

Description

Title of Invention: SILICON PRECURSOR COMPOUND, METHOD FOR PREPARING THE SAME, AND METHOD FOR PREPARING A SILICON-CONTAINING THIN FILM

Technical Field

- [1] The present invention relates to a silicon precursor compound, to a method for preparing the same, and to a method for preparing a silicon-containing thin film using the silicon precursor compound.

[2]

Background Art

- [3] Silicon-containing thin films, for example, silicon oxide, nitride, oxynitride, and carbonitride films, are one of the essential thin films for driving microelectronic devices such as non-semiconductors (logic), as well as semiconductors.
- [4] Atomic layer deposition (ALD) or chemical vapor deposition (CVD) are widely used to prepare silicon-containing thin films. Among these, atomic layer deposition (ALD) is a method of sequentially supplying a silicon compound gas and a reactant gas necessary for film formation. It has the advantage of being able to form a silicon-containing thin film with uniform thickness even on highly uneven surfaces. Thus, atomic layer deposition (ALD) is being widely used.
- [5] Chemical vapor deposition (CVD) and atomic layer deposition (ALD) are different from each other in terms of the working mechanisms. Various silicon precursors are used to prepare silicon-containing thin films according to various process conditions depending on their physical and chemical properties.
- [6] [Prior art documents]
- [7] (Patent Document 1) Korean Laid-open Patent Publication No. 2011-0017404
- [8]

Disclosure of Invention

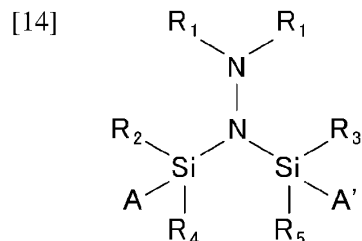
Technical Problem

- [9] An object of the present invention is to provide a novel silicon precursor compound that is advantageous for preparing silicon-containing thin films with high quality and a method for preparing the same.
- [10] In addition, another object is to provide a silicon-containing thin film using the silicon precursor compound and a method for preparing the same.
- [11]

Solution to Problem

[12] In order to accomplish the above object of the present invention, there is provided a silicon precursor compound that is characterized by being represented by the following Formula 1.

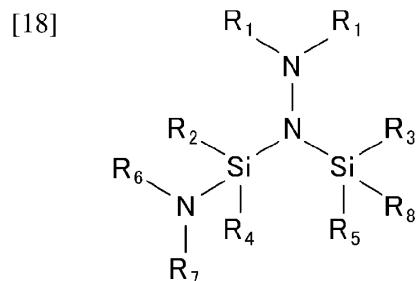
[13] [Formula 1]



[15] In Formula 1, R_1 is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R_2 to R_5 are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms; A and A' are each independently NR_6R_7 or R_8 , provided that at least one of A and A' is NR_6R_7 ; R_6 and R_7 are each independently a methyl group or an ethyl group; and R_8 is hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms.

[16] In one embodiment, the silicon precursor compound may have an asymmetric structure represented by the following Formula 2.

[17] [Formula 2]

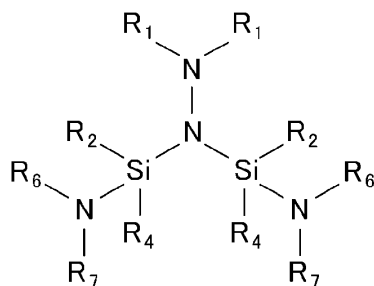


[19] In Formula 2, R_1 is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R_2 to R_5 are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms; R_6 and R_7 are each independently a methyl group or an ethyl group; and R_8 is hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms.

[20] In another embodiment, the silicon precursor compound may have a symmetric structure represented by the following Formula 3.

[21] [Formula 3]

[22]



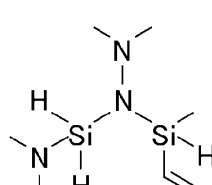
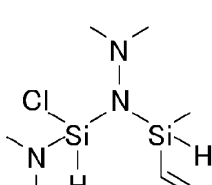
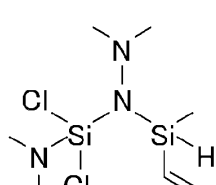
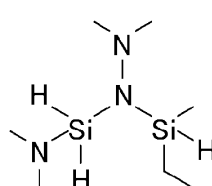
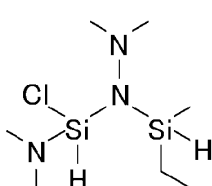
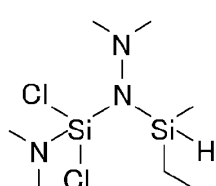
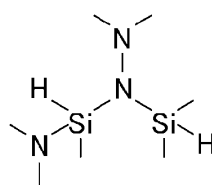
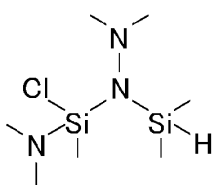
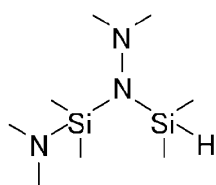
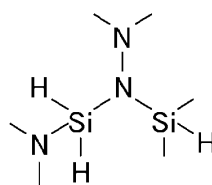
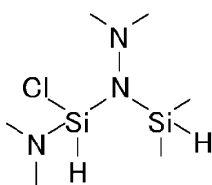
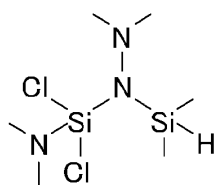
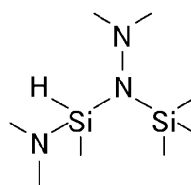
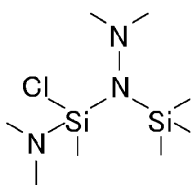
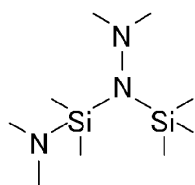
[23]

In Formula 3, R₁ is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R₂ and R₄ are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms; and R₆ and R₇ are each independently a methyl group or an ethyl group.

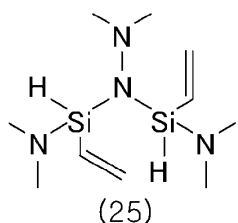
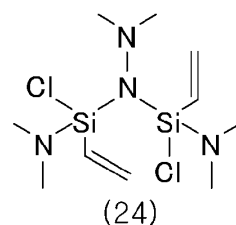
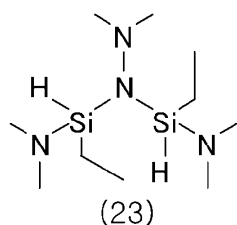
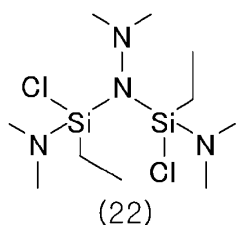
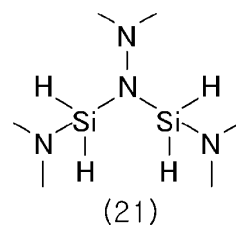
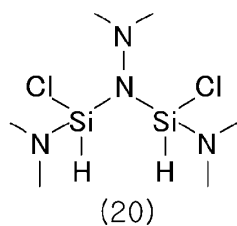
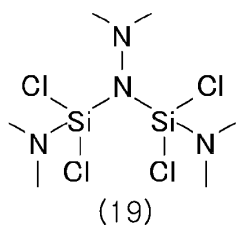
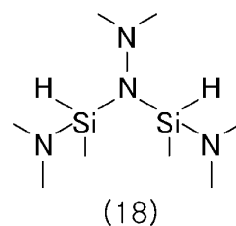
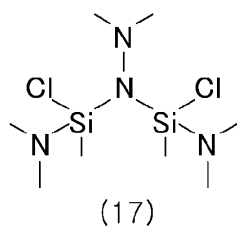
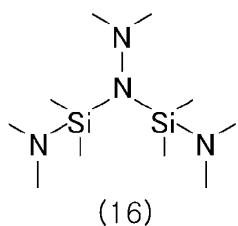
[24]

The silicon precursor compound of the present invention is preferably at least one selected from the following compounds (1) to (25).

[25]



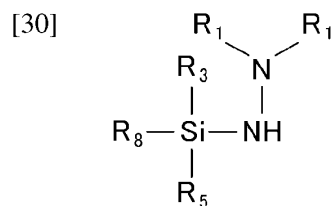
[26]



[27] In order to accomplish another object of the present invention, there is provided a method for preparing a silicon precursor compound that comprises a first step of reacting a chlorosilane derivative with any one compound selected from N,N-dimethylhydrazine, N,N-diethylhydrazine, and 1-ethyl-1-methylhydrazine as a hydrocarbyl hydrazine compound to form a first compound; a second step of reacting the first compound with an alkyl-lithium (alkyl-Li) to form a second compound containing lithium; and a third step of reacting the second compound with a silane compound to prepare a silicon precursor compound represented by Formula 2.

[28] The first compound formed in the first step is preferably a compound represented by the following Formula 9.

[29] [Formula 9]



[31] In Formula 9, R₁ is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R₃, R₅, and R₈ are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms.

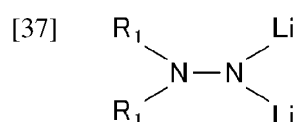
[32] In the method for preparing a silicon precursor compound of the present invention, when at least one of R₂ and R₄ in the silicon precursor compound represented by Formula 2 are halogen, the method may further comprise a fourth step of reacting it with a metal hydride as a reducing agent.

[33] In the method for preparing a silicon precursor compound of the present invention, the silane compound is produced by reacting a chlorosilane derivative with a secondary amine.

[34] Another method for preparing a silicon precursor compound of the present invention may comprise a first step of reacting a hydrocarbyl hydrazine compound with an alkyl-lithium (alkyl-Li) to form a third compound containing lithium; and a second step of reacting the third compound with a silane compound to prepare a silicon precursor compound represented by Formula 3.

[35] The third compound is preferably a compound represented by the following Formula 11.

[36] [Formula 11]



[38] In Formula 11, R₁ is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof.

[39] When at least one of R₂ and R₄ in the silicon precursor compound represented by Formula 3 are halogen, the method may further comprise a third step of reacting it with a metal hydride as a reducing agent.

[40] In order to accomplish another object of the present invention, there is provided a method for preparing a silicon-containing thin film that may comprise forming a silicon-containing thin film using the silicon precursor compound represented by Formula 1.

- [41] In the method for preparing a silicon-containing thin film of the present invention, the silicon precursor compound is preferably selected from the group consisting of compounds (1) to (25).
- [42] In the method for preparing a silicon-containing thin film of the present invention, the silicon-containing thin film may be deposited by chemical vapor deposition (CVD) or atomic layer deposition (ALD).
- [43] The silicon-containing thin film may be any one selected from the group consisting of a silicon oxide film (SiO_2), a silicon oxycarbide film (SiOC), a silicon nitride film (SiN), a silicon oxynitride film (SiON), a silicon carbonitride film (SiCN), and silicon carbonized film (SiC).
- [44] According to a particular embodiment of the method for preparing a silicon-containing thin film of the present invention, a silicon oxide film (SiO_2) may be formed by atomic layer deposition. The atomic layer deposition according to the particular embodiment comprises providing a substrate in a reactor; introducing the silicon precursor compound according to the present invention into the reactor; purging the reactor with a purge gas; introducing an oxygen-containing source into the reactor to react with the silicon precursor compound according to the present invention to form a silicon oxide film; and purging the reactor with the purge gas.
- [45] The purge gas according to a particular embodiment is used to remove unconsumed reactants and reaction by-products. Preferably, it is selected from the group consisting of nitrogen, helium, argon, and mixtures thereof, but it is not limited thereto.
- [46] The oxygen-containing source according to the particular embodiment is preferably selected from the group consisting of an oxygen, peroxide, an oxygen plasma, a water vapor, water vapor plasma, hydrogen peroxide, ozone source, and mixtures thereof, but it is not limited thereto. Preferably, the oxygen-containing source may comprise plasma, and the plasma may be generated *in situ*.
- [47] In addition, the atomic layer deposition method according to the particular embodiment may be carried out at one or more temperatures of about 500°C or lower. In such case, a person of ordinary skill in the art may appropriately select the lower limit of temperature depending on the type of the oxygen-containing source used. Preferably, a silicon oxide film may be formed at one or more temperatures of about 300°C to 450°C .
- [48] According to another particular embodiment of the method for preparing a silicon-containing thin film of the present invention, a silicon nitride (SiN) film may be formed by plasma-enhanced atomic layer deposition. The plasma-enhanced atomic layer deposition according to the particular embodiment may comprise providing a substrate in a reactor; introducing the silicon precursor compound according to the present invention into the reactor; purging the reactor with a purge gas; introducing a

nitrogen-containing plasma source and an inert gas into the reactor to react with the silicon precursor compound according to the present invention to form a silicon nitride film; and purging the reactor with the purge gas.

[49] The purge gas according to the particular embodiment is used to remove unconsumed reactants and reaction by-products. Preferably, it is selected from the group consisting of nitrogen, helium, argon, and mixtures thereof, but it is not limited thereto.

[50] The nitrogen-containing plasma source according to the particular embodiment is preferably selected from the group consisting of nitrogen plasma, nitrogen and argon mixed plasma (nitrogen and argon plasma), ammonia plasma, nitrogen and ammonia mixed plasma (nitrogen and ammonia plasma), ammonia and helium mixed plasma (ammonia and helium plasma), ammonia and argon mixed plasma (ammonia and argon plasma), ammonia and nitrogen mixed plasma (ammonia and nitrogen plasma), NF_3 plasma, organic amine plasma, and mixtures thereof, but it is not limited thereto.

[51] In addition, the plasma-enhanced atomic layer deposition method according to the particular embodiment may be carried out at one or more temperatures of about 400°C or lower. In such case, a person of ordinary skill in the art may appropriately select the lower limit of temperature depending on the type of the nitrogen-containing plasma source used. Preferably, a silicon nitride film may be formed at one or more temperatures of about 250°C to about 400°C, more preferably, at a temperature of about 350°C.

[52]

Advantageous Effects of Invention

[53] The silicon precursor compound of the present invention exhibits sufficient volatility to be applied to all of atomic layer deposition (ALD) and chemical vapor deposition (CVD) for preparing silicon-containing thin films. In particular, deposition is possible even at high temperatures in a high deposition rate, whereby a silicon-containing thin film with excellent quality can be prepared.

[54] However, the effects of the present invention are not limited to those mentioned above.

[55]

Brief Description of Drawings

[56] Fig. 1 shows a hydrogen nuclear magnetic resonance (^1H -NMR) spectrum of the silicon precursor compound prepared according to Example 1 of the present invention.

[57] Fig. 2 shows a hydrogen nuclear magnetic resonance (^1H -NMR) spectrum of the silicon precursor compound prepared according to Example 2 of the present invention.

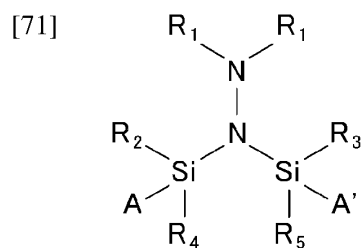
[58] Fig. 3 shows a hydrogen nuclear magnetic resonance (^1H -NMR) spectrum of the silicon precursor compound prepared according to Example 3 of the present invention.

- [59] Fig. 4 is a graph showing the results of thermogravimetric analysis (TGA) of the silicon precursor compounds prepared according to the Examples of the present invention.
- [60] Fig. 5 is a graph showing the results of measuring the vapor pressure of the silicon precursor compounds prepared according to Examples 1, 2, and 3 of the present invention.
- [61] Fig. 6 is a graph showing the sequence of each pulse in the deposition process of a silicon nitride film.
- [62] Fig. 7 is a graph showing the deposition rate of a silicon nitride film with respect to substrate temperature.
- [63] Fig. 8 is a graph showing the sequence of each pulse in the deposition process of a silicon oxide thin film.
- [64] Fig. 9 is a graph showing the deposition rate of a silicon oxide thin film with respect to substrate temperature.
- [65] Fig. 10 is an image of a silicon oxide thin film deposited according to the present invention observed using a transmission electron microscope (TEM).
- [66] Fig. 11 is a result of AES (Auger Electron Spectroscopy) analysis of the composition of a silicon oxide thin film deposited according to the present invention.
- [67]

Best Mode for Carrying out the Invention

- [68] Hereinafter, the silicon precursor compound, the method for preparing the same, the silicon-containing thin film using the silicon precursor compound, and the method for preparing the same are described in detail.
- [69] The silicon precursor compound of the present invention may be represented by the following Formula 1.

[70] [Formula 1]



- [72] In Formula 1, R₁ is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R₂ to R₅ are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms; A and A' are each independently NR₆R₇ or R₈, provided that at least one of A and A' is NR₆R₇; R₆ and R₇ are each

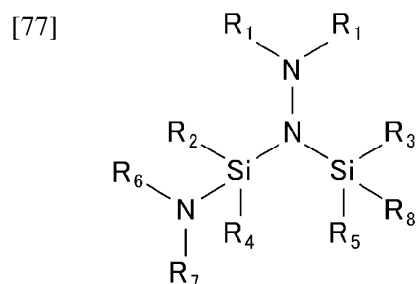
independently a methyl group or an ethyl group; and R_8 is hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms.

[73] NR_6R_7 in A and A' is an amine group. Preferably it may be any one selected from dimethylamine, ethylmethylamine, and diethylamine.

[74] The silicon precursor compound of the present invention may have an asymmetric structure or a symmetric structure.

[75] When the silicon precursor compound has an asymmetric structure, it may be represented by the following Formula 2.

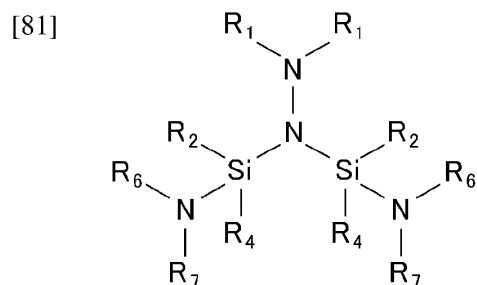
[76] [Formula 2]



[78] In Formula 2, R_1 is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R_2 to R_5 are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms; R_6 and R_7 are each independently a methyl group or an ethyl group; and R_8 is hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms.

[79] When the silicon precursor compound has a symmetric structure, it may be represented by the following Formula 3.

[80] [Formula 3]



[82] in Formula 3, R_1 is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R_2 and R_4 are each independently hydrogen (H), halogen, or a linear or branched, saturated or

unsaturated hydrocarbon group having 1 to 4 carbon atoms; and R_6 and R_7 are each independently a methyl group or an ethyl group.

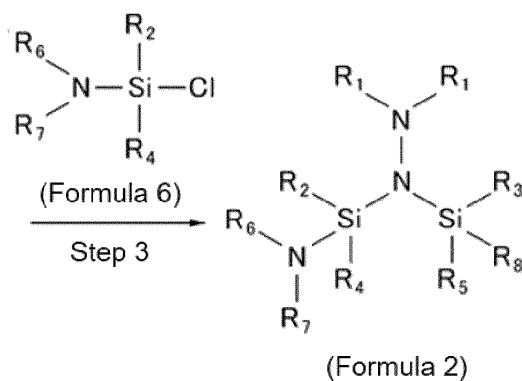
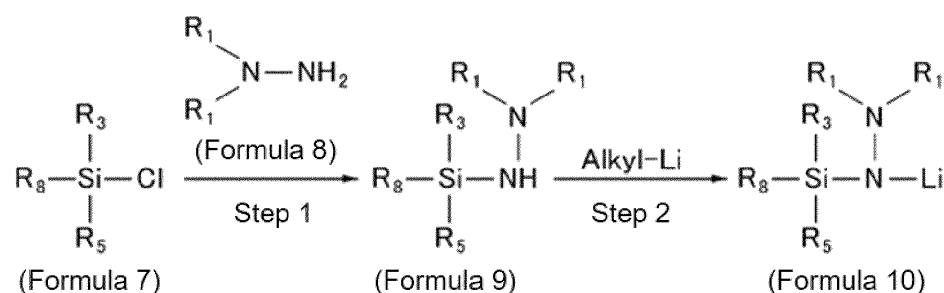
[83] The hydrocarbon groups in Formulae 1 to 3 may each independently be any one selected from the group consisting of a methyl group, an ethyl group, an n-propyl group, an iso-propyl group, a n-butyl group, an iso-butyl group, a sec-butyl group, a tert-butyl group, an ethenyl group, and isomers thereof. Preferably, any one selected from the group consisting of a methyl group, an ethyl group, and an ethenyl group may be used.

[84] The halogen may be any one selected from the group consisting of chlorine (Cl), bromine (Br), iodine (I), and fluorine (F). Chlorine (Cl) among them is preferably used.

[85] When the silicon precursor compound of the present invention has an asymmetric structure, the silicon precursor compound may be prepared by a method as shown in Reaction Scheme 1 or Reaction Scheme 2 below.

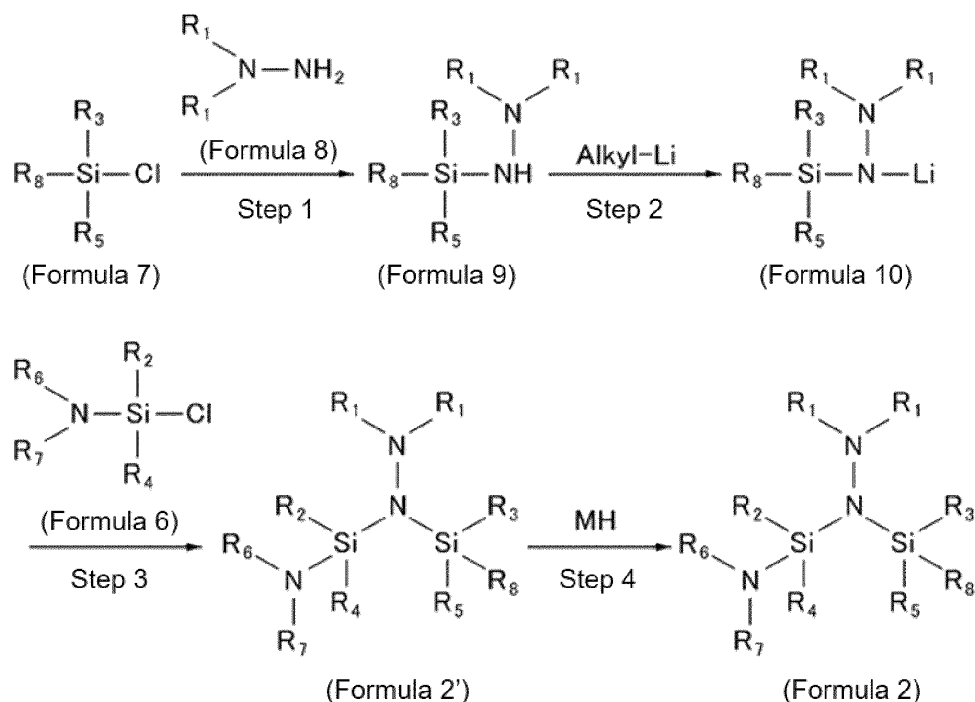
[86] [Reaction Scheme 1]

[87]



[88] [Reaction Scheme 2]

[89]

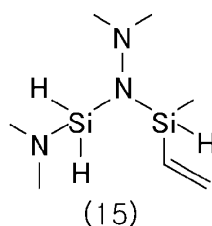
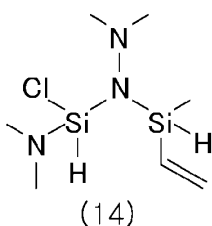
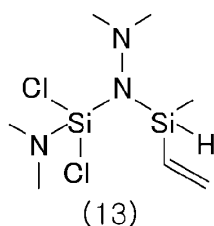
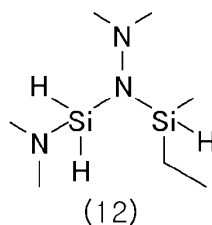
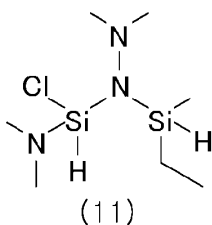
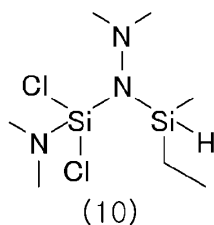
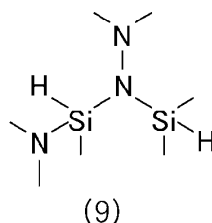
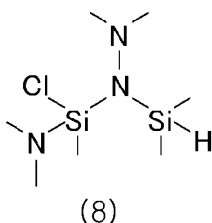
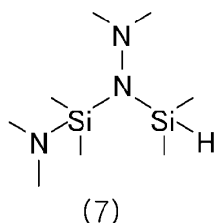
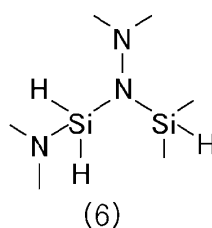
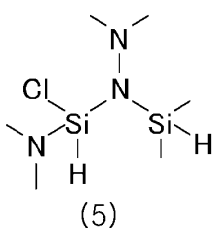
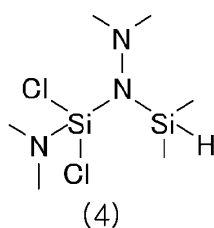
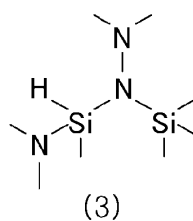
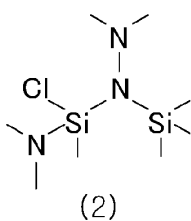
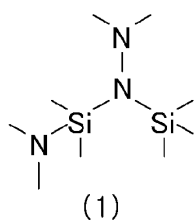


- [90] In Reaction Schemes 1 and 2, R_1 to R_8 are as defined above, and MH is a metal hydride.
- [91] In the present specification, the term the first compound, second compound, and third compound are used only for the purpose of distinguishing compounds formed in each step in the method for preparing a silicon precursor compound.
- [92] The method for preparing a silicon precursor compound according to Reaction Scheme 1 is sequentially carried out by a first step of reacting a chlorosilane derivative represented by Formula 7 with a hydrocarbyl hydrazine compound represented by Formula 8 to form a first compound represented by Formula 9; a second step of reacting the first compound with an alkyl-lithium (Alkyl-Li) to form a second compound represented by Formula 10; and a third step of reacting the second compound with a silane compound represented by Formula 6 to prepare a silicon precursor compound represented by Formula 2.
- [93] In the method for preparing a silicon precursor compound, in the first step, a triorganochlorosilane as a chlorosilane derivative is reacted with a hydrocarbyl hydrazine compound represented by Formula 8 at a low temperature of about -20°C in a non-polar solvent to substitute Cl with an amine, followed by filtration and distillation under a reduced pressure to form a compound of Formula 9.
- [94] In the second step, the compound of Formula 9 formed in the first step is reacted with an alkyl-lithium (alkyl-Li) at a low temperature of about -20°C in a non-polar solvent to perform a lithiation to form a compound of Formula 10.

- [95] The alkyl-Li is lithium containing an alkyl group having 1 to 10 carbon atoms. Examples thereof include methyl lithium, ethyl lithium, propyl lithium, butyl lithium, and isobutyl lithium, etc.
- [96] In the third step, after a reaction with a silane compound represented by Formula 6 to form a silicon precursor compound, the salt (LiCl) as a product of the reaction and unreacted materials are removed through filtration, and distillation under a reduced pressure is performed to obtain a silicon precursor compound having an asymmetric structure as represented by Formula 2.
- [97] Another method for preparing a silicon precursor compound, as shown Reaction Scheme 2, is carried out in the same manner as in Reaction Scheme 1 above, in which when at least one of R_2 and R_4 in the silicon precursor compound represented by Formula 2' in Reaction Scheme 2 are halogen selected from fluorine (F), chlorine (Cl), bromine (Br), or iodine (I), a fourth step of reacting it with a metal hydride (MH) as a reducing agent is further carried out.
- [98] The metal hydride (MH) may be at least one selected from the group consisting of lithium hydride (LiH), sodium hydride (NaH), lithium borohydride ($LiBH_4$), lithium aluminum hydride ($LiAlH_4$), sodium aluminum hydride ($NaAlH_4$), and sodium Lithium borohydride ($NaBH_4$). As an example, lithium hydride (LiH) as a metal hydride (MH) together with tetrahydrofuran (THF) may be used to perform the reduction reaction.
- [99] Preferably, the fourth step is carried out when at least one of R_2 and R_4 in Formula 2' are chlorine (Cl).
- [100] In the methods for preparing a silicon precursor compound according to Reaction Schemes 1 and 2, the silane compound of Formula 6 may be prepared as shown in Reaction Scheme 3 below. Specifically, a chlorosilane derivative represented by Formula 4 is reacted with a secondary amine represented by Formula 5 at a low temperature of about -20°C in a non-polar solvent to substitute Cl with an amine, followed by filtration and distillation under a reduced pressure to form a compound of Formula 6.
- [101] [Reaction Scheme 3]
- [102]
- The reaction scheme shows the reaction of a chlorosilane derivative (Formula 4) with a secondary amine (Formula 5) to form a silicon precursor compound (Formula 6).
 Formula 4: A central silicon atom (Si) is bonded to two chlorine atoms (Cl) and two R groups, R_2 and R_4 .
 Formula 5: A secondary amine with a central nitrogen atom (N) bonded to two R groups, R_6 and R_7 , and one hydrogen atom (H).
 The reaction arrow is labeled "(Formula 5)".
 Formula 6: The product is a silicon precursor compound where the two chlorine atoms from Formula 4 have been replaced by the R_6 and R_7 groups from Formula 5. The central silicon atom (Si) is now bonded to R_6 , R_7 , R_2 , and R_4 .
- [103] In Reaction Scheme 3, R_2 , R_4 , R_6 , and R_7 are as defined above.

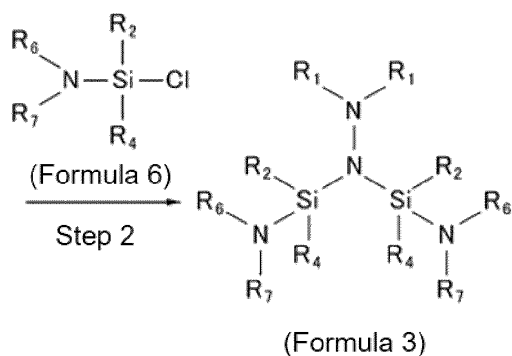
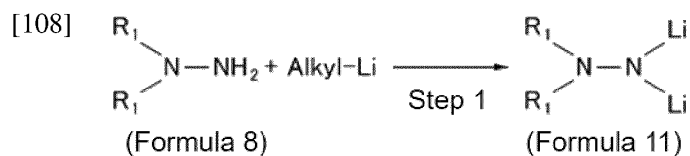
[104] The silicon precursor compound having an asymmetric structure is at least one selected from the following compounds (1) to (15).

[105]

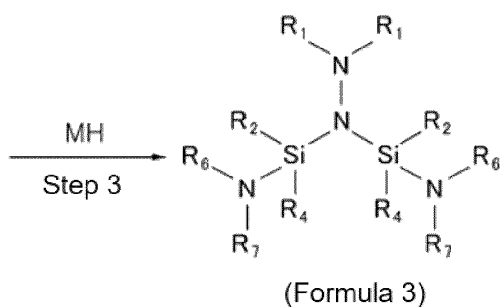
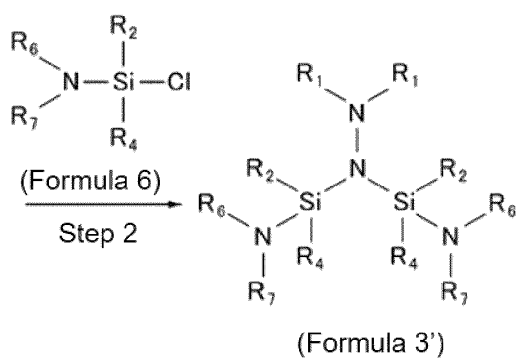
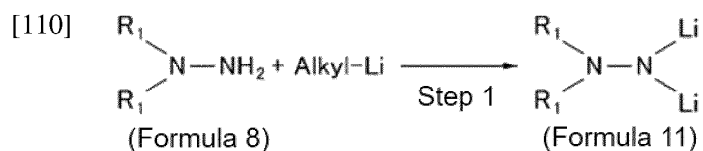


[106] When the silicon precursor compound of the present invention has a symmetric structure, the silicon precursor compound may be prepared by a method as shown in Reaction Scheme 4 or Reaction Scheme 5 below.

[107] [Reaction Scheme 4]



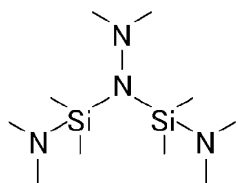
[109] [Reaction Scheme 5]



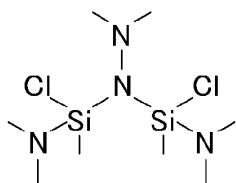
[111] In Reaction Schemes 4 and 5, R_1 to R_7 are as defined above, and MH is a metal hydride.

- [112] The method for preparing a silicon precursor compound according to Reaction Scheme 4 is sequentially carried out by a first step of reacting a hydrocarbyl hydrazine compound of Formula 8 with an alkyl-lithium (alkyl-Li) to form a third compound represented by Formula 11; and a second step of reacting the third compound with a silane compound represented by Formula 6 to prepare a silicon precursor compound represented by Formula 3.
- [113] In the method for preparing a silicon precursor compound, in the first step, a hydrocarbyl hydrazine compound of Formula 8 is reacted with an alkyl-lithium (alkyl-Li) at a low temperature of about -20°C in a non-polar solvent to perform a substitution reaction of an amine and Li to form a compound of Formula 11.
- [114] In the second step, after a reaction with a silane compound represented by Formula 6 to form a silicon precursor compound, the salt (LiCl) as a product of the reaction and unreacted materials are removed through filtration, and distillation under a reduced pressure is performed to obtain a silicon precursor compound having a symmetric structure as represented by Formula 3.
- [115] Still another method for preparing a silicon precursor compound, as shown Reaction Scheme 5, is carried out in the same manner as in Reaction Scheme 4 above, in which when at least one of R₂ and R₄ in the silicon precursor compound represented by Formula 3' in Reaction Scheme 3 are halogen selected from fluorine (F), chlorine (Cl), bromine (Br), or iodine (I), a third step of reacting it with a metal hydride (MH) as a reducing agent is further carried out.
- [116] Preferably, the third step is carried out when at least one of R₂ and R₄ in Formula 3' are chlorine (Cl).
- [117] As the non-polar solvent used in Reaction Schemes 1 to 5, hexane, n-pentane, or the like may be used, but it is not limited thereto. A non-polar solvent commonly used by a person of ordinary skill in the art can be used.
- [118] The silicon precursor compound having a symmetric structure is at least one selected from the following compounds (16) to (25).

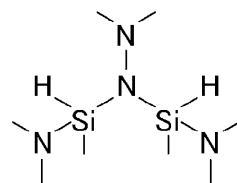
[119]



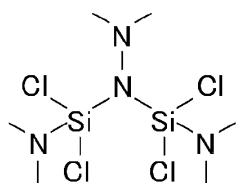
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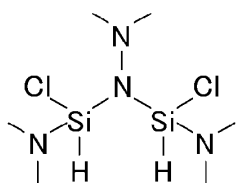
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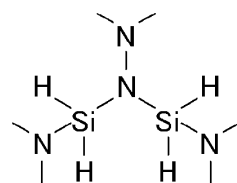
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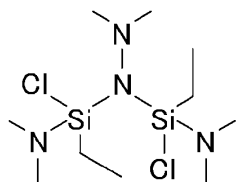
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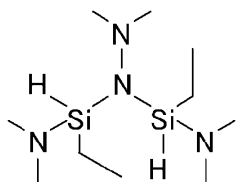
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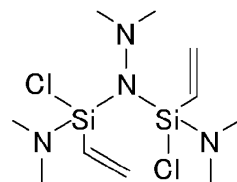
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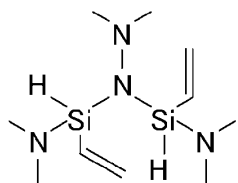
(22)



(23)



(24)



(25)

[120] In addition, in the method for preparing a silicon-containing thin film of the present invention, a silicon-containing thin film can be formed using the silicon precursor compound represented by Formula 1 by chemical vapor deposition (CVD) or atomic layer deposition (ALD).

[121] The silicon-containing thin film may be any one selected from a silicon oxide film (SiO_2), a silicon oxycarbide film (SiOC), a silicon nitride film (SiN), a silicon oxynitride film (SiON), a silicon carbonitride film (SiCN), and silicon carbonized film (SiC).

[122]

Mode for the Invention

[123] Hereinafter, the present invention will be described in more detail with reference to examples.

[124]

[125] **[Example 1]**

- [126] In Example 1, dimethyltrimethylsilylhydrazinyltrimethylsilanamine of compound (3) was prepared as a silicon precursor compound having an asymmetric structure.
- [127] First, for the preparation of a silane compound required to prepare a silicon precursor compound, a 1-liter flask in an anhydrous and inert atmosphere was charged with 360 g (3.13 moles) of dichloromethylsilane ($\text{CH}_3\text{SiHCl}_2$) and 4,516 g (62.59 moles) of n-pentane. While the temperature was maintained at about -20°C , 282 g (6.26 moles) of dimethylamine ($((\text{CH}_3)_2\text{NH})$) was slowly added thereto and stirred for 3 hours. Upon completion of the stirring, the dimethylamine hydrochloride salt ($((\text{CH}_3)_2\text{NH}_2\text{Cl})$) was removed through filtration, followed by removing the solvent under a reduced pressure and distillation to obtain 312 g (2.52 moles) of chlorodimethylaminomethylsilazane ($(\text{CH}_3)_2\text{NCH}_3\text{SiHCl}$) (yield: 80%).
- [128] $^1\text{H-NMR}(\text{C}_6\text{D}_6)$: δ 0.23(d, 3H (-Si CH_3)), 2.25(s, 6H (-N(CH_3) $_2$)), 5.0(m, 1H (-SiH))
- [129] As a first step, for the preparation of dimethyl(trimethylsilyl)hydrazine, a 1-liter flask in an anhydrous and inert atmosphere was charged with 190 g (1.75 moles) chlorotrimethylsilane ($((\text{CH}_3)_3\text{SiCl})$) and 3,387 g (46.94 moles) n-pentane. While the temperature was maintained at about -20°C , 210 g (3.50 moles) of dimethylhydrazine ($((\text{CH}_3)_2\text{NNH}_2)$) was slowly added thereto and stirred for 12 hours. Upon completion of the stirring, the dimethylhydrazine hydrochloride salt ($((\text{CH}_3)_2\text{NNH}_3\text{Cl})$) was removed through filtration, followed by removing the solvent under a reduced pressure and distillation to obtain 161 g (1.22 moles) of dimethyl(trimethylsilyl)hydrazine ($((\text{CH}_3)_2\text{NNHSi}(\text{CH}_3)_3)$) (yield: 70%).
- [130] $^1\text{H-NMR}(\text{C}_6\text{D}_6)$: δ 0.11(s, 9H (-Si(CH_3) $_3$)), 1.72(m, 1H (-NH $\text{Si}(\text{CH}_3)_2$)), 2.21(s, 6H (-NN(CH_3) $_2$))
- [131] Next, as a second step, a 1-liter flask in an anhydrous and inert atmosphere was charged with 76 g (0.58 mole) of dimethyl(trimethylsilyl)hydrazine ($((\text{CH}_3)_2\text{NNHSi}(\text{CH}_3)_3)$) and 100 g (1.16 moles) of hexane. While the temperature was maintained at about -20°C , 245 ml (1.05 moles) of 2.5 M n-BuLi was slowly added thereto, and the reaction solution was then gradually heated to room temperature and stirred at room temperature for 12 hours.
- [132] While the temperature of the mixed solution was maintained at about -20°C , 71 g (0.58 mole) of chlorodimethylaminomethylsilazane ($((\text{CH}_3)_2\text{NCH}_3\text{SiHCl})$) was slowly added thereto and stirred at room temperature for 6 hours or longer. Upon completion of the stirring, the lithium chloride (LiCl) salt was removed through filtration. The solvent was removed from the resulting solution under a reduced pressure and purified to obtain 101 g (yield: 80%) of dimethyltrimethylsilylhydrazinyltrimethylsilanamine ($((\text{CH}_3)_2\text{NCH}_3\text{SiHN}(\text{CH}_3)_2\text{NSi}(\text{CH}_3)_3)$).

- [133] Fig. 1 shows a hydrogen nuclear magnetic resonance ($^1\text{H-NMR}$) spectrum of the silicon precursor compound prepared according to Example 1 of the present invention. As shown, the silicon precursor compound prepared in Example 1 was confirmed to be dimethyltrimethylsilylhydrazinyltrimethylsilanamine.
- [134] $^1\text{H-NMR}(\text{C}_6\text{D}_6)$: δ 0.21(s, 9H (-Si(**CH**₃)₃)), 0.25(d, 3H (-SiH**CH**₃)), 2.44(s, 6H (-SiHCH₃N(**CH**₃)₂)), 2.51(s, 6H (-NN(**CH**₃)₂)), 4.90(m, 1H (-Si**H**CH₃N(CH₃)₂))
- [135]
- [136] **[Example 2]**
- [137] In Example 2, dimethylsilyldimethylhydrazinyltrimethylsilanamine of compound (9) was prepared as a silicon precursor compound having an asymmetric structure.
- [138] As a first step, for the preparation of (dimethylsilyl)dimethylhydrazine, a 1-liter flask in an anhydrous and inert atmosphere was charged with 100 g (1.06 moles) chlorodimethylsilane ((CH₃)₂SiHCl) and 100 ml of diethyl ether. While the temperature was maintained at about -20°C, 130.2 g (2.17 moles) of dimethylhydrazine ((CH₃)₂NNH₂) was slowly added thereto and stirred for 12 hours. Upon completion of the stirring, the dimethylhydrazine hydrochloride salt ((CH₃)₂NNH₃Cl) was removed through filtration, followed by removing the solvent under a reduced pressure and distillation to obtain 85 g (0.72 mole) of (dimethylsilyl)dimethylhydrazine ((CH₃)₂NNHSiH(CH₃)₂) (yield: 68%).
- [139] $^1\text{H-NMR}(\text{C}_6\text{D}_6)$: δ 0.15(d, 6H (-SiH(**CH**₃)₂)), 1.77(s, 1H (-**NH**SiH(CH₃)₂)), 2.20(s, 6H (-NN(**CH**₃)₂)), 4.71(m, 1H (-Si**H**(CH₃)₂))
- [140] Next, as a second step, a 1-liter flask in an anhydrous and inert atmosphere was charged with 121 g (0.41 mole) of (dimethylsilyl)dimethylhydrazine ((CH₃)₂NNHSiH(CH₃)₂) and 108 g (0.82 mole) of hexane. While the temperature was maintained at about -20°C, 173.3 ml (0.43 mole) of 2.5 M n-BuLi was slowly added thereto, and the reaction solution was then gradually heated to room temperature and stirred at room temperature for 12 hours.
- [141] While the temperature of the mixed solution was maintained at about -20°C, 71 g (0.41 mole) of chlorodimethylaminomethylsilazane ((CH₃)₂NCH₃SiHCl) was slowly added thereto and stirred at room temperature for 6 hours or longer. Upon completion of the stirring, the lithium chloride (LiCl) salt was removed through filtration. The solvent was removed from the resulting solution under a reduced pressure and purified to obtain 69 g (yield: 82%) of dimethylsilyldimethylhydrazinyltrimethylsilanamine ((CH₃)₂NCH₃SiHN(CH₃)₂NSiH(CH₃)₂).
- [142] Fig. 2 shows a hydrogen nuclear magnetic resonance ($^1\text{H-NMR}$) spectrum of the silicon precursor compound prepared according to Example 2 of the present invention.

As shown, the silicon precursor compound prepared in Example 2 was confirmed to be dimethylsilyldimethylhydrazinyltrimethylsilanamine.

- [143] $^1\text{H-NMR}(\text{C}_6\text{D}_6)$: δ 0.25(m, 9H ($(\text{CH}_3)_2\text{NCH}_3\text{SiHN}(\text{CH}_3)_2\text{NSiH}(\text{CH}_3)_2$)), 2.46(s, 6H ($-\text{SiHCH}_3\text{N}(\text{CH}_3)_2$)), 2.48(s, 6H ($-\text{NN}(\text{CH}_3)_2$)), 4.81(m, 1H ($-\text{SiH}(\text{CH}_3)_2$)), 4.82(m, 1H ($-\text{SiHCH}_3\text{N}(\text{CH}_3)_2$))

[144]

[145] **[Example 3]**

- [146] In Example 3, bistrimethylsilaaminodimethylhydrazine of compound (18) was prepared as a silicon precursor compound having a symmetric structure.

- [147] A 1-liter flask in an anhydrous and inert atmosphere was charged with 100 g (1.63 moles) of dimethylhydrazine ($(\text{CH}_3)_2\text{NNH}_2$) and 281 g (3.26 moles) of hexane. While the temperature was maintained at about -20°C , 1,315 ml (3.26 moles) of 2.5 M n-BuLi was slowly added thereto, and the reaction solution was then gradually heated to room temperature and stirred at room temperature for 12 hours.

- [148] While the temperature of the mixed solution was maintained at about -20°C , 403 g (3.26 moles) of chlorodimethylaminomethylsilazane ($(\text{CH}_3)_2\text{CH}_3\text{NSiHCl}$) was slowly added thereto and stirred at room temperature for 6 hours or longer. Upon completion of the stirring, the lithium chloride (LiCl) salt was removed through filtration. The solvent was removed from the resulting solution under a reduced pressure and purified to obtain 268 g (yield: 70%) of bistrimethylsilaaminodimethylhydrazine ($(\text{CH}_3)_2\text{NSiHCH}_3)_2\text{NN}(\text{CH}_3)_2$).

- [149] Fig. 3 shows a hydrogen nuclear magnetic resonance ($^1\text{H-NMR}$) spectrum of the silicon precursor compound prepared according to Example 3 of the present invention. As shown, the silicon precursor compound prepared in Example 3 was confirmed to be bistrimethylsilaaminodimethylhydrazine.

- [150] $^1\text{H-NMR}(\text{C}_6\text{D}_6)$: δ 0.30(s, 6H ($-\text{SiHCH}_3\text{N}(\text{CH}_3)_2$)), 2.50(m, 18H ($((\text{CH}_3)_2\text{NSiHCH}_3)_2\text{NN}(\text{CH}_3)_2$)), 4.79(m, 2H ($(\text{CH}_3)_2\text{NSiHCH}_3)_2\text{NN}-$))

- [151] Thermogravimetric analysis (TGA) was carried out to analyze the thermal characteristics of the silicon precursor compounds prepared in Examples 1 to 3. The results are shown in Fig. 4.

- [152] As shown in Fig. 4, the silicon precursor compounds of Examples 1 to 3 were volatile even at a temperature of 200°C or lower, confirming that they are excellent precursors capable of forming silicon-containing thin films in various temperature ranges.

- [153] The above results show that all of the silicon precursor compounds prepared according to Examples 1 to 3 show sufficient volatility to be applied to atomic layer deposition (ALD) or chemical vapor deposition (CVD).

- [154] In order to confirm that the silicon precursor compounds prepared according to Examples 1 to 3 had a vapor pressure suitable for the preparation of a silicon thin film through a deposition method, their vapor pressures were measured. The results are shown in Fig. 5.
- [155] As shown in Fig. 5, the silicon precursor compounds of Examples 1 to 3 all showed a high vapor pressure of 10 Torr at about 90°C.
- [156] The above vapor pressure results show that all of the silicon precursor compounds prepared according to Examples 1 to 3 show a high vapor pressure at a low temperature of about 90°C or lower, indicating a sufficient vapor pressure to be applied to atomic layer deposition (ALD) or chemical vapor deposition (CVD).
- [157] To evaluate silicon-containing thin films formed using the silicon precursor compounds prepared according to the present invention, bistrimethylsilylaminodimethylhydrazine, the silicon precursor compound of Example 3, was used to form a silicon nitride thin film on a silicon substrate using atomic layer deposition (ALD). In such an event, an ALD reactor in which a silicon precursor compound and a reaction gas are separately supplied in a vertical direction using a double shower head was used.
- [158] Table 1 below and Fig. 6 show the specific conditions for depositing a silicon nitride thin film.
- [159] [Table 1]

Source	Substrate temp.	Precursor heating	Precursor feeding	Purge		N ₂ plasma		Purge		No. of depositions
	(°C)	(°C)	Time (sec)	Flow rate (sccm)	Time (sec)	Flow rate (sccm)	Time (sec)	Flow rate (sccm)	Time (sec)	
Ex. 3	250-400	60	5	1,000	7	1,000	7	1,000	10	100

- [160]
- [161] The thickness of the thin film deposited using the above method was confirmed using an ellipsometer. Table 2 below and Fig. 7 show the results of analyzing the characteristics of the specific silicon nitride thin film.
- [162] [Table 2]

Source	Substrate temp.	Deposition rate	Film thickness	N/Si
	(°C)	(Å/cycle)	(Å)	Composition ratio
Ex. 3	350	0.45	70	2.47

- [163]

- [164] As shown in Table 2, the composition ratio of N/Si at a substrate temperature of 350°C indicates that a silicon-containing thin film with high purity was formed. In addition, Fig. 7 is a graph showing the deposition rate of a silicon nitride film with respect to substrate temperature. As shown in Fig. 7, the deposition rate was similar under the conditions of 250°C to 400°C.
- [165] As another example, to evaluate silicon oxide thin films (SiO₂) formed using the silicon precursor compound prepared according to the present invention, bistrimethylsilylaminodimethylhydrazine, the silicon precursor compound of Example 3, was used to form a silicon oxide thin film on a silicon substrate using atomic layer deposition (ALD). In such an event, an ALD reactor in which a silicon precursor compound and a reaction gas are separately supplied in a vertical direction using a double shower head was used.
- [166] In the present specification, SiN and SiO₂ are prepared and described as representative silicon-containing thin films, but it is not limited thereto. Silicon thin films known in the art such as SiN, SiO₂, SiON, SiCN, and SiC can be formed.
- [167] Table 3 below and Fig. 8 show the specific conditions for depositing a silicon oxide thin film.

[168] [Table 3]

Source	Substrate temp.	Precursor heating	Precursor feeding	Purge (Ar)		O ₂ plasma/ 200 W		Purge (Ar)		No. of depositions
	(°C)	(°C)	Time (sec)	Flow rate (sc cm)	Time (sec)	Flow rate (sc cm)	Time (sec)	Flow rate (sc cm)	Time (sec)	
Ex. 3	200-500	60	7	300	8	1,000	5	1,000	10	100

- [169]
- [170] The thin film deposited in the above manner was analyzed for the composition of the silicon oxide thin film by X-ray photoelectron spectroscopy and for the step coverage using transmission electron microscopy (TEM). Table 3 below and Fig. 9 show the results of analyzing the characteristics of the specific silicon oxide thin film.

[171] [Table 4]

Source	Substrate temp.	Deposition rate	Film thickness	O/Si
	(°C)	(Å/cycle)	Å	Composition ratio
Ex. 3	400	1.14	120	1.7

[172]

- [173] As shown in Table 4, the silicon oxide film formed using the silicon precursor compound of Example 3 had a thick film thickness of 120 Å. The composition ratio of O/Si indicates that a silicon-containing thin film with high purity was formed.
- [174] Fig. 9 is a graph showing the deposition rate of a silicon oxide film with respect to substrate temperature. As shown in Fig. 9, the deposition rate was similar in the temperature range of 300°C to 450°C.
- [175] Fig. 10 is a picture of a silicon oxide film deposited at a substrate temperature of 400°C using a transmission electron microscope (TEM). As shown in Fig. 10, a silicon oxide film was formed uniformly with a thickness of 120 Å.
- [176] Fig. 11 is a result of AES (Auger Electron Spectroscopy) analysis of the composition of a silicon oxide film deposited at a substrate temperature of 400°C.
- [177] As described above, the silicon precursor compound prepared according to the present invention is capable of forming an excellent silicon oxide film with high purity at a high deposition rate.
- [178] The above-described embodiments are only for the purpose of describing the preferred embodiments of the present invention. The scope of the present invention is not limited to the described embodiments. Various changes, modifications, or substitutions will be possible by those skilled in the art within the technical idea and claims of the present invention. It should be understood that such embodiments fall within the scope of the present invention.

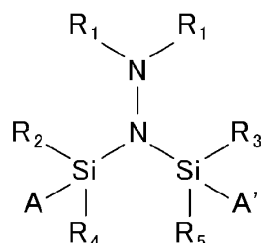
Claims

[Claim 1]

A silicon precursor compound represented by the following

Formula 1:

[Formula 1]

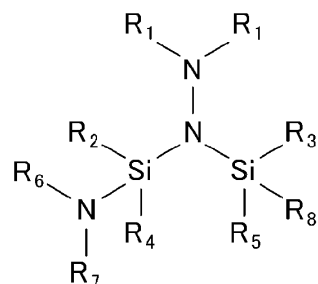


in Formula 1, R₁ is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R₂ to R₅ are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms; A and A' are each independently NR₆R₇ or R₈, provided that at least one of A and A' is NR₆R₇; R₆ and R₇ are each independently a methyl group or an ethyl group; and R₈ is hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms.

[Claim 2]

The silicon precursor compound of claim 1, wherein the silicon precursor compound has an asymmetric structure represented by the following Formula 2:

[Formula 2]



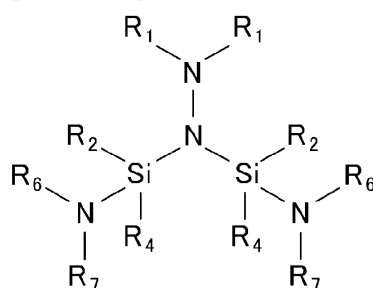
in Formula 2, R₁ is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R₂ to R₅ are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms; R₆

and R_7 are each independently a methyl group or an ethyl group;
and R_8 is hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms.

[Claim 3]

The silicon precursor compound of claim 1, wherein the silicon precursor compound has a symmetric structure represented by the following Formula 3:

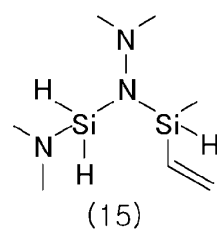
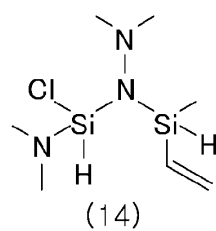
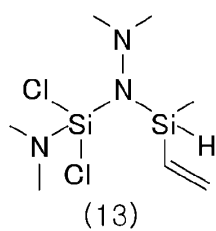
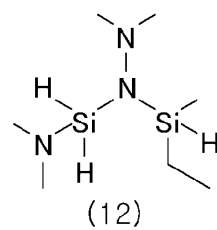
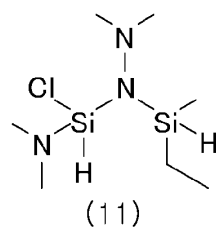
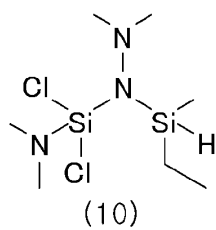
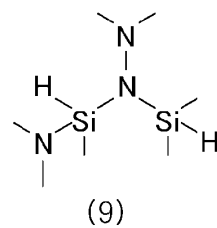
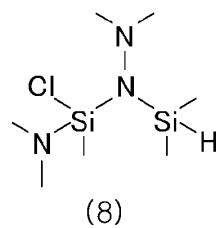
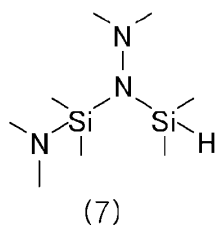
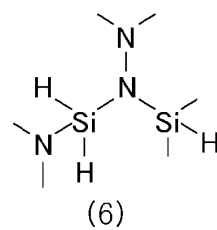
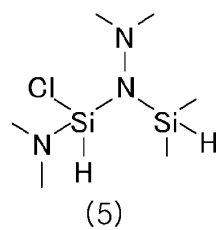
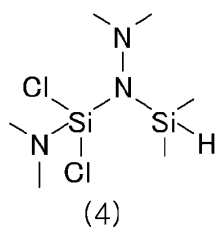
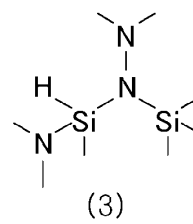
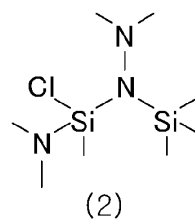
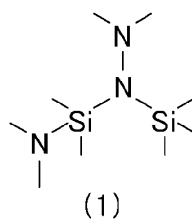
[Formula 3]

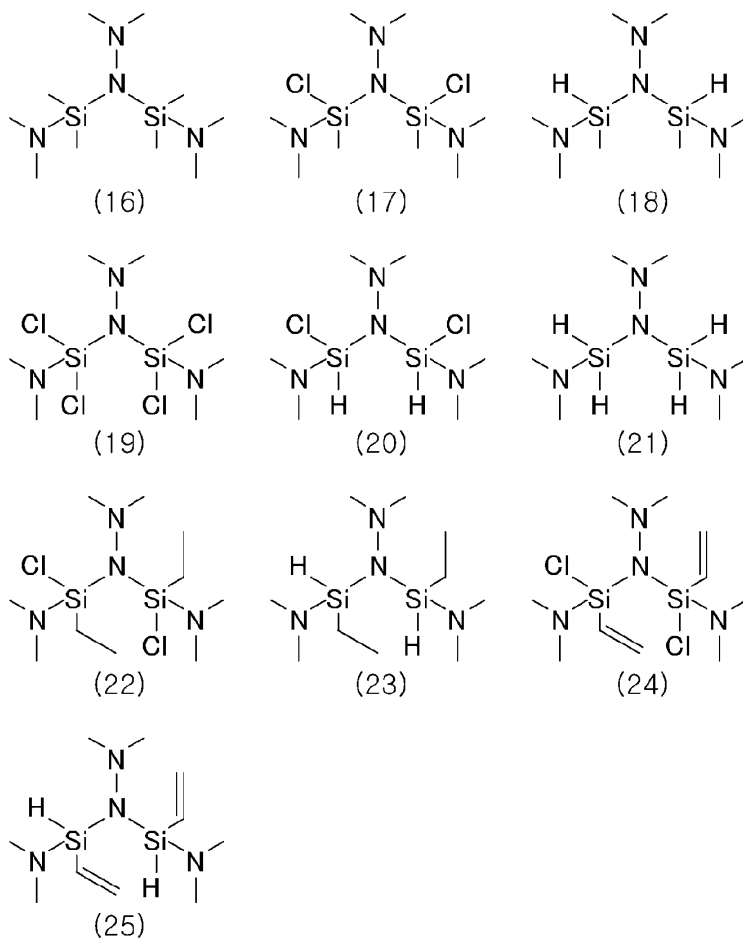


in Formula 3, R_1 is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R_2 and R_4 are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms; and R_6 and R_7 are each independently a methyl group or an ethyl group.

[Claim 4]

The silicon precursor compound of claim 1, wherein the silicon precursor compound is at least one selected from the following compounds (1) to (25).





[Claim 5]

A method for preparing a silicon precursor compound, which comprises:

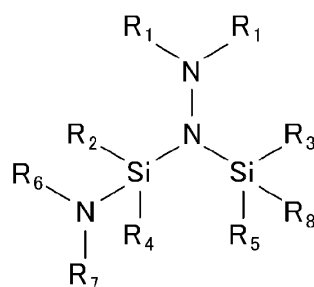
a first step of reacting a chlorosilane derivative with a hydrocarbyl hydrazine compound to form a first compound;

a second step of reacting the first compound with an alkyl-lithium (alkyl-Li) to form a second compound containing lithium; and

a third step of reacting the second compound with a silane compound to prepare a silicon precursor compound represented by the following Formula 2:

[Formula 2]

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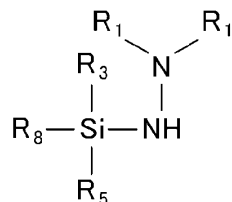


in Formula 2, R_1 is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R_2 to R_5 are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms; R_6 and R_7 are each independently a methyl group or an ethyl group; and R_8 is hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms.

[Claim 6]

The method for preparing a silicon precursor compound of claim 5, wherein the first compound is a compound represented by the following Formula 9:

[Formula 9]



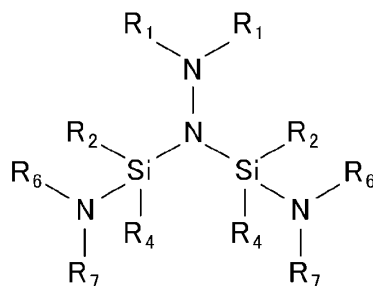
in Formula 9, R_1 is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; and R_3 , R_5 , and R_8 are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms.

[Claim 7]

The method for preparing a silicon precursor compound of claim 5, wherein, when at least one of R_2 and R_4 in the silicon precursor compound represented by Formula 2 are halogen, the method further comprises a fourth step of reacting it with a metal hydride as a reducing agent.

- [Claim 8] The method for preparing a silicon precursor compound of claim 5, wherein the silane compound is produced by reacting a chlorosilane derivative with a secondary amine.
- [Claim 9] A method for preparing a silicon precursor compound, which comprises:
 a first step of reacting a hydrocarbyl hydrazine compound with an alkyl-lithium (alkyl-Li) to form a third compound containing lithium; and
 a second step of reacting the third compound with a silane compound to prepare a silicon precursor compound represented by the following Formula 3:

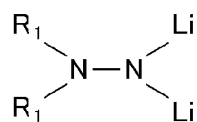
[Formula 3]



in Formula 3, R₁ is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R₂ and R₄ are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms; and R₆ and R₇ are each independently a methyl group or an ethyl group.

- [Claim 10] The method for preparing a silicon precursor compound of claim 9, wherein the third compound is a compound represented by the following Formula 11:

[Formula 11]



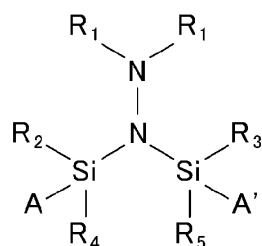
in Formula 11, R_1 is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof.

[Claim 11] The method for preparing a silicon precursor compound of claim 9, wherein, when at least one of R_2 and R_4 in the silicon precursor compound represented by Formula 3 are halogen, the method further comprises a third step of reacting it with a metal hydride as a reducing agent.

[Claim 12] The method for preparing a silicon precursor compound of claim 9, wherein the silane compound is produced by reacting a chlorosilane derivative with a secondary amine.

[Claim 13] A method for preparing a silicon-containing thin film, which comprises forming a silicon-containing thin film using a silicon precursor compound represented by the following Formula 1:

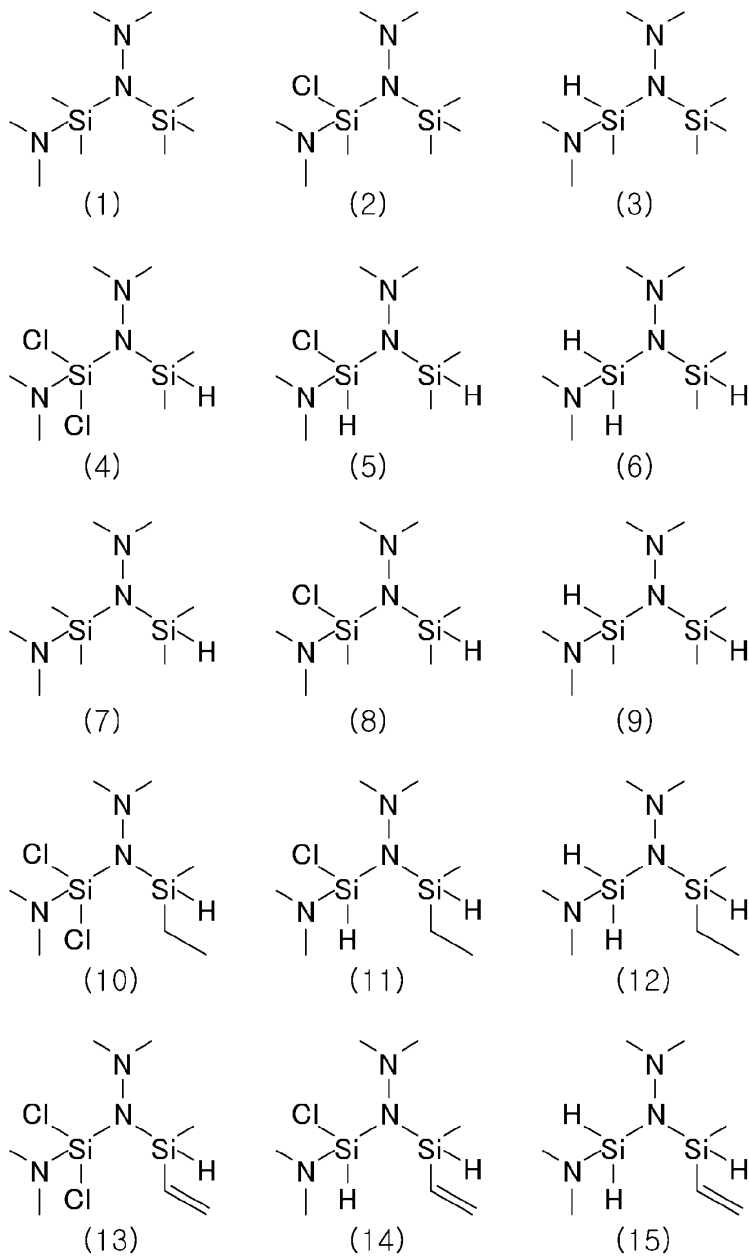
[Formula 1]

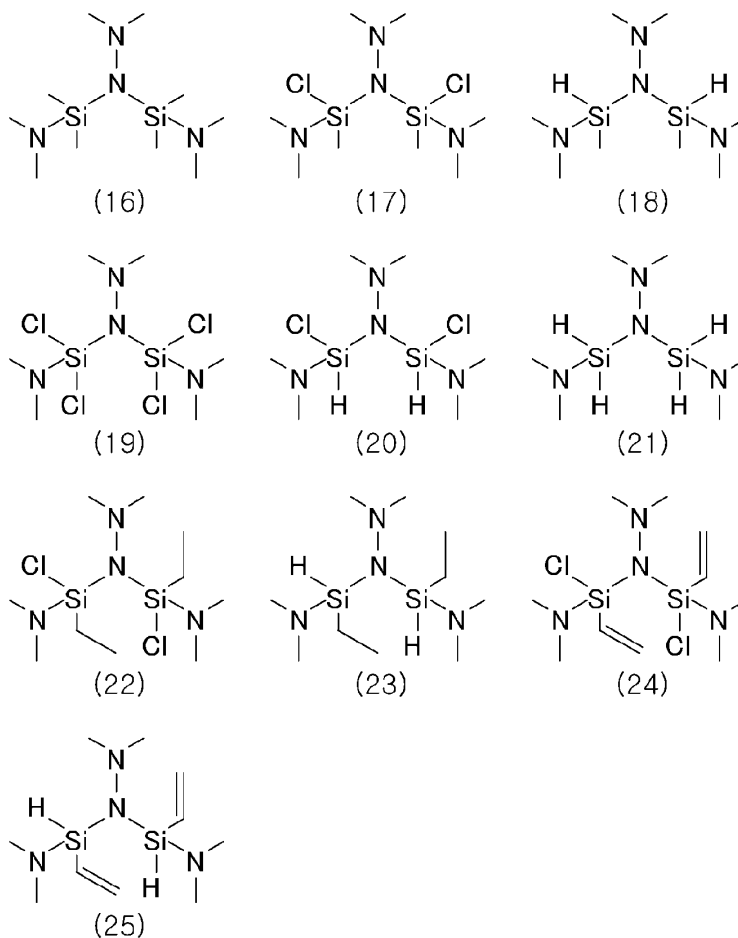


in Formula 1, R_1 is each independently a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms, or an isomer thereof; R_2 to R_5 are each independently hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms; A and A' are each independently NR_6R_7 or R_8 , provided that at least one of A and A' is NR_6R_7 ; R_6 and R_7 are each independently a methyl group or an ethyl group; and R_8 is hydrogen (H), halogen, or a linear or branched, saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms.

[Claim 14]

The method for preparing a silicon-containing thin film of claim 13, wherein the silicon precursor compound is at least one selected from the following compounds (1) to (25).





[Claim 15]

The method for preparing a silicon-containing thin film of claim 13, wherein the silicon-containing thin film is deposited by chemical vapor deposition or atomic layer deposition.

[Claim 16]

The method for preparing a silicon-containing thin film of claim 13, wherein the silicon-containing thin film is any one selected from a silicon oxide film (SiO_2), a silicon oxycarbide film (SiOC), a silicon nitride film (SiN), a silicon oxynitride film (SiON), a silicon carbonitride film (SiCN), and silicon carbonized film (SiC).

[Claim 17]

The method for preparing a silicon-containing thin film of claim 15, wherein the silicon-containing thin film is a silicon oxide film (SiO_2),

the silicon oxide film is deposited by atomic layer deposition, and the atomic layer deposition comprises providing a substrate in a reactor; introducing the silicon precursor compound into the reactor; purging the reactor with a purge gas; introducing an oxygen-containing source into the reactor to react with the silicon precursor compound to form a silicon oxide film; and purging the reactor with the purge gas.

- [Claim 18] The method for preparing a silicon-containing thin film of claim 17, wherein the purge gas is selected from the group consisting of nitrogen, helium, argon, and mixtures thereof, and the oxygen-containing source is selected from the group consisting of oxygen, peroxide, oxygen plasma, water vapor, water vapor plasma, hydrogen peroxide, ozone source, and mixtures thereof.
- [Claim 19] The method for preparing a silicon-containing thin film of claim 17, wherein the oxygen-containing source comprises plasma.
- [Claim 20] The method for preparing a silicon-containing thin film of claim 19, wherein the plasma is generated *in situ*.
- [Claim 21] The method for preparing a silicon-containing thin film of claim 17, wherein the atomic layer deposition is carried out at one or more temperatures of 500°C or lower.
- [Claim 22] The method for preparing a silicon-containing thin film of claim 15, wherein the silicon-containing thin film is a silicon nitride film (SiN), the silicon nitride film is deposited by plasma-enhanced atomic layer deposition, and the plasma-enhanced atomic layer deposition comprises providing a substrate in a reactor; introducing the silicon precursor compound into the reactor; purging the reactor with a purge gas; introducing a nitrogen-containing plasma source and an inert gas to the reactor to react with the silicon precursor compound to form a silicon nitride film; and purging the reactor with the purge gas.

[Claim 23]

The method for preparing a silicon-containing thin film of claim 22, wherein the nitrogen-containing plasma source is selected from the group consisting of nitrogen plasma, nitrogen and argon mixed plasma, ammonia plasma, nitrogen and ammonia mixed plasma, ammonia and helium mixed plasma, ammonia and argon mixed plasma, ammonia and nitrogen mixed plasma, NF_3 plasma, organic amine plasma, and mixtures thereof.

[Claim 24]

The method for preparing a silicon-containing thin film of claim 22, wherein the plasma-enhanced atomic layer deposition is carried out at one or more temperatures of 400°C or lower.