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(54) Title: PRESSURE SENSITIVE ADHESIVE COMPOSITION AND METHODS FOR ITS PREPARATION AND USE IN FLEXIBLE ORGANIC LIGHT EMITTING DIODE APPLICATIONS

(57) Abstract: A pressure sensitive adhesive composition can cure via hydrosilylation to form a pressure sensitive adhesive. The pressure sensitive adhesive composition may be coated on a substrate and cured to form a protective film. The protective film is useful in flexible OLED device fabrication processes, e.g., for protection of passivation layers.



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PRESSURE SENSITIVE ADHESIVE COMPOSITION AND METHODS FOR ITS  
PREPARATION AND USE IN FLEXIBLE ORGANIC LIGHT EMITTING DIODE  
APPLICATIONS

CROSS REFERENCE TO RELATED APPLICATIONS

[0001] This application claims the benefit of U.S. Provisional Patent Application Serial No. 63/172,737 filed on April 9, 2021 under 35 U.S.C. §119 (e). U.S. Provisional Patent Application Serial No. 63/172,737 is hereby incorporated by reference.

TECHNICAL FIELD

[0002] This invention relates to a pressure sensitive adhesive composition that is useful in processes for fabricating flexible organic light emitting diode (OLED) displays. The pressure sensitive adhesive composition cures to form a pressure sensitive adhesive with low initial adhesion, high adhesion stability over time, and low migration.

BACKGROUND

[0003] In a typical process for fabricating a flexible OLED display, an OLED is formed on a relatively rigid substrate (*e.g.*, glass or glass coated with a polyimide varnish), and a passivation layer is formed on the surface of the OLED opposite the substrate. During further processing, the rigid substrate is removed. Weak or brittle layers need to be protected from damage (*e.g.*, scratches or other shocks) during the further processing.

[0004] To protect the layers during the fabrication process, a protective film with low adhesion is desired to prevent delamination of a layer (*e.g.*, passivation layer) on a surface of an OLED module during releasing of the protective film after use. If adhesion control fails and adhesion increase with time (*i.e.*, during the fabricating process), the layer may delaminate or be damaged when the protective film is removed. Therefore, there is an industry need to provide protective films with low adhesion and high adhesion stability (where adhesion does not increase during the time needed for the fabrication process to an extent that would cause delamination or damage).

[0005] It is also important that the surface of the layer remain clean such that additional films or layers can be laminated thereto after removal of the protective film. Therefore, there is an industry need to provide a protective film with low migration (such that no or minimal amounts of adhesive from the protective film remain on the surface of the layer in the flexible OLED device after the protective film is removed).

BRIEF SUMMARY OF THE INVENTION

[0006] A pressure sensitive adhesive composition comprises:

a poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer, where the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer has silicon bonded alkyl groups with 1 to 10 carbon atoms and silicon bonded alkenyl groups with 2 to 6 carbon atoms, and where the alkenyl groups are present in an amount 0.14% to < 0.3% based on the weight of the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer,

polyalkylhydrogensiloxane having unit formula

$(R^1_2HSiO_{1/2})_2(R^1_2SiO_{2/2})_x(R^1HSiO_{2/2})_y$ , where each  $R^1$  is an independently selected alkyl group of 1 to 10 carbon atoms, subscript  $x \geq 0$ , subscript  $y \geq 0$ , and a quantity  $(x + y)$  is sufficient to give the polyalkylhydrogensiloxane a viscosity of < 20 mPa·s,

a platinum group metal catalyst, and

an acetylenic alcohol, and

an organic solvent.

[0007] A method for preparing the pressure sensitive adhesive composition, a method of using the pressure sensitive adhesive composition to form a pressure sensitive adhesive tape, and a method for fabricating a flexible organic light emitting diode device using the pressure sensitive adhesive tape are also disclosed.

#### DETAILED DESCRIPTION OF THE INVENTION

[0008] The pressure sensitive adhesive composition comprises:

A) 85 to 100 parts by weight of poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer, where the copolymer has silicon bonded alkyl groups with 1 to 10 carbon atoms and silicon bonded alkenyl groups with 2 to 6 carbon atoms, and where the alkenyl groups are present in an amount of 0.14% to < 0.3% based on the weight of the copolymer,

C) 0.1 to 5 parts by weight of a polyalkylhydrogensiloxane having unit formula  $(R^1_2HSiO_{1/2})_2(R^1_2SiO_{2/2})_x(R^1HSiO_{2/2})_y$ , where each  $R^1$  is an independently selected alkyl group of 1 to 10 carbon atoms, subscript  $x \geq 0$ , subscript  $y \geq 0$ , and a quantity  $(x + y)$  is sufficient to give the polyalkylhydrogensiloxane a viscosity of < 20 mPa·s (alternatively 8 mPa·s to 16 mPa·s),

F) an amount of a platinum group metal catalyst sufficient to provide 10 to 5000 ppm concentration of Pt group metal in the composition, and

E) 0.01 to 1 part by weight of an acetylenic alcohol, and

G) 10 weight % to 90 weight%, based on combined weights of all starting materials in the composition, of an organic solvent; with the proviso that all starting materials in the composition combined have a molar ratio of silicon bonded hydrogen atoms / silicon bonded alkenyl groups (SiH:Alkenyl ratio) of 0.5:1 to 2:1.

Starting Material A) Poly(dialkylsiloxane/alkylalkenylsiloxane) Copolymer

[0009] Starting material A) in the pressure sensitive adhesive composition is a poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer, which has silicon bonded alkyl groups with 1 to 10 carbon atoms and silicon bonded alkenyl groups with 2 to 6 carbon atoms, and where the alkenyl groups are present in an amount of 0.14% to < 0.3% based on the weight of the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer. Alternatively, starting material A) may contain at least 0.15%, alternatively > 0.20%, alternatively at least 0.21%, and alternatively at least 0.22% alkenyl groups; while at the same time starting material A) may contain up to 0.29%, alternatively up to 0.28%, alternatively up to 0.27%, alternatively up to 0.26%, alternatively up to 0.25%, alternatively up to 0.24%, alternatively up to 0.23%, alternatively up to 0.22%, and alternatively up to 0.21% of alkenyl groups, on the same basis. Alternatively, the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer may contain 0.14% to < 0.3%, alternatively > 0.20% to 0.26% alkenyl groups and alternatively 0.202% of alkenyl groups. Without wishing to be bound by theory, it is thought that if the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer contains > 0.3% alkenyl groups, the pressure sensitive adhesive prepared from the composition may cause burrs, which cause damage to the passivation layer when the pressure sensitive adhesive film is removed during fabrication of an electronic device, such as a flexible organic light emitting diode device. And, it is thought that if the alkenyl content of the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer is too low (*e.g.*, < 0.14%), migration may increase, and subsequent adhesion strength test results may become too low and/or adhesion strength may increase too much after aging (adhesion stability may become poor). Starting material A) may be free of silicon bonded aromatic hydrocarbyl groups. “Free of” in this context means that starting material A) contains no aromatic hydrocarbyl groups (such as phenyl, tolyl, xylyl or benzyl) or an amount of aromatic hydrocarbyl groups that is non-detectable by digestive gas chromatography mass spectrometry (GCMS) analysis.

[0010] Starting material A) may comprise a poly(dialkylsiloxane/alkylalkenylsiloxane) of average unit formula (I):  $(R^1_2R^2SiO_{1/2})_d(R^1_3SiO_{1/2})_e(R^1R^2SiO_{2/2})_f(R^1_2SiO_{2/2})_g$ , where subscript  $d \geq 0$ , subscript  $e \geq 0$ , subscript  $f > 0$ , subscript  $g \geq 0$ , where a quantity  $(d+e) = 2$ , and a quantity  $(d+e+f+g)$  is 3 to 10,000, alternatively 2,000 to 9,500, alternatively 4,500 to 5,000. Alternatively, subscripts  $d$ ,  $e$ ,  $f$ , and  $g$ , have values such that  $2 \geq d \geq 0$ ,  $2 \geq e \geq 0$ ,  $70 \geq f \geq 0$ ,  $10,000 \geq g \geq 0$ . Alternatively, subscript  $f$  may have a value such that  $50 \geq f \geq 0$ , alternatively that  $40 \geq f \geq 10$  and alternatively that  $37 \geq f \geq 20$ . Alternatively, subscript  $g$  may have a value such that  $9,500 \geq g \geq 0$ , alternatively  $5,500 \geq g \geq 4,000$ , alternatively  $5,000 \geq g \geq 4,500$ , and alternatively  $4,950 \geq g \geq 4,450$ . Subscripts  $d$ ,  $e$ ,  $f$ , and  $g$ , have values selected such that the

poly(dialkylsiloxane/alkylalkenylsiloxane) has the alkenyl content described above.

Alternatively, subscripts d, e, f, and g, may have values such that number average molecular weight ( $M_n$ ) of starting material A) is at least 200,000 g/mol; alternatively at least 250,000 g/mol; alternatively at least 300,000 g/mol, and alternatively at least 330,000 g/mol; while at the same time  $M_n$  may be up to 500,000 g/mol, alternatively up to 450,000 g/mol, alternatively up to 400,000 g/mol, and alternatively up to 370,000 g/mol, measured by Gel Permeation Chromatography according to the test method in U.S. Patent 9,593,209 at col. 31, Reference Example 1.

**[0011]** In unit formula (I), each  $R^1$  is independently an alkyl group of 1 to 10 carbon atoms and each  $R^2$  is independently an alkenyl group of 2 to 6 carbon atoms. Suitable alkyl groups include branched or unbranched, saturated monovalent hydrocarbon groups. Alkyl is exemplified by, but not limited to, methyl, ethyl, propyl (*e.g.*, iso-propyl and/or n-propyl), butyl (*e.g.*, isobutyl, n-butyl, tert-butyl, and/or sec-butyl), pentyl (*e.g.*, isopentyl, neopentyl, and/or tert-pentyl), hexyl, heptyl, octyl, nonyl, and decyl, as well as branched saturated monovalent hydrocarbon groups of 6 to 10 carbon atoms. Alternatively, each  $R^1$  may be selected from methyl, ethyl, and iso-propyl. Alternatively, each  $R^1$  may be methyl. Suitable alkenyl groups include branched or unbranched monovalent hydrocarbon groups having a terminal double bond. Alkenyl is exemplified by, but not limited to, vinyl, allyl, butenyl (*e.g.*, isobutenyl, n-butenyl, tert-butenyl and/or sec-butenyl), and hexenyl, including branched and linear groups of 6 carbon atoms. Alternatively, each  $R^2$  may be alkenyl. Alternatively, each  $R^2$  may be vinyl, allyl, or hexenyl. Alternatively, each  $R^2$  may be vinyl or hexenyl.

**[0012]** The alkenyl content of starting material A) may be calculated by calculating the total molecular weight of the poly(dialkylsiloxane/alkylalkenylsiloxane), calculating the molecular weights of each unit containing an alkenyl group, and dividing the combined molecular weights of each unit containing an alkenyl group by the total molecular weight. For example, in the unit formula for starting material A),  $(R^1_2R^2SiO_{1/2})_d(R^1_3SiO_{1/2})_e(R^1R^2SiO_{2/2})_f(R^1_2SiO_{2/2})_g$ , when  $R^1$  is methyl and  $R^2$  is vinyl, subscript d = 2, subscript e = 0, subscript f = 25, and subscript g = 4829, then starting material A) is a dimethylvinylsiloxo-terminated poly(dimethylsiloxane/methylvinylsiloxane) copolymer ( $M^{Vi}_2D^{Vi}_{25}D_{4829}$ ) with number average molecular weight ( $M_n$ ) and alkenyl (vinyl) content calculated as follows:

- **Number average molecular weight of the copolymer; 360,429.89 gf/mol**

If  $M_w$  of  $M^{Vi} = 93.20$  g/mol,  $M_w$  of D = 74.15 g/mol and  $M_w$  of  $D^{Vi} = 86.17$  g/mol

$$\rightarrow \text{Mn of } M^{\text{Vi}} D_{4829}^{\text{Vi}} M^{\text{Vi}} = 2*93.2 + 4829*74.15 + 25*86.17 = 360,429.89 \text{ g/mol}$$

- **The vinyl content = 0.202 %**

If Mw of  $M^{\text{Vi}}$  = 93.20 g/mol, Mw of D = 74.15 g/mol, Mw of  $D^{\text{Vi}}$  = 86.17 g/mol, and Vinyl of Mw = 27 are,

$$\rightarrow \text{Mn of } M^{\text{Vi}} D_{4829}^{\text{Vi}} M^{\text{Vi}} = (27*27)/(2*93.2 + 4829*74.15 + 25*86.17) \times 100 \% =$$

0.2022 %

**[0013]** Starting material A) is exemplified by

- i) dimethylvinylsiloxy-terminated poly(dimethylsiloxane/methylvinylsiloxane),
- ii) dimethylvinylsiloxy-terminated polymethylvinylsiloxane,
- iii) trimethylsiloxy-terminated poly(dimethylsiloxane/methylvinylsiloxane),
- iv) trimethylsiloxy-terminated polymethylvinylsiloxane,
- v) dimethylvinylsiloxy-terminated poly(dimethylsiloxane/methylvinylsiloxane),
- vi) dimethylhexenylsiloxy-terminated poly(dimethylsiloxane/methylhexenylsiloxane),
- vii) dimethylhexenylsiloxy-terminated polymethylhexenylsiloxane,
- viii) trimethylsiloxy-terminated poly(dimethylsiloxane/methylhexenylsiloxane),
- ix) trimethylsiloxy-terminated polymethylhexenylsiloxane,
- x) dimethylvinylsiloxy-terminated poly(dimethylsiloxane/methylhexenylsiloxane)
- xi) a combination of two or more of i), ii), iii), iv), v), vi), vii), viii), ix), and x).

**[0014]** Starting material A) may be one poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer or a combination of two or more poly(dialkylsiloxane/alkylalkenylsiloxane) copolymers that vary in at least one property such as selection of alkyl group, selection of alkenyl group, molecular weight, and number of dialkylsiloxane units and alkylalkenylsiloxane units. Methods of preparing poly(dialkylsiloxane/alkylalkenylsiloxane) suitable for use as starting material A) such as hydrolysis and condensation of the corresponding organohalosilanes or equilibration of cyclic polydiorganosiloxanes, are well known in the art.

#### Starting Material B) Polyalkylsiloxane Resin

**[0015]** The pressure sensitive adhesive composition may optionally further comprise starting material B) a polyalkylsiloxane resin. The polyalkylsiloxane resin is an MQ resin consisting essentially of  $R^3_3\text{SiO}_{1/2}$  (M) units and  $\text{SiO}_{4/2}$  (Q) units, where each  $R^3$  is independently selected from the group consisting of an alkyl group of 1 to 10 carbon atoms such as that described for  $R^1$ , and an alkenyl group of 2 to 6 carbon atoms such as that described above for  $R^2$ . Starting material B) may be free of silicon bonded aromatic hydrocarbon groups.

[0016] When prepared, the polyorganosilicate resin comprises the M and Q units described above, and the polyorganosilicate resin further comprises units with silanol (silicon bonded hydroxyl) groups and may comprise neopentamer of formula  $\text{Si}(\text{OSiR}^3)_4$ , where  $\text{R}^3$  is as described above.  $\text{Si}^{29}$  Nuclear Magnetic Resonance (NMR) spectroscopy, as described in U.S. Patent 9,593,209 at col. 32, Reference Example 2, may be used to measure molar ratio of M and Q units, where said ratio is expressed as

$\{\text{M}(\text{resin}) + \text{M}(\text{neopentamer})\} / \{\text{Q}(\text{resin}) + \text{Q}(\text{neopentamer})\}$  and represents the molar ratio of the total number of triorganosiloxy groups (M units) of the resinous and neopentamer portions of the polyorganosilicate resin to the total number of silicate groups (Q units) in the resinous and neopentamer portions (M:Q ratio). M:Q ratio may be 0.5:1 to 1.5:1, alternatively 0.6:1 to 1.2:1.

[0017] The  $M_n$  of the polyorganosilicate resin depends on various factors including the types of hydrocarbon groups represented by  $\text{R}^3$  that are present. The  $M_n$  of the polyorganosilicate resin refers to the number average molecular weight measured using gel permeation chromatography (GPC) according to the procedure in U.S. Patent 9,593,209 at col. 31, Reference Example 1, when the peak representing the neopentamer is excluded from the measurement. The  $M_n$  of the polyorganosilicate resin may be greater than 2,000 g/mol, alternatively > 2,000 g/mol to 10,000 g/mol, and alternatively 3,000 g/mol to 8,000 g/mol. Alternatively,  $M_n$  of the polyorganosilicate resin may be 4,000 g/mol to 7,000 g/mol.

[0018] U.S. Patent 8,580,073 at col. 3, line 5 to col. 4, line 31, is hereby incorporated by reference for disclosing silicone resins, which are suitable polyorganosilicate resins for use herein. The polyorganosilicate resin can be prepared by any suitable method, such as cohydrolysis of the corresponding silanes or by silica hydrosol capping methods. The polyorganosilicate resin may be prepared by silica hydrosol capping processes such as those disclosed in U.S. Patent 2,676,182 to Daudt, et al.; U.S. Patent 4,611,042 to Rivers-Farrell et al.; and U.S. Patent 4,774,310 to Butler, et al. The method of Daudt, et al. described above involves reacting a silica hydrosol under acidic conditions with a hydrolyzable triorganosilane such as trimethylchlorosilane, a siloxane such as hexamethyldisiloxane, or mixtures thereof, and recovering a copolymer having M units and Q units. The resulting copolymers generally contain from 2 to 5 percent by weight of silicon bonded hydroxyl (silanol) groups.

[0019] The intermediates used to prepare the polyorganosilicate resin may be triorganosilanes and silanes with four hydrolyzable substituents or alkali metal silicates. The triorganosilanes may have formula  $\text{R}^3_3\text{SiX}^1$ , where  $\text{R}^3$  is as described above and  $\text{X}^1$  represents a hydrolyzable substituent such as halogen, alkoxy, acyloxy, hydroxyl, oximo, or ketoximo; alternatively, halogen, alkoxy or hydroxyl. Silanes with four hydrolyzable substituents may have formula

$\text{SiX}_2\text{O}_4$ , where each  $\text{X}^2$  is halogen, alkoxy or hydroxyl. Suitable alkali metal silicates include sodium silicate.

**[0020]** The polyorganosilicate resin prepared as described above typically contains silicon bonded hydroxyl groups, e.g., in units of formulae,  $\text{HOSi}_{3/2}$  and/or  $\text{HOR}^3\text{SiO}_{1/2}$ . The polyorganosilicate resin may comprise up to 2% of silicon bonded hydroxyl groups. The concentration of silicon bonded hydroxyl groups present in the polyorganosilicate resin may be determined using Fourier Transform-Infra Red (FTIR) spectroscopy according to ASTM Standard E-168-16. For certain applications, it may be desirable for silicon bonded hydroxyl groups to be below 0.7%, alternatively below 0.3%, alternatively less than 1%, and alternatively 0.3% to 0.8%. Silicon bonded hydroxyl groups formed during preparation of the polyorganosilicate resin can be converted to trihydrocarbyl-siloxy groups or to a different hydrolyzable group by reacting the polyorganosilicate resin with a silane, disiloxane, or disilazane containing the appropriate terminal group. Silanes containing hydrolyzable groups may be added in molar excess of the quantity required to react with the silicon bonded hydroxyl groups on the polyorganosilicate resin.

**[0021]** Alternatively, the polyorganosilicate resin may further comprises 2% or less, alternatively 0.7% or less, and alternatively 0.3% or less, and alternatively 0.3% to 0.8% of units represented by formula  $\text{XSiO}_{3/2}$  and/or  $\text{NXR}^3\text{SiO}_{1/2}$  where  $\text{R}^3$  is as described above, and X represents a hydrolyzable substituent, as described above for  $\text{X}^1$ .

**[0022]** Alternatively, the polyorganosilicate resin may have terminal aliphatically unsaturated groups. The polyorganosilicate resin having terminal aliphatically unsaturated groups may be prepared by reacting the product of Daudt, *et al.* with an unsaturated organic group-containing endblocking agent and an endblocking agent free of aliphatic unsaturation, in an amount sufficient to provide from 3 to 30 mole percent of unsaturated organic groups in the final product. Examples of endblocking agents include, but are not limited to, silazanes, siloxanes, and silanes. Suitable endblocking agents are known in the art and exemplified in U.S. Patents 4,584,355; 4,591,622; and 4,585,836. A single endblocking agent or a mixture of such agents may be used to prepare such resin.

**[0023]** Alternatively, the polyalkylsiloxane resin may comprise unit formula (II):

$(\text{R}^1\text{R}^2\text{SiO}_{1/2})_a(\text{R}^1\text{SiO}_{1/2})_b(\text{SiO}_{4/2})_c$ , where  $\text{R}^1$  and  $\text{R}^2$  are as described above, subscripts a, b, and c represent mole fractions with values such that subscript  $a \geq 0$ , subscript  $b \geq 0$ , subscript  $c > 0$ , a quantity  $(a+b) > 0$ , and subscripts a, b, and c have values such that  $0.9 \leq (a+b)/c \leq 1.3$ .

**[0024]** The amount of starting material B) in the pressure sensitive adhesive composition depends on various factors including the desired form of the reaction product of the composition,

the quantity and hydrosilylation reactivity of the alkenyl groups of starting material A), the type and amount of catalyst, and the content of silicon bonded hydrogen atoms of starting material C). However, the amount of starting material B) may range from 0 to 15 parts by weight, per 100 parts by weight of starting material A); alternatively > 0 to 15 parts by weight on the same basis.

Starting Material C) Polyalkylhydrogensiloxane

**[0025]** The pressure sensitive adhesive composition further comprises starting material C) a polyalkylhydrogensiloxane. The polyalkylhydrogensiloxane has unit formula (III):

$(R^1_2HSiO_{1/2})_2(R^1_2SiO_{2/2})_x(R^1HSiO_{2/2})_y$ , where  $R^1$  is as described above.

**[0026]** In unit formula (III), subscript  $x \geq 0$ , subscript  $y \geq 0$ , and a quantity  $(x + y)$  is sufficient to give the polyalkylhydrogensiloxane a viscosity of < 20 mPa·s, alternatively 8 mPa·s to 16 mPa·s, where viscosity is measured by capillary viscometer at 25 deg C. Subscript x may be sufficient to give starting material C), the polyalkylhydrogensiloxane, an SiH content > 0 to < 1%, alternatively > 0 to 0.75%, based on weight of starting material C).

**[0027]** Polyalkylhydrogensiloxanes for starting material C) are exemplified by:

- a) dimethylhydrogensiloxy-terminated polydimethylsiloxane,
- b) dimethylhydrogensiloxy-terminated poly(dimethylsiloxane/methylhydrogensiloxane),
- c) dimethylhydrogensiloxy-terminated polymethylhydrogensiloxane, and
- d) a combination of two or more of a), b), and c).

**[0028]** Methods of preparing polyalkylhydrogensiloxanes suitable for use as starting material C), such as hydrolysis and condensation of alkylhalosilanes, are well known in the art.

**[0029]** The exact amount of starting material C) in the composition depends on various factors including reactivity of starting material A), whether starting material B) is present, and the type and amount of any additional starting materials (*e.g.*, starting material D)), if present. However, the amount of starting material C) in the pressure sensitive adhesive composition may be 0.1% to 5%, alternatively 0.5% to 3%, and alternatively 0.5% to 2% based on combined weights of all starting materials in the composition.

**[0030]** The alkenyl content of starting material A) and any other starting materials in the composition that contain silicon bonded alkenyl groups (*e.g.*, starting material B) and starting material C), when alkenyl functional) and the SiH content of starting material B) and any other SiH containing materials in the pressure sensitive adhesive composition (*e.g.*, starting material D) the adhesion promoter, when SiH functional) are sufficient to provide the pressure sensitive composition with a molar ratio of silicon bonded hydrogen atoms to silicon bonded alkenyl groups (SiH:Alkenyl) ratio of at least 0.5:1, alternatively up to 2:1, alternatively, 0.5:1 to 2:1, alternatively 1:1 to 1.5:1, and alternatively >1 to 1.2.

Starting Material D) – Anchorage Additive

[0031] Starting material D) is an anchorage additive that may optionally be included in the pressure sensitive adhesive composition. Without wishing to be bound by theory, it is thought that the anchorage additive will facilitate bonding to a substrate by a pressure sensitive adhesive prepared by curing the pressure sensitive adhesive composition described herein. However, the presence of the anchorage additive will not detrimentally affect the desired peel force allowing the pressure sensitive adhesive to be removed from an electronic device without damaging the device or leaving significant residue.

[0032] Suitable anchorage additives include silane coupling agents such as methyltrimethoxysilane, vinyltrimethoxysilane, allyltrimethoxysilane, 3-methacryloxypropyltrimethoxysilane, 3-aminopropyltrimethoxysilane, N-(2-aminoethyl)-3-aminopropyltrimethoxysilane, bis(trimethoxysilyl)propane, and bis(trimethoxysilyl)hexane; and mixtures or reaction mixtures of said silane coupling agents. Alternatively, the anchorage additive may be tetramethoxysilane, tetraethoxysilane, dimethyldimethoxysilane, methylphenyldimethoxysilane, methylphenyldiethoxysilane, phenyltrimethoxysilane, methyltrimethoxysilane, methyltriethoxysilane, vinyltriethoxysilane, allyltriethoxysilane, 3-glycidoxypropyltrimethoxysilane, 3-glycidoxypropyltriethoxysilane, -(3,4-epoxycyclohexyl)ethyltrimethoxysilane or 3-methacryloxypropyl trimethoxysilane.

[0033] Alternatively, the anchorage additive may be exemplified by a reaction product of an alkenyl-functional alkoxysilane (e.g., a vinyl alkoxysilane) and an epoxy-functional alkoxysilane; a reaction product of an alkenyl-functional acetoxysilane (such as vinyl acetoxysilane) and epoxy-functional alkoxysilane; and a combination (e.g., physical blend and/or a reaction product) of a polyorganosiloxane having at least one aliphatically unsaturated hydrocarbon group and at least one hydrolyzable group per molecule and an epoxy-functional alkoxysilane (e.g., a combination of a hydroxy-terminated, vinyl functional polydimethylsiloxane with glycidoxypropyltrimethoxysilane). Suitable anchorage additives and methods for their preparation are disclosed, for example, in U.S. Patent 9,562,149; U.S. Patent Application Publication Numbers 2003/0088042, 2004/0254274, 2005/0038188, 2012/0328863 at paragraph [0091], and U.S. Patent Publication 2017/0233612 at paragraph [0041]; and EP 0 556 023.

[0034] Anchorage additives are commercially available. For example, SYL-OFF™ 297, SYL-OFF™ 397, and SYL-OFF™ 9176 are available from Dow Silicones Corporation of Midland, Michigan, USA. Other exemplary anchorage additives include D-1) vinyltriacetoxysilane, D-2) glycidoxypropyltrimethoxysilane, D-3) 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane, D-4) a combination of two or more of D-1), D-2) and

D-3); and D-5) a combination of any of D-1), D-2), D-3) and/or D-4), and a polydimethylsiloxane terminated with hydroxyl groups, methoxy groups, or terminated with both a hydroxy group and a methoxy group. The combinations D-4) and D-5) may be physical blends and/or reaction products.

**[0035]** The amount of anchorage additive depends on various factors including the type of substrate to which the composition will be applied and whether a primer or other surface treatment will be used before application of the composition. However, the amount of anchorage additive may be 0 to 5%, alternatively 1% to 5%, alternatively 1% to 3%, and alternatively 0 to 1%, based on combined weights of all starting materials in the composition. The anchorage additive may be free of silicon bonded aromatic hydrocarbon groups.

#### Starting Material E) Inhibitor

**[0036]** The pressure sensitive adhesive composition further comprises starting material E) an acetylenic alcohol. The acetylenic alcohol may be added as an inhibitor or stabilizer for the pressure sensitive adhesive composition. Suitable acetylenic alcohols are exemplified by methyl butynol, ethynyl cyclohexanol (ETCH), dimethyl hexynol, and 3,5-dimethyl-1-hexyn-3-ol, 1-butyn-3-ol, 1-propyn-3-ol, 2-methyl-3-butyn-2-ol, 3-methyl-1-butyn-3-ol, 3-methyl-1-pentyn-3-ol, 3-phenyl-1-butyn-3-ol, 4-ethyl-1-octyn-3-ol, 3,5-dimethyl-1-hexyn-3-ol, and 1-ethynyl-1-cyclohexanol, and a combination thereof. Alternatively, starting material E) may be selected from the group consisting of (E-1) 1-ethynyl-1-cyclohexanol, (E-2) methyl butynol, (E-3) diallyl maleate, and (E-4) a combination of two or more of (E-1), (E-2) and (E-3). Alternatively, starting material E) is exemplified by ETCH.

**[0037]** The amount of acetylenic alcohol added to the composition will depend on various factors including the desired pot life of the pressure sensitive adhesive composition, whether the composition will be a one part composition or a multiple part composition, the particular acetylenic alcohol used, and the selection and amount of starting material C). However, when present, the amount of starting material E) may range from 0.01 to 0.5 parts by weight.

#### Starting Material F) Hydrosilylation Reaction Catalyst

**[0038]** The pressure sensitive adhesive composition further comprises starting material F) a hydrosilylation reaction catalyst. Suitable hydrosilylation reaction catalysts are known in the art and are commercially available. The hydrosilylation catalysts can be a platinum group metal selected from platinum, rhodium, ruthenium, palladium, osmium, and iridium. Alternatively, the hydrosilylation catalyst may be a compound of such a metal, for example, chloroplatinic acid, chloroplatinic acid hexahydrate, platinum dichloride, and complexes of said compounds with low molecular weight organopolysiloxanes or platinum compounds microencapsulated in a matrix or core/shell type structure. Complexes of platinum with low molecular weight

organopolysiloxanes include 1,3-diethenyl-1,1,3,3 -tetramethyldisiloxane complexes with platinum. These complexes may be microencapsulated in a resin matrix. Exemplary hydrosilylation catalysts are described in U.S. Patents 3,159,601; 3,220,972; 3,296,291; 3,419,593; 3,516,946; 3,814,730; 3,989,668; 4,784,879; 5,036,117; and 5,175,325 and EP 0 347 895 B. Microencapsulated hydrosilylation catalysts and methods of preparing them are known in the art, as exemplified in U.S. Patents 4,766,176 and 5,017,654.

[0039] The hydrosilylation reaction catalyst is added to the pressure sensitive adhesive composition in an amount sufficient to catalyze hydrosilylation reaction, typically an amount sufficient to provide 10 ppm to 5,000 ppm by weight of platinum group metal.

#### Starting Material G) Solvent

[0040] The pressure sensitive adhesive composition further comprises a solvent. The solvent can be an organic solvent. The organic solvent can be an alcohol such as methanol, ethanol, isopropanol, butanol, or n-propanol; a ketone such as acetone, methylethyl ketone, or methyl isobutyl ketone; an aromatic hydrocarbon such as benzene, toluene, or xylene; an aliphatic hydrocarbon such as heptane, hexane, or octane; a glycol ether such as propylene glycol methyl ether, dipropylene glycol methyl ether, propylene glycol n-butyl ether, propylene glycol n-propyl ether, or ethylene glycol n-butyl ether, a halogenated hydrocarbon such as dichloromethane, 1,1,1-trichloroethane or methylene chloride; chloroform; dimethyl sulfoxide; dimethyl formamide, acetonitrile; tetrahydrofuran; white spirits; mineral spirits; naphtha; n-methyl pyrrolidone; or a combination thereof. Alternatively, the solvent may be selected from the group consisting of benzene, toluene, heptane, ethylbenzene, xylene, and a combination of two or more thereof.

[0041] The amount of solvent will depend on various factors including the type of solvent selected and the amount and type of other starting materials selected for the pressure sensitive adhesive composition. However, the amount of solvent may range from 10% to 90%, alternatively 20% to 60%, based on combined weights of all starting materials in the pressure sensitive adhesive composition. The solvent can be added during preparation of the pressure sensitive adhesive composition, for example, to aid mixing and delivery. All or a portion of the solvent may be added with one of the other starting materials. For example, starting material A), and when present starting material B), may be dissolved in a solvent, such as an aromatic hydrocarbon, before combination with the other starting materials in the composition. All or a portion of the solvent may optionally be removed after the pressure sensitive adhesive composition is prepared.

#### Starting Material H) Additional Polyalkylhydrogensiloxane

[0042] The pressure sensitive adhesive composition may optionally further comprise up to 20%, based on weight of starting material C), of starting material H) an additional polyalkylhydrogensiloxane having unit formula  $(R^1)_3SiO_{1/2})_2(R^1HSiO_{2/2})_z$ , where  $R^1$  is as described above, and subscript  $z$  is 200 to 250. Without wishing to be bound by theory, it is thought that starting material H) may be used to adjust SiH: Alkenyl ratio to the ranges described herein.

[0043] Methods of preparing polyalkylhydrogensiloxanes suitable for use as starting material H), such as hydrolysis and condensation of alkylhalosilanes, are well known in the art. Examples of suitable additional polyalkylhydrogensiloxanes for starting material H) include H-1) trimethylsiloxy-terminated poly(methylhydrogensiloxane), H-2) trimethylsiloxy-terminated poly(dimethylsiloxane/methylhydrogensiloxane), and H-3) a combination of both H-1) and H-2).

[0044] When selecting starting materials for the pressure sensitive adhesive composition described above, there may be overlap between types of starting materials because certain starting materials described herein may have more than one function. For example, certain alkoxysilanes may be useful as filler treating agents and as adhesion promoters. Certain particulates may be useful as fillers and as pigments, and even as flame retardants, *e.g.*, carbon black. When adding additional starting materials to the composition, the additional starting materials are distinct from one another and from the required starting materials in the composition.

[0045] The pressure sensitive adhesive composition can be prepared by a method comprising combining all starting materials by any convenient means such as mixing at ambient or elevated temperature. Starting material E) the acetylenic alcohol may be added before starting material F) the platinum group metal catalyst, for example, when the pressure sensitive adhesive composition will be prepared at elevated temperature and/or the composition will be prepared as a one part composition.

[0046] Alternatively, the pressure sensitive composition may be prepared as a multiple part composition, for example, when the composition will be stored for a long period of time before use. In the multiple part composition, starting material F) the platinum group metal catalyst is stored in a separate part from any starting material having a silicon bonded hydrogen atom, for example starting material C) the polyalkylhydrogensiloxane and starting material H) (when present) and the parts are combined shortly before use of the pressure sensitive adhesive composition. For example, a two part composition may be prepared by combining starting materials comprising A) the alkenyl-containing polydialkylsiloxane, C) the polyalkylhydrogensiloxane (and starting material H), when present) and G) the organic solvent,

and optionally one or more other additional starting materials described above to form a base part, by any convenient means such as mixing. A curing agent may be prepared by combining starting materials comprising A) the alkenyl-containing polydialkylsiloxane, F) the platinum group metal catalyst and G) the organic solvent, and optionally one or more other additional starting materials described above by any convenient means such as mixing. The starting materials may be combined at ambient or elevated temperature. Starting material E) the acetylenic alcohol may be included in one or more of the base part, the curing agent part, or a separate additional part. Starting material D) anchorage additive, when used, may be added to the base part, or may be added as a separate additional part. Starting material B) the polyalkylsiloxane resin, when used, may be added to the base part, the curing agent part, or a separate additional part. When a two part composition is used, the weight ratio of amounts of base part to curing agent part may range from 1:1 to 10:1. The pressure sensitive adhesive composition will cure via hydrosilylation reaction to form a pressure sensitive adhesive.

**[0047]** The method described above may further comprise one or more additional steps. The pressure sensitive adhesive composition prepared as described above may be used to form an adhesive article, *e.g.*, a pressure sensitive adhesive (prepared by curing the pressure sensitive adhesive composition described above) on a substrate. The method described above may, therefore, further comprise applying the pressure sensitive adhesive composition to a substrate.

**[0048]** Applying the pressure sensitive adhesive curable composition to the substrate can be performed by any convenient means. For example, the pressure sensitive adhesive curable composition may be applied onto a substrate by gravure coater, offset coater, offset-gravure coater, roller coater, reverse-roller coater, air-knife coater, or curtain coater.

**[0049]** The substrate can be any material that can withstand the curing conditions (described below) used to cure the pressure sensitive adhesive curable composition to form the pressure sensitive adhesive on the substrate. For example, any substrate that can withstand heat treatment at a temperature equal to or greater than 120°C, alternatively 150°C is suitable. Examples of materials suitable for such substrates including plastic films such as polyimide (PI), polyetheretherketone (PEEK), polyethylene naphthalate (PEN), liquid-crystal polyarylate, polyamideimide (PAI), polyether sulfide (PES), or polyethylene terephthalate (PET), or PE (polyethylene), or PP (polypropylene). Alternatively, the substrate may be a metal foil such as aluminum foil or copper foil. The thickness of the substrate is not critical, however, the thickness may range from 5 micrometers to 300 micrometers.

**[0050]** To improve bonding of the pressure sensitive adhesive to the substrate, the method may optionally further comprise treating the substrate before applying the pressure sensitive

adhesive composition. Treating the substrate may be performed by any convenient means, such as applying a primer, or subjecting the substrate to corona-discharge treatment, etching, or plasma treatment before applying the pressure sensitive adhesive composition to the substrate.

**[0051]** An adhesive article such as a protective film may be prepared by applying the pressure sensitive adhesive composition described above onto the substrate described above. The method may optionally further comprise removing all, or a portion, of the solvent before and/or during curing. Removing solvent may be performed by any convenient means, such as heating at a temperature that vaporizes the solvent without fully curing the pressure sensitive adhesive composition, *e.g.*, heating at a temperature of 70°C to 120°C, alternatively 50°C to 100°C, and alternatively 70°C to 80°C for a time sufficient to remove all or a portion of the solvent (*e.g.*, 30 seconds to 1 hour, alternatively 1 minute to 5 minutes). The method then further comprises curing the pressure sensitive adhesive composition (which may have some or all of the solvent removed when the drying step is performed) room temperature or by heating at a temperature of 140°C to 220°C, alternatively 150°C to 220°C, alternatively 160°C to 200°C, and alternatively 165°C to 180°C for a time sufficient to cure the pressure sensitive adhesive curable composition (*e.g.*, for 30 seconds to an hour, alternatively 1 to 5 minutes). This forms a pressure sensitive adhesive on the substrate. Drying and/or curing may be performed by placing the substrate in an oven. The amount of the pressure sensitive adhesive composition to be applied to the substrate depends on the specific application, however, the amount may be sufficient such that after curing thickness of the pressure sensitive adhesive may be 5 micrometers to 200 micrometers, and for protective film the thickness may be 10 micrometers to 50 micrometers, alternatively 20 micrometers to 40 micrometers, and alternatively 30 micrometers.

**[0052]** The method described herein may optionally further comprise applying a removable release liner to the pressure sensitive adhesive opposite the substrate, *e.g.*, to protect the pressure sensitive adhesive before use of the adhesive article.

**[0053]** The adhesive article (*e.g.*, protective film) prepared as described above is suitable for use in flexible OLED device fabrication processes as a protective film with low adhesion, high adhesion stability, and/or low migration.

**[0054]** For example, a method for fabricating a flexible OLED device may include forming an OLED module on a surface of a substrate, *e.g.*, a passivation layer on a surface of the OLED module opposite the substrate, and applying a protective film prepared as described herein to a surface of the passivation layer opposite the OLED module.

## EXAMPLES

[0055] These examples are intended to illustrate some embodiments of the invention and should not be interpreted as limiting the scope of the invention set forth in the claims. Starting materials used in these examples are described in Table 1.

Table 1 – Starting Materials Used in the Examples

| Starting Material | Chemical Description and Source                                                                                                                                                                                                                                                | Vinyl Content (wt%) | SiH Content (wt%) |
|-------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------|-------------------|
| A1                | A dimethylvinylsiloxo-terminated poly(dimethylsiloxane/methylvinylsiloxane) copolymer with average unit formula: $(\text{ViMe}_2\text{SiO}_{1/2})_2(\text{MeViSiO}_{2/2})_{25}(\text{Me}_2\text{SiO}_{2/2})_{4829}$ , where Me is methyl and Vi from Dow Silicones Corporation | 0.202               | None              |
| G1                | Toluene                                                                                                                                                                                                                                                                        | N/A                 | N/A               |
| H1                | $\text{Me}_3\text{SiO}(\text{HMeSiO})_x\text{SiMe}_3$ , where x is 35; having a viscosity of 20 mPa·s measured by capillary viscometer at 25 deg C from Dow Silicones Corporation                                                                                              | None                | 1.6               |
| H2                | $\text{Me}_3\text{SiO}(\text{HMeSiO})_x\text{SiMe}_3$ , where x is 223; having a viscosity of 230 mPa·s measured by capillary viscometer at 25 deg C from Dow Silicones Corporation                                                                                            | None                | 1.6               |
| H3                | $\text{Me}_3\text{SiO}(\text{HMeSiO})_x(\text{Me}_2\text{SiO})_y\text{SiMe}_3$ , where $(x + y) = 11$ ; having a viscosity of 33 mPa·s measured by capillary viscometer at 25 deg C from Dow Silicones Corporation, DOWSIL™ 6-3570 Polymer                                     | None                | 0.76              |
| H4                | $\text{Me}_3\text{SiO}(\text{HMeSiO})_x(\text{Me}_2\text{SiO})_y\text{SiMe}_3$ , where $(x + y) = 57$ ; having a viscosity of 31 mPa·s measured by capillary viscometer at 25 deg C from Dow Silicones Corporation                                                             | None                | 1.05              |
| C1                | $\text{HMe}_2\text{SiO}(\text{HMeSiO})_x(\text{Me}_2\text{SiO})_y\text{SiMe}_2\text{H}$ , where x and y are sufficient to give an SiH content of 0.43% and viscosity of 12.3 mPa·s measured by capillary viscometer at 25 deg C from Dow Silicones Corporation                 | None                | 0.43              |
| E1                | Ethynyl cyclohexanol from BASF                                                                                                                                                                                                                                                 | N/A                 | N/A               |
| F1                | Karstedt's catalyst, SYL-OFF™ 4000 Catalyst                                                                                                                                                                                                                                    | Not tested          | N/A               |
| D1                | Anchorage additive containing 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane, SYL-OFF™ 9176                                                                                                                                                                                      | N/A                 | N/A               |

[0056] In the table above, 'N/A' means not applicable, and 'Me' means methyl. DOWSIL™, SILASTIC™, and SYL-OFF™ products are commercially available from Dow Silicones Corporation of Midland, Michigan, USA.

## Reference Example 1 – Preparation of Pressure Sensitive Adhesive Composition Samples

[0057] Pressure sensitive adhesive composition samples were prepared by combining the starting materials shown in Table 1. First, 100 parts by weight of starting material A1)

dimethylvinylsiloxyl-terminated poly(dimethylsiloxane/methylvinylsiloxane) copolymer was mixed with starting material G1) toluene solvent to make a 25 weight % solution. A polyalkylhydrogensiloxane (one or more of H1) to H4) and C1)) was added to the mixture in amounts shown below in Table 2 and Table 3. The amount of polyalkylhydrogensiloxane was sufficient to make the SiH/Vi molar ratio in the mixture 0.5, 1 or 2. This ratio included vinyl present in the vinyl containing siloxane complexed platinum catalyst F1), which was added at the end of this procedure. An inhibitor E1) was added, and the resulting combination was mixed well. After homogenizing the mixture, starting material (F1) was added to the mixture. The mixture was immediately mixed again carefully not to generate heat. The resulting pressure sensitive adhesive composition sample was then ready for use. The amount of each starting material (in weight parts) in the samples are shown in Table 2 (comparative) and Table 3 (Working), below. Tables 2 and 3 also show SiH/Vi ratio, total Pt content, and % solids (based on weight of all starting materials excluding solvent).

#### Reference Example 2 – Preparation of a PSA Sheet

**[0058]** The pressure sensitive adhesive composition samples were prepared as described in Reference Example 1 were applied onto a 50 um polyethylene terephthalate (PET) film to form a pressure-sensitive adhesive layer (which after curing had a thickness of 30 um). A pressure-sensitive adhesive sheet was produced by heating the coated film for 2 min. at 150°C. The substrate film (PET) was commercially available; sold under the trade name of Toray Advanced Material Korea (Trade name SH-86, TAMK).

#### Reference Example 3 - Initial Adhesion measurement

**[0059]** Pressure sensitive adhesive tapes were formed by cutting the sheet obtained according to Reference Example 2 into 1 inch-wide strips. Adhesion strength to a stainless-steel plate (SUS304) was measured according to a 180-degree peel test specified in ASTM D1000. Each tape was peeled from the peelable polyethylene terephthalate films and adhered under pressure of 2 kgf developed by rubber rollers to an adherend in the form of a mirror-surface stainless steel plate (SUS304). Initial adhesion higher than 2 gf/inch was defined as Fail.

#### Reference Example 4 - Adhesion at 85°C/85% (Adhesion Stability)

**[0060]** Pressure sensitive adhesive tapes were formed by cutting the sheet obtained according to Reference Example 2 into 1 inch-wide strips. Adhesion strength to a stainless steel plate (SUS304) was measured according to a 180-degree peel test specified in the ASTM D1000 after aging, as follows. Each tape was peeled from the peelable polyethylene terephthalate films and adhered under pressure of 2 kgf developed by rubber rollers to an adherend in the form of a mirror-surface stainless steel plate (SUS304). The tapes were aged for 1 day (24hr) and 3 days (72hr) in an oven at 85°C /85% moisture. After cooling to room temperature, the adhesion force

was measured by pulling the pressure-sensitive adhesive tape off from the adherend at an angle of 180° relative to the surface of the adherend with the use of a tensile tester at a constant speed of 12inch/min. (Test equipment; AR-1000 (Cheminstrument Company)). Results are shown in Tables 2 and 3, below. For some applications, it may be desirable for adhesion after 24 h to be < 9 gf/in, and < 15 gf/ in after 72 h.

**Reference Example 5 - Simple scratch test (burr test)**

**[0061]** A pressure sensitive adhesive sheet was prepared according to Reference Example 2. A cut was made on the pressure sensitive adhesive surface by a cutter, and the cross section was observed by naked eye and microscope. If edge of cut looks clean and no particle was generated, the result was pass. If a particle was generated and observed in the edge, or a burr was shown in the edge, the result was Fail.

**Reference Example 6 - Simple Rub-off test (anchorage test)**

**[0062]** A pressure sensitive adhesive coated on PET was prepared according to Reference Example 2, and anchorage of the pressure sensitive adhesive was evaluated as follows. A cut was made: cross-mark 10cmX10cm only on PSA by cutter (being careful not to cut PET on bottom). The marked surface was rubbed vigorously by finger. After rubbing 30 sec., the surface was observed to determine whether the PSA was rubbed off or not. If PSA was rubbed off, the result was Fail.

**Reference Example 7 - Residual Adhesive-Remaining Property and Silicone-Transfer Property after exposure to 85°C/85% condition (Residual Adhesion)**

**[0063]** Pressure sensitive adhesive tapes were formed by cutting the sheet obtained according to Reference Example 2 into 1 inch-wide strips. Each tape was peeled from the peelable polyethylene terephthalate films and adhered under pressure of 2 kgf developed by rubber rollers to an adherend in the form of a mirror-surface stainless steel plate (SUS304). They are aged for 1 day (24hrs) and 3 days (72hrs) in an oven at 85°C/85%. After cooling to room temperature, PSA tapes were removed from a mirror-surface stainless steel plate. Adhere Nitto 31B by 2KG roller on the place that Si tape removed. After 30 minutes, the adhesion force was measured by pulling the pressure sensitive adhesive tape off from the adherend at an angle of 180° relative to the surface of the adherend with the use of a tensile tester at a constant speed of 12 inch/min. Results for each sample are also shown below in Tables 2 and 3. 'AF' in the tables denotes adhesive failure and is undesirable for some applications.

**Reference Example 8 – Burr Test**

**[0064]** A pressure sensitive adhesive sheet sample prepared according to Reference Example 2 was used. A cutter was used to make a cut on the pressure sensitive adhesive surface, which was visually observed by naked eye and microscope. If the edge of the cut looked clean and no

particle was generated, the sample passed. If a particle or fragment was generated in the edge or a burr was observed, the sample failed. Results for each sample are also shown below in Tables 2 and 3.

Table 2

| Start Material               | 1         | 2         | 3         | 4         | 5         | 6         | 7         | 8         | 9         | 10        | 11        | 12        | 21        |
|------------------------------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| A1                           | 10<br>0   | 10<br>0   | 10<br>0   | 10<br>0   | 10<br>0   | 10<br>0   | 10<br>0   | 10<br>0   | 10<br>0   | 10<br>0   | 10<br>0   | 10<br>0   | 10<br>0   |
| H1                           | 0.2<br>8  | 0.5<br>5  | 1.1<br>1  | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         |
| H2                           | 0         | 0         | 0         | 0.2<br>8  | 0.5<br>5  | 1.1<br>1  | 0         | 0         | 0         | 0         | 0         | 0         | 0         |
| H3                           | 0         | 0         | 0         | 0         | 0         | 0         | 0.5<br>7  | 1.1<br>4  | 2.2<br>8  | 0         | 0         | 0         | 0         |
| H4                           | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0.4<br>2  | 0.8<br>4  | 1.6<br>9  | 0         |
| C1                           | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 2.4<br>97 |
| E1                           | 0.7       | 0.7       | 0.7       | 0.7       | 0.7       | 0.7       | 0.7       | 0.7       | 0.7       | 0.7       | 0.7       | 0.7       | 0.7       |
| F1                           | 2.0       | 2.0       | 2.0       | 2.0       | 2.0       | 2.0       | 2.0       | 2.0       | 2.0       | 2.0       | 2.0       | 2.0       | 2.0<br>56 |
| G1                           | 30<br>0   | 30<br>0   | 30<br>0   | 30<br>0   | 30<br>0   | 30<br>0   | 30<br>0   | 30<br>0   | 30<br>0   | 30<br>0   | 30<br>0   | 30<br>0   | 30<br>0   |
| D1                           | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 0         | 1.6<br>67 |
|                              |           |           |           |           |           |           |           |           |           |           |           |           |           |
| SiH/Vi                       | 0.5<br>0  | 1.0<br>0  | 2.0<br>0  | 0.5<br>0  | 1.0<br>0  | 2.0<br>0  | 0.5<br>0  | 1.0<br>0  | 2.0<br>0  | 0.5<br>0  | 1.0<br>0  | 2.0<br>0  | 1         |
| Pt ppm (total mixture based) | 25.<br>1  | 25.<br>1  | 25.<br>1  | 25.<br>1  | 25.<br>1  | 25.<br>1  | 25.<br>1  | 25.<br>1  | 25.<br>2  | 25.<br>1  | 25.<br>1  | 25.<br>1  | 25.<br>3  |
| Solid %                      | 26        | 26        | 26        | 26        | 26        | 26        | 26        | 26        | 26        | 26        | 26        | 26        | 26        |
|                              |           |           |           |           |           |           |           |           |           |           |           |           |           |
| Initial Adhesion Stability   | 2.6       | 2.0       | 1.5       | 7.4       | 1.8       | 1.6       | 1.4       | 1.2       | 1.4       | 2.2       | 1.0       | 0.9       | 0.6       |
| 24 h Adhesion Stability      | 14.<br>4  | 11.<br>1  | 7.7       | 59.<br>1  | 13.<br>4  | 10.<br>2  | 6.5       | 6.0       | 6.7       | 9.4       | 6.2       | 7.0       | 7.1       |
| 72 h Adhesion Stability      | 41.<br>0  | 30.<br>9  | 17.<br>9  | n/d       | 75.<br>5  | 43.<br>6  | n/d       | n/d       | n/d       | 14<br>5.0 | 30.<br>1  | 31.<br>5  | 13.<br>4  |
| 24 h Residual Adhesion       | 38<br>7.5 | 41<br>4.0 | 42<br>0.0 | 10<br>8.2 | 20<br>7.5 | 16<br>0.5 | 38<br>7.5 | 41<br>4.0 | 42<br>0.0 | 10<br>8.2 | 20<br>7.5 | 16<br>0.5 | 31<br>4   |
| 72 h Residual Adhesion       | n/d       | n/d       | n/d       | 18<br>5.0 | 11<br>6.0 | 12<br>1.0 | n/d       | n/d       | n/d       | 18<br>5.0 | 11<br>6.0 | 12<br>1.0 | 24<br>7.4 |
| Burr Test                    | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | Pas<br>s  |
| Initial Anchorage            | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  |
| 24 h Anchorage               | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  |
| 72 h                         | AF        | AF        | AF        | pas<br>s  | pas<br>s  | pas<br>s  | AF        | AF        | AF        | pas<br>s  | pas<br>s  | pas<br>s  | pas<br>s  |

Table 3

| Starting Material            | 13    | 14    | 15    | 16    | 17    | 18    | 19    | 20    |
|------------------------------|-------|-------|-------|-------|-------|-------|-------|-------|
| A1                           | 100   | 100   | 100   | 100   | 100   | 100   | 100   | 100   |
| H1                           | 0     | 0     | 0     | 0     | 0     | 0     | 0     | 0     |
| H2                           | 0     | 0     | 0     | 0.087 | 0.150 | 0.237 | 0     | 0     |
| H3                           | 0     | 0     | 0     | 0     | 0     | 0     | 0     | 0     |
| H4                           | 0     | 0     | 0     | 0     | 0     | 0     | 0     | 0     |
| C1                           | 1.02  | 2.05  | 4.10  | 1.733 | 1.500 | 1.183 | 2.140 | 2.317 |
| E1                           | 0.7   | 0.7   | 0.7   | 0.7   | 0.7   | 0.7   | 0.7   | 0.7   |
| F1                           | 2.0   | 2.0   | 2.0   | 2.012 | 2.013 | 2.014 | 2.017 | 2.022 |
| G1                           | 300   | 300   | 300   | 300   | 300   | 300   | 300   | 300   |
| D1                           | 0     | 0     | 0     | 0     | 0     | 0     | 0.333 | 1.000 |
|                              |       |       |       |       |       |       |       |       |
| SiH/Vi                       | 0.50  | 1.00  | 2.00  | 1     | 1     | 1     | 1     | 1     |
| Pt ppm (total mixture based) | 25.1  | 25.2  | 25.3  | 25.2  | 25.2  | 25.2  | 25.2  | 25.3  |
| Solid %                      | 26    | 26    | 26    | 26    | 26    | 26    | 26    | 26    |
|                              |       |       |       |       |       |       |       |       |
| Initial Adhesion Stability   | 1.3   | 0.9   | 0.7   | 1.2   | 1.1   | 1.0   | 0.6   | 0.6   |
| 24 h Adhesion Stability      | 7.7   | 5.2   | 4.5   | 5.9   | 5.9   | 13.5  | 3.4   | 5.0   |
| 72 h Adhesion Stability      | 8.7   | 6.4   | 7.3   | 7.4   | 8.1   | 23.2  | 7.4   | 7.8   |
| 24 h Residual Adhesion       | 339.0 | 406.5 | 388.5 | 387.8 | 385.3 | 345.9 | 397.0 | 355.0 |
| 72 h Residual Adhesion       | 293.0 | 351.0 | 342.0 | 307.6 | 301.9 | 132.5 | 324.9 | 312.0 |
| Burr Test                    | pass  | pass  | pass  | pass  | pass  | pass  | pass  | pass  |
| Initial Anchorage            | pass  | pass  | pass  | pass  | pass  | pass  | pass  | pass  |
| 24 h Anchorage               | pass  | pass  | pass  | pass  | pass  | pass  | pass  | pass  |
| 72 h                         | RF    | RF    | RF    | pass  | pass  | pass  | pass  | pass  |

**[0065]** In Tables 2 and 3, 'N/D' means Not detected due to adhesive transfer; 'AF' means Adhesive failure, i.e., that adhesive is transferred to substrate, 'RF' means Rubbing failure, i.e., that adhesive is rubbed off by stress, and 'RF' means result is 'not applicable' to invention.

Example 9 (Comparative)

**[0066]** Samples were prepared according to the procedure of Reference Example 1, except the composition contained 100 weight parts of starting material A2) a comparative dimethylvinylsiloxyl-terminated poly(dimethylsiloxane/methylvinylsiloxane) copolymer having both pendant and terminal vinyl groups but with a vinyl content = 0.41% and DP > 5,000, 300 weight parts toluene, 0.748 weight parts of starting material H4), 0.15 weight parts of starting material E1) and 1.5 weight parts of starting material F1). A silicone pressure sensitive adhesive prepared by curing this composition failed the burr test when tested according to Reference Example 8. Without wishing to be bound by theory, it is thought that when vinyl content of

starting material A) in the pressure sensitive adhesive composition was too high, pressure sensitive adhesives prepared therefrom failed the burr test.

#### Industrial Applicability

[0067] The pressure sensitive adhesive composition described herein cures to form a pressure sensitive adhesive suitable for use in flexible OLED device fabrication processes. The examples above show that the pressure sensitive adhesive prepared by curing the pressure sensitive adhesive composition described above may have initial adhesion of < 2 gf/ inch on stainless steel (SUS) (as tested according to Reference Example 3); adhesion after 24 h < 9 gf/in and adhesion after 72 h < 15 gf/in as tested according to Reference Example 4. The pressure sensitive adhesive composition described herein may also cure to form a pressure sensitive adhesive with residual adhesion on SUS > 300 gf/in after 24 h and > 250 g/in after 72 h (as tested according to Reference Example 7). SUS represents an industry standard to develop and test pressure sensitive adhesives. Therefore, without wishing to be bound by theory, it is thought that adhesion properties on SUS will provide comparable results on substrates used in OLED fabrication processes, such as passivation layers, and it is expected that this combination of properties allows a pressure sensitive adhesive film to be used as a protective film on a passivation layer during fabrication of a flexible OLED device.

[0068] Furthermore, it was surprisingly found that when the pressure sensitive adhesive composition contained an poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer with an alkenyl content of 0.14% to < 0.3%, the resulting pressure sensitive adhesive passed the burr test (when comparative pressure sensitive adhesives containing alternative functional polydialkylsiloxanes with alkenyl content > 0.3% failed the burr test, as shown in Example 8).

#### Usage of Terms

[0069] All amounts, ratios, and percentages are by weight unless otherwise indicated by the context of the specification. The articles 'a', 'an', and 'the' each refer to one or more, unless otherwise indicated by the context of the specification. The disclosure of ranges includes the range itself and also anything subsumed therein, as well as endpoints. For example, disclosure of a range of 0.14 to < 0.3 includes not only the range of 0.14 to < 0.3, but also 0.15, 0.20, 0.26 and < 0.3 individually, as well as any other number subsumed in the range. Furthermore, disclosure of a range of, for example, 0.15 to < 0.3 includes the subsets of, for example, 0.15 to 0.21, 0.15 to 0.26, 0.202 to 0.26, and 0.202 to < 0.3 as well as any other subset subsumed in the range. Similarly, the disclosure of Markush groups includes the entire group and also any individual members and subgroups subsumed therein. For example, disclosure of the Markush group a vinyl, allyl or hexenyl includes the member vinyl individually; the subgroup vinyl and hexenyl; and any other individual member and subgroup subsumed therein.

**[0070]** Abbreviations used herein have the following meanings: '°C' and 'deg C' each mean degrees Celsius; 'DP' means degree of polymerization; 'ETCH' means ethynyl cyclohexanol; 'GPC' means gel permeation chromatography, 'GCMS' means gas chromatography mass spectrometry; 'gf' means grams force; 'hr' means hours; 'KG' or 'kg' means kilograms; 'kgf' means kilograms force; 'min' means minutes; 'Me' means methyl, 'mPa·s' means milliPascal seconds, 'OLED' means organic light emitting diode; 'um' means micrometer, and 'Vi' means vinyl.

#### Embodiments of the Invention

**[0071]** In a first embodiment, a method for fabricating a flexible organic light emitting diode device comprises:

- 1) applying a pressure sensitive adhesive composition to a surface of a substrate,
  - 2) curing the pressure sensitive adhesive composition to form a pressure sensitive adhesive film, and
  - 3) applying the pressure sensitive adhesive film to a passivation layer in the device;
- wherein the pressure sensitive adhesive composition comprises.

A) 85 to 100 parts by weight of a poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer, where the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer has silicon bonded alkyl groups with 1 to 10 carbon atoms and silicon bonded alkenyl groups with 2 to 6 carbon atoms, and where the alkenyl groups are present in an amount 0.14% to < 0.3% based on the weight of the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer,

C) 0.1 to 5 parts by weight of a polyalkylhydrogensiloxane having unit formula  $(R^1_2HSiO_{1/2})_2(R^1_2SiO_{2/2})_x(R^1HSiO_{2/2})_y$ , where each  $R^1$  is an independently selected alkyl group of 1 to 10 carbon atoms, subscript  $x \geq 0$ , subscript  $y \geq 0$ , and a quantity  $(x + y)$  is sufficient to give the polyalkylhydrogensiloxane a viscosity of < 20 mPa·s measured at 25 °,

F) an amount of a platinum group metal catalyst sufficient to provide 10 to 5000 ppm concentration of Pt group metal in the composition, and

E) 0.01 to 1 part by weight of an acetylenic alcohol, and

G) 10 weight % to 90 weight%, based on combined weights of all starting materials in the composition, of an organic solvent; with the proviso that all starting materials in the composition combined have a molar ratio of silicon bonded hydrogen atoms / silicon bonded alkenyl groups (SiH:Alkenyl ratio) of 0.5:1 to 2:1.

**[0072]** In a second embodiment, in the method of the first embodiment, starting material A) has a property selected from the group consisting of: i) number average molecular weight of

200,000 g/mol to 500,000 g/mol, ii) alkenyl group content 0.20% to 0.26%, and iii) both i) and ii).

[0073] In a third embodiment, in the method of the first embodiment or the second embodiment, starting material A) comprises average unit formula:

$(R^1_2R^2SiO_{1/2})_d(R^1_3SiO_{1/2})_e(R^1R^2SiO_{2/2})_f(R^1_2SiO_{2/2})_g$ , where each  $R^1$  is independently an alkyl group of 1 to 10 carbon atoms; each  $R^2$  is independently selected from  $R^1$ , OH, and alkenyl groups of 2 to 6 carbon atoms; subscript  $d \geq 0$ , subscript  $e \geq 0$ , subscript  $f > 0$ , subscript  $g \geq 0$ , where a quantity  $(d+e) = 2$ , and a quantity  $(d+e+f+g)$  is 4,500 to 5,000.

[0074] In a fourth embodiment, in the method of any one of the first to third embodiments, starting material C) has a property selected from the group consisting of i) viscosity at 25 deg C of 8 mPa·s to 16 mPa·s, ii) SiH content of  $> 0\%$  to  $< 1\%$ , iii) both i) and ii).

[0075] In a fifth embodiment, in the method of any one of the first to fourth embodiments, starting material F) is selected from the group consisting of (F-1) a platinum group metal selected from platinum, rhodium, ruthenium, palladium, osmium, and iridium; (F-2) a compound of the platinum group metal; (F-3) a complex of the compound with a low molecular weight organopolysiloxane; and (F-4) the compound (F-2), or the complex (F-3), microencapsulated in a matrix or core/shell type structure.

[0076] In a sixth embodiment, in the method of any one of the first to fifth embodiments, starting material E) is selected from the group consisting of (E-1) 1-ethynyl-1-cyclohexanol, (E-2) methyl butynol, (E-3) diallyl maleate, and (E-4) a combination of two or more of (E-1), (E-2) and (E-3). starting material E) comprises ethynyl cyclohexanol.

[0077] In a seventh embodiment, in the method of any one of the first to sixth embodiments, starting material G) is selected from the group consisting of benzene, toluene, heptane, ethylbenzene, xylene, and a combination of two or more thereof.

[0078] In an eighth embodiment, in the method of any one of the first to seventh embodiments, the pressure sensitive adhesive composition further comprises up to 20%, based on weight of starting material C), of H) an additional polyalkylhydrogensiloxane having unit formula

$(R^1_3SiO_{1/2})_2(R^1HSiO_{2/2})_z$ , where subscript  $z$  is 200 to 250.

[0079] In a ninth embodiment, in the method of any one of the first to eighth embodiments, the pressure sensitive adhesive composition further comprises up to 15 parts by weight of B) a polyalkylsiloxane resin comprising units of formula  $(R^1_2R^2SiO_{1/2})_a(R^1_3SiO_{1/2})_b(SiO_{4/2})_c$ , where each  $R^1$  is independently an alkyl group of 1 to 10 carbon atoms; each  $R^2$  is independently selected from  $R^1$ , OH, and alkenyl groups of 2 to 6 carbon atoms; subscript  $a \geq 0$ ,

subscript  $b \geq 0$ , subscript  $c > 0$ , a quantity  $(a+b) > 0$ , and subscripts  $a$ ,  $b$ , and  $c$  have values such that  $0.9 \leq (a+b)/c \leq 1.2$ .

**[0080]** In a tenth embodiment, in the method of any one of the first to ninth embodiments, the pressure sensitive adhesive composition further comprises up to 1 part by weight of D) an anchorage additive.

**[0081]** In an eleventh embodiment, in the method of the tenth embodiment, starting material D) is selected from the group consisting of (D-1) vinyltriacetoxysilane, (D-2) glycidoxypolytrimethoxysilane, (D-3) a combination of (D-1) and (D-2), and (D-4) a combination of (D-3) and a polydimethylsiloxane terminated with hydroxyl groups, methoxy groups, or terminated with both a hydroxy group and a methoxy group.

**[0082]** In a twelfth embodiment, the method of any one of the first to eleventh embodiments further comprises: removing all or a portion of the solvent from the pressure sensitive adhesive composition before step 2).

**[0083]** In a thirteenth embodiment, the method of any one of the first to twelfth embodiments further comprises: treating the substrate before step 1).

**[0084]** In a fourteenth embodiment, starting material A) is free of silicon bonded aromatic hydrocarbyl groups, starting material B), when present, is free of silicon bonded aromatic hydrocarbyl groups, starting material C) is free of silicon bonded aromatic hydrocarbyl groups, starting material D), when present, is free of silicon bonded aromatic hydrocarbyl groups or two or more thereof in the nineteenth embodiment.

## Claims:

## 1. A pressure sensitive adhesive composition comprising:

A) 85 to 100 parts by weight of a poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer, where the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer has silicon bonded alkyl groups with 1 to 10 carbon atoms and silicon bonded alkenyl groups with 2 to 6 carbon atoms, and where the alkenyl groups are present in an amount 0.14% to < 0.3% based on the weight of the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer,

C) 0.1 to 5 parts by weight of a polyalkylhydrogensiloxane having unit formula  $(R^1_2HSiO_{1/2})_2(R^1_2SiO_{2/2})_x(R^1HSiO_{2/2})_y$ , where each  $R^1$  is an independently selected alkyl group of 1 to 10 carbon atoms, subscript  $x \geq 0$ , subscript  $y \geq 0$ , and a quantity  $(x + y)$  is sufficient to give the polyalkylhydrogensiloxane a viscosity of < 20 mPa·s measured at 25 °C with a capillary viscometer,

F) an amount of a platinum group metal catalyst sufficient to provide 10 to 5000 ppm concentration of Pt group metal in the composition, and

E) 0.01 to 1 part by weight of an acetylenic alcohol, and

G) 10 weight % to 90 weight%, based on combined weights of all starting materials in the composition, of an organic solvent; with the proviso that all starting materials in the composition combined have a molar ratio of silicon bonded hydrogen atoms / silicon bonded alkenyl groups (SiH:Alkenyl ratio) of 0.5:1 to 2:1.

2. The composition of claim 1, where the composition further comprises up to 20%, based on weight of starting material C), of H) an additional polyalkylhydrogensiloxane having unit formula  $(R^1_3SiO_{1/2})_2(R^1HSiO_{2/2})_z$ , where subscript  $z$  is 200 to 250.

3. The composition of claim 1 or claim 2, where starting material C) has a silicon bonded hydrogen (SiH) content > 0 to < 1 weight %.

4. The composition of any one of claims 1 to 3, further comprising: B) up to 15 parts by weight of a polyalkylsiloxane resin comprising units of formula

$(R^1_2R^3SiO_{1/2})_a(R^1_3SiO_{1/2})_b(SiO_{4/2})_c$ , where each  $R^1$  is independently an alkyl group of 1 to 10 carbon atoms; each  $R^3$  is independently selected from  $R^1$ , OH, and alkenyl groups of 2 to 6 carbon atoms; subscript  $a \geq 0$ , subscript  $b \geq 0$ , subscript  $c > 0$ , a quantity  $(a+b) > 0$ , and subscripts  $a$ ,  $b$ , and  $c$  have values such that  $0.9 \leq (a+b)/c \leq 1.2$ .

5. The composition of any one of claims 1 to 4, further comprising: up to 1 part by weight of D) an anchorage additive.

6. The composition of claim 5, where starting material D) is selected from the group consisting of D-1) vinyltriacetoxysilane, D-2) glycidoxypolytrimethoxysilane, D-3) 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane, D-4) a combination of two or more of D-1), D-2) and D-3); and D-5) a combination of any of D-1), D-2), D-3) and/or D-4), and a polydimethylsiloxane terminated with hydroxyl groups, methoxy groups, or terminated with both a hydroxy group and a methoxy group.

7. The composition of any one of claims 1 to 6, where A) the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer has > 0.2% to 0.26% alkenyl groups.

8. The composition of any one of claims 1 to 7, where A) the poly(dialkylsiloxane/alkylalkenylsiloxane) copolymer comprises unit formula:

$(R^1_2R^2SiO_{1/2})_d(R^1_3SiO_{1/2})_e(R^1R^2SiO_{2/2})_f(R^1_2SiO_{2/2})_g$ , where each  $R^1$  is independently an alkyl group of 1 to 10 carbon atoms; each  $R^2$  is independently an alkenyl group of 2 to 6 carbon atoms; subscript  $d \geq 0$ , subscript  $e \geq 0$ , subscript  $f > 0$ , subscript  $g \geq 0$ , where a quantity  $(d+e) = 2$ , and a quantity  $(d+e+f+g)$  is 4,500 to 5,000.

9. The composition of any one of claims 1 to 8, where starting material F) is selected from the group consisting of (F-1) a platinum group metal selected from platinum, rhodium, ruthenium, palladium, osmium, and iridium; (F-2) a compound of the platinum group metal; (F-3) a complex of the compound with a low molecular weight organopolysiloxane; and (F-4) the compound (F-2), or the complex (F-3), microencapsulated in a matrix or core/shell type structure.

10. The composition of any one of claims 1 to 9, where starting material E) is selected from the group consisting of (E-1) 1-ethynyl-1-cyclohexanol, (E-2) methyl butynol, (E-3) diallyl maleate, and (E-4) a combination of two or more of (E-1), (E-2) and (E-3).

11. The composition of any one of claims 1 to 10, where starting material G) is selected from the group consisting of benzene, toluene, heptane, ethylbenzene, xylene, and a combination of two or more thereof.

12. The composition of any one of claims 1 to 11, where the composition is a multiple part composition comprising a base part and a curing agent part, where the base part comprises starting materials A), C), and G); and the curing agent part comprises starting materials A), F) and G); and the composition further comprises starting material E) in one or more of the base part or a separate third part.

13. A method of preparing a pressure sensitive adhesive tape comprising a pressure sensitive adhesive film on a surface of a substrate, the method comprising:

optionally 1) treating the surface of the substrate,

2) applying a pressure sensitive adhesive composition according to any one of claims 1 to 12 to the surface of the substrate,

optionally 3) removing all or portion of the solvent, and

4) curing the pressure sensitive adhesive composition to form the pressure sensitive adhesive film on the surface of the substrate.

14. A method for fabricating a flexible organic light emitting diode device, where the method comprises: performing the method steps of claim 13, thereby preparing a pressure sensitive adhesive tape, and applying the pressure sensitive adhesive tape to a passivation layer in the device.

15. Use of the pressure sensitive adhesive tape prepared by the method of claim 13 for protection of a passivation layer in a flexible organic light emitting diode device.

# INTERNATIONAL SEARCH REPORT

International application No  
**PCT/US2022/016542**

|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      |                                                                                                                                                                                                                                                                                   |                       |
|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------|
| <b>A. CLASSIFICATION OF SUBJECT MATTER</b><br><b>INV. H01L23/29</b><br><b>ADD.</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |                                                                                                                                                                                                                                                                                   |                       |
| According to International Patent Classification (IPC) or to both national classification and IPC                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |                                                                                                                                                                                                                                                                                   |                       |
| <b>B. FIELDS SEARCHED</b><br>Minimum documentation searched (classification system followed by classification symbols)<br><b>C08G H01L C08L C09D C08K</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |                                                                                                                                                                                                                                                                                   |                       |
| 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)<br><br><b>EPO-Internal</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |                                                                                                                                                                                                                                                                                   |                       |
| <b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |                                                                                                                                                                                                                                                                                   |                       |
| Category*                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            | Citation of document, with indication, where appropriate, of the relevant passages                                                                                                                                                                                                | Relevant to claim No. |
| <b>X</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                             | <b>US 2020/392384 A1 (JANG WONBUM [KR] ET AL)</b><br><b>17 December 2020 (2020-12-17)</b><br><b>examples P3, P14</b><br><b>paragraphs [0002], [0021], [0046],</b><br><b>[0047], [0049], [0056] - [0059]</b><br><b>table 0</b><br><br><div style="text-align: center;">-----</div> | <b>1-15</b>           |
| <div style="display: flex; justify-content: space-between;"> <div> <input type="checkbox"/> Further documents are listed in the continuation of Box C.         </div> <div> <input checked="" type="checkbox"/> See patent family annex.         </div> </div>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       |                                                                                                                                                                                                                                                                                   |                       |
| <div style="display: flex;"> <div style="flex: 1;"> <p>* Special categories of cited documents :</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier application or patent but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="flex: 1;"> <p>"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</p> <p>"X" document of particular relevance;; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</p> <p>"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</p> <p>"&amp;" document member of the same patent family</p> </div> </div> |                                                                                                                                                                                                                                                                                   |                       |
| Date of the actual completion of the international search                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            | Date of mailing of the international search report                                                                                                                                                                                                                                |                       |
| <b>24 May 2022</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | <b>08/06/2022</b>                                                                                                                                                                                                                                                                 |                       |
| Name and mailing address of the ISA/<br>European Patent Office, P.B. 5818 Patentlaan 2<br>NL - 2280 HV Rijswijk<br>Tel. (+31-70) 340-2040,<br>Fax: (+31-70) 340-3016                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | Authorized officer<br><br><b>Queste, Sébastien</b>                                                                                                                                                                                                                                |                       |

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

**PCT/US2022/016542**

| Patent document<br>cited in search report | Publication<br>date | Patent family<br>member(s) | Publication<br>date |
|-------------------------------------------|---------------------|----------------------------|---------------------|
| <b>US 2020392384 A1</b>                   | <b>17-12-2020</b>   | <b>CN 111032814 A</b>      | <b>17-04-2020</b>   |
|                                           |                     | <b>EP 3645651 A1</b>       | <b>06-05-2020</b>   |
|                                           |                     | <b>JP 2021500417 A</b>     | <b>07-01-2021</b>   |
|                                           |                     | <b>KR 20200087743 A</b>    | <b>21-07-2020</b>   |
|                                           |                     | <b>US 2020392384 A1</b>    | <b>17-12-2020</b>   |
|                                           |                     | <b>WO 2019079366 A1</b>    | <b>25-04-2019</b>   |
| -----                                     |                     |                            |                     |