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(54) Titre : COMPOSITIONS DE REVETEMENT DE SOL-GEL ET PROCEDES ASSOCIES
(54) Title: SOL-GEL COATING COMPOSITIONS AND RELATED PROCESSES

(57) **Abrégé/Abstract:**

An alkoxysilane is contacted with water and an inorganic acid to form a first composition. A zirconium alkoxide is contacted with an organic acid to form a second composition. One or more alkoxysilanes and an organic acid are contacted with a mixture of the first and second compositions to form a sol-gel composition, to which a photoinitiator is added. The sol-gel composition has a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from about 2 to about 10. The sol-gel composition is applied on a substrate (e.g., an aluminum alloy substrate) multiple times to form multiple sol-gel layers, and at least one of the sol-gel layers is cured by UV radiation. The multiple sol-gel layers are then thermally cured.

ABSTRACT

An alkoxysilane is contacted with water and an inorganic acid to form a first composition. A zirconium alkoxide is contacted with an organic acid to form a second composition. One or more alkoxysilanes and an organic acid are contacted
5 with a mixture of the first and second compositions to form a sol-gel composition, to which a photoinitiator is added. The sol-gel composition has a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from about **2** to about **10**. The sol-gel composition is applied on a substrate (e.g., an aluminum alloy substrate) multiple times to form multiple sol-gel layers, and at least one of the
10 sol-gel layers is cured by UV radiation. The multiple sol-gel layers are then thermally cured.

SOL-GEL COATING COMPOSITIONS AND RELATED PROCESSES

BACKGROUND

1. Technical Field

The present disclosure relates to coating compositions and processes and,
5 more particularly, to sol-gel coating compositions and related processes.

2. Related Art

High strength alloys such as aluminum alloys are widely used in various
industries such as the aerospace industry due to their high strength to weight ratio.
However these alloys are prone to corrosion due to the presence of alloying
10 materials.

In order to protect these alloys from the environment, a chrome conversion
coating may be provided on a surface of an alloy followed by application of primer
and a top coat. Although organic paint systems applied on the surface provide
good barrier properties against corrosion, even small defects formed in the organic
15 paint ensure pathways for the ingress of electrolyte to the metallic surface, which
initiates localized corrosion. Therefore, chromium based conversion coatings have
been used in anti-corrosion pretreatments before application of organic coatings.
However, hexavalent chromium compounds have harmful effects.

Thus, there is a need for coating compositions and processes that are
20 chromium-free and provide a coating that is corrosion-resistant.

SUMMARY

In accordance with embodiments of the present disclosure, various methods
and formulations are provided relating to sol-gel coating of substrates such as an
25 aluminum substrate, an aluminum alloy substrate (e.g., AA **2024**, AA **6061**, or AA
7075), or other substrate. The sol-gel coating provided on a substrate may provide
corrosion protection. Further, the sol-gel coating provided on the substrate may
provide enhanced adhesion between the substrate and a paint system (e.g., primer
and paint).

In one exemplary aspect, a method for preparing a sol-gel composition includes contacting a first alkoxy silane with water and an inorganic acid to form a first composition, contacting a zirconium alkoxide with a first organic acid to form a second composition, contacting the first composition with the second composition to form a mixture, contacting a phenyl alkoxy silane, an aminopropyl alkoxy silane, a glycidoxypropyl alkoxy silane, and a second organic acid with the mixture to form the sol-gel composition that has a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from about **2** to about **10**, and adding a photoinitiator to the sol-gel composition. The method may also include diluting the sol-gel composition with a solvent and stirring.

In an additional exemplary aspect, a method for providing a corrosion-resistant coating on a substrate (e.g., an aluminum substrate, an aluminum alloy substrate, or other substrate) includes forming a plurality of sol-gel layers on the substrate using the sol-gel composition to provide the corrosion-resistant coating. The method may further include contacting the sol-gel composition with the substrate for each sol-gel layer, curing at least one of the multiple sol-gel layers by UV radiation, and thermally curing the multiple sol-gel layers.

In another exemplary aspect, a sol-gel composition and/or a corrosion resistant coating including multiple sol-gel layers is provided by one of the methods described above.

In a further exemplary aspect, a corrosion resistant coated product includes a plurality of UV-cured sol-gel layers on a substrate, each of the UV-cured sol-gel layers including a photoinitiator and a polymer composite of a methacryloxyalkyl alkoxy silane, an aryl alkoxy silane, an aminoalkyl alkoxy silane, a glycidoxyalkyl alkoxy silane, a zirconium alkoxide, and an organic acid, and each of the UV-cured sol-gel layers having a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from about **2** to about **10**.

In another exemplary aspect, a method of preparing a sol-gel composition includes contacting a first alkoxy silane with water and an inorganic acid to form a

first composition; contacting a zirconium alkoxide with a first organic acid to form a second composition; contacting the first composition with the second composition to form a mixture; contacting one or more second alkoxysilanes and a second organic acid with the mixture to form a sol-gel composition, the sol-gel composition having a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from about **2** to about **10**; aging the sol-gel composition by adding an organic solvent and stirring for about **1** hour to about **24** hours; and adding a photoinitiator to the sol-gel composition.

In a further exemplary aspect, a method of preparing a sol-gel composition includes contacting a first alkoxysilane with water and an inorganic acid to form a first composition; contacting a zirconium alkoxide with a first organic acid to form a second composition; contacting the first composition with the second composition to form a mixture; contacting one or more second alkoxysilanes and a second organic acid with the mixture to form a sol-gel composition, the sol-gel composition having a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from about **2** to about **10**; aging the sol-gel composition by adding an organic solvent in a weight ratio of about **1:1**; and adding a photoinitiator to the sol-gel composition.

In a further exemplary aspect, a method of sol-gel coating an aluminum or aluminum alloy substrate includes: (i) contacting a first alkoxysilane with water and an inorganic acid to form a first composition; (ii) contacting a zirconium alkoxide with a first organic acid to form a second composition; (iii) contacting the first composition with the second composition to form a mixture; (iv) contacting one or more second alkoxysilanes and a second organic acid with the mixture to form a sol-gel composition, the sol-gel composition having a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from 5 to 10; (v) adding a photoinitiator to the sol-gel composition; (vi) applying the sol-gel composition on an aluminum or aluminum alloy substrate a plurality of times to form a plurality of sol-gel layers; (vii) curing at least one of the plurality of sol-gel layers by UV radiation; and (viii) thermally curing the plurality of sol-gel layers to form a corrosion-resistant coating comprising the plurality of sol-gel layers.

A better understanding of the methods and formulations for sol-gel coating of the present disclosure, as well as an appreciation of the above and additional advantages thereof, will be afforded to those of skill in the art by a consideration of the following detailed description of one or more example embodiments thereof. In
5 this description, reference is made to the various views of the appended sheets of

drawings, which are briefly described below, and within which, like reference numerals are used to identify like ones of the elements illustrated therein.

BRIEF DESCRIPTION OF THE DRAWINGS

5 FIG. 1 illustrates an example process for preparing a sol-gel composition in accordance with an embodiment of the present disclosure.

 FIG. 2 illustrates an example process for forming a corrosion-resistant coating that includes one or more sol-gel layers in accordance with an embodiment of the present disclosure.

10 FIGS. 3A-C illustrate example corrosion-resistant coatings that include one or more sol-gel layers in accordance with embodiments of the present disclosure.

 FIG. 4 is a scanning electron microscopy (SEM) image of a surface of a sol-gel layer coating a substrate.

 FIG. 5A is an image of a sol-gel coated substrate formed by the process of
15 FIG. 2 after a corrosion-resistance test.

 FIG. 5B is an image of a hexavalent chrome conversion coated substrate after a corrosion-resistance test.

 FIG. 5C is an image of an uncoated substrate after a corrosion-resistance test.

20 FIG. 6 is an image of a primer applied sol-gel coated substrate formed by the process of FIG. 2 after an adhesion test.

DETAILED DESCRIPTION


 The following description is merely exemplary in nature and is not intended
25 to limit the present disclosure, application, or uses.

 The terms “substituent”, “radical”, “group”, “moiety,” and “fragment” may be used interchangeably.

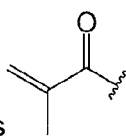
 Singular forms “a” and “an” may include plural reference unless the context clearly dictates otherwise.

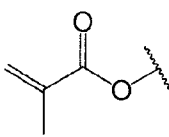
The number of carbon atoms in a substituent can be indicated by the prefix “C_{A-B}” where A is the minimum and B is the maximum number of carbon atoms in the substituent.

The term “alkyl” embraces a linear or branched acyclic alkyl radical containing from **1** to about **15** carbon atoms. In some embodiments, alkyl is a C₁₋₁₀ alkyl, C₁₋₆ alkyl, or C₁₋₃ alkyl radical. Examples of alkyl include, but are not limited to, methyl, ethyl, propyl, isopropyl, butyl, isobutyl, *tert*-butyl, *sec*-butyl, pentan-**3**-yl

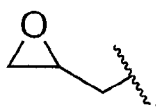
(i.e., ) , and the like.

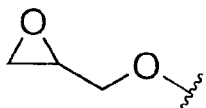
The term “alkoxy” is RO- where R is alkyl. Non-limiting examples of alkoxy include methoxy, ethoxy, propoxy, n-butyloxy, and *tert*-butyloxy. The terms “alkyloxy”, “alkoxy,” and “alkyl-O-” may be used interchangeably.

The term “methacryl” is 

The term “methacryloxy” is 

The term “methacryloxyalkyl” embraces alkyl substituted with methacryloxy. Non-limiting examples of methacryloxyalkyl include methacryloxyethyl, methacryloxypropyl, and methacryloxybutyl.

The term “glycidyl” is 

The term “glycidyloxy” is . The terms “glycidyloxy” and “glycidoxy” may be used interchangeably.

The term “glycidoxyalkyl” embraces alkyl substituted with glycidoxy. Non-limiting examples of glycidoxyalkyl include, glycidoxyethyl, and glycidoxypropyl, and

glycidoxybutyl. The terms "glycidylalkyl" and "glycidoxyalkyl" may be used interchangeably.

The term "aminoalkyl" embraces an amino radical attached to a parent molecular scaffold through an alkyl radical (e.g., NH₂-alkyl-scaffold).

5 The term "aryl" refers to any monocyclic, bicyclic, or tricyclic cyclized carbon radical, wherein at least one ring is aromatic. An aromatic radical may be fused to a non-aromatic cycloalkyl or heterocyclyl radical. Aryl may be substituted or unsubstituted. Examples of aryl include phenyl and naphthyl.

10 The term "aralkyl" embraces aryl attached to a parent molecular scaffold through alkyl and may be used interchangeably with the term "arylalkyl." Examples of aralkyl include benzyl, diphenylmethyl, triphenylmethyl, phenylethyl, and diphenylethyl. The terms "benzyl" and "phenylmethyl" may be used interchangeably.

The term "silane" is a compound containing silicon.

15 The term "organosilane" is a silane having at least one silicon to carbon bond.

The term "alkoxysilane" is a silane having at least one silicon to alkoxy bond.

The term "organoalkoxysilane" is a silane having at least one silicon to carbon bond and at least one silicon to alkoxy bond.

20 The term "about," as used herein when referring to a measurable value such as an amount, concentration, time and the like, is meant to encompass variations of $\pm 20\%$, $\pm 10\%$, $\pm 5\%$, $\pm 1\%$, $\pm 0.5\%$, or even $\pm 0.1\%$ of the specified value.

25 Compositions and processes relating to sol-gel coating of substrates such as metal or metal alloy substrates (e.g., aluminum substrates, aluminum alloy substrates (e.g., AA **2024**, AA **6061**, or AA **7075**), or other substrates) are provided. Sol-gel coating may be used as a chrome-free pretreatment on substrates prior to the application of organic coatings such as primer and paint. The pretreatment may be performed by applying a layer of a sol-gel composition that is obtained as a product of hydrolysis and condensation of a mixture of organosilanes and a metal
30 alkoxide. Ultraviolet (UV) radiation is used to densify the sol-gel layer in addition to,

or instead of, thermal curing the sol-gel layer. Thermal curing may include exposing the sol-gel layer to a high temperature (e.g., in a hot air circulated oven). Alternatively, or in addition, thermal curing may include exposing the sol-gel layer to infrared (IR) radiation or near IR radiation, which reduces curing time.

5 FIG. 1 illustrates an example process 100 for preparing a sol-gel composition. A low temperature curable matrix sol is synthesized in two parts (Composition A and Composition B), the two parts are mixed together, and additional compounds are added and stirred to complete synthesis and obtain a sol-gel composition.

10 At block 102, Composition A is prepared from an alkoxy silane such as an organoalkoxy silane. An alkoxy silane is contacted with water and an inorganic acid (e.g., HCl, HNO₃, H₃PO₄, or other inorganic acid) to form Composition A.

For example, an alkoxy silane is mixed with water and stirred, and an inorganic acid is added to the solution of the alkoxy silane and water and stirred in
 15 an ice bath until the solution turns transparent. The ratio of the number of moles of the alkoxy silane (which is equal to the number of moles of silicon from the alkoxy silane) to the number of moles of water (n_{Si}/n_{water}) in Composition A ranges from about 0.5 to about 2. The ratio may be, for example, about 0.5, 0.6, 0.7, 0.8, 0.9, 1.0, 1.1, 1.2, 1.3, 1.4, 1.5, 1.6, 1.7, 1.8, 1.9, or 2.0, where any value may form
 20 an upper end point or a lower end point, as appropriate.

In an aspect, an alkoxy silane of Formula I is used as Precursor A:



wherein;

R_A is methacryloxyalkyl or glycidoxyalkyl; and

25 R_B is alkoxy.

In another aspect, R_A is methacryloxyalkyl (e.g., methacryloxymethyl, methacryloxyethyl, methacryloxypropyl, methacryloxybutyl, or other methacryloxyalkyl) or glycidoxyalkyl (e.g., glycidoxymethyl, glycidoxyethyl, glycidoxypropyl, glycidoxybutyl); and each R_B is independently alkoxy (e.g.,
 30 methoxy, ethoxy, propoxy).

Specific examples of $R_A\text{-Si-(R}_B)_3$ include 3-methacryloxypropyltrimethoxysilane, 3-methacryloxypropyltriethoxysilane, 3-glycidioxypropyltrimethoxysilane, and 3-glycidioxypropylethoxysilane.

In some aspects, an alkoxysilane used as Precursor A of a sol-gel composition includes methacryloxyalkyl alkoxysilane (an alkoxysilane of Formula I in which R_A is methacryloxyalkyl) and/or a glycidioxyalkyl alkoxysilane (an alkoxysilane of Formula I in which the R_A is glycidioxyalkyl). The methacryloxyalkyl alkoxysilane and/or the glycidioxyalkyl alkoxysilane are used, for example, to facilitate polymerization of the sol-gel composition when exposed to UV radiation.

At block 104, Composition B is prepared from a transition metal alkoxide such as a zirconium alkoxide. A zirconium alkoxide is contacted with an organic acid such as a carboxylic acid (e.g., methacrylic acid (MAA) or other carboxylic acid) to form Composition B.

For example, the zirconium alkoxide is mixed with methacrylic acid and stirred. The ratio of the number of moles of the zirconium alkoxide (which is equal to the number of moles of zirconium from the zirconium alkoxide) to the ratio of the number of moles of methacrylic acid ((n_{Zr}/n_{MAA}) ranges from about 0.5 to about 2. The ratio may be, for example, about 0.5, 0.6, 0.7, 0.8, 0.9, 1.0, 1.1, 1.2, 1.3, 1.4, 1.5, 1.6, 1.7, 1.8, 1.9, or 2.0, where any value may form an upper end point or a lower end point, as appropriate.

In an aspect, a zirconium alkoxide of Formula II is used as Precursor B:



wherein;

R_C is alkoxy.

In another aspect, each R_C is independently alkoxy (methoxy, ethoxy, n-propoxy, isopropoxy, n-butoxy, tert-butoxy, or other alkoxy).

Specific examples of $\text{Zr-(R}_C)_4$ include zirconium ethoxide, zirconium n-propoxide, zirconium isopropoxide, zirconium n-butoxide, and zirconium tert-butoxide.

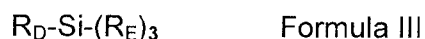
In some aspects, a zirconium alkoxide is used as Precursor B of a sol-gel composition, for example, to match the coefficient of thermal expansion of the sol-gel composition with a substrate. The zirconium alkoxide may be used in an amount such that the coefficient of thermal expansion of the sol-gel composition is equal to or about the coefficient of thermal expansion of the substrate.

At block 106, Composition A and Composition B are mixed together. For example, Composition B is added to Composition A under stirring to avoid agglomeration, and the mixture of Composition A and Composition B is stirred in an ice bath and then stirred at room temperature so that the temperature of the mixture reaches room temperature.

At block 108, one or more alkoxysilanes such as one or more organoalkoxysilanes are added to the mixture of Composition A and Composition B. One or more alkoxysilanes and an organic acid such as a carboxylic acid (e.g., methacrylic acid or other carboxylic acid) are contacted with the mixture of Composition A and Composition B to form a sol-gel composition.

For example, each of one or more alkoxysilanes are added to the mixture and stirred. Then, methacrylic acid is added to the resulting mixture and stirred. Optionally, an inorganic acid is added before, together with, or after the organic acid.

In an aspect, one or more alkoxysilane of Formula III is used as Precursor C:



wherein;

R_D is aryl, aralkyl, glycidoxyalkyl, or aminoalkyl; and

R_E is alkoxy.

In another aspect, R_D is aryl (e.g., phenyl or other aryl), aralkyl (e.g., benzyl, phenylethyl, phenylpropyl, or other aralkyl), glycidoxyalkyl (e.g., glycidomethyl, glycidoxyethyl, glycidoxypropyl, glycidoxybutyl, or other glycidoxyalkyl), or aminoalkyl (e.g., aminomethyl, aminoethyl, aminopropyl, aminobutyl, or other aminoalkyl); and each R_E is independently alkoxy (e.g., methoxy, ethoxy, propoxy).

Specific examples of $R_D\text{-Si-(R}_E)_3$ include phenyltrimethoxysilane, phenyltriethoxysilane, **3**-glycidoxypropyltrimethoxysilane, **3**-glycidoxypropyltriethoxysilane, **3**-aminopropyltrimethoxysilane, and **3**-aminopropyltriethoxysilane.

5 In some aspects, one or more alkoxysilanes used as Precursor C of a sol-gel composition include an aryl alkoxysilane (an alkoxysilane of Formula III in which R_D is aryl), a glycidylalkyl alkoxysilane (an alkoxysilane of Formula III in which R_D is glycidylalkyl), and/or an aminoalkyl alkoxysilane (an alkoxysilane of Formula III in which R_D is aminoalkyl). The aryl alkoxysilane is used, for example, to improve the
10 barrier properties of a coating formed from the sol-gel composition. The glycidylalkyl alkoxysilane is used, for example, to generate a thick coating. The aminoalkyl alkoxysilane is used, for example, to improve the adhesion of the sol-gel composition to a substrate when deposited.

In an example, an aryl alkoxysilane is added to the mixture and stirred.
15 Then, an aminoalkyl alkoxysilane is added to the mixture and stirred. Then, a glycidylalkyl alkoxysilane is added to the mixture. Then, methacrylic acid is added and stirred. An inorganic acid may also be added. The order of the alkoxysilanes that are added may be changed in other examples.

The total amount of the alkoxysilanes, which includes the alkoxysilane used
20 in block **102** and the one or more alkoxysilanes used in block **108**, and the amount of the zirconium alkoxide used in block **104** are such that the sol-gel composition has a ratio of a number of moles of alkoxysilanes (which is equal to the number of moles of silicon from the alkoxysilanes) to a number of moles of zirconium alkoxide (which is equal to the number of moles of zirconium from the zirconium alkoxide)
25 (n_{Si}/n_{Zr}) ranging from about **2** to about **10**. The ratio of the number of moles of silicon to the number of moles of zirconium (n_{Si}/n_{Zr}) may be about **2, 3, 4, 5, 6, 7, 8, 9, or 10**, where any value may form an upper end point or a lower end point, as appropriate.

In some examples, one or more of the stirring performed in blocks **102, 104,**
30 **106, and/or 108** may be performed for a time period ranging from about **10 min** to

about **120** min. The stirring performed in blocks **102**, **104**, **106**, and/or **108** may be performed for a time period of about **10**, **20**, **30**, **40**, **50**, **60**, **70**, **80**, **90**, **100**, **110**, or **120** min, where any value may form an upper end point or a lower end point, as appropriate.

5 At block **110**, the sol-gel composition formed by block **108** is diluted with a solvent such as alcohol (e.g., isopropanol or other solvent) and stirred. The dilution of the sol-gel composition, the stirring to age the sol-gel composition, or both (e.g., block **110** entirely) may be omitted in some embodiments.

10 For example, the sol-gel composition is diluted with isopropanol in a weight ratio of about **1:1**. The diluted sol-gel composition, or the sol-gel composition formed by block **108** if dilution is omitted, is stirred to age the sol-gel composition for a time period ranging from **1** to about **24** hours (h). The stirring to age the sol-gel composition may be performed for a time period of about **1**, **2**, **3**, **4**, **5**, **6**, **9**, **12**, **15**, **18**, **21**, or **24** h, where any value may form an upper end point or a lower end point,
15 as appropriate.

 At block **112**, a photoinitiator is added to the sol-gel composition formed by block **110** (or by block **108** for embodiments in which block **110** is omitted) and stirred to form a sol-gel composition that includes the photoinitiator.

20 For example, a photoinitiator in an amount ranging from about **0.5** to about **3** parts by weight per **100** parts by weight of the sol-gel composition (the weight of the sol-gel with the photoinitiator to be added or, alternatively, the weight of the sol-gel before adding the photoinitiator) is added, and the sol-gel composition with the photoinitiator is stirred. The amount of the photoinitiator may be about **0.5**, **1**, **1.5**, **2**, **2.5**, or **3** parts by weight per **100** parts of the sol-gel composition, where any
25 value may form an upper end point or a lower end point, as appropriate. The stirring may be performed for a time period ranging from about **10** to about **60** min. The stirring may be performed for a time period of about **10**, **20**, **30**, **40**, **50**, or **60** min, where any value may form an upper end point or a lower end point, as appropriate. Once the photoinitiator is added, exposure of the sol-gel composition

to light may be avoided by covering a container for the sol-gel composition (e.g., using aluminum foil) and/or storing in an amber-colored container.

Example 1.

Composition A was synthesized by mixing **171.5 g** of
5 **3-methacryloxypropyltrimethoxysilane** and **17.0 g** of water in a glass jar loaded on a magnetic stirrer. **5.5 grams** of **0.1 N HCl** was further added to the mixture. The solution was stirred in an ice bath till the solution turned transparent. Although **3-methacryloxypropyltrimethoxysilane** was used in this example, one or more other alkoxysilanes of Formula I may be used in place of, or in addition to,
10 **3-methacryloxypropyltrimethoxysilane** in other examples. Also, although HCl was used in this example, one or more other inorganic acids may be used in place of, or in addition to, HCl in other examples.

Composition B was synthesized by mixing **11.8 g** of methacrylic acid and
15 **45.2 g** of zirconium n-propoxide under vigorous stirring. Stirring was continued for about **2 h**. Although zirconium n-propoxide was used in this example, one or more other zirconium alkoxides of Formula II may be used in place of, or in addition to, zirconium n-propoxide in other examples.

Composition B was added to Composition A under vigorous stirring to avoid agglomeration by placing the mixture in an ice bath, and the mixture was stirred for
20 about **1 h**. The jar containing the mixture was removed from the ice bath and stirred at room temperature for at least **1 h** for the mixture to come to room temperature.

Then **100 g** of phenyltrimethoxysilane was added to the mixture of Composition A and Composition B and stirred for about **1 h**, and then **100 g** of **3-aminopropyltrimethoxysilane** was added and stirred for about **1 h**. After completion
25 of the **1 h** of stirring with **3-aminopropyltrimethoxysilane**, **25 grams** of **3-glycidoxypropyltrimethoxysilane** was added. Finally, **10 grams** of methacrylic acid was added followed by **4 g** of **0.1 N HCl** and stirred for a further duration of **1 h**. Although phenyltrimethoxysilane, **3-aminopropyltrimethoxysilane**, and **3-glycidoxypropyltrimethoxysilane** were used in this example, one or more other
30 alkoxysilanes of Formula III may be used in place of, or in addition to,

phenyltrimethoxysilane, **3**-aminopropyltrimethoxysilane, and/or **3**-glycidoxypropyltrimethoxysilane in other examples.

The resulting mixture was diluted with isopropanol in a weight ratio of about **1:1** and stirred for about **3** h at room temperature for aging. Although the mixture was stirred for about **3** h, the mixture may be aged for a different time period in other examples, such as by stirring overnight. Approximately **1** kg of sol-gel composition ready for coating application was formed. A photoinitiator, IRGACURE[®] **184**, in the amount of about **2** % by weight per **100** % of the final sol-gel composition (including the photoinitiator) was added and stirred for **30** min. Although IRGACURE[®] **184** was used in this example, one or more other photoinitiators may be used in place of, or in addition to, IRGACURE[®] **184** in other examples. After adding IRGACURE[®] **184**, the sol-gel composition was kept away from light to avoid the sol-gel composition from interacting with light.

FIG. **2** illustrates an example process **200** for forming a corrosion-resistant coating that includes one or more sol-gel layers (e.g., one or more sol-gel coatings) on a substrate such as a panel (e.g., an aluminum substrate, an aluminum alloy substrate, or other substrate). One or more layers of the sol-gel composition are applied to a substrate, each of the one or more layers is cured by UV light, and then the one or more layers of the sol-gel composition are thermally cured.

At block **202**, the sol-gel composition including the photoinitiator prepared by process **100** of FIG. **1** is applied to a substrate. The sol-gel composition is contacted with the substrate, for example, by dipping the substrate in the sol-gel composition, by immersing the substrate in the sol-gel composition, by spraying the sol-gel composition on the substrate, and/or by other methods of applying the sol-gel composition to the substrate. If dip coating is used, sol-gel layers can be deposited using a withdrawal speed ranging from about **1** to about **15** mm/s (e.g., about **5** to about **12** mm/s, about **10** mm/s, or other withdrawal speed). The withdrawal speed may be about **1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, or 15** mm/s, where any value may form an upper end point or a lower end point, as appropriate.

At block **204**, the sol-gel layer formed by block **202** is cured by UV radiation. For example, the UV radiation has a light dose ranging from about **500** to about **1000** mJ/cm². The UV radiation may have a light dose of about **500, 550, 600, 650, 700, 750, 800, 850, 900, 950, or 1000** mJ/cm², where any value may form an upper end point or a lower end point, as appropriate. The curing by UV radiation may be performed for a time period ranging from about **0.5** to about **30** min. The time period may be about **0.5, 1, 2, 3, 4, 5, 10, 15, 20, 25, or 30** min, where any value may form an upper end point or a lower end point, as appropriate.

At block **206**, if additional sol-gel layers are to be formed, blocks **202** and **204** are repeated. Blocks **202** and **204** may be repeated until the desired number of sol-gel layers is formed. Blocks **202** and **204** are performed for each of the sol-gel layer. In other embodiments, block **204** may be omitted for at least one of the sol-gel layers (e.g., at least one of the sol-gel layers may be air dried or thermally cured instead of curing using UV radiation). For example, curing using UV radiation may be omitted for the final, top-most sol-gel layer among the desired sol-gel layers.

At block **208**, the sol-gel layers are thermally cured. For example, the one or more sol-gel layers are thermally cured at a temperature ranging from about **70** to about **90** °C. The one or more sol-gel layers may be thermally cured at about **70, 75, 80, 85, or 90** °C, where any value may form an upper end point or a lower end point, as appropriate. The thermal curing may be performed for a time period ranging from about **40** to about **120** minutes. The time period may be about **40, 50, 60, 70, 80, 90, 100, 110, or 120** min, where any value may form an upper end point or a lower end point, as appropriate. In an example, the thermal curing is performed in a hot air circulated oven. Alternatively, or in addition to, thermal curing at a high temperature, the thermal curing includes exposing the sol-gel layers to infrared (IR) radiation, near IR radiation, and/or microwave radiation. For example, the sol-gel layers are exposed to IR and/or near IR radiation for a time period ranging from about **10** to about **60** min (e.g., **30** min or other time period). The time period of exposure to IR and/or near IR may be about **10, 15, 20, 25, 30, 35, 40, 45,**

50, 55, or 60 min, where any value may form an upper end point or a lower end point, as appropriate.

At block 210, primer and/or paint is applied on the sol-gel layer of the substrate. For example, the primer is applied on the top-most sol-gel layer, and the paint is applied on the primer. The cured sol-gel layers not only may provide corrosion resistance to the substrate but may also facilitate adherence of the primer and/or paint to the substrate.

Example 2.

Each sol-gel layer of a substrate was UV cured using a conveyORIZED UV curing unit. UV curing was performed on both sides of the substrate using three-medium-pressure-mercury lamp conveyORIZED UV curing unit. The lamps provided an output of about 120 W/cm with a total wattage/lamp (1 m long) = 12 kW. The belt speed was maintained at about 2 m/min during curing. The light dose as measured by a UV radiometer was 871 mJ/cm² in the UV-C region. After UV curing for about 5 minutes, the sol-gel layer coated substrate was subjected to thermal curing in an air circulated oven at 80°C for 1 h.

FIGS. 3A-C illustrate example corrosion-resistant coatings that include one or more sol-gel layers, which may be formed, for example, by process 200 of FIG. 2. FIG. 3A shows a substrate 302 with one sol-gel layer 304. A sol-gel composition is contacted with substrate 302 to provide sol-gel layer 304. Sol-gel layer 304 is UV cured, and then thermally cured.

FIG. 3B shows a substrate 312 with two sol-gel layers 314 and 316. Sol-gel layer 314 is formed on substrate 312, and sol-gel layer 316 is formed on sol-gel layer 314 of substrate 312. A sol-gel composition is contacted with substrate 312 to provide sol-gel layer 314 and sol-gel layer 314 is UV cured. Then, the sol-gel composition is contacted with sol-gel layer 314 to provide sol-gel layer 316 on sol-gel layer 314, and sol-gel layer 316 is UV cured. Sol-gel layer 316 is not UV cured in other examples. Then, sol-gel layers 314 and 316 are thermally cured.

FIG. 3C shows a substrate 322 with three sol-gel layers 324, 326, and 328. Sol-gel layer 324 is formed on substrate 322, sol-gel layer 326 is formed on sol-gel

layer **324** of substrate **322**, and sol-gel layer **328** is formed on sol-gel layer **326** of substrate **322**. A sol-gel composition is contacted with substrate **322** to provide sol-gel layer **324** and sol-gel layer **324** is UV cured. Then, the sol-gel composition is contacted with sol-gel layer **324** to provide sol-gel layer **326** on sol-gel layer **324**, and sol-gel layer **326** is UV cured. Then, the sol-gel composition is contacted with sol-gel layer **326** to provide sol-gel layer **328**, and sol-gel layer **328** is UV cured. Sol-gel layer **328** is not UV cured in other examples. Then, sol-gel layers **324**, **326**, and **328** are thermally cured. Although multiple sol-gel layers including two layers (FIG. 3B) and three layers (FIG. 3C) are shown, multiple sol-gel layers may include more layers in other examples.

FIG. 4 is a scanning electron microscopy (SEM) image of a surface **400** of a sol-gel layer coating a substrate. Sol-gel layer surface **400** is amorphous and no features are present except agglomerates of silica particles.

FIG. 5A is an image of a sol-gel coated substrate **500**, formed by the process of FIG. 2, after a corrosion-resistance test. Sol-gel coated substrate **500** with two sol-gel layers was exposed to a 5% salt spray. Sol-gel coated substrate **500** was able to with stand more than 168 h of salt spray. FIG. 5A shows sol-gel coated substrate **500** after 336 h of the salt spray test.

FIG. 5B is an image of a hexavalent chrome conversion coated substrate **510**, formed by conversion coating, after a corrosion-resistance test. Chromated substrate **510** was exposed to a 5% salt spray. FIG. 5B shows chrome conversion coated substrate **510** after 336 h of the salt spray test. Chrome conversion coating followed by applying a primer and paint is currently considered the state of the art for corrosion-resistant coatings for metal or metal alloy substrates. Sol-gel coated substrate **500** shown in FIG. 5A may have a corrosion resistance comparable to or better than that of hexavalent chrome conversion coated substrate **510**.

FIG. 5C is an image of an uncoated aluminum alloy substrate **520** after a corrosion-resistance test. Uncoated substrate **520** was exposed to a 5% salt spray. FIG. 5C shows uncoated substrate **520** after 336 h of the salt spray test, which was severely corroded.

FIG. 6 is an image of a primer applied sol-gel coated substrate **600** formed by process **200** of FIG. 2 after an adhesion test. A scribed wet tape adhesion test on primer applied sol-gel coated substrate **600** was performed as follows. Primer applied sol-gel coated substrate **600** was immersed in tap water for **24** h. After **24** h, primer applied sol-gel coated substrate **600** was removed and blot dry with cheesecloth. Using a metal cutting tool (e.g., a razor blade, a scalpel, a knife, or other cutting tool), two parallel **2** inch long scratches were made, **3/4** to **1** inch apart, through the coating and to the substrate. The parallel scratches were joined with two intersecting lines, or an “X” pattern. An adhesive tape was applied over the scratched pattern, and the tape was pressed against the test surface with firm hand pressure. (Alternatively, adhesive tape may be applied using a roller). One end of the tape was lifted such that a length of about **2** inches on that end of the tape was not in contact with the test surface. The minimum remaining length of the tape in contact with the test area was approximately **4** inches. The lifted-up **2** inch segment of the tape was positioned to create an approximate **45** degree angle with the test surface. The remainder of the tape was removed by pulling up and back with an abrupt motion. For this adhesion test, if the primer comes off the substrate, it implies poor adhesion. If the primer does not come off the substrate, it implies good adhesion. As shown by the result in FIG. 6, the primer of sol-gel coated substrate **600** did not come off. Thus, the sol-gel layers formed by process **200** of FIG. 2 may have good adhesion to organic paint such as primer and/or paint.

When introducing elements of the embodiments described or exemplary aspects thereof, the articles “a,” “an,” “the,” and “said” are intended to mean that there are one or more of the elements. The terms “comprising,” “including,” and “having” are intended to be inclusive and mean that there may be additional elements other than the listed elements. Although specific embodiments have been described, the details of these embodiments are not to be construed as limitations. Different aspects, embodiments and features are defined in detail herein. Each aspect, embodiment or feature so defined may be combined with any other aspect(s), embodiment(s) or feature(s) (preferred, advantageous or otherwise) unless clearly indicated to the contrary.

EMBODIMENTS IN WHICH AN EXCLUSIVE PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

1. A method of preparing a sol-gel composition, the method comprising:
 - 5 contacting a first alkoxy silane with water and an inorganic acid to form a first composition;
 - contacting a zirconium alkoxide with a first organic acid to form a second composition;
 - contacting the first composition with the second composition to form a mixture;
 - 10 contacting a phenyl alkoxy silane, an aminopropyl alkoxy silane, a glycidoxypropyl alkoxy silane, and a second organic acid with the mixture to form the sol-gel composition, the sol-gel composition having a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from about 2 to about 10; and
 - 15 adding a photoinitiator to the sol-gel composition.

2. The method of claim 1, further comprising:
 - diluting the sol-gel composition with a solvent and stirring.

- 20 3. The method of claim 1, further comprising:
 - aging the sol-gel composition by adding only an organic solvent and stirring for about 1 hour to about 24 hours.

- 25 4. The method of any one of claims 1 to 3, wherein the contacting to form the first composition comprises mixing the first alkoxy silane having the formula $R_A-Si-(R_B)_3$ with the water and the inorganic acid, wherein the R_A is methacryloxyalkyl or glycidylalkyl, and wherein the R_B is methoxy or ethoxy.

5. The method of claim 4, wherein the mixing comprises:
- adding the water to a methacryloxypropyl alkoxysilane, a ratio of a number of moles of silicon to a number of moles of water ($n_{\text{Si}}/n_{\text{water}}$) ranging from about 0.5 to about 2; and
- adding the inorganic acid and stirring.
6. The method of any one of claims 1 to 5, wherein the contacting to form the second composition comprises mixing the zirconium alkoxide having the formula $\text{Zr}-(\text{R}_C)_4$ with methacrylic acid (MAA), and wherein the R_C is ethoxy, n-propoxy, isopropoxy, n-butyloxy, or tert-butyloxy.
7. The method of claim 6, wherein the mixing comprises adding MAA to zirconium n-propoxide and stirring, a ratio of a number of moles of zirconium to a number of moles of MAA ($n_{\text{Zr}}/n_{\text{MAA}}$) ranging from about 0.5 to about 2.
8. The method of any one of claims 1 to 7, wherein the contacting to form the sol-gel composition comprises:
- adding the phenyl alkoxysilane, the aminopropyl alkoxysilane, and the glycidoxypropyl alkoxysilane; and
- adding MAA and stirring.
9. The method of any one of claims 1 to 8, wherein the adding the photoinitiator comprises adding the photoinitiator in an amount ranging from about 0.5 to about 3 parts by weight per 100 parts by weight of the sol-gel composition.

10. The method of any one of claims 1 to 9, further comprising:
- applying the sol-gel composition on a substrate a plurality of times to form a plurality of sol-gel layers;
 - curing at least one of the plurality of sol-gel layers by UV radiation; and
 - 5 thermally curing the plurality of sol-gel layers to form a corrosion-resistant coating comprising the plurality of sol-gel layers.
11. The method of claim 10, wherein the curing the at least one of the plurality of sol-gel layers by the UV radiation comprises exposing the at least one of the plurality of sol-gel layers to the UV radiation having a light dose ranging from about 500 to about 1000 mJ/cm² for a time period ranging from about 0.5 to about 30 min.
12. The method of claim 10, wherein the thermally curing the plurality of sol-gel layers comprises curing the plurality of sol-gel layers at a temperature ranging from about 70 to about 90 °C for a time period ranging from about 40 to about 120 minutes in a hot air circulated oven.
13. The method of claim 10, wherein the thermally curing the plurality of sol-gel layers comprises exposing the plurality of sol-gel layers to infrared (IR) radiation, near IR radiation, microwave radiation, or a combination thereof.
14. The method of any one of claims 10 to 13, further comprising applying primer and/or paint on the plurality of sol-gel layers on the substrate, the plurality of sol-gel layers facilitating adherence of the primer and/or the paint to the substrate, wherein the substrate comprises an aluminum alloy substrate selected from the group comprising 2024 aluminum alloy, 6061 aluminum alloy, and 7075 aluminum alloy.

15. The method of any one of claims 1 to 9, further comprising:
- contacting the sol-gel composition with a substrate to form a first layer of the sol-gel composition on the substrate;
 - 5 exposing the first layer to UV radiation;
 - contacting the sol-gel composition with the first layer to form a second layer of the sol-gel composition on the first layer;
 - exposing the second layer to the UV radiation; and
 - 10 thermally curing the first layer and the second layer to form a corrosion-resistant coating comprising the first layer and the second layer.
16. The method of claim 15, further comprising:
- contacting the sol-gel composition with the second layer to form a third layer of the sol-gel composition on the second layer; and
 - 15 exposing the third layer to the UV radiation;
- wherein the corrosion-resistant coating further comprises the third layer.
17. The sol-gel composition prepared by the method of any one of claims 1 to 9.
- 20 18. The corrosion-resistant coating formed by the method of any one of claims 10 to 16.
19. A corrosion-resistant coated product, comprising:
- 25 a plurality of ultraviolet (UV)-cured sol-gel layers on a substrate, each of the UV-cured sol-gel layers comprising a photoinitiator and a polymer composite of a methacryloxyalkyl alkoxy silane, an aryl alkoxy silane, an aminoalkyl alkoxy silane, a glycidioxyalkyl alkoxy silane, a

zirconium alkoxide, and an organic acid, and each of the UV-cured sol-gel layers having a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from about 2 to about 10.

- 5 20. The corrosion-resistant coated product of claim 19, wherein the organic acid comprises methacrylic acid (MAA).
21. A method of preparing a sol-gel composition, the method comprising:
- 10 contacting a first alkoxy silane with water and an inorganic acid to form a first composition;
- contacting a zirconium alkoxide with a first organic acid to form a second composition;
- contacting the first composition with the second composition to form a mixture;
- 15 contacting one or more second alkoxy silanes and a second organic acid with the mixture to form a sol-gel composition, the sol-gel composition having a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from about 2 to about 10;
- aging the sol-gel composition by adding an organic solvent and stirring for about 1 hour to about 24 hours; and
- 20 adding a photoinitiator to the sol-gel composition.
22. The method of claim 21, wherein the organic solvent comprises an alcohol.
- 25 23. The method of claim 22, wherein the alcohol comprises isopropanol.

24. The method of any one of claims 21 to 23, wherein the contacting to form the first composition comprises mixing the first alkoxy silane having the formula $R_A-Si-(R_B)_3$ with the water and the inorganic acid, wherein the R_A is methacryloxyalkyl or glycidioxyalkyl, and wherein the R_B is methoxy or ethoxy.

5

25. The method of claim 24, wherein the mixing comprises:

adding the water to a methacryloxypropyl alkoxy silane, a ratio of a number of moles of silicon to a number of moles of water (n_{Si}/n_{water}) ranging from about 0.5 to about 2; and

10

adding the inorganic acid and stirring.

26. The method of any one of claims 21 to 25, wherein the contacting to form the second composition comprises mixing the zirconium alkoxide having the formula $Zr-(R_C)_4$ with methacrylic acid (MAA), and wherein the R_C is ethoxy, n-propoxy, isopropoxy, n-butyloxy, or tert-butyloxy.

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27. The method of claim 26, wherein the mixing comprises adding MAA to zirconium n-propoxide and stirring, a ratio of a number of moles of zirconium to a number of moles of MAA (n_{Zr}/n_{MAA}) ranging from about 0.5 to about 2.

20

28. The method of any one of claims 21 to 27, wherein the contacting to form the sol-gel composition comprises:

adding a phenyl alkoxy silane, an aminopropyl alkoxy silane, and a glycidioxypropyl alkoxy silane; and

25

adding MAA and stirring.

29. The method of any one of claims 21 to 28, wherein the adding the photoinitiator comprises adding the photoinitiator in an amount ranging from about 0.5 to about 3 parts by weight per 100 parts by weight of the sol-gel composition.
- 5 30. The method of any one of claims 21 to 29, further comprising:
 applying the sol-gel composition on a substrate a plurality of times to form a plurality of sol-gel layers;
 curing at least one of the plurality of sol-gel layers by UV radiation; and
 thermally curing the plurality of sol-gel layers to form a corrosion-
10 resistant coating comprising the plurality of sol-gel layers.
31. The method of claim 30, wherein the curing the at least one of the plurality of sol-gel layers by the UV radiation comprises exposing the at least one of the plurality of sol-gel layers to the UV radiation having a light dose ranging from
15 about 500 to about 1000 mJ/cm² for a time period ranging from about 0.5 to about 30 min.
32. The method of claim 30, wherein the thermally curing the plurality of sol-gel layers comprises curing the plurality of sol-gel layers at a temperature
20 ranging from about 70 to about 90° C for a time period ranging from about 40 to about 120 minutes in a hot air circulated oven.
33. The method of claim 30, wherein the thermally curing the plurality of sol-gel layers comprises exposing the plurality of sol-gel layers to infrared (IR)
25 radiation, near IR radiation, microwave radiation, or a combination thereof.
34. The method of any one of claims 30 to 33, further comprising applying primer and/or paint on the plurality of sol-gel layers on the substrate, the plurality of

sol-gel layers facilitating adherence of the primer and/or the paint to the substrate, wherein the substrate comprises an aluminum alloy substrate selected from the group comprising 2024 aluminum alloy, 6061 aluminum alloy, and 7075 aluminum alloy.

5

35. The method of any one of claims 21 to 29, further comprising:

contacting the sol-gel composition with a substrate to form a first layer of the sol-gel composition on the substrate;

exposing the first layer to UV radiation;

10

contacting the sol-gel composition with the first layer to form a second layer of the sol-gel composition on the first layer;

exposing the second layer to the UV radiation; and

thermally curing the first layer and the second layer to form a corrosion-resistant coating comprising the first layer and the second layer.

15

36. The method of claim 35, further comprising:

contacting the sol-gel composition with the second layer to form a third layer of the sol-gel composition on the second layer; and

exposing the third layer to the UV radiation;

20

wherein the corrosion-resistant coating further comprises the third layer.

37. The sol-gel composition prepared by the method of any one of claims 21 to 29.

38. The corrosion-resistant coating formed by the method of any one of claims 30 to 36.

25

39. A method of preparing a sol-gel composition, the method comprising:

- contacting a first alkoxy silane with water and an inorganic acid to form a first composition;
- contacting a zirconium alkoxide with a first organic acid to form a second composition;
- contacting the first composition with the second composition to form a mixture;
- contacting one or more second alkoxy silanes and a second organic acid with the mixture to form a sol-gel composition, the sol-gel composition having a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from about 2 to about 10;
- aging the sol-gel composition by adding an organic solvent in a weight ratio of about 1:1; and
- adding a photoinitiator to the sol-gel composition.

15

40. The method of claim 39, wherein the organic solvent comprises an alcohol.

41. A method of sol-gel coating an aluminum or aluminum alloy substrate, the method comprising:

20

(i) contacting a first alkoxy silane with water and an inorganic acid to form a first composition;

(ii) contacting a zirconium alkoxide with a first organic acid to form a second composition;

25

(iii) contacting the first composition with the second composition to form a mixture;

(iv) contacting one or more second alkoxy silanes and a second organic acid with the mixture to form a sol-gel composition, the sol-gel

composition having a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from 5 to 10;

- (v) adding a photoinitiator to the sol-gel composition;
- (vi) applying the sol-gel composition on an aluminum or aluminum alloy substrate a plurality of times to form a plurality of sol-gel layers;
- (vii) curing at least one of the plurality of sol-gel layers by UV radiation; and
- (viii) thermally curing the plurality of sol-gel layers to form a corrosion-resistant coating comprising the plurality of sol-gel layers.

10 42. The method of claim 41, wherein the sol-gel composition has a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from 6 to 10.

15 43. The method of claim 41, wherein the sol-gel composition has a ratio of a number of moles of silicon to a number of moles of zirconium (n_{Si}/n_{Zr}) ranging from 8 to 10.

20 44. The method of any one of claims 41 to 43, wherein in step (i) the contacting to form the first composition comprises mixing the first alkoxy silane having the formula $R_A-Si-(R_B)_3$ with the water and the inorganic acid, wherein the R_A is methacryloxyalkyl or glycidylalkyl, and wherein the R_B is methoxy or ethoxy.

45. The method of claim 44, wherein step (i) comprises:

25 adding the water to a methacryloxypropyl alkoxy silane, a ratio of a number of moles of silicon to a number of moles of water (n_{Si}/n_{water}) ranging from 0.5 to 2; and

adding the inorganic acid and stirring.

46. The method of any one of claims 41 to 45, wherein the contacting to form the second composition comprises mixing the zirconium alkoxide having the formula $Zr-(R_C)_4$ with methacrylic acid (MAA), and wherein the R_C is ethoxy, n-propoxy, isopropoxy, n-butyloxy, or tert-butyloxy.
47. The method of claim 46, wherein step (ii) comprises adding MAA to zirconium n-propoxide and stirring, the ratio of a number of moles of zirconium to a number of moles of MAA (n_{Zr}/n_{MAA}) ranging from 0.5 to 2.
48. The method of any one of claims 41 to 47, wherein the contacting to form the sol-gel composition in step (iv) comprises:
- adding the one or more second alkoxy silanes each having the formula $R_D-Si-(R_E)_3$ to the mixture, wherein the R_D is aryl, aminoalkyl, or glycidoxyalkyl, and wherein the R_E is methoxy or ethoxy; and
 - adding MAA and stirring.
49. The method of any one of claims 41 to 48, wherein the adding the one or more second alkoxy silanes in step (iv) comprises:
- adding a phenyl alkoxy silane and stirring;
 - adding an aminopropyl alkoxy silane and stirring; and
 - adding a glycidoxypropyl alkoxy silane.
50. The method of any one of claims 41 to 49, wherein the adding the photoinitiator comprises adding the photoinitiator in an amount ranging from 0.5 to 3 parts by weight per 100 parts by weight of the sol-gel composition.

51. The method of any one of claims 41 to 50, wherein the curing the at least one of the plurality of sol-gel layers by the UV radiation in step (vii) comprises exposing the at least one of the plurality of sol-gel layers to the UV radiation having a light dose ranging from 500 to 1000 mJ/cm² for a time period ranging from 0.5 to 30 min.
52. The method of any one of claims 41 to 50, wherein the thermally curing the plurality of sol-gel layers in step (viii) comprises curing the plurality of sol-gel layers at a temperature ranging from 70 to 90 °C for a time period ranging from 40 to 120 minutes in a hot air circulated oven.
53. The method of any one of claims 41 to 52, wherein the thermally curing the plurality of sol-gel layers in step (viii) comprises exposing the plurality of sol-gel layers to infrared (IR) radiation, near IR radiation, microwave radiation, or a combination thereof.
54. The method of any one of claims 41 to 53, further comprising applying primer and/or paint on the plurality of sol-gel layers on the substrate, the plurality of sol-gel layers facilitating adherence of the primer and/or the paint to the substrate, wherein the substrate comprises an aluminum alloy substrate selected from the group comprising 2024 aluminum alloy, 6061 aluminum alloy, and 7075 aluminum alloy.
55. The method of any one of claims 41 to 54, further comprising:
- contacting the sol-gel composition with the substrate to form a first layer of the sol-gel composition on the substrate;
 - exposing the first layer to UV radiation;

contacting the sol-gel composition with the first layer to form a second layer of the sol-gel composition on the first layer;

exposing the second layer to the UV radiation; and

5 thermally curing the first layer and the second layer to form a corrosion-resistant coating comprising the first layer and the second layer.

56. The method of any one of claims 41 to 55, further comprising:

contacting the sol-gel composition with the second layer to form a third layer of the sol-gel composition on the second layer; and

10 exposing the third layer to the UV radiation;

wherein the corrosion-resistant coating further comprises the third layer.

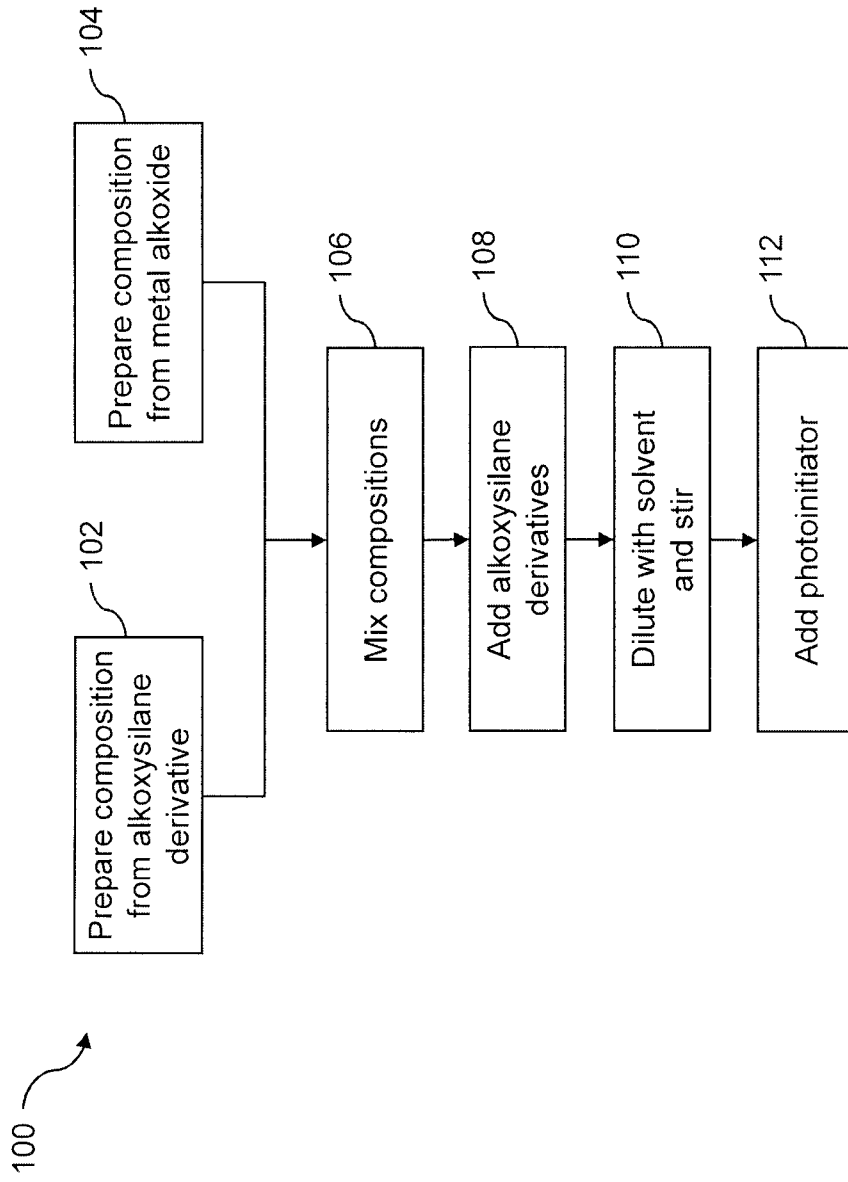


FIG. 1

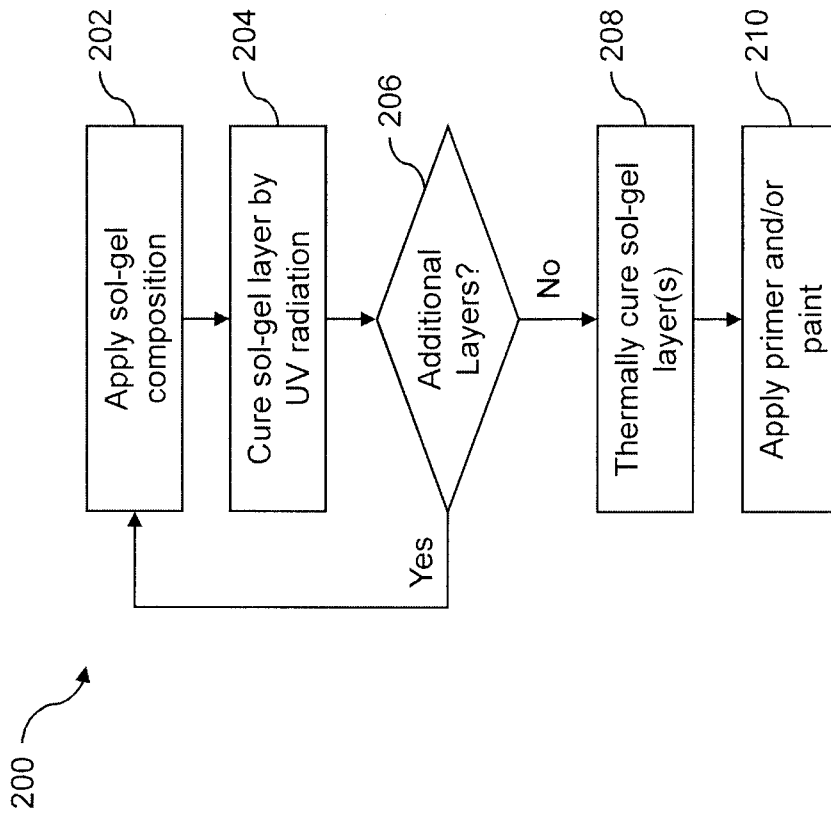


FIG. 2

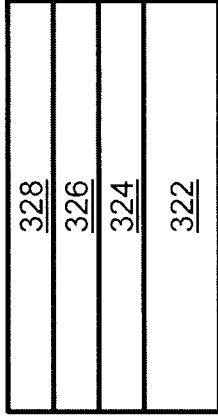


FIG. 3C

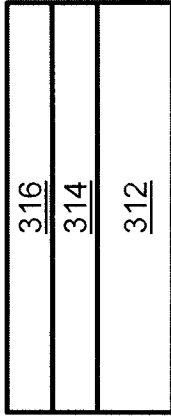


FIG. 3B

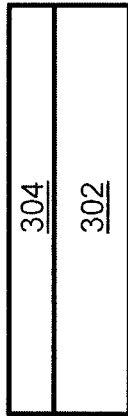


FIG. 3A

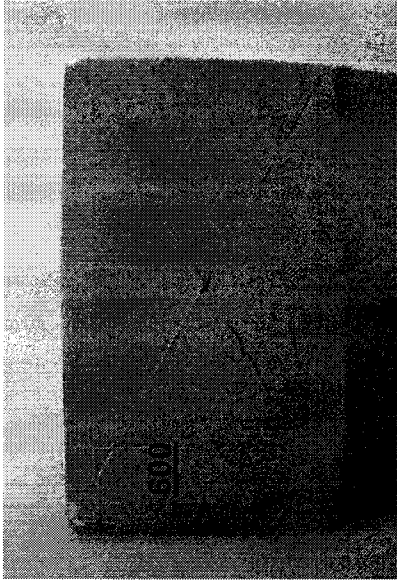


FIG. 6

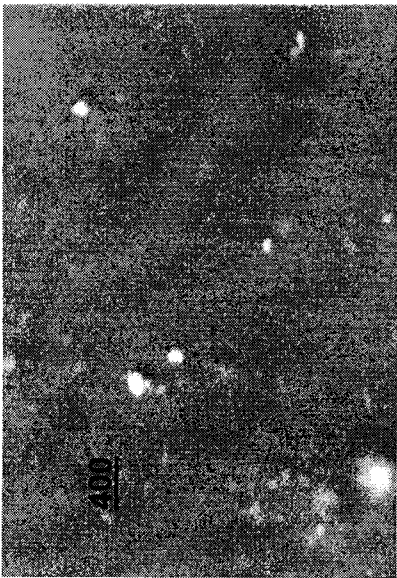


FIG. 4



FIG. 5C

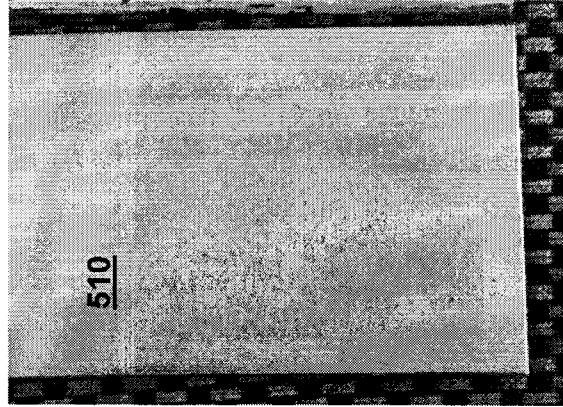


FIG. 5B

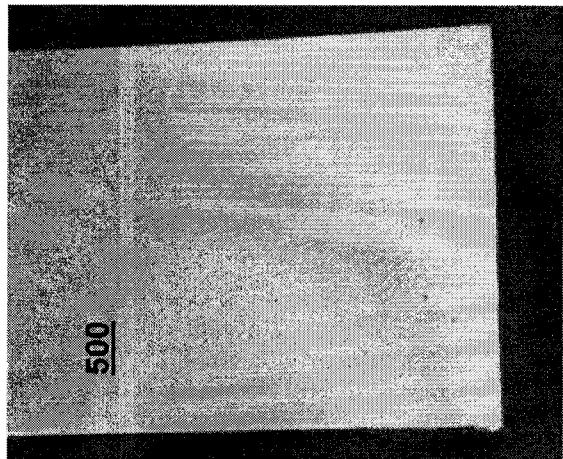


FIG. 5A