 3GEH; DR-5) and DL-6 (75 phr of 3GEH; DR-5), reducing the plasticizer content in layers formed by this resin increases the glass transition temperature by only 1°C (from 1°C to 2°C), while increasing the refractive index by 0.004 (from 1.484 to 1.488).

**Example 3: Preparation of Interlayers using High Refractive Index Resins**

[0153] Several of the Comparative and Disclosed Interlayers formed in Example 2 and respectively summarized in Tables 4 and 5, above, were used to create several Comparative and Disclosed Multilayer Interlayers. Each multilayer interlayer included a pair of outer “skin” layers, having a total thickness of 28 mils, sandwiching an inner “core” layer having a thickness of 5 mils, usually formed of a resin having a lower residual hydroxyl content. The composition and several properties of the interlayers including refractive index, glass transition temperature, mottle, and loss factor, were measured as described above, and the results for the Comparative Multilayer Interlayers (CI-1 to CI-16) and Disclosed Multilayer Interlayers (DI-1 to DI-29) are summarized in Tables 6 and 7, below.

Table 6: Properties of Several Comparative Multilayer Interlayers

Interlayer	Polymer layer				T <sub>g</sub> (°C)		Refractive Index			$\Delta$ Refractive Index (Skin- Core)	Refractive Index of Interlayer (nD25)	Mottle	Loss Factor at 20°C
	Skin 1	Core	Skin 2	Skin 1	Core	Skin 2	Skin 1	Core	Skin 2				
CI-1	CL-2	CL-7	CL-2	30	2	30	1.477	1.467	1.477	0.010	1.475	4	0.29
CI-2	CL-2	CL-11	CL-2	30	-2	30	1.477	1.466	1.477	0.011	1.475	5	0.33
CI-3	CL-2	CL-8	CL-2	30	-4	30	1.477	1.464	1.477	0.013	1.475	5	nd
CI-4	CL-1	CL-9	CL-1	36	-3	36	1.478	1.466	1.478	0.012	1.477	5	nd
CI-5	CL-1	CL-8	CL-1	36	-4	36	1.478	1.464	1.478	0.014	nd	5	0.36
CI-6	CL-2	CL-9	CL-2	30	-3	30	1.477	1.466	1.477	0.011	nd	5	nd
CI-7	CL-1	CL-11	CL-1	36	-2	36	1.478	1.466	1.478	0.012	1.477	5	0.35
CI-8	CL-2	CL-10	CL-2	30	-3	30	1.477	1.465	1.477	0.012	nd	5	0.31
CI-9	CL-1	CL-7	CL-1	36	2	36	1.478	1.467	1.478	0.011	nd	5	nd
CI-10	CL-1	CL-10	CL-1	36	-3	36	1.478	1.465	1.478	0.013	nd	5	nd
CI-11	CL-2	CL-13	CL-2	30	nd	30	nd	nd	nd	nd	nd	> 5	nd
CI-12	CL-2	CL-14	CL-2	30	nd	30	nd	nd	nd	nd	nd	> 5	nd
CI-13	CL-1	CL-13	CL-1	36	nd	36	nd	nd	nd	nd	nd	> 5	nd
CI-14	CL-1	CL-14	CL-1	36	nd	36	nd	nd	nd	nd	nd	> 5	nd
CI-15	CL-2	DL-7	CL-2	30	2	30	1.477	1.490	1.477	-0.013	1.479	> 5	nd
CI-16	CL-2	DL-8	CL-2	30	3	30	1.477	1.506	1.477	-0.029	1.481	> 5	nd

nd = not determined

Table 7: Properties of Some Disclosed Multilayer Interlayers

Interlayer	Polymer layer		T <sub>g</sub> (°C)			Refractive Index (nD25)			Δ Refractive Index (Skin-Core)	Refractive Index of Interlayer	Mottle	
	Skin 1	Core	Skin 2	Skin 1	Core	Skin 2	Skin 1	Core	Skin 2			
DI-1	CL-2	DL-1	CL-2	30	1	30	1.477	1.473	1.477	0.004	1.476	1
DI-2	CL-2	DL-4	CL-2	30	0	30	1.477	1.477	1.477	0.000	1.477	1
DI-3	CL-5	DL-6	CL-5	30	1	30	1.477	1.482	1.477	-0.005	1.478	nd
DI-4	CL-2	DL-6	CL-2	30	1	30	1.477	1.482	1.477	-0.005	nd	< 1
DI-5	CL-1	DL-6	CL-1	36	1	36	1.478	1.482	1.478	-0.004	nd	< 1
DI-6	CL-2	DL-3	CL-2	30	0	30	1.477	1.475	1.477	0.002	nd	< 1
DI-7	CL-2	DL-5	CL-2	30	1	30	1.477	1.480	1.477	-0.003	nd	< 1
DI-8	CL-2	DL-6	CL-1	30	1	36	1.477	1.482	1.478	-0.005	nd	< 1
DI-9	CL-2	DL-6	CL-5	30	1	30	1.477	1.482	1.477	-0.005	nd	< 1
DI-10	CL-2	DL-16	CL-2	30	nd	30	1.477	nd	1.477	nd	nd	< 1
DI-11	CL-2	DL-17	CL-2	30	nd	30	1.477	nd	1.477	nd	nd	< 1
DI-12	CL-1	DL-16	CL-1	36	nd	36	1.478	nd	1.478	nd	nd	< 1
DI-13	CL-1	DL-17	CL-1	36	nd	36	1.478	nd	1.478	nd	nd	< 1
DI-14	CL-2	DL-18	CL-2	30	nd	30	1.477	nd	1.477	nd	nd	< 1
DI-15	CL-1	DL-18	CL-1	36	nd	36	1.478	nd	1.478	nd	nd	< 1

nd = not determined

Table 7 (Continued): Properties of Some Disclosed Multilayer Interlayers

Interlayer	Polymer layer		T <sub>g</sub> (°C)		Refractive Index (nD25)				Δ Refractive Index (Skin-Core)	Refractive Index of Interlayer	Mottle
	Skin 1	Core	Skin 1	Skin 2	Core	Skin 1	Skin 2	Core			
DI-16	DL-19	DL-24	DL-19	30	1	1.477	1.488	1.477	-0.005	1.478	< 1
DI-17	CL-2	DL-9	CL-2	30	12	1.477	1.480	1.477	-0.003	nd	< 1
DI-18	CL-2	DL-10	CL-2	30	11	1.477	1.478	1.477	-0.001	nd	< 1
DI-19	CL-2	DL-11	CL-2	30	7	1.477	1.477	1.477	0.000	nd	< 1
DI-20	CL-2	DL-12	CL-2	30	22	1.477	1.480	1.477	-0.003	nd	< 1
DI-21	CL-2	DL-13	CL-2	30	12	1.477	1.476	1.477	0.001	nd	< 1
DI-22	CL-2	DL-14	CL-2	30	17	1.477	1.480	1.477	-0.003	nd	< 1
DI-23	CL-2	DL-15	CL-2	30	12	1.477	1.479	1.477	-0.002	nd	< 1
DI-24	DL-20	DL-7	DL-20	nd	2	1.487	1.490	1.487	-0.003	nd	< 1
DI-25	DL-21	DL-7	DL-21	nd	2	1.501	1.490	1.501	-0.011	nd	> 5
DI-26	DL-22	DL-7	DL-22	nd	2	1.525	1.490	1.525	-0.035	nd	> 5
DI-27	DL-20	DL-8	DL-20	nd	3	1.487	1.506	1.487	-0.009	nd	3
DI-28	DL-21	DL-8	DL-21	nd	3	1.501	1.506	1.501	-0.005	nd	< 1
DI-29	DL-22	DL-8	DL-22	nd	3	1.525	1.506	1.525	-0.019	nd	> 5

[0154] As shown in Table 6, above, interlayers formed from skin and core layers having refractive index differences of 0.010 or more exhibited optical defects, as indicated by the mottle values greater than 5. As shown in Table 7, however, interlayers formed from skin and core layers having refractive index differences of less than 0.010 exhibited low mottle values of 1 or less. Additionally, as shown in Table 7, such low mottle values were achievable by interlayers having a core layer with a higher or lower refractive index than the skin layer, as long as the absolute value of the difference between the refractive indices of adjacent layers was less than 0.010. Also as shown in Table 7, interlayers formed from skin and core layers both having high RI aldehyde residues and having refractive index differences of greater than 0.010 exhibited high mottle values of 5 or greater.

#### Example 4: Stability of Multilayer Interlayers

[0155] Two Comparative Multilayer Interlayers, CI-2 and CI-7, and two Disclosed Multilayer Interlayers, DI-4 and DI-5, prepared as described in Examples 1-3 above, were tested to determine the relative stability of the interlayers over time. Net plasticizer migration was measured by comparing the glass transition temperatures of each layer at an initial time ( $t=0$ ) and after the layers have achieved equilibrium. The results are summarized in Table 8, below.

**Table 8: Net Plasticizer Migration and Properties of Exemplary Comparative and Disclosed Interlayers**

Interlayer	T <sub>g</sub> at t=0 (°C)		T <sub>g</sub> at Equilibrium (°C)		$\Delta T_g$ (°C)		$\Delta$ Refractive Index (Skin-Core)	Mottle
	T <sub>g</sub> Skin	T <sub>g</sub> Core	T <sub>g</sub> Skin	T <sub>g</sub> Core	$\Delta T_g$ Skin	$\Delta T_g$ Core		
CI-2	36	4.2	36	4.6	0.0	0.4	0.011	> 5
DI-4	36	1.4	36.4	1.4	0.4	0.0	-0.005	< 1
CI-7	42.5	2.2	42.5	1.4	0.0	-0.8	0.012	> 5
DI-5	40.9	0.2	41.7	-1.8	0.8	-2.0	-0.004	< 1



[0156] Disclosed Interlayer DI-4 (as well as Comparative Interlayers CI-2 and CI-7) exhibited minimal changes in the glass transition temperature of both the skin and core layers of the interlayer at equilibrium. This indicates a minor amount of plasticizer migration between the skin and core layers of each of Interlayers CI-2, CI-7, and DI-4. Although Comparative Interlayers CI-2 and CI-7 may be relatively stable, both exhibited a mottle value greater than 5, which would be unacceptable for most optical applications. In contrast, the mottle value exhibited by Disclosed Interlayer DI-4 was less than 1.

[0157] The slight decrease in glass transition temperature exhibited by Disclosed Interlayer DI-5 at equilibrium indicated that a small amount of plasticizer migrated from the skin layer to the core layer. Such migration could be alleviated by utilizing a smaller amount of plasticizer in the skin layer or a larger amount in the core. Even so, the refractive index of the core layer and the skin layer of Disclosed Interlayer DI-5 differed by only 0.004 and, as a result, the interlayer also exhibited a mottle value less than 1.

#### **Example 5: Blended Poly(vinyl acetal) Resins**

[0158] Several Comparative and Disclosed Resins, prepared as described in Example 1 above, were mixed and melt-blended together with 38 phr of 3GEH plasticizer to form Comparative Blended Layers CBL-16 and CBL-17 and Disclosed Blended Layers DBL-27 and DBL-28. Comparative Polymer Layer CL-2 is listed in Table 4. Comparative Polymer Layer CL-15 was formulated with Comparative Resin CR-10 and 38 phr of 3GEH, while Disclosed Polymer layers DL-27 and DL-28 were formulated with Disclosed Resins DR-3 and DR-5, respectively, and 38 phr of 3GEH. The haze and percent visual transmittance ( $T_{vis}$ ) for each blended resin interlayer were measured, along with the haze and percent visual transmittance. The results are provided in Table 9, below.

**Table 9: Haze and Percent Visual Transmittance of Several Polymer Layers**

Interlayer	Layer Composition				Plasticizer content (phr)	Property	
	CR-2 (wt%)	CR-10 (wt%)	DR-3 (wt%)	DR-5 (wt%)		Haze (%)	% T <sub>vis</sub>
CL-2	100	-	-	-	38	0.5	87.7
CL-15	-	100	-	-	38	0.5	87.8
CBL-16	96	4	-	-	38	2.1	84.3
CBL-17	89	11	-	-	38	14.7	80.2
DBL-27	96	-	4	-	38	0.8	87.8
DBL-28	89	-	-	11	38	0.6	87.8
DL-27	-	-	100	-	38	0.5	87.7
DL-28	-	-	-	100	38	0.5	87.8

[0159] As shown in Table 9 above, Comparative Blended Interlayers CBL-16 and CBL-17, which were formed from a blend of Comparative Resins CR-2 and CR-10, exhibited high haze values and lower percent visual transmittance than single polymer layers of Comparative Resin CR-2 (Comparative Layer CL-2) or CR-10 (Comparative Interlayer CL-15). In contrast, Disclosed Blended Interlayers DBL-27 and DBL-28, which were formed from a blend of Comparative Resin CR-2 and a high refractive index Disclosed Resin (Resin DR-3 in Layer DBL-27 or Resin DR-5 in Layer DBL-28), exhibited substantially the same haze and percent visual transmittance as Comparative Interlayer CL-2, which was formulated with Comparative Resin CR-2 alone. Thus, addition of inventive high refractive index resins to a comparative interlayer does not reduce the optical quality of the resulting interlayer.

#### **Example 6: Preparation of Interlayers having High Refractive Index Additives**

[0160] Several poly(vinyl acetal) resins were prepared by acetalizing polyvinyl alcohol with n-butyraldehyde. The resins, which had different residual hydroxyl contents, were melt blended with varying amounts of a 3GEH plasticizer and used to form various layers of multilayer interlayers. Each interlayer had an inner “core” layer having a thickness of 5 mils sandwiched between two outer “skin” layers, each having a thickness of 14 mils. The poly(vinyl butyral) resin used to form the core layers had a hydroxyl content of 11 weight percent and the resin used for the skin layers had a

hydroxyl content of 19 weight percent. Both resins had a residual acetate content of about 2 weight percent.

[0161] Comparative Interlayers CI-17 through CI-19 were formed with polymer layers plasticized with 3GEH, which was present in varying amounts in the core and skin layers. In addition to the 3GEH, Disclosed Interlayers DI-30 through DI-38 also included varying amounts of two different high refractive index additives, Benzoflex™ 2-45 (diethyleneglycol dibenzoate; commercially available from Eastman Chemical Company, Kingsport, Tennessee) (Additive A-1), which had a melting point of 28°C and a refractive index of 1.542; and Benzoflex™ 352 (1,4-cyclohexane dimethanol dibenzoate; commercially available from Eastman Chemical Company) (Additive A-2), which had a melting point of 118°C and a refractive index of 1.554. The refractive index and glass transition temperatures of each of the layers of Comparative Interlayers CI-17 through CI-19 and Disclosed Interlayers DI-30 through DI-38 were measured and the results are summarized in Table 10 below.

Table 10: Properties of Several Comparative and Disclosed Interlayers

Interlayer	Skin Layers				Core Layer										$\Delta$ Refractive Index (Skin-Core)
	Plasticizer Content (P) (phr)	High RI Additive Type	High RI Additive (A) (phr)	Total A+P	A:P Ratio	RI	T <sub>g</sub> (°C)	Plasticizer Content (P) (phr)	High RI Additive Type	High RI Additive (A) (phr)	Total A+P	A:P Ratio	RI	T <sub>g</sub> (°C)	
CI-17	40	-	-	-	-	1.476	30	65.0	-	-	-	-	1.467	2.9	0.008
CI-18	40	-	-	-	-	1.476	30	75.0	-	-	-	-	1.466	-2.0	0.010
CI-19	40	-	-	-	-	1.476	30	85.0	-	-	-	-	1.465	-3.1	0.011
DI-30	20	A-1	20	40	1.0	1.492	29	32.5	A-1	32.5	65	1.0	1.488	4.2	0.004
DI-31	20	A-1	20	40	1.0	1.492	29	37.5	A-1	37.5	75	1.0	1.488	-0.6	0.004
DI-32	20	A-1	20	40	1.0	1.489	29	42.5	A-1	42.5	85	1.0	1.488	-3.3	0.004
DI-33	17.2	A-1	22.8	40	1.3	1.496	30	22.8	A-1	42.2	65	1.9	1.494	5.2	0.002
DI-34	17.2	A-1	22.8	40	1.3	1.496	30	26.2	A-1	48.8	75	1.9	1.494	2.4	0.002
DI-35	17.2	A-1	22.8	40	1.3	1.496	30	29.8	A-1	55.2	85	1.9	1.494	-2.1	0.002
DI-36	20	A-2	20	40	1.0	1.493	34	27.5	A-2	27.5	55	1.0	1.490	15.9	0.003
DI-37	20	A-2	20	40	1.0	1.493	34	35.0	A-2	35.0	70	1.0	1.490	8.4	0.003
DI-38	20	A-2	20	40	1.0	1.493	34	42.5	A-2	42.5	85	1.0	1.491	1.8	0.002

[0162] As shown in Table 10, above, increasing the plasticizer content of the core layer of an interlayer that only included a 3GEH plasticizer reduced the glass transition temperature of the layer, which, ultimately, would have improved its acoustic performance. However, such an increase also widened the difference between the refractive indices of the skin and core layers, thereby reducing the optical quality of the interlayer. As shown by comparison with Disclosed Interlayers DI-30 through DI-38 in Table 10, the refractive index of core layers formulated with an additional high refractive index additive, remained fairly constant with increased plasticizer loading, while still exhibiting a similar reduction in glass transition temperature. The result was an interlayer having core and skin layers with nearly the same refractive index, which greatly reduced optical defects such as mottle. At the same time, the core layer also exhibited a sufficiently low glass transition temperature, indicating that the resin also had acoustic properties.

**Example 7: Preparation of Core Layers having Reactive High Refractive Index Additives**

[0163] Several polymer layers, used to simulate the inner core layer of a multilayer interlayer, were formed by melt blending a polyvinyl n-butyral resin having a residual hydroxyl content of 11 weight percent and a residual acetate content of about 2 weight percent with varying amounts of 3GEH plasticizer. Comparative Layer CL-16 included 75 phr of 3GEH, while Disclosed Layers DL-29 through DL-31 were formulated with various mixtures of 3GEH and a reactive high refractive index additive (reactive high RI additive). The reactive high RI additive used in Disclosed Layers DL-29 and DL-30 (Additive A) was diphenyldimethoxysilane (commercially available as SID4535.0 from Gelest, Inc., Morrisville, Pennsylvania), and the reactive high RI additive used in Disclosed Layer DL-31 (Additive B) was phthalic anhydride (commercially available from Sigma Aldrich Co., St. Louis, Missouri). The refractive index of Comparative Layer CL-16 and each of Disclosed Layers DL-29 through DL-31 was measured and the results are provided in Table 11, below.

**Table 11: Refractive Index of Comparative and Disclosed Core Polymer Layers**

<b>Polymer Layer</b>	<b>Plasticizer Content (phr)</b>	<b>Additive A Content (phr)</b>	<b>Additive B Content (phr)</b>	<b>Refractive Index (nD25)</b>
CL-16	75	-	-	1.466
DL-29	72	3	-	1.468
DL-30	72	6	-	1.470
DL-31	72	-	3	1.470

[0164] As shown in Table 11, polymer layers formed using 3GEH in combination with one or more reactive high refractive index additives had a higher refractive index than polymer layers formulated with only 3GEH. As a result, when employed as an inner core layer in a multilayer interlayer, the Disclosed Layers DL-29 through DL-31 had a refractive index that more closely matched the refractive index of a skin layer formed of polyvinyl n-butyral (RI=1.477). As a result, multilayer interlayers formed with Disclosed Layers DL-29 through DL-31 as a core layer exhibit fewer optical defects than multilayer interlayers formed with Comparative Layer CL-16 as an inner core layer.

#### **Example 8: Various Interlayers with Resin Blends having a High Refractive Index Plasticizer**

[0165] Two polyvinyl n-butyral resins, R-1 and R-2, were prepared according to the procedure described above in Example 1. Resin R-1 had a residual hydroxyl content of 19 weight percent, while resin R-2 had a residual hydroxyl content of 11 weight percent. Both resins had residual acetate contents of 2 weight percent. Several resin blends were prepared that included varying amounts of resins R-1 and R-2 in order to simulate various blending rates. The blends were combined with 38 phr of a plasticizer selected from 3GEH (plasticizer P-1; RI = 1.442), dioctyl phthalate (plasticizer P-2; RI = 1.485), a blend of 30 weight percent 3GEH and 70 weight percent Benzoflex® 2088, which is commercially available from Eastman Chemical Company, Kingsport, Tennessee, (plasticizer P-3; RI = 1.506), and nonylphenyl tetraethylene glycol (plasticizer P-4; RI = 1.500). The resulting plasticized resins were then formed

into single sheets that included both resins and the plasticizer. The refractive index, haze, and percent visual transmittance was determined for each sheet and the results are provided in Table 12, below.

Table 12: Properties of Several Resin Blends

Resin A (wt %)	Resin B (wt %)	Plasticizer P-1			Plasticizer P-2			Plasticizer P-3			Plasticizer P-4		
		RI	Haze	T <sub>vis</sub>	RI	Haze	T <sub>vis</sub>	RI	Haze	T <sub>vis</sub>	RI	Haze	T <sub>vis</sub>
		Interlayer	(%)	(%)	Interlayer	(%)	(%)	Interlayer	(%)	(%)	Interlayer	(%)	(%)
100	0	1.477	0.4	88.5	1.494	0.4	88.5	1.495	0.3	88.4	1.493	0.3	88.5
98.9	1.1	1.477	0.6	87.9	1.494	0.4	88.6	1.495	0.3	88.8	1.493	0.3	88.7
97.8	2.2	1.476	1.2	87.1	1.498	0.3	88.4	1.495	0.3	88.7	1.493	0.2	88.7
95.6	4.4	1.477	1.9	84.6	1.495	0.4	88.4	1.495	0.3	88.6	1.492	0.4	88.5
91.2	8.8	1.476	5.4	81.7	1.495	0.5	88.3	1.495	0.4	88.5	1.493	0.3	88.6
88.9	11.1	1.476	12.9	80.6	1.494	0.6	88.2	1.495	0.4	88.2	1.492	0.4	88.2



[0166] As shown in Table 12, above, although blended polymer layers formulated with plasticizer P-1 maintained a substantially constant refractive index with increasing amounts of the lower hydroxyl content resin R-2, the optical properties of these resin blends having high levels of R-2 worsened as the amount of R-2 increased. For example, as shown in Table 12, the haze of blends that included more than 1.1 percent of resin R-2 increased, while the percent visual transmittance of these blends decreased from 88.5 percent to 81.7 percent.

[0167] In contrast, the resin blends including more than 2.2 percent of resin R-2 that were plasticized with higher refractive index plasticizers P-2 through P-4, each exhibited substantially the same haze value and percent visual transmittance as blends having lower amounts of resin R-2. Therefore, it can be concluded that the resin blends utilizing higher refractive index plasticizers, such as plasticizers P-2 through P-4, may permit higher amounts of lower hydroxyl content resins without adversely impacting the optical properties of the final blend.

**Example 9: Poly(vinyl butyral) Layers including a High Refractive Index Plasticizer**

[0168] Several poly(vinyl n-butyral) layers were formed by combining and melt blending three different poly(vinyl n-butyral) resins (PVB-1 through PVB-3) with different types and amounts of plasticizer. Each of the resins PVB-1 through PVB-3 had a different residual hydroxyl content, ranging from 11 to 20.4 weight percent, and all three resins had a residual vinyl acetate content of 1 weight percent. Comparative Layers CL-17 through CL-19 were formulated with varying amounts of triethylene glycol di-(2-ethylhexanoate) ("3GEH"; RI=1.442), while Disclosed Layers DL-32 through DL-37 included a mixture of 3GEH with Benzoflex™ 354 (commercially available from Eastman Chemical Company, Kingsport, Tennessee)(RI=1.53). The refractive index of each layer was measured and the results are summarized in Table 13, below.

**Table 13: Several Poly(vinyl) Butyral Layers with Various Plasticizers**

Polymer Layer	Residual hydroxyl content (wt%)	Plasticizer Content			Total plasticizer content (phr)	Refractive Index (nD25)
		3GEH content (phr)	Benzoflex™ 354 (phr)	Ratio of 3GEH to Benzoflex™ 354		
CL-17	19	38	-	-	38	1.477
CL-18	19	38	-	-	38	1.477
CL-19	11	75	-	-	38	1.466
DL-32	20.4	30.1	12.9	30/70	43	1.484
DL-33	20.4	25.8	17.2	40/60	43	1.485
DL-34	20.4	21.5	21.5	50/50	43	1.488
DL-35	11	59.5	25.5	30/70	85	1.479
DL-36	11	51	34	40/60	85	1.481
DL-37	11	42.5	42.5	50/50	85	1.486

[0169] As shown in Table 13, above, polymer layers that included a high refractive index plasticizer exhibited a higher refractive index than those that included only a low refractive index plasticizer.

#### **Example 10: Preparation of Interlayers having High Refractive Index Additives**

[0170] Several of the Comparative and Disclosed Interlayers formed in Example 9 and summarized in Table 13, above, were used to create several Comparative and Disclosed Multilayer Interlayers. Each multilayer interlayer included a pair of outer “skin” layers, each having a thickness of 14 mils, sandwiching an inner “core” layer, having a thickness of 5 mils, formed of a resin having a lower residual hydroxyl content. The composition and several properties of the multilayer interlayers, including total plasticizer content, refractive index, glass transition temperature, mottle, and loss factor, were measured as described above, and the results for the Comparative Multilayer Interlayers CI-20 and CI-21 and Disclosed Multilayer Interlayers DI-39 through DI-41 are summarized in Table 14, below.

Table 14: Properties of Several Comparative and Disclosed Interlayers

Interlayer No	Skin Layer 1	Core Layer	Skin Layer 2	Total plasticizer content (phr)	$\Delta$		Refractive Index (nD25)	$T_g$ (°C)		Mottle	Loss Factor at 20°C
					Refractive Index (Skin- Core)			Skin	Core		
Cl-20	CL-17	CL-18	CL-17	38	0.000		1.477	30	-	0	0.02
Cl-21	CL-17	CL-19	CL-17	42.5	0.011		1.475	35	3	5	0.32
DI-39	DL-32	DL-35	DL-32	48	0.005		1.484	38.8	2.6	0.4	0.41
DI-40	DL-33	DL-36	DL-33	48	0.004		1.486	39.6	4.7	0.4	0.39
DI-41	DL-34	DL-37	DL-34	48	0.002		1.488	40.9	8.7	0.4	0.33

[0171] As shown in Table 14 above, interlayers formed from skin and core layers having a refractive index difference greater than 0.010 exhibited more optical defects, as shown by the mottle value of 5. Additionally, Disclosed Interlayers DI-39 through DI-41, which utilized a high refractive index plasticizer, exhibited a higher overall refractive index as compared to Comparative Interlayers CI-20 and CI-21, which only utilized a plasticizer having a refractive index less than 1.460.

**Example 11: Measurement of Double Image Separation Distance for Comparative and Disclosed Multiple Layer Panels with Varying Thickness and Wedge Angle**

[0172] Several interlayers were formed by combining poly(vinyl butyral) resin having a residual hydroxyl content of 18.7 weight percent with 38 phr of the plasticizer triethylene glycol bis(2-ethylhexanoate) (3GEH), and extruding the resulting mixture to form single layer sheets. A flat interlayer sheet, i.e., with no tapered zone, was formed with a uniform thickness of about 30 mils (about 0.76 mm) and was cut to form several comparative interlayers having a wedge angle of 0°.

[0173] Several of the sheets were formed with tapered zones each having a different constant wedge angle, and each sheet was cut to form several tapered monolithic interlayers. The tapered interlayers each had a wedge angle ranging from 0.30 to 0.73 mrad. Each of the interlayer samples was then laminated between pairs of 6 inch by 12 inch glass sheets, with each glass/interlayer/glass multiple layer panel having a thickness of 3.0 mm, 4.32 mm, or 5.0 mm. The specific configuration of each of the Comparative Multiple Layer Panels (CP-1 through CP-3) and Disclosed Multiple Layer Panels (DP-1 through DP-15) are shown in Table 15, below.

**Table 15: Double Image Separation of Several Comparative & Disclosed Laminates**

Laminate	Wedge Angle (mrad)	Laminate Thickness (mm)	Double Image Separation (arc min)
CP-1	0.00	3.0	5.459
CP-2	0.00	4.3	7.389
CP-3	0.00	5.0	8.491
DP-1	0.20	3.0	3.305

Laminate	Wedge Angle (mrad)	Laminate Thickness (mm)	Double Image Separation (arc min)
DP-2	0.20	4.3	5.362
DP-3	0.20	5.0	6.527
DP-4	0.30	3.0	2.228
DP-5	0.30	4.3	4.411
DP-6	0.30	5.0	5.576
DP-7	0.41	3.0	1.088
DP-8	0.41	4.3	3.271
DP-9	0.41	5.0	4.436
DP-10	0.50	3.0	-0.618
DP-11	0.50	4.3	1.789
DP-12	0.50	5.0	3.042
DP-13	0.73	3.0	-2.714
DP-14	0.73	4.3	-0.403
DP-15	0.73	5.0	0.888

[0174] Each of the laminates listed in Table 15 was subjected to the double image separation test using the apparatus 100 depicted in FIG. 9. As shown in FIG. 9, test apparatus 100 includes a projector 130, an adjustable mirror 140, and a detector 120. The distance between projector 130 and adjustable mirror 140, shown in FIG. 9 as line A, was 43.8 cm, the distance between adjustable mirror 140 and panel 110, shown in FIG. 9 as line B, was 31.7 cm, and the distance between panel 110 and detector 120, shown in FIG. 9 as line C, was 47 cm. Additionally, the incident angle at which the light from projector 130 hit adjustable mirror 140, shown in FIG. 9 as  $\theta_1$ , was  $30.11^\circ$ , and panel 110 was arranged so that the incident angle of the image from mirror 140 onto panel 110, shown in FIG. 9 as  $\theta_2$ , and the light reflected from panel 110 to detector 120, shown as  $\theta_2'$ , were each about the same and equal to  $24.8^\circ$ . Finally, as shown in FIG. 9 by  $\theta_3$ , panel 110 was oriented from the vertical by an angle of  $30.5^\circ$ .

[0175] To measure the double image separation distance of panel 110 using apparatus 100 shown in FIG. 9, an image was generated using a standard thin film transistor display and was reflected by a flat surface of mirror 140 to panel 110. The

image was reflected off of one or more surfaces of panel 110 and projected to detector 120. The image was then recorded using a digital camera to form a captured image, as shown in FIG. 10, which was then analyzed to form a profile by converting the digital image to a matrix having elements that represent the intensity (grey scale level) of each pixel of the digital image. The matrix was then plotted, column by column, as a function of pixel number, as shown in FIG. 11. The higher intensity peaks shown in FIG. 11 represented the primary image reflected from the panel, while the shorter, less intense peaks corresponded to the secondary or “ghost” image. The separation distance, in pixels, between the primary and secondary peaks was then determined used to calculate the double image separation distance (D) for each panel, in minutes (arc min), according to the following equation:

$$D = \frac{\text{peak separation (pixels)}}{\text{Virtual Image distance (mm)}} \times 3427.75$$

[0176] Using the above-described procedure, the double image separation distance for each of the Comparative Panels (CP-1 through CP-3) and each of the Disclosed Panels (DP-1 through DP-15) was measured, and the results are provided in Table 15, above. The results are also summarized graphically in FIG. 12.

[0177] Additionally, several additional Comparative Multiple Layer Panels (CP-4 through CP-8) and Disclosed Multiple Layer Panels (DP-16 through DP-29) were constructed in a similar manner as described above, but with varying thicknesses at several different wedge angles. The above-described double image separation test was then performed on each of the Comparative Panels CP-4 through CP-8 and Disclosed Panels DP-16 through DP-29, and the results are provided in Table 16, below. FIG. 13 shows the double image separation distance as a function of wedge angle for Comparative Panels CP-4 through CP-8 and Disclosed Panels DP-16 through DP-29 at varying glass configurations.

**Table 16: Double Image Separation of Several Comparative and Disclosed Laminates**

Laminate	Wedge Angle (mrad)	Laminate Thickness (mm)	Double Image Separation (arc min)
CP-4	0.0	2.16	3.97
CP-5	0.0	3.06	5.45
CP-6	0.0	3.97	6.83
CP-7	0.0	4.66	7.96
CP-8	0.0	5.36	9.02
DP-16	0.20	3.19	3.49
DP-17	0.20	4.07	4.82
DP-18	0.20	5.45	7.06
DP-19	0.41	3.39	0.55
DP-20	0.41	4.12	2.88
DP-21	0.41	5.47	4.44
DP-22	0.50	4.16	1.22
DP-23	0.50	4.64	2.39
DP-24	0.50	2.20	-2.10
DP-25	0.50	5.44	3.43
DP-26	0.73	2.30	-4.27
DP-27	0.73	3.20	-2.59
DP-28	0.73	3.90	-0.53
DP-29	0.73	4.60	-0.04

**Example 12: Determination of Equivalent Refractive Index ( $n_{eq}$ ) for Several Comparative and Disclosed Multiple Layer Panels**

[0178] The equivalent refractive index,  $n_{eq}$ , of a Comparative Multiple Layer Panel and a Disclosed Multiple Layer Panel are determined in the following example. A Comparative Panel (CP-A) is formed by laminating a PVB interlayer having a uniform thickness of 0.76 mm and a refractive index of 1.475 between glass panels, one of which has a thickness of 1.6 mm and the other of which has a thickness of 2.3 mm. The glass panels each have a refractive index of 1.52, measured according to ASTM D542 at a wavelength of 589 nm and 25°C. At an incident angle ( $\theta_2$ ) of 60°, the equivalent refractive index of the Comparative Panel is determined, according to the above equations (1) through (4), to be 1.512.

[0179] Similarly, a Disclosed Multiple Layer Panel (DP-B) is formed by laminating another PVB interlayer having a uniform thickness of 0.76 mm between two glass panels, each panel having the same thickness as the panels used to form the Comparative Multiple Layer Panel. However, the PVB interlayer used to form the Disclosed Multiple Layer Panel (DP-B) has a refractive index of 1.65. Using the formulas above, the equivalent refractive index of the Disclosed Multiple Layer Panel is calculated to be 1.540. This Disclosed Multiple Layer Panel DP-B may represent the practical maximum (or practical limit) of equivalent refractive index achievable in some cases. The results are summarized in Table 17, below.

**Table 17: Refractive Index Properties of Interlayers (at incident angle of 60°)**

Sample	Thickness (mm)				Refractive Index		
	Glass Panel 1	Glass Panel 2	Interlayer	Total	Glass	Interlayer	$n_{eq}$
Glass Only	4.66	-	-	4.66	1.52	-	1.520
Comparative CP-A	1.6	2.3	0.76	4.66	1.52	1.475	1.512
Disclosed DP-B	1.6	2.3	0.76	4.66	1.52	1.65	1.540

**Example 13: Effect of Refractive Index on Double Image Separation for Various Laminates**

[0180] Several glass/air/glass laminates were formed to simulate multiple layer panels having different equivalent refractive indices. Each laminate was constructed by spacing several glass sheets of varying thickness apart from one another using appropriately-sized metal shims in order to create and maintain air gaps between the glass sheets. The overall thickness of the resulting glass/air/glass laminates was maintained at between 4 and 5 mm, and the equivalent refractive index for each was calculated according to Formulas (1) through (4), above. The specific configuration and values for the parameters of Formulas (1) through (4), above, for two exemplary panels, AGL-1 and AGL-2, are summarized in respective Tables 18 and 19, below.



**Table 18: Configuration and Equivalent Refractive Index Parameters for AGL-1**

Layer	Material	Thickness (mm)	$n_1$	$\Theta_1$	$n_2$	$\Theta_2$	$x_i$ (mm)
Layer 1	Air		1	65.20			
Layer 2	Glass	0.7	1	65.20	1.52	36.67	0.521217
Layer 3	Air	3.6	1.52	36.67	1.00	65.20	7.791114
Layer 4	Glass	0.7	1	65.20	1.52	36.67	0.521217
Layer 5	Air		1.52	36.67	1.00	65.20	
<b>Equivalent</b>		<b>5</b>			<b>1.04</b>	<b>60.49</b>	<b>8.833547</b>

**Table 19: Configuration and Equivalent Refractive Index Parameters for AGL-2**

Layer	Material	Thickness (mm)	$n_1$	$\Theta_1$	$n_2$	$\Theta_2$	$x_i$ (mm)
Layer 1	Air		1	65.20			
Layer 2	Glass	2.3	1	65.20	1.52	36.67	1.712569
Layer 3	Air	0.4	1.52	36.67	1.00	65.20	0.865679
Layer 4	Glass	2.3	1	65.20	1.52	36.67	1.712569
Layer 5	Air		1.52	36.67	1.00	65.20	
<b>Equivalent</b>		<b>5</b>			<b>1.39</b>	<b>40.63</b>	<b>4.290818</b>

[0181] Each of the glass/air/glass panels formed as described above was then subjected to a double image separation test similar to the one described in Example 11 above using the apparatus shown in FIG. 9, except that the captured image formed when analyzing these panels included three double images, instead of one, due to the large difference between the refractive indices of glass and air. For this analysis, the separation between the primary image and the last double image were analyzed when determining the double image separation distance for each panel. The results of these analyses are summarized graphically in FIG. 14.

[0182] While the invention has been disclosed in conjunction with a description of certain embodiments, including those that are currently believed to be the preferred embodiments, the detailed description is intended to be illustrative and

should not be understood to limit the scope of the present disclosure. As would be understood by one of ordinary skill in the art, embodiments other than those described in detail herein are encompassed by the present invention. Modifications and variations of the described embodiments may be made without departing from the spirit and scope of the invention

[0183] It will further be understood that any of the ranges, values, or characteristics given for any single component of the present disclosure can be used interchangeably with any ranges, values or characteristics given for any of the other components of the disclosure, where compatible, to form an embodiment having defined values for each of the components, as given herein throughout. For example, an interlayer can be formed comprising poly(vinyl butyral) having a residual hydroxyl content in any of the ranges given in addition to comprising a plasticizers in any of the ranges given to form many permutations that are within the scope of the present disclosure, but that would be cumbersome to list. Further, ranges provided for a genus or a category, such as phthalates or benzoates, can also be applied to species within the genus or members of the category, such as dioctyl terephthalate, unless otherwise noted.

**CLAIMS**

What is claimed is

1. A tapered interlayer comprising at least one polymer layer comprising a poly(vinyl acetal) resin and at least one plasticizer, wherein said polymer layer has a refractive index of at least 1.480, and wherein said interlayer comprises a tapered zone having an overall wedge angle of not more than 0.85 mrad.
2. The interlayer of claim 1, wherein said tapered zone has an overall wedge angle of less than 0.55 mrad and wherein said polymer layer has a refractive index of at least 1.500.
3. The interlayer of claim 1, wherein said interlayer is a monolithic interlayer.
4. The interlayer of claim 1, wherein said interlayer is a multiple layer interlayer comprising said polymer layer and another polymer layer adjacent to said polymer layer, wherein the difference between the glass transition temperature of said polymer layer and the glass transition temperature of said another polymer layer is at least 3°C.
5. The interlayer of claim 4, wherein said another polymer layer comprises a second poly(vinyl acetal) resin and at least one plasticizer, wherein the difference between the residual hydroxyl content of said poly(vinyl acetal) resin in said polymer layer and the residual hydroxyl content of said second poly(vinyl acetal) resin in said another polymer layer is at least 2 weight percent.
6. The interlayer of claim 1, wherein said tapered zone comprises at least one variable angle zone having a curved thickness profile and a continuously varying wedge angle.

7. The interlayer of claim 1, wherein said tapered zone comprises at least one constant angle zone having a linear thickness profile and a constant wedge angle.

8. A multiple layer panel comprising a pair of rigid substrates and the interlayer of claim 1, wherein said interlayer is disposed between said pair of rigid substrates and wherein said rigid substrates have a combined thickness of less than 4.0 mm.

9. A multiple layer panel comprising:  
a pair of rigid substrates; and  
an interlayer disposed between said substrates, wherein said interlayer comprises a tapered zone having an overall wedge angle of less than 0.85 mrad,  
wherein the equivalent refractive index of said multiple layer panel is at least 0.010 higher than the refractive index of each of said rigid substrates.

10. The panel of claim 9, wherein the equivalent refractive index of said multiple layer panel is at least 1.500.

11. The panel of claim 9, wherein the refractive index of each of said substrates is at least 20 percent lower than the refractive index of said interlayer.

12. The panel of claim 9, wherein the refractive index of said interlayer is at least about 1.600.

13. The panel of claim 9, wherein said tapered zone has an overall wedge angle of less than 0.55 mrad.

14. The panel of claim 9, wherein said interlayer comprises at least one polymer layer comprising a poly(vinyl acetal) resin and at least one plasticizer, wherein said combined thickness of each of said rigid substrates is not more than 4.0 mm.

15. The panel of claim 14, wherein said interlayer comprises a multiple layer interlayer comprising two or more polymer layers each comprising a poly(vinyl acetal) resin and at least one plasticizer, wherein said interlayer comprises at least two glass transition temperatures, wherein the difference between said two glass transition temperatures is at least about 3°C.

16. A multiple layer panel comprising  
a pair of rigid substrates; and  
a tapered interlayer disposed between said substrates, wherein said tapered interlayer comprises at least one polymeric resin,  
wherein the refractive index of said interlayer is at least 5 percent higher than the refractive index of each of said rigid substrates.

17. The panel of claim 16, wherein said tapered comprises a tapered zone having a wedge angle of not more than 0.85 mrad and wherein said interlayer comprises at least one polymer layer having a refractive index of at least about 1.480.

18. The panel of claim 16, wherein said panel has an equivalent refractive index higher than said refractive index of each of said rigid substrates.

19. The panel of claim 16, wherein said interlayer is a multiple layer interlayer comprising at least a first polymer layer and a second polymer layer adjacent to said first polymer layer, wherein first polymer layer comprises a first poly(vinyl acetal) resin and at least one plasticizer and said second polymer layer comprises a second poly(vinyl acetal) resin and at least one plasticizer, wherein the

difference between the glass transition temperature of said first polymer layer and the glass transition temperature of said second polymer layer is at least 3°C, and wherein the difference between the residual hydroxyl content of said first poly(vinyl acetal) resin and the residual hydroxyl content of said second poly(vinyl acetal) resin is at least 2 weight percent.

20. The panel of claim 16, wherein said interlayer has a minimum thickness in the range of from about 0.25 to about 1.2 mm and a maximum thickness in the range of from about 0.38 to about 2.2 mm, wherein the difference between the minimum thickness of said interlayer and the maximum thickness of said interlayer is at least about 0.13 mm, and wherein the combined thickness of each of said rigid substrates is less than about 4.0 mm.

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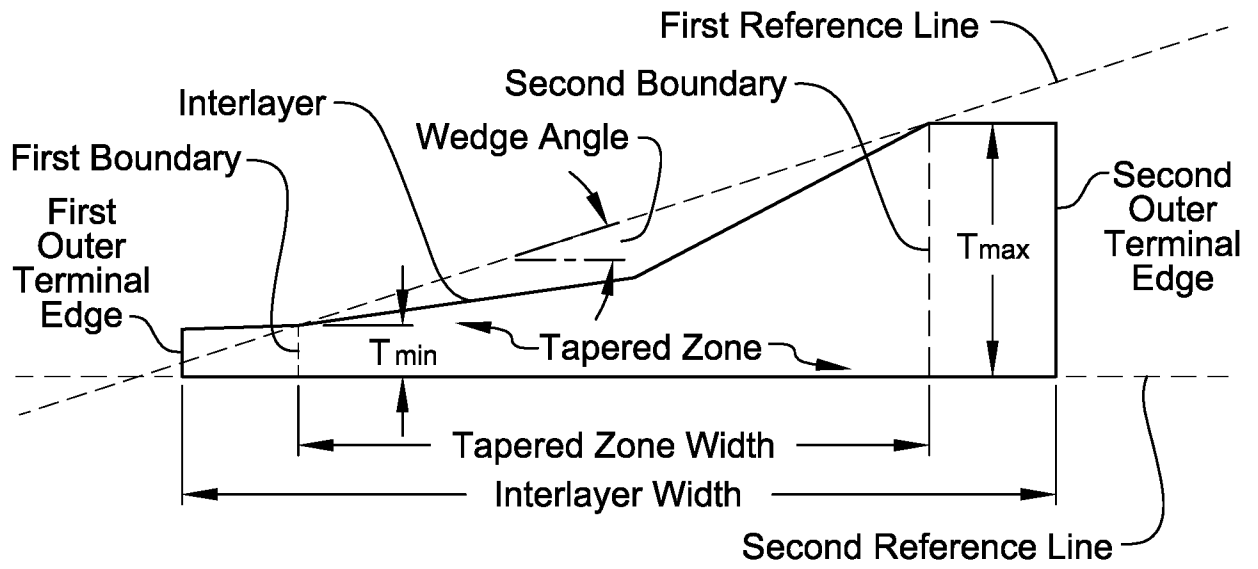


FIG. 1

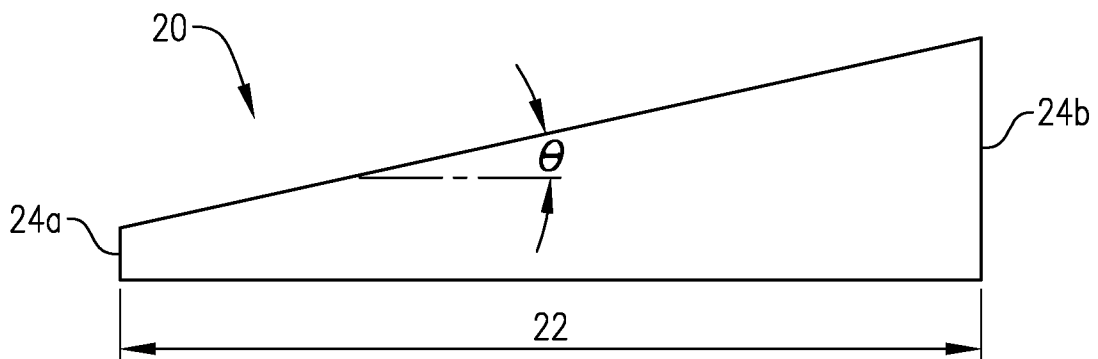


FIG. 2

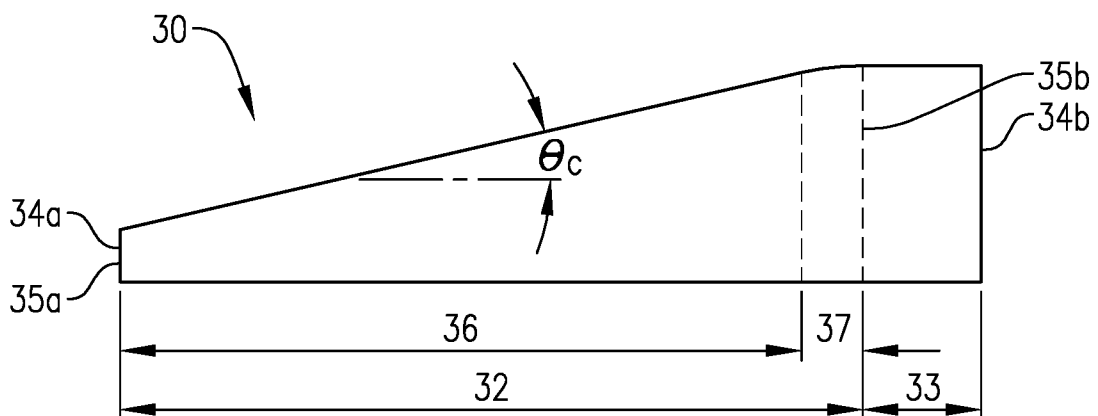


FIG. 3

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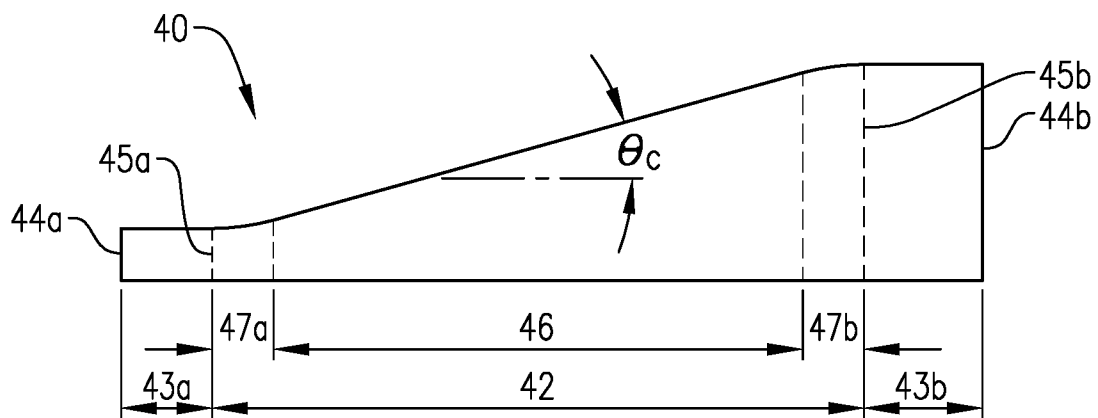


FIG. 4

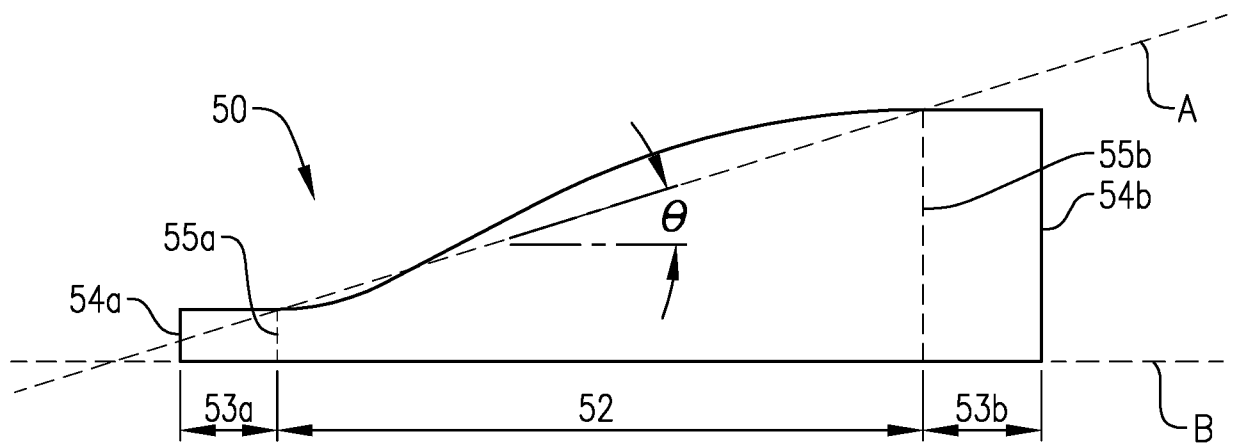


FIG. 5

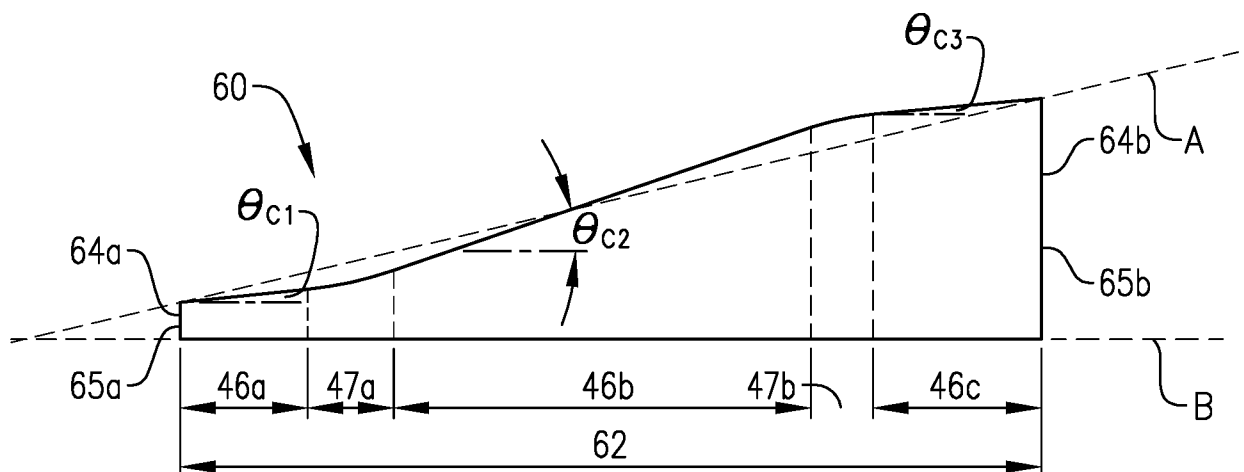


FIG. 6



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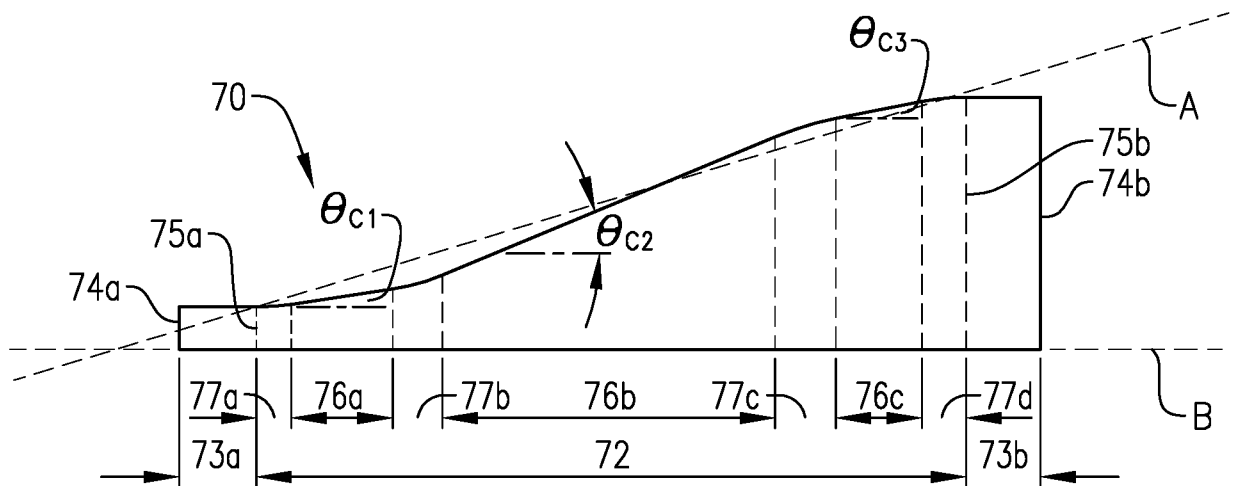


FIG. 7

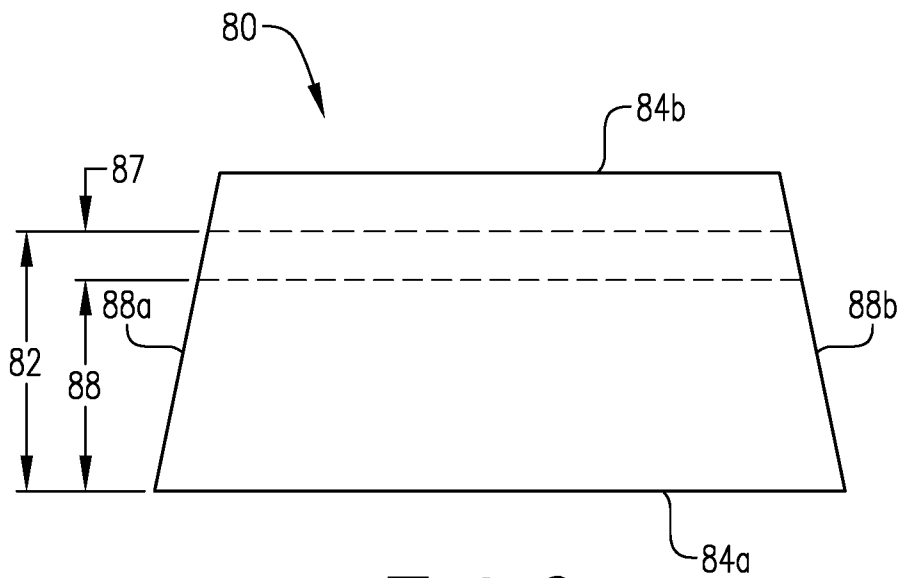


FIG. 8a

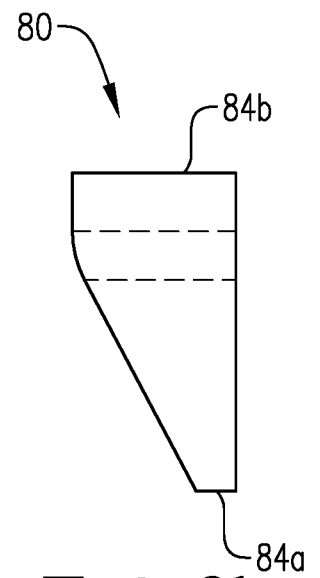


FIG. 8b

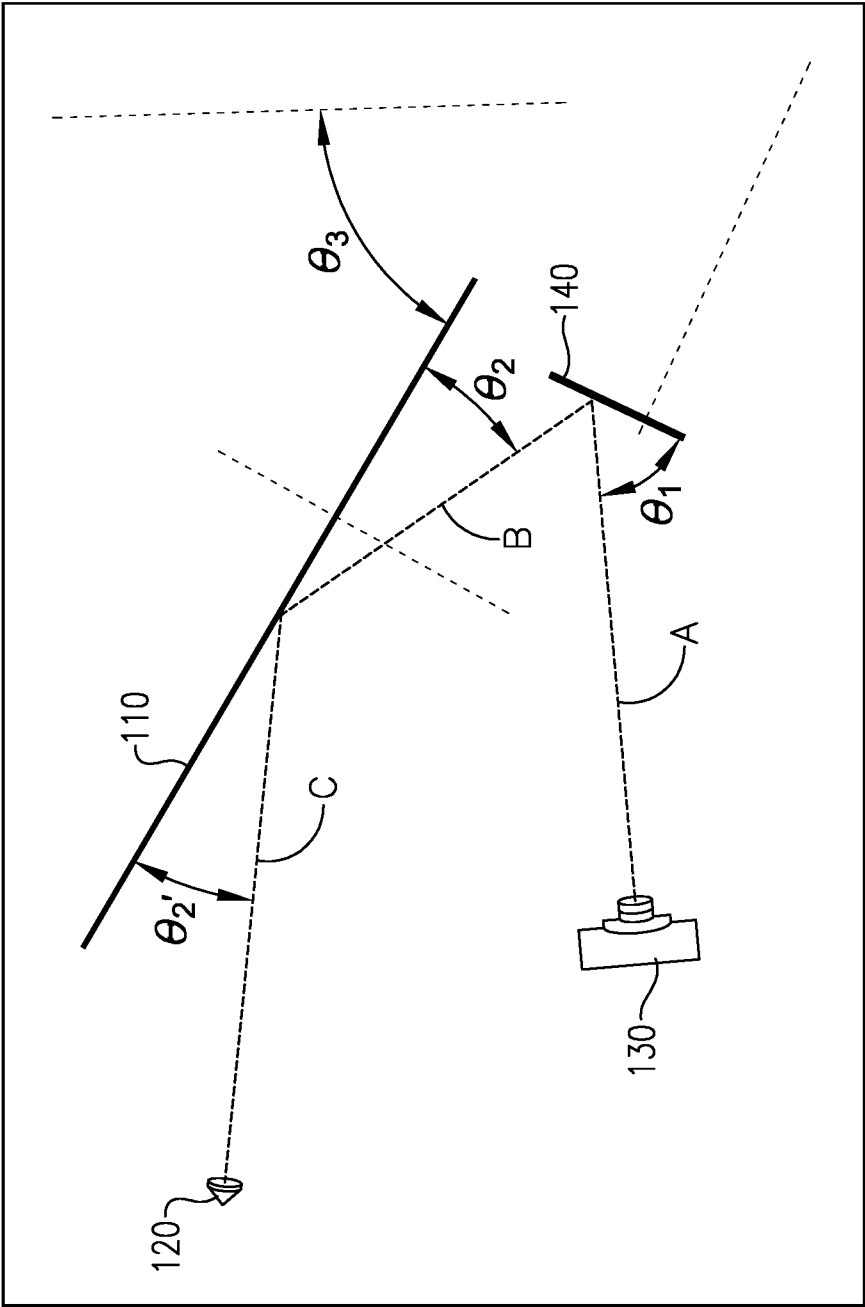


FIG. 9

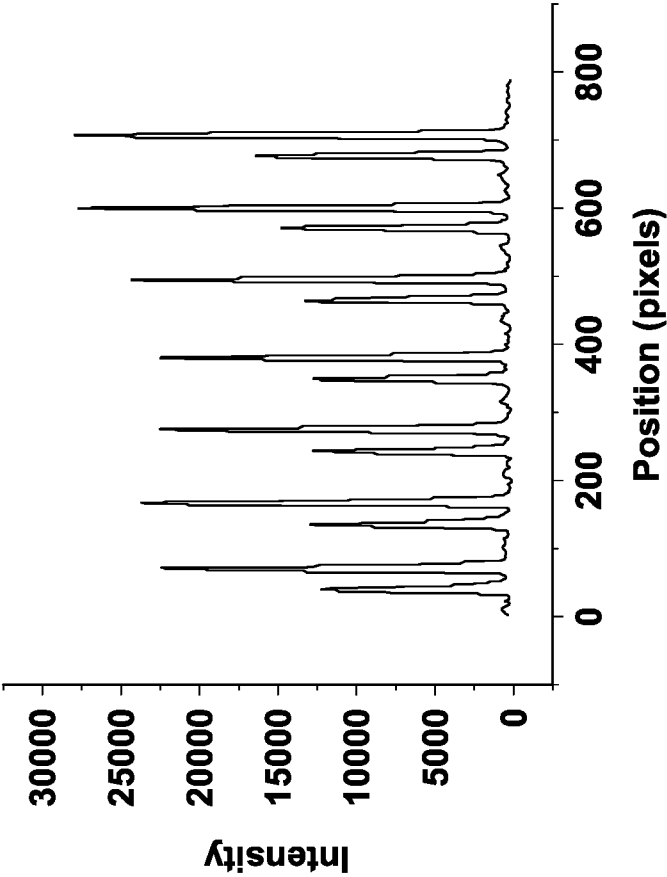


FIG. 11

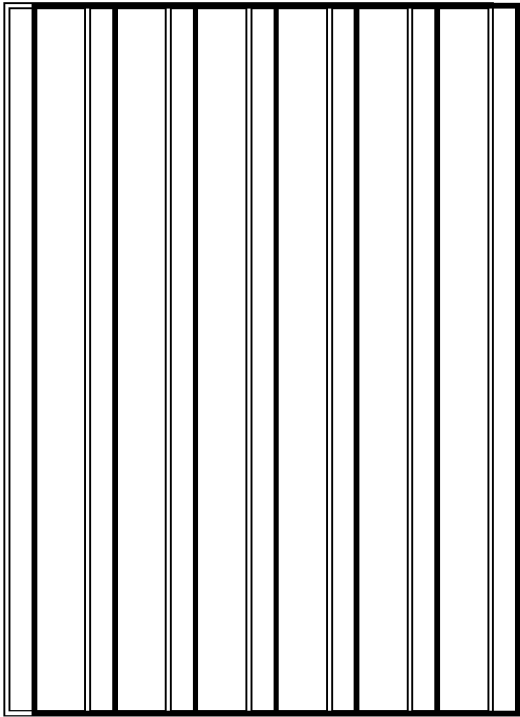


FIG. 10

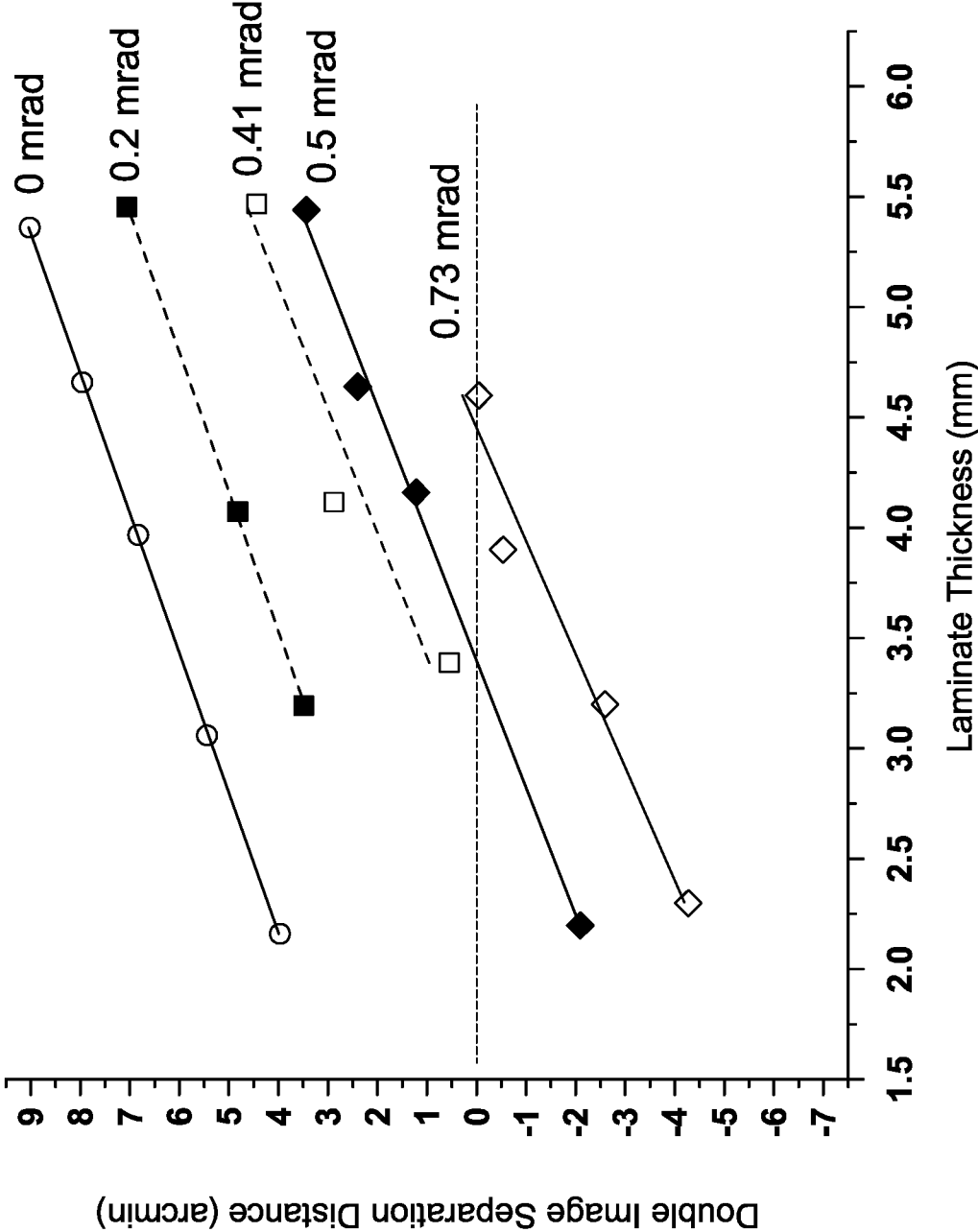


FIG.12

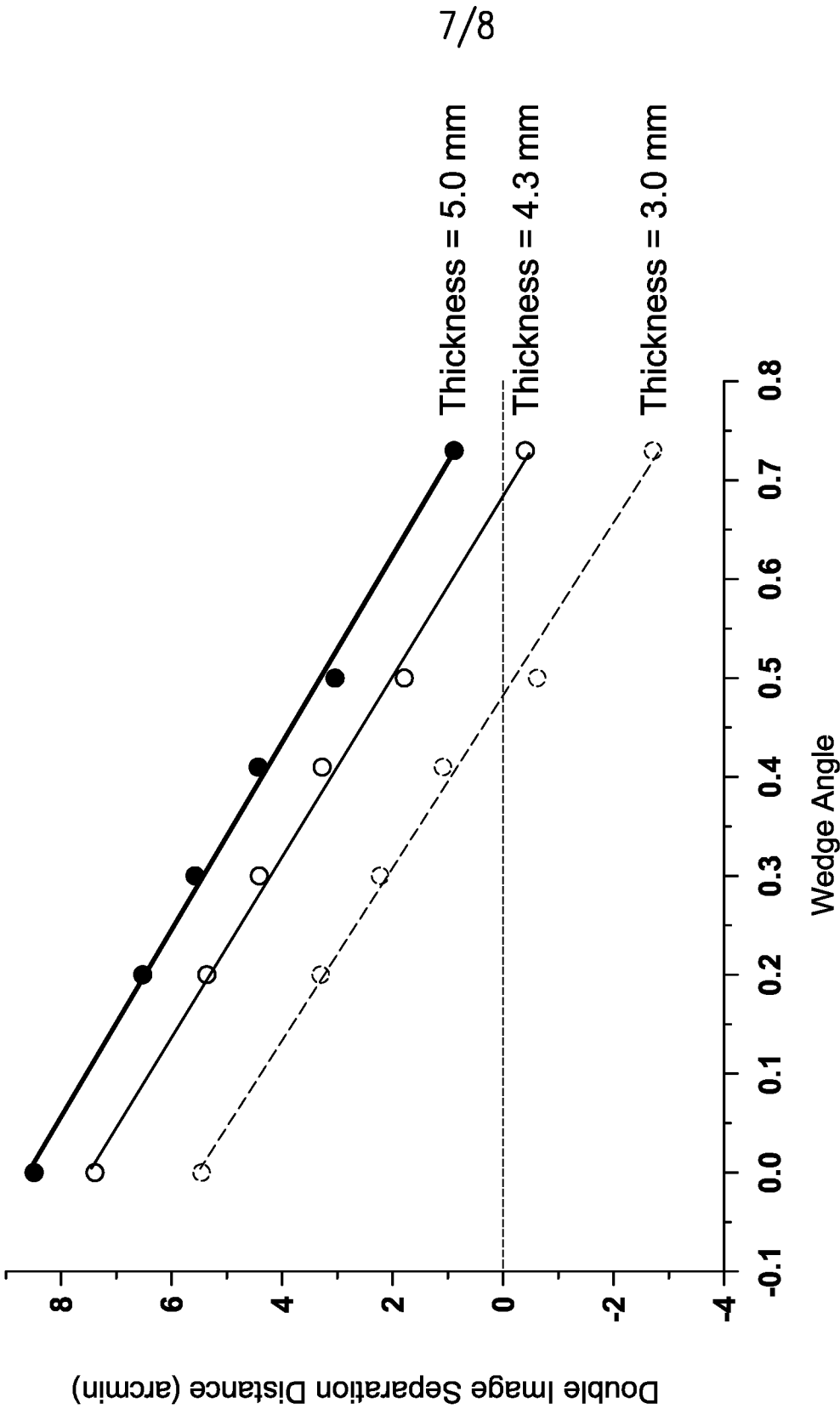


FIG.13

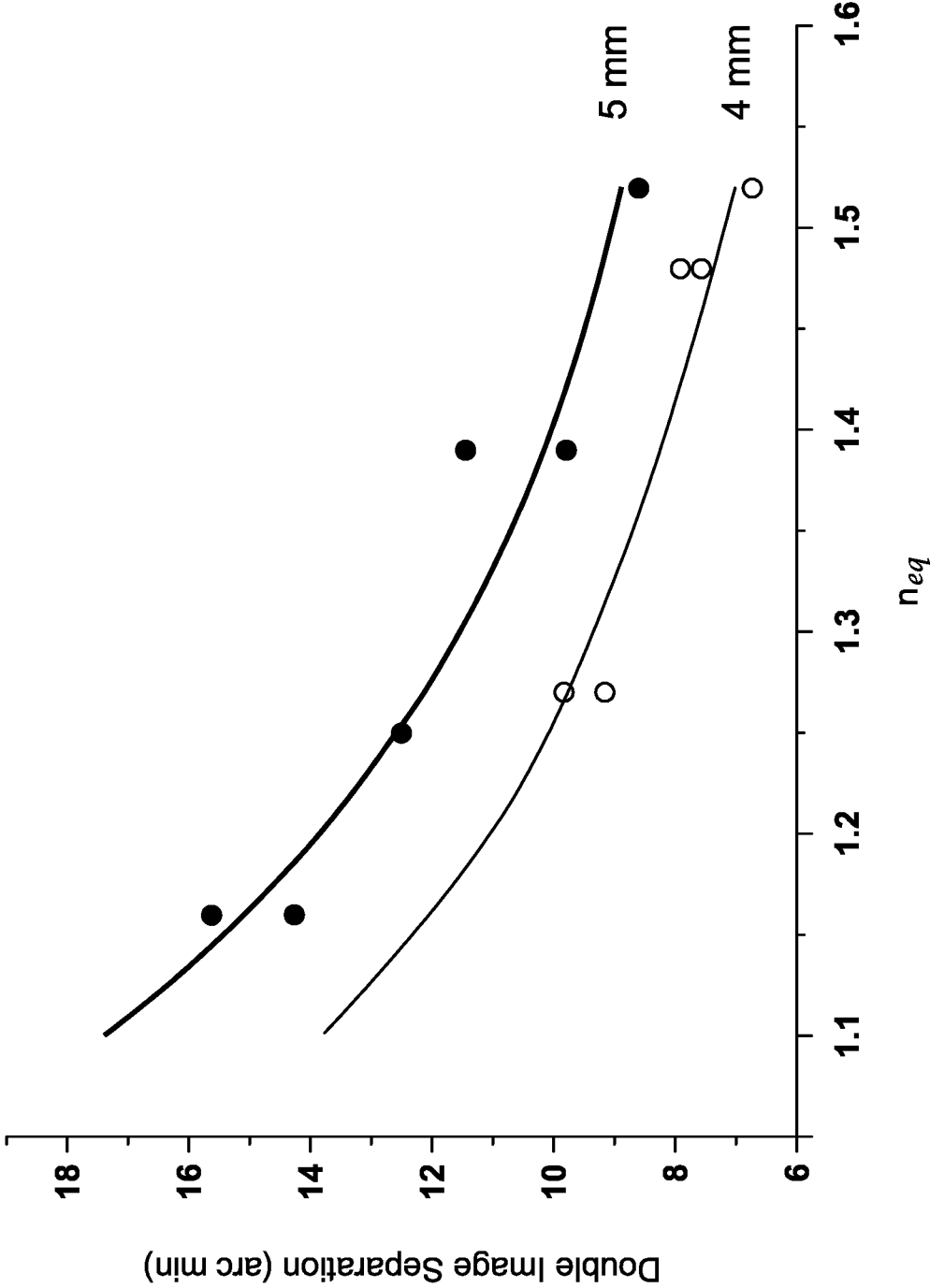


FIG. 14

## INTERNATIONAL SEARCH REPORT

International application No  
PCT/US2017/016898

A. CLASSIFICATION OF SUBJECT MATTER  
INV. B32B17/10 B32B27/30  
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)  
B32B

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.
Y	US 2015/258747 A1 (MIYAI JIRO [JP]) 17 September 2015 (2015-09-17) paragraphs [0008], [0009], [0061], [0062], [0072] -----	1-20
Y	US 2014/363651 A1 (LU JUN [US] ET AL) 11 December 2014 (2014-12-11) paragraphs [0009], [0010], [0017], [0059], [0132], [0134] -----	1-20
A	US 2007/148472 A1 (MASAKI YUUGI [JP] ET AL) 28 June 2007 (2007-06-28) paragraphs [0068], [0070], [0071], [0075], [0078] -----	1-20



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

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

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"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

27 April 2017

Date of mailing of the international search report

09/05/2017

Name and mailing address of the ISA/

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Authorized officer

Somerville, Fiona

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/US2017/016898

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 2015258747	A1	17-09-2015	BR PI0711462 A2 08-11-2011
		CA 2646038 A1 22-11-2007	
		CN 101443287 A 27-05-2009	
		CN 102515572 A 27-06-2012	
		CN 102976637 A 20-03-2013	
		EP 2017237 A1 21-01-2009	
		JP 5576019 B2 20-08-2014	
		JP 5782084 B2 24-09-2015	
		JP 2014037347 A 27-02-2014	
		JP 2014231472 A 11-12-2014	
		JP 2016164119 A 08-09-2016	
		KR 20090010952 A 30-01-2009	
		RU 2008148968 A 20-06-2010	
		US 2009294212 A1 03-12-2009	
		US 2011094825 A1 28-04-2011	
		US 2012003428 A1 05-01-2012	
		US 2014178651 A1 26-06-2014	
		US 2015258747 A1 17-09-2015	
		US 2016341960 A1 24-11-2016	
		WO 2007132777 A1 22-11-2007	
US 2014363651	A1	11-12-2014	AU 2014278387 A1 24-12-2015
		AU 2014278391 A1 19-11-2015	
		CA 2912575 A1 18-12-2014	
		CA 2912592 A1 18-12-2014	
		CN 105283314 A 27-01-2016	
		CN 105358321 A 24-02-2016	
		EP 3007898 A2 20-04-2016	
		EP 3007899 A1 20-04-2016	
		JP 2016523222 A 08-08-2016	
		JP 2016528314 A 15-09-2016	
		KR 20160018772 A 17-02-2016	
		KR 20160019927 A 22-02-2016	
		US 2014363651 A1 11-12-2014	
		US 2014363652 A1 11-12-2014	
		US 2014364549 A1 11-12-2014	
		US 2014364550 A1 11-12-2014	
		US 2016101605 A1 14-04-2016	
		US 2016102181 A1 14-04-2016	
		WO 2014200989 A2 18-12-2014	
		WO 2014200993 A1 18-12-2014	
US 2007148472	A1	28-06-2007	CN 103786391 A 14-05-2014
		EP 1800855 A1 27-06-2007	
		JP 2007223883 A 06-09-2007	
		KR 20070068291 A 29-06-2007	
		US 2007148472 A1 28-06-2007	