

TRANSPARENT ADHESIVE

FIELD OF INVENTION

The present invention generally relates to a transparent adhesive. In particular, the present invention relates to a transparent adhesive suitable for joining transparent ceramics.

BACKGROUND

The following discussion of the background to the invention is intended to facilitate an understanding of the present invention only. It should be appreciated that the discussion is not an acknowledgement or admission that any of the material referred to was published, known or part of the common general knowledge of the person skilled in the art in any jurisdiction as at the priority date of the invention.

Optically transparent materials are widely used in various applications such as, but not limited to transparent ballistic protective windows, blast-resistant windows, windows for machining, mining and construction operations, transparent components in the watch-making industry, windows for high temperature applications, ultra-high index lenses and touch panels for smart devices. Consequently, various optically transparent materials have been developed. These include glass, polymers, alkali hydrides, single crystals such as sapphire (Al_2O_3) and transparent ceramics.

Transparent ceramics have various technical advantages over the other optically transparent materials. However, it is challenging to manufacture large profiles of transparent ceramics with sizes comparable to that of transparent glass. In order to create large profiles, transparent ceramics have to be bonded together.

The joining (or bonding) of transparent ceramics to form large profiles is not straightforward. For example, the bond strength should be high enough to provide a good structural integrity. This is heavily dependent on the type of bond formed, as well as the crystallographic compatibility between the bonded surfaces. More importantly, from an optical perspective, the final bond interface achieved has to be effectively seamless. To achieve this, the bonding technique has to result in an optically transparent interface (equal to or greater than the inherent transparency of the bonded

transparent ceramic). Furthermore, the bond formed should achieve effective optical coupling. This means that the refractive index of the bond achieved has to be equal to or greater than the refractive index of the abutting surfaces. Otherwise, the physical phenomenon of total internal reflection will occur and mirror sheens will become
5 largely visible, thereby rendering optical coupling impossible.

Consequently, although optically transparent glues are widely used in joining transparent glass to manufacture larger profiles, they are unsuitable for joining transparent ceramics. This is due to the significantly higher refractive index of transparent ceramics over transparent glass. Specifically, a transparent glass may
10 have a refractive index in the range of 1.3 to 1.5, while a transparent ceramic may have a refractive index in the range of greater than 1.7. Thus, the use of optically transparent glues having a refractive index in the range of 1.3 to 1.6 is compatible with transparent glass because of the comparable refractive index. However, when these
15 optically transparent glues, having a lower refractive index than transparent ceramics, are used to bond transparent ceramics, total internal reflection and mirror sheens would likely occur due to the incompatibility.

In light of the above, there exists a need to develop a transparent adhesive for joining transparent ceramics that ameliorates or overcomes at least one of the
20 aforementioned disadvantages.

SUMMARY OF INVENTION

The present invention seeks to provide a transparent adhesive for joining transparent ceramics that addresses the aforementioned need at least in part. In particular, the present invention seeks to provide a transparent adhesive having refractive index
25 matching the refractive index of at least a portion or surface of the transparent ceramic the transparent adhesive is applied to. The present invention also seeks to provide a method of making a transparent adhesive, a method of applying the transparent adhesive, a transparent ceramic comprising a transparent ceramic surface coated with a transparent adhesive and a transparent panel formed from a plurality of transparent
30 ceramic pieces.

The inventors are motivated by the need to achieve multiple objectives such as structural integrity and optical coupling between a plurality of transparent ceramic pieces.

5 In a first aspect of the present invention, there is provided a transparent adhesive for joining transparent ceramics, the transparent adhesive having the chemical formula:



wherein M is a metal selected from the group consisting of aluminium, lead, titanium, yttrium and zirconium; and wherein M is in an amount of about 50 to about 85 wt%.

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Preferably, the metal M is derived from a metal oxide.

Preferably, the metal oxide has a mean particle size of about 0.1 to about 10 μm .

15 Preferably, the silicon is derived from silicon oxide.

Preferably, the silicon oxide has a mean particle size of about 1 to about 200 nm.

Preferably, the silicon is in an amount of about 8 to about 15 wt%.

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Preferably, the oxygen is in an amount of about 10 to about 30 wt%.

Preferably, the transparent adhesive is mixed with a liquid to form a paste.

25 Preferably, the liquid is one or more selected from the list consisting of an alcohol, a ketone, and a mixture thereof.

Preferably, the transparent adhesive further comprises a binder.

30 Preferably, the binder is one or more selected from the group consisting of polyacrylates, polyethylene glycol, paraffin, polyvinyl alcohol and lignosulfonate.

Preferably, the binder is in an amount of about 0.01 to about 5 wt%.

Preferably, the transparent adhesive further comprises a metal N, a metal N oxide or a mixture thereof.

- 5 Preferably, the metal N, the metal N oxide or the mixture thereof is in an amount of less than about 1.4 wt%.

Preferably, the transparent adhesive has a refractive index of about 1.7 to about 1.9.

- 10 In a second aspect of the present invention, there is provided a method of making a transparent adhesive, the method comprising:

(a) preparing a composition having the chemical formula:



- 15 wherein M is a metal selected from the group consisting of aluminium, lead, titanium, yttrium and zirconium;

wherein M is in an amount of about 50 to about 85 wt%;

(b) heating the composition; and

(c) grinding the composition to form a transparent adhesive.

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Preferably, preparing the composition further comprises mixing a metal oxide comprising the metal M with a silicon oxide.

- 25 Preferably, the metal oxide comprising the metal M is added in an amount of about 50 to about 85 wt%.

Preferably, the heating is carried out at a temperature ranging from about 700°C to about 950°C.

- 30 Preferably, the grinding is carried out using high energy ball milling.

Preferably, the high energy ball milling uses a milling media selected from the group consisting of oxides, nitrides and carbides of aluminium, silicon or zirconium.

Preferably, the transparent adhesive has a particle size in the range of about 0.1 to about 20 μm .

5 Preferably, the method further comprises adding a liquid selected from the group consisting of an alcohol, a ketone and a mixture thereof to form a paste.

10 Preferably, the method further comprises adding a binder in an amount of about 0.01 to about 5 wt%, wherein the binder is one or more selected from the group consisting of polyacrylates, polyethylene glycol, paraffin, polyvinyl alcohol and lignosulfonate.

10 Preferably, the method further comprises adding a metal N, a metal N oxide or a mixture thereof.

15 Preferably, the metal N, the metal N oxide or the mixture thereof is in an amount of less than about 1.4 wt%.

In a third aspect of the present invention, there is provided a method of applying a transparent adhesive, comprising the steps of:

20 (a) polishing a surface of a first transparent ceramic and a surface of a second transparent ceramic;

(b) applying a transparent adhesive as described above to the surface of the first transparent ceramic;

(c) applying the transparent adhesive as described above to the surface of the second transparent ceramic; and

25 (d) contacting the surface of the first transparent ceramic with the surface of the second transparent ceramic to form an adjoining surface.

Preferably, the method further comprises thermal conditioning the adjoining surface.

30 Preferably, the thermal conditioning comprises heating the adjoining surface at a temperature ranging from about 400°C to about 650°C, followed by heating the adjoining surface at a temperature ranging from about 700 °C to about 950°C.

In a fourth aspect of the present invention, there is provided a transparent ceramic comprising a transparent ceramic surface coated with a transparent adhesive as described above.

- 5 In a fifth aspect of the present invention, there is provided a transparent panel formed from a plurality of transparent ceramic pieces, each transparent ceramic piece having a surface joined to a surface of an adjacent transparent ceramic piece to form an adjoining surface, wherein the adjoining surface is formed by application of a transparent adhesive having the chemical formula:

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wherein M is a metal selected from the group consisting of aluminium, lead, titanium, yttrium and zirconium; and wherein M is in an amount of about 50 to about 85 wt%.

- 15 Preferably, the plurality of transparent ceramic pieces are planar transparent ceramic pieces.

Preferably, each planar transparent ceramic piece comprises a planar surface and a side surface, and wherein the surface joined to the surface of the adjacent transparent
20 ceramic piece is the side surface.

Preferably, the x, y, and z parameter of the transparent adhesive is adjustable to achieve a refractive index between about 1.7 to about 1.9.

BRIEF DESCRIPTION OF THE DRAWINGS

- 25 The present invention will now be described, by way of example only, with reference to the accompanying drawings, in which:

Fig. 1 is a microscopic view of a transparent adhesive in accordance with an embodiment of the invention, wherein the transparent adhesive is in dry form;

- Fig. 2 is a microscopic view of the microstructure of a transparent adhesive in
30 accordance with an embodiment of the invention, after the transparent adhesive has been applied to a transparent ceramic;

Fig. 3(a) is a planar view of a transparent ceramic in accordance with an embodiment of the invention, the transparent ceramic having a planar surface and four side surfaces; Fig. 3(b) is another planar view of the transparent ceramic of Fig. 3(a), illustrating another planar surface;

- 5 Fig. 4 is a photograph showing two transparent ceramics joined using (left) a transparent adhesive in accordance with an embodiment of the invention and (right) a conventional adhesive as the control;

Fig. 5(a) is a perspective view of two transparent ceramics joined using a transparent adhesive in accordance with an embodiment of the invention, each transparent ceramic having a planar surface and four side surfaces; Fig. 5(b) is a planar view of
10 the two transparent ceramics of Fig. 5(a); Fig. 5(c) is a cross-sectional view of the two transparent ceramics of Fig. 5(a) as viewed from V_1 of Fig. 5(a); Fig. 5(d) is a cross-sectional view of the two transparent ceramics of Fig. 5(a) as viewed from V_2 of Fig. 5(a);

- 15 Fig. 6 is a photograph showing two transparent ceramics joined using a transparent adhesive in accordance with an embodiment of the invention to form a bonding interface having refractive index matching that of the transparent ceramics, each transparent ceramic having a planar surface and four side surfaces;

Fig. 7 is a graph depicting the relationship between the force at failure vs. maximum
20 deflection of transparent ceramics joined using transparent adhesives in accordance with embodiments of the invention and conventional adhesives as the control;

Fig. 8 is a graph depicting the relationship between the force at failure vs. amount of the metal N oxide in transparent adhesives in accordance with an embodiment of the invention.

- 25 Other arrangements of the invention are possible and, consequently, the accompanying drawings are not to be understood as superseding the generality of the preceding description of the invention.

DESCRIPTION OF EMBODIMENTS OF THE INVENTION

Definitions

The following words and terms used herein shall have the meaning indicated:

- 5 Unless otherwise indicated, all percentages and proportions given herein are by weight, i.e. weight percentage (wt%), based on the total weight of the composition. For instance, when metal M is in an amount of about 15 wt%, the proportion is based on the total weight of the composition or in other words, the total weight of the transparent adhesive in dry form.
- 10 Unless specified otherwise, the terms “comprising”, “comprise”, and grammatical variants thereof, are intended to represent “open” or “inclusive” language such that they include recited elements but also permit inclusion of additional, unrecited elements. The terms “including”, “include”, and grammatical variants thereof are construed similarly.
- 15 As used herein, the term “planar” refers to a transparent ceramic having a substantially level surface. However, this does not mean that a planar transparent ceramic cannot have any protrusions or indentations. An object may still be considered planar if a greater extent of the object has a substantially level surface. As illustrated in Fig. 3(a) and Fig. 3(b), each transparent ceramic has a planar surface P1 and P2.
- 20 As used herein, the term “transparent adhesive” refers to an adhesive that may be optically clear to the naked eye after bonding a transparent ceramic to another transparent ceramic. As illustrated in Fig. 6, the bonding interface is seamless and clear to the human eye. In other words, the adhesive has a refractive index matching or approximately matching to that of the transparent ceramics it is applied to. It is to
- 25 be appreciated that the above term “transparent adhesive” can also be extended to include other selected regions of the electromagnetic spectrum (e.g. infrared) which may not be visible to the naked eye.

As used herein, the term “about” typically means +/- 5% of the stated value, more typically +/- 4% of the stated value, more typically +/- 3% of the stated value, more typically +/- 2% of the stated value, even more typically +/- 1% of the stated value, and even more typically +/- 0.5% of the stated value.

5 Throughout this disclosure, certain embodiments may be disclosed in a range format. It should be understood that the description in range format is merely for convenience and brevity and should not be construed as a limitation on the scope of the disclosed ranges. Accordingly, the description of a range should be considered to have specifically disclosed all the possible sub-ranges as well as individual numerical values
10 within that range. For example, description of a range such as from 1 to 6 should be considered to have specifically disclosed sub-ranges such as from 1 to 3, from 1 to 4, from 1 to 5, from 2 to 4, from 2 to 6, from 3 to 6 etc., as well as individual numbers within that range, for example, 1, 2, 3, 4, 5, and 6. Ranges are not limited to integers, and can include decimal measurements where applicable. This applies regardless of
15 the breadth of the range.

Provided herein is a transparent adhesive for joining optically transparent materials having a high refractive index of greater than 1.7, such as, transparent ceramics. Exemplary, non-limiting embodiments of the composition of the transparent adhesive will be described.

20 In an aspect of the present invention, there is provided a transparent adhesive for joining transparent ceramics, the transparent adhesive having the chemical formula:



25 wherein M is a metal selected from the group consisting of aluminium, lead, titanium, yttrium and zirconium; and
wherein M is in an amount of about 50 to about 85 wt%.

In some embodiments, the transparent adhesive has good bond strength and allows
30 the bonding of transparent ceramics, which have a refractive index of greater than 1.7. In some embodiments, the transparent adhesive results in minimizing or avoiding total internal reflection at the optical boundaries, thereby eliminating seam lines at the adjoining surfaces.

The optical refractive index of the transparent adhesive can be tuned by the choice and quantity of components, such as the metal oxide comprising the metal M, in order to suit the properties of the transparent ceramic to be bonded. Consequently and in a preferred embodiment, the transparent adhesive has a refractive index of between about 1.7 to about 1.9. Preferably, the metal M is derived from a metal oxide. A chemical/physical property of the metal oxide is to increase the refractive index of the transparent adhesive, such that it is suitable for joining transparent ceramics.

10 Preferably, the silicon is derived from silicon oxide.

The mean particle size of the metal oxide comprising the metal M and the mean particle size of the silicon oxide is selected to give an optimum homogenization of the raw materials prior to reaction and formation of the transparent adhesive. In various embodiments, the metal oxide comprising the metal M has a mean particle size of about 0.1 to about 10 μm , about 0.1 to about 9 μm , about 0.1 to about 8 μm , about 0.1 to about 7 μm , about 0.1 to about 6 μm , about 0.1 to about 5 μm or about 0.5 to about 10 μm . In various embodiments, the silicon oxide has a mean particle size of about 1 to about 200 nm, about 1 to about 150 nm, about 1 to about 100 nm, about 1 to about 90 nm, about 1 to about 80 nm, about 10 to about 200 nm, about 50 to about 200 nm, about 100 to about 200 nm or about 150 to about 200 nm.

Depending on the choice of metal M, the amount of silicon and oxygen may vary. In various embodiments, silicon is in an amount of about 8 to about 15 wt%. In various embodiments, oxygen is in an amount of about 10 to about 30 wt%.

In various embodiments, the x, y, and z parameter of the transparent adhesive may be an integer or a non-integer.

30 The transparent adhesive may be in dry form. Fig. 1 is a microscopic view of a transparent adhesive in accordance with an embodiment of the invention, wherein the transparent adhesive is in dry form. In various embodiments, the transparent adhesive may be mixed with a liquid in a mechanical mixer or other suitable mixer, to form a paste or slurry. In a preferred embodiment, the transparent adhesive is mixed with a

liquid to form a paste. The liquid may be one or more selected from the list consisting of an alcohol, a ketone, and a mixture thereof. The alcohol may be selected from the list consisting of tert-amyl alcohol, benzyl alcohol, 1,4-butanediol, 1,2,4-butanetriol, butanol, 2-butanol, N-butanol, tert-butyl alcohol, ethanol, ethylene glycol, 2-ethylhexanol, furfuryl alcohol, glycerol, isobutanol, isopropyl alcohol, methanol, 2-(2-methoxyethoxy)ethanol, 2-methyl-1-butanol, 2-methyl-1-pentanol, 3-methyl-2-butanol, neopentyl alcohol, 2-pentanol, 1,3-propanediol, 1-propanol and propylene glycol. The ketone may be selected from the group consisting of acetone, acetophenone, butanone, cyclopentanone, cyclohexanone, dimethyl ketone, diisopropyl ketone, ethyl isopropyl ketone, 2-hexanone, isophorone, mesityl oxide, methyl amyl ketone, methyl ethyl ketone, methyl isobutyl ketone, methyl isopropyl ketone, methyl propyl ketone, 3-methyl-2-pentanone, 2-pentanone and 3-pentanone. In a preferred embodiment, the liquid has a moderate evaporation rate and low viscosity. In particular, the evaporation rate should not be too high to the point where it becomes hard to work with or difficult to use. For instance, if the evaporation rate is too high, the transparent adhesive, which may be a suspension may be dried too rapidly before it is effectively spread to cover the entire surface of the abutting interfaces. In a preferred embodiment, the evaporation rate is lower than about 0.1 cubic centimetres per second (cm^3/s). In addition, the evaporation rate should not be too low because use of a liquid with low evaporation rate may cause the transparent adhesive to run prematurely, such as prior to heat treatment. Consequently, the resultant transparent adhesive will not be ideal in terms of coating homogeneity. Furthermore, a low viscosity facilitates good flowability and spread.

In various embodiments, the transparent adhesive further comprises a binder. Advantageously, the binder may increase the compatibility of the transparent adhesive with the transparent ceramic, such that a seamless bond may be formed. The binder may be one or more selected from the group consisting of polyacrylates such as (but not limited to) poly(methyl acrylate) and poly(ethyl acrylate), polyethylene glycol, paraffin, polyvinyl alcohol and lignosulfonate. In some embodiments, the transparent adhesive does not comprise a binder.

In various embodiments, the binder is in an amount of about 0.01 to about 5 wt%.

In various embodiments, the transparent adhesive further comprises a metal N and/or an oxide of the metal N (i.e. a metal N oxide). The metal N may be a metal selected from the group consisting of aluminium, titanium, yttrium and zirconium. In a preferred embodiment, the transparent adhesive comprises an oxide of aluminium (i.e. aluminium oxide). Consequently, the transparent adhesive may exhibit enhanced mechanical properties, such as greater bond strength. Preferably, the metal N, the metal N oxide or the mixture thereof is in an amount of less than about 1.4 wt%.

In various embodiments, the metal N is a different metal from the metal M.

In another aspect of the present invention, there is provided a method of making a transparent adhesive, the method comprising:

(a) preparing a composition having the chemical formula:



wherein M is a metal selected from the group consisting of aluminium, lead, titanium, yttrium and zirconium;

wherein M is in an amount of about 50 to about 85 wt%;

(b) heating the composition; and

(c) grinding the composition to form a transparent adhesive.

In various embodiments, preparing the composition further comprises mixing a metal oxide comprising the metal M with a silicon oxide.

In various embodiments, the metal oxide comprising the metal M is added in an amount of about 15 to about 50 wt%.

In various embodiments, the heating is carried out at a temperature ranging from about 700°C to about 950°C.

In various embodiments, the grinding is carried out using high energy ball milling. Advantageously, grinding results in reduction of the particle size of the components of the transparent adhesive. The high energy ball milling uses a milling media selected

from the group consisting of oxides, nitrides, carbides of the elements aluminium, silicon or zirconium.

5 In various embodiments, the transparent adhesive has a particle size in the range of about 0.1 to about 20 μm , about 0.1 to about 9 μm , about 0.1 to about 8 μm , about 0.1 to about 7 μm , about 0.1 to about 6 μm , about 0.1 to about 5 μm or about 0.5 to about 10 μm .

10 In various embodiments, the method further comprises adding a liquid or fluid selected from the group consisting of an alcohol, a ketone and a mixture thereof to form a paste. The alcohol may be selected from the list consisting of tert-amyl alcohol, benzyl alcohol, 1,4-butanediol, 1,2,4-butanetriol, butanol, 2-butanol, N-butanol, tert-butyl alcohol, ethanol, ethylene glycol, 2-ethylhexanol, furfuryl alcohol, glycerol, isobutanol, isopropyl alcohol, methanol, 2-(2-methoxyethoxy)ethanol, 2-methyl-1-butanol, 2-
15 methyl-1-pentanol, 3-methyl-2-butanol, neopentyl alcohol, 2-pentanol, 1,3-propanediol, 1-propanol and propylene glycol. The ketone may be selected from the group consisting of acetone, acetophenone, butanone, cyclopentanone, cyclohexanone, dimethyl ketone, diisopropyl ketone, ethyl isopropyl ketone, 2-hexanone, isophorone, mesityl oxide, methyl amyl ketone, methyl ethyl ketone, methyl
20 isobutyl ketone, methyl isopropyl ketone, methyl propyl ketone, 3-methyl-2-pentanone, 2-pentanone and 3-pentanone. In a preferred embodiment, the liquid has a moderate evaporation rate and low viscosity. In particular, the evaporation rate should not be too high to the point where it becomes hard to work with or difficult to use. For instance, if the evaporation rate is too high, the transparent adhesive, which may be a suspension
25 may be dried too rapidly before it is effectively spread to cover the entire surface of the abutting interfaces. In a preferred embodiment, the evaporation rate is lower than about 0.1 cubic centimetres per second (cm^3/s). In addition, the evaporation rate should not be too low because use of a liquid with low evaporation rate would cause the transparent adhesive to run prematurely, such as prior to heat treatment.
30 Consequently, the resultant transparent adhesive will not be ideal in terms of coating homogeneity. Furthermore, a low viscosity facilitates good flowability and spreading.

In various embodiments, the method further comprises adding a binder in an amount of about 0.01 to about 5 wt%, wherein the binder is one or more selected from the

group consisting of polyacrylates such as (but not limited to) poly(methyl acrylate) and poly(ethyl acrylate), polyethylene glycol, paraffin, polyvinyl alcohol and lignosulfonate. In some embodiments, the transparent adhesive does not comprise a binder.

- 5 In various embodiments, the binder is added when grinding the composition. In particular, the binder is added when adding the liquid.

In various embodiments, the method further comprises adding a metal N and/or an oxide of the metal N (i.e. a metal N oxide). For instance, the metal N and/or metal N
10 oxide is added when grinding the composition, such as by high energy ball milling. The amount and type of metal N and/or the metal N oxide may be dependent on the type of milling media used. The metal N may be a metal selected from the group consisting of aluminium, titanium, yttrium and zirconium. In a preferred embodiment, aluminium oxide is added. Consequently, the transparent adhesive may exhibit enhanced
15 mechanical properties, such as greater bond strength. Preferably, the metal N and/or metal N oxide is added in an amount of less than about 1.4 wt%.

In various embodiments, the metal N is a different metal from the metal M.

- 20 In another aspect of the invention, there is provided a method of applying a transparent adhesive, comprising the steps of:

- (a) polishing a surface of a first transparent ceramic and a surface of a second transparent ceramic;
- (b) applying a transparent adhesive of the present invention to the surface
25 of the first transparent ceramic;
- (c) applying the transparent adhesive of the present invention to the surface of the second transparent ceramic; and
- (d) contacting the surface of the first transparent ceramic with the surface of the second transparent ceramic to form an adjoining surface.

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In various embodiments and as illustrated in Fig. 2, microstructures may be observed after the transparent adhesive has been applied to the surface of the first transparent ceramic and/or the surface of the second transparent ceramic.

Advantageously, polishing of the surface of the first transparent ceramic and the surface of the second transparent ceramic aids in achieving an optically clear finish. In various embodiments, the method further comprises a step of thermal conditioning the adjoining surface. The thermal conditioning comprises the step of heating the adjoining surface at a temperature ranging from about 400°C to about 650°C, followed by heating the adjoining surface at a temperature ranging from about 700°C to about 950°C. Thermal conditioning makes use of highly specialized and customized jigs. When undergoing heating at a temperature ranging from about 400°C to about 650°C, the optional binders would be pyrolysed. When undergoing heating at a temperature ranging from about 700°C to about 950°C, the transparent adhesive undergoes melting and coalesces to wet the entire surface area of the adjoining surfaces, thereby leading to the desired optical coupling of transparent ceramics. Advantageously, instead of producing a crystallographic continuum, only an optical continuum is created. Consequently, the method of the present invention can effectively join transparent ceramics having high refractive index and simultaneously create a high refractive index adjoining surface.

Furthermore, the method of the present invention has advantages over prior art methods for bonding transparent ceramics. For instance, the method of the present invention has technical advantages over diffusion bonding. Diffusion bonding, which uses the diffusion of atoms, ions and molecules to create a bond typically under a combination of high pressure and temperature, has a tendency of creating significant strain at the adjoining surface due to the high pressure being applied in order to diffuse the atoms across the abutting interfaces. In contrast, the method of the present invention does not generate significant strain at the adjoining surface. Furthermore, the method of the present invention does not require long treatment times compared to diffusion bonding, which requires very high energy and is a time-consuming method. Consequently, the method of the present invention results in a significant reduction in the operating costs compared to diffusion bonding.

In addition, the method of the present invention has technical advantages over adhesive bonding. Adhesive bonding, which is widely applied in touch panels and display devices, are usually polymeric in nature and makes use of an optically clear liquid adhesive or an optically clear non-liquid adhesive tape to bond materials

together. Advantageously, the method of the present invention can generate a significantly greater bond strength than adhesive bonding.

In addition, the method of the present invention has technical advantages over thermal bonding. Thermal bonding makes use of compatible materials (binders) to create an interface between abutting surfaces. For the joining of ceramics, such materials often include some type of compatible ceramic binder as a component of the composition. This aids in achieving a crystallographic continuum between abutting surfaces, thereby achieving the seamless bond required. Often, however, the ideal of achieving a crystallographic continuum (EG epitaxial growth) is often unachievable as a result of non-ideal densification conditions. Consequently, optical scattering defects such as pores and secondary phases tend to be present, thereby resulting in an undesired opaque interface. Advantageously, as the method of the present invention does not require high temperature ceramics in the composition of the transparent adhesive, the treatment time and temperature required is significantly lower than that of thermal bonding, which requires ceramic binders. Consequently, the method of the present invention results in a significant reduction in the operating costs compared to thermal bonding.

In another aspect of the invention, there is provided a transparent ceramic comprising a transparent ceramic surface coated with a transparent adhesive of the present invention. As illustrated in Fig. 3(a) and Fig. 3(b), each transparent ceramic piece is preferably planar, and comprises planar surfaces P1 and P2 and side surfaces S1, S2, S3, S4, such that the transparent ceramic surface coated with a transparent adhesive of the present invention is one or more of the four side surfaces S1, S2, S3, S4.

In another aspect of the invention, there is provided a transparent panel formed from a plurality of transparent ceramic pieces, each transparent ceramic piece having a surface joined to a surface of an adjacent transparent ceramic piece to form an adjoining surface, wherein the adjoining surface is formed by application of a transparent adhesive having the chemical formula:



wherein M is a metal selected from the group consisting of aluminium, lead, titanium, yttrium and zirconium; and wherein M is in an amount of about 50 to about 85 wt%.

In a preferred embodiment, the plurality of transparent ceramic pieces are planar transparent ceramic pieces. As illustrated in Fig. 3(a) and Fig. 3(b), each planar transparent ceramic piece 10 comprises planar surfaces P1 and P2 and side surfaces S1, S2, S3 and S4. Preferably, the surface joined to the surface of the adjacent transparent ceramic piece is one of the side surfaces S1, S2, S3 and S4. Consequently, a transparent panel may be formed from two transparent ceramic pieces, such that the adjoining surface is formed by contacting one of the side surfaces, such as S4 of the transparent ceramic piece 10, to another side surface, such as S2' of another transparent ceramic piece 10' (left side of Fig. 4).

In another embodiment and as illustrated in Fig. 5(a) to Fig. 5(d), a planar transparent ceramic piece 100 comprising planar surface P1 and side surfaces S10 is joined to a planar transparent piece 100' comprising planar surface P1 and side surfaces S10'. As illustrated in Fig. 5(b), the side surfaces S10 and S10' each have a length L and when viewed from V₁ of Fig. 5(a) or viewed from V₂ of Fig. 5(a), the transparent ceramic piece 100 and 100' each has a thickness W (Fig. 5(c) and Fig. 5(d)). Consequently, a transparent panel may be formed from two transparent ceramic pieces, such that the adjoining surface is formed by contacting one of the side surfaces, such as S10 of the transparent ceramic piece 100, to another side surface, such as S10' of another transparent ceramic piece 100' (Fig. 6). In various embodiments, the length L may be about 50 mm and the thickness W may be about 10 mm. Advantageously, the distance D between the transparent ceramic piece 100 and the transparent ceramic piece 100' is less than or equal to 0.05 mm. More advantageously and as illustrated in Fig. 6, the adjoining surface appears seamless to the human eye, which is a result of the matching between the refractive index of the adhesive and the adjoin surface.

In a preferred embodiment, the x, y, and z parameter of the transparent adhesive is adjustable to achieve a refractive index between about 1.7 to about 1.9.

Examples

Example 1:

A transparent adhesive in accordance with embodiments of the present invention was prepared. Fig. 1 is a microscopic view of the transparent adhesive in dry form, while Fig. 2 is a microscopic view of the microstructure of the transparent adhesive after application to a transparent ceramic.

As illustrated in Fig. 4 and 6, the transparent adhesive results in minimizing or avoiding total internal reflection at the optical boundaries, thereby eliminating seam lines at the adjoining surfaces. Specifically, when two transparent ceramics are joined using a transparent adhesive in accordance with an embodiment of the invention, the word "ST" can be clearly seen due to the minimization or avoidance of total internal reflection at the optical boundaries (left of Fig. 4). In contrast, when two transparent ceramics are joined using a conventional adhesive, the word "inetics" could not be clearly seen due to significant total internal reflection at the optical boundaries (right of Fig. 4).

Example 2: Comparative Study

Two series of transparent adhesives in accordance with embodiments of the present invention were prepared, namely STK OCA Series 1 and STK OCA Series 2 (Figure 7). Variations in processing parameters were studied against a series of control adhesives labelled 'OCAX-1', 'OCAX-2', 'OCAX-3', and 'OCAX- w/o wetting' comprising a thioacrylate and a polymerization initiator, and having a high refractive index ranging from 1.7 to 1.73. The control material was selected specifically for its claimed primary application in bonding transparent ceramics.

All bonded samples tested in the comparative study used identical transparent ceramic plates as substrates, and were subjected to the same test performed on the same day. Samples were subjected to 3 point bend testing with the bonding adhesive being in the centre. The results of the tests reveal that not only do the transparent adhesives of the present invention exhibit a markedly greater adhesive strength as compared to the control, they exhibited greater deflections, making the bonding system described

in this invention significantly tougher as an adhesive for transparent ceramics as compared to the control. As such, the transparent adhesives of the present invention has good bond strength and allows the bonding of transparent ceramics, which have a high refractive index of greater than 1.7.

5 Example 3

A series of experiments incorporating different amounts of aluminium oxide as the metal N oxide into the transparent adhesive was carried out. As illustrated in Fig. 8, the resultant transparent adhesive may exhibit enhanced mechanical properties, particularly when the metal N oxide (N) is in an amount of less than about 1.4 wt%.

10 It will be apparent that various other modifications and adaptations of the invention will be apparent to the person skilled in the art after reading the foregoing disclosure without departing from the spirit and scope of the invention. It is intended that all such modifications and adaptations come within the scope of the appended claims.

15 Further, it is to be appreciated that features from various embodiment(s), may be combined to form one or more additional embodiments.

Claims

1. A transparent adhesive for joining transparent ceramics, the transparent adhesive having the chemical formula:

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wherein M is a metal selected from the group consisting of aluminium, lead, titanium, yttrium and zirconium;

wherein M is in an amount of 50 to 85 wt%;

wherein Si is in an amount of 8 to 15 wt%;

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wherein O is in an amount of 10 to 30 wt%;

wherein each of x, y and z is > 0; and

wherein the transparent adhesive has a particle size in the range of 0.1 to 20 µm and a refractive index of 1.7 to 1.9.

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2. The transparent adhesive of claim 1, wherein the metal M is derived from a metal oxide.

3. The transparent adhesive of claim 2, wherein the metal oxide has a mean particle size of 0.1 to 10 µm.

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4. The transparent adhesive of any one of the preceding claims, wherein the silicon is derived from silicon oxide.

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5. The transparent adhesive of claim 4, wherein the silicon oxide has a mean particle size of 1 to 200 nm.

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6. The transparent adhesive of any one of the preceding claims, wherein the transparent adhesive is mixed with a liquid to form a paste.

7. The transparent adhesive of claim 6, wherein the liquid is one or more selected from the list consisting of an alcohol, a ketone, and a mixture thereof.

8. The transparent adhesive of any one of the preceding claims, further comprising a binder.

9. The transparent adhesive of claim 8, wherein the binder is one or more selected from the group consisting of polyacrylates, polyethylene glycol, paraffin, polyvinyl alcohol and lignosulfonate.

10. The transparent adhesive of claim 8 or 9, wherein the binder is in an amount of 0.01 to 5 wt%.

11. The transparent adhesive of any one of the preceding claims, further comprising a metal N, a metal N oxide or a mixture thereof.

12. The transparent adhesive of claim 11, wherein the metal N, the metal N oxide or the mixture thereof is in an amount of less than 1.4 wt%.

13. A method of making a transparent adhesive, the method comprising:

(a) preparing a composition having the chemical formula:



wherein M is a metal selected from the group consisting of aluminium, lead, titanium, yttrium and zirconium;

wherein M is in an amount of 50 to 85 wt%; wherein Si is in an amount of 8 to 15 wt%; wherein O is in an amount of 10 to 30 wt%; wherein each of x, y and z is > 0;

- (b) heating the composition; and
- (c) grinding the composition to form a transparent adhesive.

- 5 14. The method of claim 13, wherein preparing the composition further comprises mixing a metal oxide comprising the metal M with a silicon oxide.
- 15 15. The method of claim 14, wherein the metal oxide comprising the metal M is added in an amount of 50 to 85 wt%.
- 10 16. The method of any one of claims 13 to 15, wherein the heating is carried out at a temperature ranging from 700°C to 950°C.
- 15 17. The method of any one of claims 13 to 15, wherein the grinding is carried out using high energy ball milling.
18. The method of claim 17, wherein the high energy ball milling uses a milling media selected from the group consisting of oxides, nitrides and carbides of aluminium, silicon or zirconium.
- 20 19. The method of any one of claims 13 to 18, wherein the transparent adhesive has a particle size in the range of 0.1 to 20 µm.
- 25 20. The method of any one of claims 13 to 19, further comprising adding a liquid selected from the group consisting of an alcohol, a ketone and a mixture thereof to form a paste.
- 30 21. The method of claim 20, further comprising adding a binder in an amount of 0.01 to 5 wt%, wherein the binder is one or more selected from the group consisting of polyacrylates, polyethylene glycol, paraffin, polyvinyl alcohol and lignosulfonate.
22. The method of any one of claims 13 to 21, further comprising adding a metal N, a metal N oxide or a mixture thereof.

23. The method of claim 22, wherein the metal N, the metal N oxide or the mixture thereof is in an amount of less than 1.4 wt%.

24. A method of applying a transparent adhesive, comprising the steps of:

5 (a) polishing a surface of a first transparent ceramic and a surface of a second transparent ceramic;

(b) applying a transparent adhesive of any one of claims 1 to 12 to the surface of the first transparent ceramic;

10 (c) applying the transparent adhesive of any one of claims 1 to 12 to the surface of the second transparent ceramic; and

(d) contacting the surface of the first transparent ceramic with the surface of the second transparent ceramic to form an adjoining surface.

15 25. The method of claim 24, further comprising thermal conditioning the adjoining surface.

26. The method of claim 25, wherein the thermal conditioning comprises heating the adjoining surface at a temperature ranging from 400°C to 650°C, followed by heating the adjoining surface at a temperature ranging from 700 °C to 950°C.

20 27. A transparent ceramic comprising a transparent ceramic surface coated with a transparent adhesive of any one of claims 1 to 12.

25 28. A transparent panel formed from a plurality of transparent ceramic pieces, each transparent ceramic piece having a surface joined to a surface of an adjacent transparent ceramic piece to form an adjoining surface, wherein the adjoining surface is formed by application of a transparent adhesive having the chemical formula:



wherein M is a metal selected from the group consisting of aluminium, lead, titanium, yttrium and zirconium; wherein M is in an amount of 50 to 85 wt%; wherein Si is in an amount of 8 to 15 wt%; wherein O is in an amount of 10 to 30

wt%; wherein each of x, y and z is > 0 ; and wherein the transparent adhesive has a particle size in the range of 0.1 to 20 μm and a refractive index of 1.7 to 1.9.

5 29. The transparent panel of claim 28, wherein the plurality of transparent ceramic pieces are planar transparent ceramic pieces.

30. The transparent panel of claim 29, wherein each planar transparent ceramic piece comprises a planar surface and a side surface, and wherein the surface joined to the surface of the adjacent transparent ceramic piece is the side surface.

10 31. The transparent panel of any one of claims 28 to 30, wherein the x, y, and z parameter of the transparent adhesive is adjustable to achieve the refractive index of 1.7 to 1.9.

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