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(54) Title: CONTINUOUS PROCESS FOR PRODUCING HOT MELT ADHESIVE COMPOSITIONS

(57) Abstract: A continuous method for producing moisture curable hot melt adhesive compositions includes feeding a molten mixture of an organopolysiloxane, silicone resin, silane crosslinker, catalyst and solvent into an extruder and removing the volatiles. By using a continuous process it is possible to control the non-volatile content and produce a consistent product. Upon exposure to moisture the holt melt adhesive composition cures resulting in adhesion between two substrates.



WO 2004/037941 A2

DESCRIPTION

CONTINUOUS PROCESS FOR PRODUCING HOT MELT ADHESIVE COMPOSITIONS

CROSS REFERENCE

- 5 [0001] This application claims priority to U.S. Provisional Patent Application Serial Number 60/420,575, which is hereby incorporated by reference in its entirety.

FIELD OF THE INVENTION

- 10 [0002] This invention pertains to a continuous method for producing moisture curable hot melt adhesive compositions. The method comprises feeding a mixture of an organopolysiloxane, silicone resin, silane crosslinker and solvent into an extruder and removing the volatiles. By using a continuous process it is possible to control the non-volatile content and produce a consistent product. Upon exposure to moisture the hot melt adhesive composition cures resulting in adhesion between at least two substrates.

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BACKGROUND OF THE INVENTION

- [0003] Moisture curable organosiloxane compositions find use in various applications, for example as sealant compositions that can be applied to a joint between elements and cured to provide an elastomeric seal between them. These compositions cure at room temperature and are particularly attractive for use as sealant compositions for sealing, for example highway joints, joints in articles such as vehicle headlights and joints in buildings and in glazing applications, because no special heating or other cure conditions are generally required to produce a seal of desired quality.

- 20 [0004] Many moisture curable organosiloxane compositions have been proposed and are generally formed from an at least one substantially linear polyorganosiloxane containing at least two silanol groups, a crosslinker capable of reaction with the polyorganosiloxane to yield a crosslinked network, and catalyst materials. These compositions cure by a condensation reaction promoted by moisture.

- 25 [0005] The crosslinker in moisture curable organosiloxane compositions is generally selected from polyfunctional silanes that readily hydrolyze. Commonly employed crosslinkers are triacetoxysilanes, trialkoxysilanes, triamino silanes and trioximo silanes. It is believed that the condensation reaction proceeds via a capping of the polyorganosiloxane

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with, for example, dialkoxyalkylsilyl groups followed by interaction of the alkoxy groups of the end caps and or silanol groups to yield a crosslinked structure.

[0006] While some curing of the composition during manufacture and storage is acceptable, it is important that this curing does not proceed too far prior to application at its intended work site, at which it is intended to cure under influence of atmospheric moisture. Thus the exposure of the composition to moisture should be kept to a uniform, acceptably low extent from batch to batch during manufacture and storage, otherwise the composition cures to an extent that renders it impractical for its intended purpose.

[0007] Moisture curable compositions based on organosilicon compounds generally contain finely divided fillers. The fillers generally used are those that strengthen the cured material, reduce the cost of the product or otherwise confer a desired combination of properties.

[0008] Typical fillers include but are not limited to high surface area silicas, ground quartz, iron oxide, zinc oxide, carbon black, calcium carbonate and diatomaceous earth. Moisture curable organosiloxane compositions can be manufactured using a batch or continuous process during which the filler and polyorganosiloxane are mixed together, the crosslinker and catalyst are added to the mixture and the resultant composition is then packaged in containers such as cartridges, which are then sealed in an airtight manner to prevent ingress of moisture.

[0009] Silicone pressure-sensitive adhesives (hereinafter also referred to as PSAs) typically contain at least two primary components, namely a linear siloxane polymer and a tackifier resin consisting essentially of triorganosiloxane units (i.e., $R_3SiO_{1/2}$ units, in which R denotes a monovalent organic group) and silicate units (i.e., $SiO_{4/2}$ units). In addition to the above two ingredients, some silicone PSA compositions contain some crosslinking means (e.g., peroxide or hydrosilylation cure systems) in order to optimize various properties of the final adhesive product. In view of the high viscosity imparted by the polymer component, these PSA compositions are typically dispersed in an organic solvent for ease of application. Some of these PSAs contain reactive groups, which allow the compositions to be cured by exposure to moisture. When the proportions of the above described resin and polymer and other parameters are adjusted similar combinations can be formulated into coating compositions. Under certain other conditions, hot melt PSAs can be obtained.

SUMMARY OF THE INVENTION

[0010] This invention pertains to continuous process for producing a moisture curable hot melt silicone pressure sensitive adhesive composition. The process comprises combining an organopolysiloxane, silicone resin, silane crosslinker and solvent; feeding the combination through an extrusion device to remove volatiles; and recovering a hot melt adhesive composition having a non-volatile content of 95 wt% or more. The use of the continuous process allows for more efficient production of the hot melt adhesive composition and the production of a more consistent product. This method of processing is advantageous from the standpoints of cost, convenience and product consistency.

DETAILED DESCRIPTION OF THE INVENTION

[0011] This invention pertains to a continuous process for producing hot melt adhesive compositions. The process comprises combining an organopolysiloxane, silicone resin, silane crosslinker and solvent; feeding the combination through an extrusion device to remove volatiles; and recovering a hot melt adhesive composition having a non-volatile content of 95 wt% or more.

[0012] The silicone resin useful herein contains monofunctional units represented by $R^1_3SiO_{1/2}$ and tetrafunctional units represented by $SiO_{4/2}$. R^1 represents a substituted or unsubstituted monovalent hydrocarbon radical. Silicone resins of this type are well known in the art as one of the ingredients present in organosiloxane compositions used as pressure sensitive adhesives.

[0013] The silicone resin is soluble in liquid hydrocarbons such as benzene, toluene, xylene, heptane and the like or in liquid organosilicon compounds such as a low viscosity cyclic and linear polydiorganosiloxanes.

[0014] In the $R^1_3SiO_{1/2}$ unit, R^1 is typically a monovalent hydrocarbon radical containing up to 20 carbon atoms, typically from 1 to 10 carbon atoms. Examples of suitable hydrocarbon radicals for R^1 include alkyl radicals, such as methyl, ethyl, propyl, pentyl, octyl, undecyl and octadecyl; alkenyl radicals, such as vinyl, allyl and 5-hexenyl; cycloaliphatic radicals, such as cyclohexyl and cyclohexenylethyl; and aryl radicals such as phenyl, tolyl, xylyl, benzyl and 2-phenylethyl. Non-reactive substituents that can be present on R^1 include but are not limited to halogen and cyano. Typical substituted hydrocarbon radicals that can be represented by R^1 include but are not limited to chloromethyl and 3,3,3-trifluoropropyl.

[0015] At least one-third, alternatively at least two-thirds of the R^1 radicals in the $R^1_3SiO_{1/2}$ unit are methyl radicals. Examples of $R^1_3SiO_{1/2}$ units include but are not limited to $Me_3SiO_{1/2}$, $PhMe_2SiO_{1/2}$ and $Me_2ViSiO_{1/2}$ where Me, Ph and Vi denote methyl, phenyl and vinyl, respectively. The silicone resin may contain two or more of these units.

5 [0016] The molar ratio of the $R^1_3SiO_{1/2}$ and $SiO_{4/2}$ units in the silicone resin is typically from 0.5/1 to 1.5/1, preferably from 0.6/1 to 0.9/1. These mole ratios are conveniently measured by Si^{29} n.m.r. spectroscopy. This technique is capable of quantitatively determining the concentration of $R^1_3SiO_{1/2}$ ("M") and $SiO_{4/2}$ ("Q") units derived from the silicone resin and from the neopentamer, $Si(Me_3SiO)_4$, present in the initial silicone resin, in
10 addition to the total hydroxyl content of the silicone resin.

[0017] For the purposes of the present invention the $R^1_3SiO_{1/2}$ to $SiO_{4/2}$ ratio can be expressed as $\{M(\text{resin})+M(\text{neopentamer})\}/\{Q(\text{resin})+Q(\text{neopentamer})\}$ and represents the ratio of the total number of triorganosiloxy groups of the resinous and neopentamer portions of the silicone resin to the total number of silicate groups in the resinous and neopentamer
15 portions.

[0018] The silicone resin contains 2.0 wt% or less, alternatively 0.7 wt% or less, alternatively 0.3 wt% or less, of terminal units represented by the formula $XSiO_{3/2}$, where X represents hydroxyl or a hydrolyzable group such as alkoxy such as methoxy and ethoxy; alkenyloxy such as isopropenyloxy; ketoximo such as methylethylketoximo; carboxy such as
20 acetoxo; amidoxo such as acetamidoxo; and aminoxo such as N,N-dimethylaminoxo. The concentration of silanol groups present in the silicone resin can be determined using Fourier transform infrared spectrophotometry (FTIR).

[0019] The number average molecular weight, M_n , required to achieve the desired flow characteristics of the silicone resin will depend at least in part on the molecular weight of the
25 silicone resin and the type(s) of hydrocarbon radicals, represented by R^1 , that are present in this ingredient. M_n as used herein represents the molecular weight measured using gel permeation chromatography, when the peak representing the neopentamer is excluded from the measurement. The M_n of the silicone resin is typically greater than 3,000, more typically from 4500 to 7500. Typically the thermal hold (i.e. the ability of an adhesive to retain its
30 adhesion at elevated temperatures) above 150°C, becomes appreciable when the M_n exceeds 3000.

[0020] The silicone resin can be prepared by any suitable method. Silicone resins of this type have reportedly been prepared by cohydrolysis of the corresponding silanes or by silica hydrosol capping methods known in the art. The silicone resin is preferably prepared by the silica hydrosol capping processes of Daudt, et al., U.S. Pat. No. 2,676,182; of Rivers-Farrell et al., U.S. Pat. No. 4,611,042; and of Butler, U.S. Pat. No. 4,774,310.

[0021] The intermediates used to prepare the silicone resin are typically triorganosilanes of the formula R^1_3SiX' , where X' represents a hydrolyzable group, and either a silane with four hydrolyzable groups such as halogen, alkoxy or hydroxyl, or an alkali metal silicate such as sodium silicate.

[0022] It is desirable that the silicon-bonded hydroxyl groups (i.e. $HOR^1SiO_{1/2}$ or $HOSiO_{3/2}$ groups) in the silicone resin be below 0.7% by weight of the total weight of the silicone resin, alternatively below 0.3%. Silicon-bonded hydroxyl groups formed during preparation of the silicone resin are converted to trihydrocarbylsiloxy groups or a hydrolyzable group by reacting the silicone resin with a silane, disiloxane or disilazane containing the appropriate terminal group. Silanes containing hydrolyzable groups are typically added in excess of the quantity required to react with the silicon-bonded hydroxyl groups of the silicone resin.

[0023] The organopolysiloxane useful herein is comprised of difunctional units of the formula R^2R^3SiO and terminal units of the formula $R^4_aX'_{3-a}SiG-$ wherein R^2 is an alkoxy group or a monovalent unsubstituted or substituted hydrocarbon radical; R^3 is a unsubstituted or substituted monovalent hydrocarbon radical; R^4 is aminoalkyl or R^1 group X' is a hydrolyzable group; G is a divalent group linking the silicon atom of the terminal unit with another silicon atom and a is 0 or 1. The organopolysiloxane can optionally contain up to about 20 percent, based on total of trifunctional units of the formula $R^3SiO_{3/2}$ where R^3 is as described previously. At least 50 percent, typically at least 80 percent, of the radicals represented by R^2 and R^3 in the R^2R^3SiO units are lower alkyl such as methyl.

[0024] The terminal units present on the organopolysiloxane are represented by the formula $R^4_aX'_{3-a}SiG-$, where X' is a hydrolyzable group, R^4 is aminoalkyl or R^1 , G is a divalent group linking the silicon atom of the terminal unit with another silicon atom and a is 0 or 1.

Typically the organopolysiloxane contains an average of two or more hydrolyzable(X') groups per molecule in order to form a crosslinked product. Typical hydrolyzable groups represented by X' include but are not limited to hydroxy, alkoxy such as methoxy and

ethoxy, alkenyloxy such as isopropenyloxy, ketoximo such as methylethylketoximo, carboxy such as acetoxo, amidoxy such as acetamidoxy and aminoxy such as N,N-dimethylaminoxy.

[0025] In the terminal groups when a is 0 the groups represented by X' can be alkoxy, ketoximo, alkenyloxy, carboxy, aminoxy or amidoxy. When a is 1 X' is typically alkoxy and R^5 is alkyl such as methyl or ethyl, or aminoalkyl such as aminopropyl or 3-(2-aminoethylamino)propyl. The amino portion of the aminoalkyl radical can be primary, secondary or tertiary.

[0026] In the formula for the terminal unit G is a divalent group or atom that is hydrolytically stable. By hydrolytically stable it is meant that it is not hydrolyzable and links the silicon atom(s) of the terminal unit to another silicon atom in the organopolysiloxane such that the terminal unit is not removed during curing of the composition and the curing reaction is not adversely affected. Hydrolytically stable linkages represented by G include but are not limited to oxygen, hydrocarbylene such as alkylene and phenylene, hydrocarbylene containing one or more hetero atoms selected from oxygen, nitrogen and sulfur, and combinations of these linking groups. G can represent a silalkylene linkage such as $-(OSiMe_2)CH_2CH_2-$, $-(CH_2CH_2SiMe_2)(OSiMe_2)CH_2CH_2-$, $-(CH_2CH_2SiMe_2)O-$, $(CH_2CH_2SiMe_2)OSiMe_2O-$, $-(CH_2CH_2SiMe_2)CH_2CH_2-$ and $-CH_2CH_2-$, a siloxane linkage such as $-(OSiMe_2)O-$ or, more preferably, an oxygen atom.

[0027] Specific examples of preferred terminal units include, but are not limited to, $(MeO)_3SiCH_2CH_2-$, $(MeO)_3SiO-$, $Me(MeO)_2SiO-$, $H_2NCH_2CH_2N(H)(CH_2)_3SiO-$, $(EtO)_3SiO-$, $(MeO)_3SiCH_2CH_2SiMeCH_2SiMeCH_2CH_2SiMe_2O-$, $Me_2NOSiO-$, $MeC(O)N(H)SiO-$ and $CH_2=C(CH_3)OSiO-$. Me in these formulae represents methyl and Et represents ethyl.

[0028] When X' contains an alkoxy group, it may be desirable to separate this X' group from the closest siloxane unit by an alkylene radical such as ethylene. In this instance $R^4_a X'_3$.

$_aSiG-$ would be $(MeO)_3SiCH_2CH_2Si(Me_2)O-$. Methods for converting alkoxy groups to trialkoxysilylalkyl groups are described in the prior art. For example, moisture reactive groups having the formulae $(MeO)_3SiO-$ and $Me(MeO)_2SiO-$ can be introduced into a silanol-terminated polyorganosiloxane by compounds having the formulae $(MeO)_4Si$ and $Me(MeO)_3Si$, respectively. Alternatively, compounds having the formulae $(MeO)_3SiH$ and $Me(MeO)_2SiH$, respectively, can be used when the polyorganosiloxane contains silanol groups or alkenyl radicals such as vinyl and a platinum group metal or a compound thereof as

a hydrosilylation reaction catalyst. It will be understood that other hydrolyzable groups such as dialkylketoximo, alkenyloxy and carboxy can replace the alkoxy group.

[0029] The organopolysiloxane used in the hot melt adhesive is preferably a polydimethylsiloxane containing three alkoxy or ketoximo groups, two ketoximo groups or two alkoxy groups together with either an alkyl or aminoalkyl radical.

[0030] The viscosity of the organopolysiloxane should be in the range of 0.02 Pa·s to 100 Pa·s at 25 °C, typically 0.35 to about 60 Pa·s.

[0031] The silicone resin and organopolysiloxane are present in amounts to provide 55 to 75% resin solids based on the amount of silicone resin and organopolysiloxane. Higher amounts of resin can be used however, higher application temperatures will be needed to apply the moisture curable hot melt adhesive composition to a substrate.

[0032] The silane crosslinker is represented by the formula $R^1_nSiZ_{(4-n)}$, where R^1 is as described previously and Z is a hydrolyzable group that reacts with the terminal groups of at least the organopolysiloxane under ambient conditions to form a cured material and n is 0, 1 or 2. Typically R^1 is an alkyl and/or a phenyl group. Suitable hydrolyzable groups represented by Z include but are not limited to alkoxy containing from 1 to 4 carbon atoms, carboxy such as acetoxy, ketoximo such as methylethylketoximo and aminoxy. When n=2 in the silane crosslinker, the organopolysiloxane typically contain 3 X' groups (e.g., a = 0).

[0033] Suitable silane crosslinkers include but are not limited to methyltrimethoxysilane, isobutyltrimethoxysilane, methyltris(methylethylketoximo)silane, methyltriethoxysilane, isobutyltriethoxysilane, methyltriacetoxysilane and alkyl orthosilicates such as ethyl orthosilicate.

[0034] The amount of silane crosslinker used is in the range of 0.5 to 15 parts per hundred based on the amount of silicone resin and polymer, typically 1 to 8 pph. If too much silane crosslinker is present, the green strength and/or cure rate of the hot melt adhesive will decrease. If the silane crosslinker is volatile it may be necessary to use an excess amount to achieve the 1.5 to 15 pph in the final hot melt adhesive composition. One skilled in the art will be able to determine the amount need to produce a composition with 1.5 to 15 pph.

[0035] A titanate catalyst is typically used in the hot melt adhesive formulation except when the organopolysiloxane and/or the silane crosslinker contains ketoxime functional groups. The titanate catalyst is an organotitanium compound such as tetrabutyl titanate and partially chelated derivatives of these salts with chelating agents such as acetoacetic acid

esters and beta-diketones. The amount of titanate catalyst used is in the range of 0.01-2 pph based on the amount of resin and polymer, typically in the range of 0.05-1. If too much titanate catalyst is added then the cure of composition will be impaired. Additionally, as the amount of catalyst is increased the viscosity of the hot melt adhesive increases resulting in higher melt temperature required to apply the material.

[0036] The hot melt adhesive composition may contain 0.05-2 pph based on resin and polymer of an adhesion promoter. Adhesion promoters are known in the art and are typically silanes having the formula $R^5_c R^6_d Si(OR)_{4-(c+d)}$ where R^5 is independently a substituted or unsubstituted, monovalent hydrocarbon group having at least 3 carbon atoms and R^6 contains at least one SiC bonded group having an adhesion-promoting group, such as amino, epoxy, mercapto or acrylate groups, c has the value of 0 to 2 and d is either 1 or 2 and the sum of c+d is not greater than 3. The adhesion promoter can also be a partial condensate of the above silane.

[0037] The hot melt adhesive composition may contain 0.1 to 40 wt% based on total adhesive of a treated and/or untreated filler. Examples of suitable fillers include calcium carbonates, fumed silica, silicate, metal oxides, metal hydroxides, carbon blacks, sulfates or zirconates.

[0038] Solvent is typically used in producing the hot melt adhesive. Solvent aids with the flow and introduction of the silicone resin and organopolysiloxane polymers. However, essentially all of the solvent is removed in the continuous process for producing the hot melt adhesive. By essentially it is meant that the hot melt adhesive composition should contain no more than 0.05 to 5 wt.%, preferably less than 0.5 % solvent based on the weight of the hot melt adhesive. If too much solvent is present the viscosity of the hot melt adhesive will be too low and the product performance will be hindered.

[0039] Solvents used herein are those that help fluidize the components used in producing the hot melt adhesive but essentially do not react with any of the components in the hot melt adhesive. Suitable solvents are organic solvents such as toluene, xylene, methylene chloride, naphtha mineral spirit and low molecular weight siloxanes.

[0040] The silicone resin, organopolysiloxane, silane crosslinker, solvent and any optional ingredients are fed into a continuous mixing device. The order of addition into the continuous mixing device is not critical to produce the hot melt adhesive composition. If the

resin has typically more than 0.7 wt% silanol it is desirable to add the silane crosslinker and/or catalyst and resin together to allow for any reaction to take place and the reaction product (i.e. volatiles) to be removed. The continuous mixing device should be capable of mixing the ingredients and should include means for removing the solvent. Typically an
5 extrusion device is used and more typically a twin-screw extrusion device is used.

[0041] When using a extrusion device the components are fed into the extruder and heated to a temperature in the range of 50 to 250 C, alternatively 80 to 150 °C. By heating the composition in the extruder the viscosity is lowered to allow for adequate mixing the ingredients. Typically in the extrusion device the silicone resin and organopolysiloxane and
10 solvent are fed into the device. The silane crosslinker and optional catalyst may also be added at this point or they may be added further downstream in the device after some mixing has taken place. The continuous process of hot melt adhesives on a co-rotating twin-screw extruder is described in T. Peitz, "Continuous Processing of Hot Melt Adhesives on Co-Rotating Twin Screw Extruders", 1996 Hot Melt Symposium, p. 37-45.

[0042] The solvent is removed during the continuous mixing process. Typically vacuum is applied on the continuous mixing device to facilitate removal of the solvent and any other volatile components that may be in the hot melt adhesive composition. Vacuum may be applied in a single or multiple stages on the continuous mixing device. It has been found that the use of multiple vacuum stages provides improved removal to the solvent. Because the
20 silane crosslinker may be volatile, it is preferable to add the silane crosslinker after most of the solvent has been removed to prevent removal of the crosslinker with the solvent.

[0043] The hot melt adhesive composition can be used adhere at least two substrates together. Typically the hot melt adhesive composition is used as a layer between the two substrates to produce a laminate of the first substrate, the cured hot melt adhesive and the
25 second substrate. The laminate structure produced herein is not limited to these three layers. Additional layers of cured adhesive and substrate may be applied. The layer of hot melt adhesive composition in the laminate may be continuous or discontinuous. For example, a continuous layer may be used to form a laminate a such as the window 100 shown below in Figure 1. Alternatively, a discontinuous layer may be used to form a laminate such as the
30 housing 200 with a lid seal shown below in Figure 2.

[0044] Further there is no limitation on the material that may be used as the substrate. Suitable substrates to which the hot melt adhesive composition, or cured product thereof, may

be applied include, but are not limited to, glass; metals, such as aluminum, copper, gold, nickel, silicon, silver, stainless steel alloys, and titanium; ceramic materials; plastics including engineered plastics such as epoxies, polycarbonates, poly(butylene terephthalate) resins, polyamide resins and blends thereof, such as blends of polyamide resins with syndiotactic polystyrene such as those commercially available from The Dow Chemical Company, of Midland, Michigan, U.S.A., acrylonitrile-butadiene-styrenes, styrene-modified poly(phenylene oxides), poly(phenylene sulfides), vinyl esters, polyphthalamides, and polyimides; cellulosic substrates such as paper, fabric, and wood; and combinations thereof. When more than one substrate will be used, there is no requirement for the substrates to be made of the same material. For example, it is possible to form a laminate of a glass and metal substrate or glass and plastic substrate.

[0045] One method for producing the laminate structure is to apply a film of the hot melt adhesive composition on the surface of the first substrate. A surface of the second substrate is then contacted with the hot melt adhesive composition and the first and second surfaces are pressed together. Conventional application methods suitable for use with molten materials include, but are not limited to, dipping, spraying, coextrusion, and spreading using heated doctor blades, draw-down bars and calendar rolls. Typically the hot melt adhesive composition is applied by heating at a temperature of 80 to 150 °C and applied by pressure through a hose. Equipment of this type is known in the art and commercially available. Upon exposure to moisture the hot melt adhesive composition cures. The hot melt adhesive composition may be exposed to moisture by contacting moisture in the air or by direct introduction of moisture such as from contacting the laminate with steam or placing the laminate in a humidity chamber.

[0046] It is also possible to form a laminate by forming a film of the hot melt adhesive composition and then curing the film of the hot melt adhesive composition. A first surface of the cured film is contacted with a surface of the first substrate. A surface of the second substrate is then contacted with the other surface of the cured film and the first and second surfaces are pressed together to form the laminate.

[0047] Another alternative to producing the laminate is to apply a film of the hot melt adhesive composition on the surface of the first substrate. The hot melt adhesive composition is then cured by exposure to moisture to produce the cured film. The second substrate is then

contacted with the cured film on the first substrate and the first and second surfaces are pressed together to form the laminate.

[0048] The cured films prepared by curing the hot melt adhesive compositions according to this invention find utility in various industries such as automotive, electronic, construction, space, and medical. The cured films may provided bonds that are resistant to hostile environments such as heat and moisture. For example, the cured films may be used as conformal coatings for substrates such as printed circuit boards and other substrates containing electrical or electronic components.

[0049] Various laminates may be prepared according to this invention using the hot melt adhesive compositions. For example, the laminate may be a portion of an air bag, a car interior, a window, or a lid seal. Figure 1 shows a portion of a window prepared according to this invention. The window includes a sash 100 on which a film of the hot melt adhesive composition 101 is applied. An insulating glass unit 102 comprised of two panes of glass 103, 104 separated by a spacer 105 is applied to the film of the hot melt adhesive composition 101. The film of hot melt adhesive composition 101 may be cured before application to the sash 100, after application to the sash 100, before application to the insulating glass unit 102, or after application to the insulating glass unit 102. The two panes of glass 103, 104 may be attached to the spacer 105 by films of the hot melt adhesive composition 106. The films of hot melt adhesive composition 106 may be cured before application to the pane of glass 103 or 104, after application to the pane of glass 103 or 104, before application to the spacer 105 or after application to the spacer 105.

[0050] Figure 2 shows a schematic representation of a housing 200 for an electronic component 204 with a lid seal prepared according to this invention. The housing 200 may be prepared by, for example,

- (1) applying the hot melt adhesive composition 201 described above onto the rim of a first substrate shown here as a container 202,
- (2) placing a second substrate shown here as a lid 203 over the container 202 such that the edges of the lid 203 are in contact with the hot melt adhesive composition 201, and
- (3) curing the hot melt adhesive composition 201 to form a lid seal between the container 202 and the lid 203.

[0051] One skilled in the art would recognize that the hot melt adhesive composition may be applied to the edges of the lid 203 first and thereafter the lid 203 placed onto the container 202. One skilled in the art would recognize that curing the hot melt adhesive composition 201 may be performed before, during, or after application to one or both of the substrates (lid 203 and container 202). Alternatively, the hot melt adhesive composition or hot melt adhesive formed by curing the hot melt adhesive composition may be applied at the edges of the lid after the lid has been placed onto the container.

EXAMPLES

10 Materials:

Silicone Resins

[0052] Resin A: a xylene soluble resinous copolymer containing triorganosiloxy units and SiO₂ units in the molar ratio of 0.8 mol of triorganosiloxy units per mol of SiO₂ units, where the triorganosiloxy units are trimethylsiloxy and dimethylvinylsiloxy and the copolymer contains 1.9 weight percent of vinyl radicals and 1.5 wt% Si bonded hydroxy groups. The resin is dissolved in xylene to produce a solution of 70 wt% solids

[0053] Resin B: a xylene soluble resinous copolymer containing triorganosiloxy units and SiO₂ units in the molar ratio of 0.8 . The resin is capped with trimethylsiloxy groups to produce a resin with 0.7 % by weight of Si bonded hydroxyl groups. The resin is dissolved in xylene to produce a solution of 60 wt% solids.

Organopolysiloxanes

[0054] Polymer A: a primarily linear polydimethylsiloxane polymer of approximate viscosity 70,000 cs, terminated by -CH₂CH₂Si(OMe)₃ end groups.

[0055] Polymer B: a primarily linear polydimethylsiloxane polymer of approximate viscosity 70,000 cs, approximately 80 % of the ends terminated by —CH₂CH₂SiOSiCH₂CH₂Si(OCH₃)₃, with the remaining ends terminated by vinyl groups.

[0056] Polymer C: a primarily linear hydroxyl endblocked polydimethylsiloxane fluid having a viscosity of 50 Pa·s and 25 °C.

Crosslinkers

[0057] Alkoxy silane A : i-Bu Si(OMe)₃, isobutyltrimethoxysilane

[0058] Alkoxy silane B : MeSi(OMe)₃, methyltrimethoxysilane

[0059] Oxime silane C: Methyltri(ethylmethylketoxime)silane

5 [0060] Oxime silane D: Vinyltri(methylethylketoxime)silane

Optional Ingredients

[0061] Alkoxy silane C : NH₂CH₂CH₂NHCH₂CH₂CH₂Si(OMe)₃

[0062] Alkoxy silane D : reaction mixture of 74 parts by weight (pbw) of
10 glycidoxypyltrimethoxy silane and 26 pbw of aminopropyltrimethoxysilane prepared by
mixing the silanes and allowing to stand for 16 hrs at room temperature.

CaCO₃ – a ground calcium carbonate of average 2 micron particle size, treated with stearate.
Available from Georgia Marble Co., Kennesaw GA. USA .

15 Titanate Catalysts

[0063] Titanate catalyst A : Ti(OtBu)₄ , tetra - tertiary butyl titanium

[0064] Titanate catalyst B : Ti(Obu)₄, tetra – n butyl titanium

[0065] Titanate catalyst C : Ti(iOPr)₂(EAA)₂ – diisopropoxy bis-ethylacetoacetato titanium

[0066] All these titanate catalysts are commercially available from DuPont , Inc. Deepwater
20 NJ, USA.

[0067] Tin catalyst D: Dibutyl tin dilaurate

Testing Methods :

25 [0068] NVC : The non-volatile content (NVC) is determined by placing approximately 1 g
of the hot melt adhesive into an aluminum weighing dish, heating at 110 °C for 16 hrs and
recording the weight change. (note; example 10 uses 1 hr / 150 C conditions)

[0069] Viscosity : The equipment used is a Bohlin Instruments VOR Rheometer attached
30 with a Mitutoyo micrometer and temperature controller. Parallel plate geometry is used with
1.5 mm gap between the plates. Testing is done in the Oscillation mode with using a

frequency = 1Hz and Amplitude = 25% (strain = 0.0518). A metal cartridge containing the hot melt is placed in oven at 110 °C to obtain the sample for viscosity testing. The sample is then placed between the plates and the plates are brought together until a gap of 1.5mm is reached. During this process, the sample is heated up to 160 °C to 180 °C, and any excess
5 sample is cleaned off from the sides of the plates. Once the desired temperature and gap are reached, the sample is ramped down from 160 –180 °C to 25 °C at a cooling rate = 3 °C/2min, while collecting data every 2min.

Examples 1 - 9

10 [0070] Resin and polymer were mixed in a 5-gallon metal pail, then a pre-mixed slurry of silane and catalyst slurry was added and mixed. The mixture was pumped into a 30mm twin-screw extruder at a flow rate of about 10 lbs./hr. The barrel temperature (bl temp), screw speed (rpm) and vacuum conditions (vacuum mmHg) are provided in the tables.

[0071] In the twin screw extruder before each vacuum stage, were two reversing elements
15 (10/10 left handed) that were used to create seal before the vacuum. Under each vacuum port, 3-4 long, conveying elements (42/42) were used so that the material can have more surface area and longer residence time exposed to vacuum.

[0072] When resin B was used, the mixture (resin, polymer, silane, and catalyst) viscosity was much lower. Using a flow rate of 10 lbs/hr, significant surging was observed, and the
20 material could not be well stripped. To solve this problem conveying elements with less pumping capability were used (e.g. use 42/42 and 28/28 instead of 20/20) and add one reversing element was added between the second and third vacuum

Example 1

25 [0073] Example 1 shows the impact of vacuum and rpm on nvc and viscosity.

Table 1A: Example 1 Ingredients and Processing Conditions

Run #	parts	parts	pph	pph	Vacuum			rpm	bl temp °F
	resin A	polymer A	silane A	catalyst A	1	2	3		
1-1	66	34	8	0.55	17	15	13.6	300	220
1-2	66	34	8	0.55	17	15	14.6	300	220
1-3	66	34	8	0.55	17	15	11.5	400	220
1-4	66	34	8	0.55	17	15	13.4	400	220
1-5	66	34	8	0.55	17	15	14.7	400	220
1-6	66	34	8	0.55	17	15	11.5	500	220
1-7	66	34	8	0.55	17	15	13.5	500	220
1-8	66	34	8	0.55	17	15	14.7	500	220

Table 1B: Example 1 Results

Run #	nvc %	Viscosity (P)		xylene gc%
		at 75°C	at 120°C	
1-1	98.0	35000	2200	0.25
1-2	99.3	45000	3300	not dissol.
1-3	98.2	17000	1100	0.55
1-4	98.6	22000	1300	0.23
1-5	99.4	89000	6700	0.12
1-6	98.3	39000	2200	0.74
1-7	99.1	30000	1800	0.42
1-8	99.4	120000	7200	0.34

5 Example 2

[0074] Example 2 show the impact of varying catalyst levels on viscosity at two different resin contents.

Table 2A: Example 2 Ingredients and Processing Conditions

Run #	parts	parts	pph	pph	Vacuum			rpm	bl temp
	resin A	polymer A	silane A	catalyst A	1	2	3		°F
2-1	68	32	12	0.10	17	17	14.6	400	220
2-2	68	32	12	1.00	17	17	14.6	400	220
2-3	64	36	4	0.10	17	17	14.6	400	220
2-4	64	36	4	1.00	17	17	14.6	400	220

Table 2B: Example 2 Results

Run #	nvc %	Viscosity (P)	
		at 75C	at 120C
2-1	99.3	33000	1500
2-2	99.3	250000	16000
2-3	99.1	31000	4100
2-4	99.6	88000	7800

5 Example 3

[0075] Examples 3 shows the impact of crosslinker volatility on nvc and viscosity.

Table 3A: Example 3 Ingredients and Processing Conditions

Run #	parts	parts	pph	pph	vacuum			rpm	bl temp
	resin A	polymer A	silanes A/B	catalyst A	1	2	3		°F
3-1	65	35	0 / 8	1	18	15	14.2	400	220
3-2	65	35	4 / 4	1	15	15	13.3	400	220

10 Table 3B: Example 3 Results

Run #	nvc %	Viscosity (P)	
		at 75C	at 120C
3-6	99.0	67000	6800
3-7	98.0	32000	2000

Example 4

[0076] Example 4 shows that a partially capped polymer can be used to make the hot melt adhesive.

Table 4: Example 4 Ingredients and Processing Conditions

Run #	parts resin A	parts polymer B	pph silane A	pph catalyst A	vacuum			rpm	bl temp F
					1	2	3		
4-1	66	34	8	0.55	17	17	14.6	400	220

5

Table 4B: Example 4 Results

Run #	nvc %	Viscosity (P)	
		at 75C	at 120C
4-1	98	60000	4100

Example 5

[0077] Example 5 shows primarily the impact of vacuum on nvc. In these cases the Me₃ capped resin is used. The last 3 samples are replicates of each other.

10

Table 5A: Example 5 Ingredients and Processing Conditions

Run #	parts Resin B	parts Polymer A	pph silane A	pph catalyst A	vacuum			rpm	bl temp F
					1	2	3		
5-1	65	35	8	0.55	20	20	14.5	400	220
5-2	65	35	8	0.55	20	20	13.6	400	220
5-3	65	35	8	0.1	20	20	14.5	400	220
5-4	65	35	8	0.1	20	20	13.6	400	220
5-5	65	35	12	0.1	20	20	14.5	400	220
5-6	65	35	12	0.1	20	20	13.6	400	220
5-7	65	35	12	0.1	20	20	14	400	220
5-8	65	35	12	0.1	20	20	14	400	220
5-9	65	35	12	0.1	20	20	14	400	220

Table 5B: Example 5 Results

Run #	nvc %	Viscosity (P)	
		at 75C	at 120C
5-1	99		
5-2	97.7		
5-3	99		
5-4	97.3		
5-5	99.3	48000	2100
5-6	97.4		
5-7	98.1	28000	1000
5-8	98.1		
5-9	98.5		

Example 6

[0078] This example shows the impact of catalyst loading on viscosity, resin is Me₃

5 capped.

Table 6A: Example 6 Ingredients and Processing Conditions

Run #	parts Resin B	parts Polymer A	pph silane A	pph catalyst A	vacuum			rpm	bl temp F
					1	2	3		
6-1	65	35	8	0.1	20	20	14	400	220
6-2	65	35	8	0.55	20	20	14	400	220
6-3	65	35	12	0.1	20	20	14	400	220

Table 6B: Example 6 Results

Run #	nvc %	Viscosity (P)	
		at 75C	at 120C
6-1	98	34000	1400
6-2	97.7	53000	2500
6-3	97.8	28000	1000

Example 7

[0079] Example 7 shows that other titanate catalysts can be used to make the hot melt adhesives.

5 Table 7A: Example 7 Ingredients and Processing Conditions

Run #	parts Resin B	parts Polymer A	pph silane A	pph catalyst	vacuum			rpm	bl temp F
					1	2	3		
7-1	65	35	8	0.55 - cat B	20	20	14	400	220
7-2	65	35	8	0.1 - cat B	20	20	14	400	220
7-3	65	35	8	0.65 - cat C	17	20	14	400	220
7-4	65	35	8	0.55 - cat. A	20	20	14	400	220
7-5	66	34	8	0.55 - cat. A	17	20	14	400	220

Table 7B: Example 7 Results

Run #	nvc %	Viscosity (P)	
		at 75C	at 120C
7-1	97.7	n/a	n/a
7-2	97.5	15000	680
7-3	n/a	54000	3000
7-4	n/a	53000	2500
7-5	n/a	69000	3400

Example 8

10 [0080] Example 8 shows that fillers and adhesion promoters may be used.

Table 8A: Example 8 Ingredients and Processing Conditions

Run #	parts Resin B	parts Polymer A	pph silane A	pph catalyst A	pph other	vacuum			rpm	bl temp F
						1	2	3		
8-1	65	35	8	0.55	CaCO ₃ - 2	17	20	14	400	220
8-2	65	35	8	0.55	silane C - 0.2	17	20	14	400	220
8-3	65	35	8	0.55	silane D - 0.2	20	20	14	400	220

Table 8B: Example 8 Results

Run #	nvc %	Viscosity (P)	
		at 75 °C	at 120 °C
8-1	98.3	20000	1100
8-2	97.9		
8-3	97.2		

5 Example 9

[0081] Example 9 shows that useful hot melt materials may be prepared with oxime cure.

Table 9A: 9 Ingredients and Processing Conditions and results

	parts	parts	pph	pph	vacuum				bl temp	nvc
Run #	Resin B	Polymer C	oxime silane	catalyst D	1	2	3	rpm	°F	%
9-1	65	35	5 of C	0.2	12	15	14	400	200	96.7
9-2	65	35	5 of D	0.2	12	15	14	400	200	95.9

10 Example 10

[0082] A 50mm twin-screw compounder with L/D=50 was used for these samples. The resin and polymer were pre-mixed in tank, then transferred to another tank which was used to feed the compounder (both tanks can be heated, the resin and polymer mixture feed

15 temperature is noted below); three vacuum stages were used to remove solvent from the

process, the three vacuum stages were at 13.93D, 20.86D, and 30.87D; a mixture of crosslinker and catalyst was loaded at 35.87D; a final vacuum was applied at 45.86D.

[0083] Example 10 shows that a hot melt adhesive can be made by feeding the resin and polymer separately from the crosslinker and catalyst. Also shows that the resin and polymer stream can be heated before introducing to the compounder.

Table 10A: Example 10 Ingredients and Processing Conditions

Run #	parts Resin B	parts Poly A	pph Silane A	pph Cat. A	Feed Temp C	rpm	vacuum in hg			
10-1	65	35	1	0.1	50	400	18.5	19	23.5	21.5
10-2	65	35	2.5	0.25	80	500	19	20	22	20

Table 10B: Example 10 Results

Run #	nvc %	Viscosity (P)	
		120 C	75 C
10-1	99.3	3100	54000
10-2	98.7	1700	39000

Example 11

[0084] Resin B (65 pbw) and Polymer A (35 pbw) were mixed for 30 minutes. A pre-mixed slurry of Alkoxy silane A (8 pbw) and Titanate catalyst A (0.1 pbw) was added into the mixture of resin and polymer and mixed for 10 minutes. The resulting mixture was pumped through a twin screw extruder under heat and vacuum to produce a hot melt sealant.

[0085] The hot melt sealant was used to prepare a cured film by preparing a film having a 0.254 centimeter (cm) thickness and curing at room temperature for 21 days. The cured film was cut into 2.54 x 2.54 cm pieces. Each piece was applied on a first substrate and a second substrate was pressed on top of the cured film piece to prepare a cured film laminate.

[0086] The hot melt sealant was heated to 121 °C and applied on a first substrate. A second substrate was pressed on top of the hot melt to produce a 0.254 cm thickness between the first

and second substrates to form a wet film laminate. All laminates were cured for at least 21 days at room temperature.

[0087] Table 11 shows the laminate number, substrates used in each laminate, and the shear force measured by Sintech shear testing instrument on each of the laminates after curing. In Table 11, G represents glass, A represents aluminum, PC represents polycarbonate, and W represents wood.

Table 11 - Results of Example 11

Laminates of silicone hot melt			
Laminate #	Substrate	Shear Force	
		Peak Stress, psi(MPa)	
		Wet Film	Cured Film
11-1	G-G	372(2.6)	450(3.1)
11-2	G-W	635(4.4)	551(3.8)
11-3	G-A	628(4.3)	629(4.3)
11-4	G-PC	567(3.9)	485(3.3)
11-5	W-W	729(5.0)	571(3.9)
11-6	W-PC	685(4.7)	648(4.5)

- 10 [0088] Example 11 shows that a hot melt adhesive composition made by the method of this invention effectively laminates a variety of substrates.

Comparative Example 1

A sample is prepared by mixing the components in Table 11A.

- 15 Table 11A - Components in Comparative Example 1

Component	Amount (parts)
Hydroxyl terminated dimethylsiloxane	70
Fumed silica	9
Silicone plasticizer	15
Oxime silane crosslinker	5.8
Dibutyltin dilaurate catalyst	0.1

[0089] Laminates are prepared using the compositions in Comparative Example 1 and Example 11. Lap shear is measured on the laminates at 0.0847 cm/sec with 6.45 square cm overlap and thickness of 0.254 cm. Lap shear is measured several times as the compositions cure over 14 days. The results are in Table 12.

5

Table 12 - Lap Shear psi (MPa)

Cure time	15 min.	1 hrs	2 hrs	24 hrs	72 hrs	7 days	14 days
Example 12	38(2.6)	47(3.2)	64(4.4)	72(5.0)	84(5.8)	150(10.3)	165(11.4)
Comparative Example 1	0(0)	0(0)	0(0)	6(0.4)	28(1.9)	148(10.2)	191(13.2)

Industrial Applicability

10 [0090] The hot melt adhesive compositions and hot melt adhesives prepared by curing the hot melt adhesive compositions find use in the construction and electronics industries. The hot melt adhesive compositions and hot melt adhesives may be used in the construction industry, for example, in window applications. The hot melt adhesive compositions and hot melt adhesives may be used in the electronics industry, for example, in lid seal applications.

DRAWINGS

[0091] Figure 1 is a portion of a window prepared according to this invention.

[0092] Figure 2 is a schematic representation of a housing with a lid seal prepared according to this invention.

5		<u>Reference Numerals</u>
	100	sash
	101	hot melt adhesive composition
	102	insulating glass unit
	103	glass pane
10	104	glass pane
	105	spacer
	106	hot melt adhesive composition
	200	housing
	201	hot melt adhesive composition
15	202	container
	203	lid
	204	electronic component

CLAIMS

1. A continuous process for producing a hot melt adhesive composition, wherein the process comprises:

(A) feeding into a continuous mixing device

- 5 (1) a silicone resin having a silanol content of less than 2 wt% and comprised of monofunctional units represented by $R^1_3SiO_{1/2}$ and tetrafunctional units represented by $SiO_{4/2}$ where R^1 is a substituted or unsubstituted monovalent hydrocarbon radical,
- 10 (2) an organopolysiloxane comprised of difunctional units of the formula R^2R^3SiO and terminal units of the formula $R^4_aX'_{3-a}SiG-$ wherein R^2 is an alkoxy group or a monovalent unsubstituted or substituted hydrocarbon radical; R^3 is a monovalent unsubstituted or substituted hydrocarbon radical; R^4 is aminoalkyl or R^1 group, X' is a hydrolyzable group; G is a divalent group linking the silicon atom of the terminal unit with another
- 15 silicon atom and a is 0 or 1,
- (3) 0.5 to 15 parts of a silane crosslinker per hundred per part of silicone resin and organopolysiloxane and
- (4) solvent;
- (B) creating a mixture of (1) - (4) in the continuous mixing device;
- 20 (C) sufficiently removing volatiles from the mixture in the continuous mixing device until the mixture has a non-volatile content of at least 97 weight %; and
- (D) recovering the hot melt adhesive composition.

25 2. The process as claimed in claim 1 wherein the mixture of (1) - (4) is created at a temperature of 50 to 250 °C.

3. The process as claimed in claim 1 wherein the silane crosslinker is added to the mixture after the volatiles have been removed.

30 4. The process as claimed in claim 1 wherein the silane crosslinker and catalyst are added to the mixture after the volatiles have been removed.

5. The process as claimed in claim 1 wherein the mixture additionally comprises at least one of (5) an adhesion promoter and (6) a filler.

6. The process as claimed in claim 1 wherein the continuous mixing device is an extruder.

5

7. The process as claimed in claim 6 wherein the extruder is a twin screw extrusion device.

8. The process as claimed in claim 1 wherein vacuum is applied to the continuous mixing device to assist with the removal of any volatiles.

10

9. The process as claimed in claim 1 wherein the hot melt adhesive composition has a viscosity of 200 to 10,000 poise at 120 °C.

10. A process for producing a laminate wherein the process comprises

15

(A) applying a film of the hot melt adhesive composition produced in claim 1 to a surface of a first substrate;

(B) contacting a surface of a second substrate with the film of the hot melt adhesive composition;

(C) pressing the first and second substrate together

20

(D) exposing the hot melt adhesive composition to moisture to cure the hot melt adhesive composition and produce the laminate.

11. A process for producing a laminate wherein the process comprises

(A) producing a thin film of the hot melt adhesive composition produced in claim 1;

25

(B) exposing the hot melt adhesive composition to moisture to cure the hot melt adhesive composition to form a cured film;

(C) applying the cured film to a surface of a first substrate;

(D) contacting a surface of a second substrate with the cured film;

(E) pressing the first and second substrate together to produce the laminate.

30

12. The process as claimed in claim 10 or claim 11 wherein the first substrate and the second substrate are each independently selected from glass, metal, ceramic materials, plastic, and cellulosic substrates.

5 13. The process as claimed in claim 12 wherein the first and second substrate are the same.

14. A hot melt adhesive composition prepared by the process of claim 1.

15. A laminate prepared by the process of claim 10 or claim 11.

10

16. Use of the hot melt adhesive composition of claim 14 in construction industry applications or electronics industry applications.

Figure 1

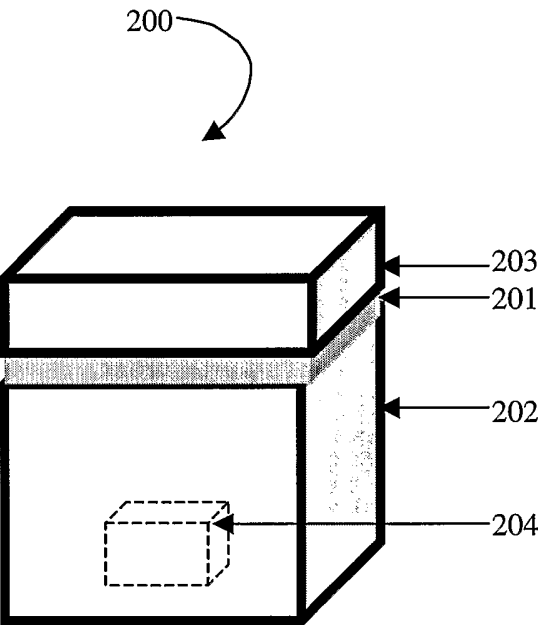
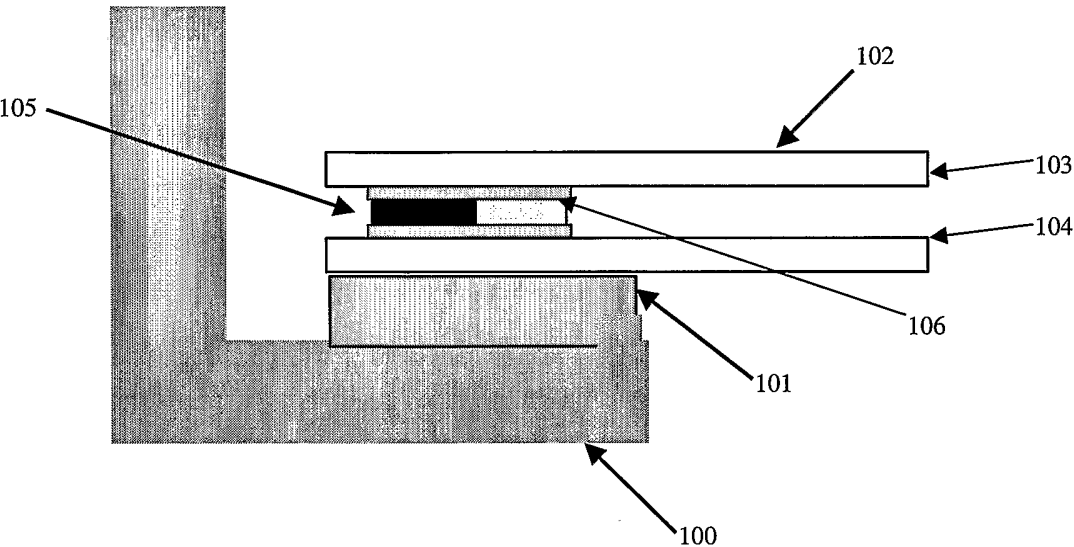


Figure 2