(54) **LAMINATE**

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(57) **ABSTRACT**

A laminate comprising a substrate, an underlayer disposed on the substrate, and a surface-treating layer disposed on the underlayer, wherein the underlayer is a layer formed of (A) polysilazane represented by the following formula (P) or (B) aminosilane represented by the following formula (Q) and the surface-treating layer is a layer formed of a fluorine-containing silane compound:

$$\left[\begin{array}{c} R^{61} \\ | \\ Si-N \\ | \quad | \\ R^{62} \quad R^{63} \end{array} \right]_n \qquad (P)$$
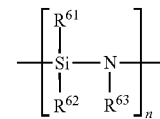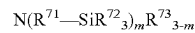
$$N(R^{71}-SiR^{72}_3)_m R^{73}_{3-m} \qquad (Q).$$

LAMINATE

CROSS REFERENCE TO RELATED APPLICATIONS

[0001] This application is a Rule 53 (b) Continuation of International Application No. PCT/JP2023/024465 filed Jun. 30, 2023, which claims priority from Chinese Patent Application No. 202210801338.1 filed Jul. 8, 2022, the respective disclosures of all of the above are incorporated herein by reference in their entirety.

TECHNICAL FIELD

[0002] The present disclosure relates to a laminate.

BACKGROUND ART

[0003] Certain types of fluorine-containing silane compounds are known to be capable of providing excellent water-repellency, oil-repellency, antifouling properties, and the like when used in surface treatment of a substrate. A layer obtained from a surface-treating agent containing a fluorine-containing silane compound (hereinafter, also referred to as a "surface-treating layer") is applied as a so-called functional thin film to a large variety of substrates such as glass, plastics, fibers, sanitary articles, and building materials (Patent Literature 1).

## CITATION LIST

Patent Literature

[0004] Patent Literature 1: International Publication No. WO 2018-056413

SUMMARY

[0005] The present disclosure includes the following embodiments. [1] A laminate comprising a substrate, an underlayer disposed on the substrate, and a surface-treating layer disposed on the underlayer, wherein

[0006] the underlayer is a layer formed of:
(A) polysilazane represented by the following formula (P):

[Formula 1]

$$\left[\begin{array}{c} R^{61} \\ | \\ -Si-N- \\ | \quad | \\ R^{62} \quad R^{63} \end{array} \right]_n$$
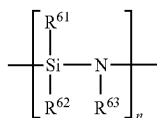
wherein

[0007] R^{61} is a hydrogen atom or a C_{1-6} alkyl group,

[0008] R^{62} is a hydrogen atom, a C_{1-6} alkyl group, or a group having a functional group,

[0009] R^{63} is a hydrogen atom, a C_{1-6} alkyl group, or a group having a functional group, and

[0010] n is an integer of 2 to 100; or
(B) aminosilane represented by the following formula (Q):

$$N(R^{71}-SiR^{72}_3)_m R^{73}_{3-m} \qquad \text{[Formula 2]}$$

wherein

[0011] R^{71} is each independently a C_{1-6} alkylene group,

[0012] R^{72} is each independently a hydrolyzable group,

[0013] R^{73} is a hydrogen atom or a C_{1-6} alkyl group, and

[0014] m is 2 or 3; and

[0015] the surface-treating layer is a layer formed of a fluorine-containing silane compound.

Advantageous Effect

[0016] According to the present disclosure, a laminate having a surface-treating layer with greater ultraviolet resistance can be provided by disposing an underlayer formed of polysilazane or aminosilane between a substrate and the surface-treating layer.

DESCRIPTION OF EMBODIMENTS

[0017] The term "monovalent organic group" as used herein refers to a monovalent group containing carbon. The monovalent organic group is not limited, and may be a hydrocarbon group or a derivative thereof. The derivative of a hydrocarbon group refers to a group that has one or more of N, O, S, Si, amide, sulfonyl, siloxane, carbonyl, carbonyloxy, and the like at the terminal or in the molecular chain of the hydrocarbon group. The term simply referred to as an "organic group" means a monovalent organic group. The term "divalent organic group" refers to a divalent group containing carbon. Such a divalent organic group may be, for example, a divalent group obtained by further removing one hydrogen atom from an organic group. The trivalent or higher organic group similarly refers to a group obtained by removing a predetermined number of hydrogen atoms from an organic group.

[0018] The term "hydrocarbon group", as used herein, refers to a group that contains carbon and hydrogen and that is obtained by removing a hydrogen atom from a hydrocarbon. Such a hydrocarbon group is not limited, and examples include C_{1-20} hydrocarbon groups such as an aliphatic hydrocarbon group and an aromatic hydrocarbon group. The above "aliphatic hydrocarbon group" may be either linear, branched, or cyclic, and may be either saturated or unsaturated. The hydrocarbon group may contain one or more ring structures. The hydrocarbon group may be substituted with one or more substituents.

[0019] The substituent of the "hydrocarbon group" as used herein is not limited, and examples include a halogen atom; and one or more groups selected from a C_{1-6} alkyl group, a C_{2-6} alkenyl group, a C_{2-6} alkynyl group, a C_{3-10} cycloalkyl group, a C_{3-10} unsaturated cycloalkyl group, a 5- to 10-membered heterocyclyl group, a 5- to 10-membered unsaturated heterocyclyl group, a C_{6-10} aryl group, and a 5- to 10-membered heteroaryl group, each of which is optionally substituted with one or more halogen atoms.

[0020] The term "hydrolyzable group" as used herein refers to a group capable of undergoing a hydrolysis reaction, i.e., refers to a group that can be eliminated from the main backbone of a compound by a hydrolysis reaction. Examples of the hydrolyzable group include $-OR^j$, $-OCOR^j$, $-O-N=CR^j_2$, $-NR^j_2$, $-NHR^j$, $-NCO$, and a halogen (in these formulae, R^j represents a substituted or unsubstituted alkyl group having 1 to 4 carbon atoms), and $-OR^j$ (that is, an alkoxy group) is preferable. Examples of R^j include unsubstituted alkyl groups such as a methyl group, an ethyl group, a propyl group, an isopropyl group, a n-butyl group, and an isobutyl group; and substituted alkyl groups such as a chloromethyl group. Among such groups,

an alkyl group, in particular an unsubstituted alkyl group, is preferable, and a methyl group or an ethyl group is more preferable.

(Laminate)

[0021] The laminate of the present disclosure comprises a substrate, an underlayer disposed on the substrate, and a surface-treating layer disposed on the underlayer. The underlayer is a layer formed of:
(A) polysilazane represented by the following formula (P):

[Formula 8]

$$\left[\begin{array}{c} R^{61} \\ | \\ Si\text{---}N \\ | \quad | \\ R^{62} \quad R^{63} \end{array} \right]_{n} \tag{P}$$
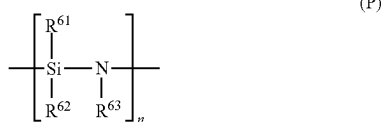
wherein
   [0022] R^{61} is a hydrogen atom or a C_{1-6} alkyl group,
   [0023] R^{62} is a hydrogen atom, a C_{1-6} alkyl group, or a group having a functional group,
   [0024] R^{63} is a hydrogen atom, a C_{1-6} alkyl group, or a group having a functional group, and
   [0025]   n is an integer of 2 to 100; or
(B) aminosilane represented by the following formula (Q):

[Formula 9]

$$N(R^{71}\text{---}SiR^{72}{}_{3})_{m}R^{73}{}_{3-m} \tag{Q}$$

wherein
   [0026] R^{71} is each independently a C_{1-6} alkylene group,
   [0027] R^{72} is each independently a hydrolyzable group,
   [0028] R^{73} is a hydrogen atom or a C_{1-6} alkyl group, and
   [0029]   m is 2 or 3.
The surface-treating layer is a layer formed of a fluorine-containing silane compound.
[0030] The laminate of the present disclosure has the underlayer formed of polysilazane represented by formula (A) for aminosilane represented by formula (B) between the substrate and t the surface-treating and layer, accordingly the ultraviolet resistance of the surface-treating layer is increased. The friction durability of the surface-treating layer can be increased as well.

(Substrate)

[0031] The material constituting the substrate used in the laminate of the present disclosure is not limited, and is selected from the group consisting of, for example, resin, metal, metal oxide, ceramic, and glass. The material constituting the substrate is preferably metal, ceramic, or glass, and is more preferably glass.
[0032] The resin is not limited, and examples include acrylic resin, polycarbonate resin, ABS (acrylonitrile-buta-diene-styrene) resin, polyester resin, polyimide resin, polyamide resin, and polyolefin resin.
[0033] The metal is not limited, and examples include aluminum, copper, iron, stainless steel, and alloy thereof.
[0034] The metal oxide is not limited, and examples include ITO (indium tin oxide) and anodized aluminum.

[0035] The ceramic is not limited, and examples include aluminum oxide, silicon oxide, and zirconium.
[0036] Glass is not limited, and is preferably, for example, soda-lime glass, alkali aluminosilicate glass, borosilicate glass, alkali-free glass, crystal glass, or quartz glass, including chemically strengthened soda-lime glass, chemically strengthened alkali aluminosilicate glass, and chemically bonded borosilicate glass.
[0037] The form of the substrate is not limited, and the substrate may be, for example, a film or a plate.

(Polysilazane)

[0038] Polysilazane used in the formation of the underlayer of the laminate of the present disclosure is represented by the following formula (P):

[Formula 10]

$$\left[\begin{array}{c} R^{61} \\ | \\ Si\text{---}N \\ | \quad | \\ R^{62} \quad R^{63} \end{array} \right]_{n} \tag{P}$$
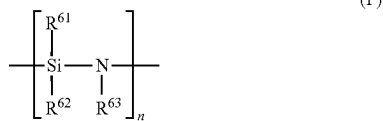
wherein
   [0039] R^{61} is a hydrogen atom or a C_{1-6} alkyl group,
   [0040] R^{62} is a hydrogen atom, a C_{1-6} alkyl group, or a group having a functional group,
   [0041] R^{63} is a hydrogen atom, a C_{1-6} alkyl group, or a group having a functional group, and
   [0042]   n is an integer of 2 to 100.
[0043] In one embodiment, R^{61} is a hydrogen atom.
[0044] In another embodiment, R^{61} is a C_{1-6} alkyl group.
[0045] In one embodiment, R^{62} is a hydrogen atom.
[0046] In another embodiment, R^{62} is a C_{1-6} alkyl group.
[0047] In another embodiment, R^{62} is a group having a functional group.
[0048] In one embodiment, R^{63} is a hydrogen atom.
[0049] In another embodiment, R^{63} is a C_{1-6} alkyl group.
[0050] In another embodiment, R^{63} is a group having a functional group.
[0051] The C_{1-6} alkyl group may be linear or may be branched. The C_{1-6} alkyl group is preferably linear.
[0052] The C_{1-6} alkyl group is preferably a C_{1-4} alkyl group, more preferably a methyl group or an ethyl group, and even more preferably a methyl group.
[0053] The group having the functional group is preferably —R^{64}—R^{65}. R^{64} is a single bond or a divalent organic group, and R^{65} is a functional group.
[0054] The divalent organic group of R^{64} is preferably a C_{1-6} alkyl group.
[0055] The functional group is preferably a carbonyl group, a carboxyl group, or a hydroxyl group.
[0056] In one embodiment, R^{61} is a hydrogen atom or a C_{1-6} alkyl group, R^{62} is a hydrogen atom or a C_{1-6} alkyl group, and R^{63} is a hydrogen atom.
[0057] In a preferable embodiment, R^{61} is a hydrogen atom or a C_{1-6} alkyl group, R^{62} is a hydrogen atom, and R^{63} is a hydrogen atom.
[0058] In a more preferable embodiment, R^{61} is a hydrogen atom, R^{62} is a hydrogen atom, and R^{63} is a hydrogen atom.

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[0059] n is preferably an integer of 5 to 80, and more preferably an integer of 10 to 50.

[0060] The number average molecular weight of the polysilazane is preferably 1,000 to 10,000, more preferably 2,000 to 8,000, and even more preferably 3,000 to 6,000.

[0061] The number average molecular weight of the polysilazane can be determined by GPC (gel permeation chromatography).

[0062] The underlayer formed of the polysilazane can be formed by treating the substrate surface with a composition (A) containing polysilazane. For example, the underlayer can be obtained by applying the composition (A) to the substrate to form a film of the composition (A), and leaving the film to stand still or subjecting the film to a heat treatment to cure the film of the composition (A).

[0063] The composition (A) may contain a solvent.

[0064] Examples of the solvent include cyclic ethers such as THE (tetrahydrofuran), tetrahydropyran, and dioxane; acyclic ethers such as diethyl ether, diisopropyl ether, dibutyl ether, monoglyme, diglyme, and triglyme; aromatic compounds such as HMPA (hexamethylphosphamide), DMPU (dimethylpropylene), TMEDA (tetramethylethylene-diamine), toluene, xylene, and benzotrifluoride; amides such as N, N-dimethylformamide, N, N-dimethylacetamide, and N-methylpyrrolidone; and mixtures thereof.

[0065] The concentration of polysilazane in the composition (A) may be preferably 0.1% by mass to 20% by mass, more preferably 0.2% by mass to 10% by mass, and even more preferably 0.5% by mass to 5% by mass, based on the entirety of the composition.

[0066] The composition (A) may contain a catalyst.

[0067] Examples of the catalyst include acids (such as acetic acid and trifluoroacetic acid), bases (such as ammonia, triethylamine, and diethylamine), transition metals (such as Ti, Ni, and Sn), and metal chelates (such as tris(2,4-pentanedionato)aluminum (III)).

[0068] The polysilazane may be a commercially available product or a synthesized product. For example, the composition (A) may be of the Durazane® series.

[0069] The film of the composition (A) can be formed by applying the composition (A) to the substrate surface so as to coat the surface.

[0070] The coating method is not limited, and examples include dip coating, spin coating, flow coating, spray coating, roll coating, gravure coating, and similar methods.

[0071] The time for leaving the film to stand still may be, for example, 1 minute to 12 hours and preferably 5 minutes to 5 hours at room temperature.

[0072] The temperature of the heat treatment may be, for example, 50° C. to 500° C. and preferably 100° C. to 300° C. The heating time may be, for example, 1 minute to 6 hours and preferably 5 minutes to 3 hours.

(Aminosilane)

[0073] Aminosilane used in the formation of the underlayer of the laminate of the present disclosure is represented by the following formula (Q):

$$N(R^{71}\text{—}SiR^{72}{}_3)_m R^{73}{}_{3-m} \qquad \text{[Formula 11]}$$

wherein

[0074] R^{71} is each independently a C_{1-6} alkylene group,

[0075] R^{72} is each independently a hydrolyzable group,

[0076] R^{73} is a hydrogen atom or a C_{1-6} alkyl group, and

[0077] m is 2 or 3.

[0078] The C_{1-6} alkylene group may be linear or may be branched. The C_{1-6} alkylene group is preferably linear.

[0079] The C_{1-6} alkylene group is preferably a C_{2-6} alkylene group, and more preferably a C_{2-4} alkylene group.

[0080] R^{72} is preferably —OR^j, wherein R^j is a substituted or unsubstituted alkyl group having 1 to 4 carbon atoms. R^{72} is preferably —OCH_3 or —OC_2H_5, more preferably —OCH_3.

[0081] In one embodiment, R^{73} is a hydrogen atom.

[0082] In one embodiment, R^{73} is a C_{1-6} alkyl group.

[0083] The C_{1-6} alkyl group of R^{73} may be linear or may be branched. The C_{1-6} alkyl group is preferably linear.

[0084] The C_{1-6} alkyl group of R^{73} is preferably a C_{1-4} alkyl group, more preferably a methyl group or an ethyl group, and even more preferably a methyl group.

[0085] m is preferably 2.

[0086] In a preferable embodiment, aminosilane is a compound represented by the following formula (Q-1):

$$HN(R^{71}\text{—}SiR^{72}{}_3)_2 \qquad \text{[Formula 12]}$$

wherein

[0087] R^{71} is each independently a C_{1-6} alkylene group, preferably a C_{2-6} alkylene group, and more preferably a C_{2-4} alkylene group, and

[0088] R^{72} is each independently a hydrolyzable group, preferably —OR^j (wherein R^j is a substituted or unsubstituted alkyl group having 1 to 4 carbon atoms), more preferably —OCH_3 or —OC_2H_5, and even more preferably —OCH_3.

[0089] The underlayer formed of aminosilane can be formed by treating the substrate surface with a composition (B) containing aminosilane. For example, the underlayer can be obtained by applying the composition (B) to the substrate to form a film of the composition (B).

[0090] The composition (B) may contain a solvent.

[0091] Examples of the solvent include cyclic ethers such as THE (tetrahydrofuran), tetrahydropyran, and dioxane; acyclic ethers such as diethyl ether, diisopropyl ether, dibutyl ether, monoglyme, diglyme, and triglyme; aromatic compounds such as HMPA (hexamethylphosphamide), DMPU (dimethylpropylene), TMEDA (tetramethylethylene-diamine), toluene, xylene, and benzotrifluoride; amides such as N, N-dimethylformamide, N, N-dimethylacetamide, and N-methylpyrrolidone; and mixtures thereof.

[0092] The concentration of aminosilane in the composition (B) may be preferably 0.1% by mass to 20% by mass, more preferably 0.2% by mass to 10% by mass, and even more preferably 0.5% by mass to 5% by mass, based on the entirety of the composition.

[0093] The composition (B) may contain a catalyst.

[0094] Examples of the catalyst include acids (such as acetic acid and trifluoroacetic acid), bases (such as ammonia, triethylamine, and diethylamino), and transition metals (such as Ti, Ni, and Sn).

[0095] The film of the composition (B) can be formed by applying the composition (B) to the substrate surface so as to coat the surface.

[0096] The coating method is not limited, and examples include dip coating, spin coating, flow coating, spray coating, roll coating, gravure coating, and similar methods.

[0097] The formed film is preferably left to stand still at room temperature for, for example, 1 minute to 12 hours and preferably 5 minutes to 5 hours.

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(Fluorine-Containing Silane Compound)

[0098] The fluorine-containing silane compound used in the formation of the surface-treating layer of the laminate of the present disclosure is a compound containing a fluorine-containing group and a hydrolyzable silane group.

[0099] In one embodiment, the fluorine-containing silane compound is a fluorine-containing silane compound represented by the following formula (A1) or (A2):

[Formula 13]

$$R^{F1}_{\alpha 1}-X^A-R^{Si}_{\beta 1} \tag{A1}$$

$$R^{Si}_{\gamma 1}-X^A-R^{F2}-X^A-R^{Si}_{\gamma 1} \tag{A2}$$

wherein

[0100] R^{F1} is each independently Rf^1- or $Rf^1-R^F-O_q-$,

[0101] R^{F2} is $-Rf^2-$ or $-Rf^2_p-R^F-O_q-$,

[0102] R^F is each independently a divalent fluoropolyether group,

[0103] Rf^1 is each independently a C_{1-16} alkyl group optionally substituted with one or more fluorine atoms,

[0104] Rf^2 is a C_{1-6} alkylene group optionally substituted with one or more fluorine atoms,

[0105] p is 0 or 1,

[0106] q is each independently 0 or 1,

[0107] R^{Si} is each independently a monovalent group containing a Si atom to which a hydroxyl group, a hydrolyzable group, a hydrogen atom, or a monovalent organic group is bonded,

[0108] at least one R^{Si} is a monovalent group containing a Si atom to which a hydroxyl group or a hydrolyzable group is bonded,

[0109] X^A is each independently a single bond or a di- to decavalent organic group,

[0110] $\alpha 1$ is an integer of 1 to 9,

[0111] $\beta 1$ is an integer of 1 to 9, and

[0112] $\gamma 1$ is each independently an integer of 1 to 9.

[0113] In the formula (A1), R^{F1} is each independently Rf^1- or $Rf^1-R^F-O_q-$.

[0114] In a preferable embodiment, R^{F1} is each independently $Rf^1-R^F-O_q-$.

[0115] In the formula (A2), R^{F2} is $-Rf^2-$ or $-Rf^2_p-R^F-O_q-$.

[0116] In a preferable embodiment, R^{F2} is $-Rf^2_p-R^F-O_q-$.

[0117] In the formula, Rf^1 is each independently a C_{1-16} alkyl group optionally substituted with one or more fluorine atoms.

[0118] In the C_{1-16} alkyl group optionally substituted with one or more fluorine atoms, the "C_{1-16} alkyl group" may be linear or branched, and is preferably a linear or branched C_{1-6} alkyl group, in particular C_{1-3} alkyl group, and more preferably a linear C_{1-6} alkyl group, in particular C_{1-3} alkyl group.

[0119] Rf^1 is preferably a C_{1-16} alkyl group substituted with one or more fluorine atoms, more preferably a CF_2H-C_{1-15} perfluoroalkylene group, and even more preferably a C_{1-16} perfluoroalkyl group.

[0120] The C_{1-16} perfluoroalkyl group may be linear or branched, and is preferably a linear or branched C_{1-6} perfluoroalkyl group, in particular C_{1-3} perfluoroalkyl group, more preferably a linear C_{1-6} perfluoroalkyl group, in par-

ticular C_{1-3} perfluoroalkyl group, and specifically $-CF_3$, $-CF_2CF_3$, or $-CF_2CF_2CF_3$.

[0121] In the above formula, Rf^2 is a C_{1-6} alkylene group optionally substituted with one or more fluorine atoms.

[0122] In the C_{1-6} alkylene group optionally substituted with one or more fluorine atoms, the "C_{1-6} alkylene group" may be linear or branched, and is preferably a linear or branched C_{1-3} alkylene group, and more preferably a linear C_{1-3} alkylene group.

[0123] Rf^2 is preferably a C_{1-6} alkylene group substituted with one or more fluorine atoms, more preferably a C_{1-6} perfluoroalkylene group, and even more preferably a C_{1-3} perfluoroalkylene group.

[0124] The C_{1-6} perfluoroalkylene group may be linear or branched, and is preferably a linear or branched C_{1-3} perfluoroalkylene group, more preferably a linear C_{1-3} perfluoroalkylene group, and specifically $-CF_2-$, $-CF_2CF_2-$, or $-CF_2CF_2CF_2-$.

[0125] In the above formulae, p is 0 or 1. In one embodiment, p is 0. In another embodiment, p is 1.

[0126] In the above formulae, q is each independently 0 or 1. In one embodiment, q is 0. In another embodiment, q is 1.

[0127] In the formulae (A1) and (A2), R^F is each independently a divalent fluoropolyether group.

[0128] R^F is preferably a group represented by the following formula:

$$-(OC_6F_{12})_a-(OC_5F_{10})_b-(OC_4F_8)_c-(OC_3R^{Fa}_6)_d-(OC_2F_4)_e-(OCF_2)_f-$$
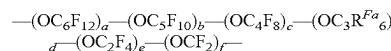
wherein

[0129] R^{Fa} is each independently a hydrogen atom, a fluorine atom, or a chlorine atom;

[0130] a, b, c, d, e, and f are each independently an integer of 0 to 200, and the sum of a, b, c, d, e, and f is 1 or more; and the occurrence order of the respective repeating units enclosed in parentheses provided with a, b, c, d, e, or f is not limited in the formula; provided that when all R^{Fa} groups are hydrogen atoms or chlorine atoms, at least one of a, b, c, e, and f is 1 or more.

[0131] R^{Fa} is preferably a hydrogen atom or a fluorine atom, and more preferably a fluorine atom, provided that when all R^{Fa} groups are hydrogen atoms or chlorine atoms, at least one of a, b, c, e, and f is 1 or more.

[0132] a, b, c, d, e, and f may be preferably each independently an integer of 0 to 100.

[0133] The sum of a, b, c, d, e, and f is preferably 5 or more, is more preferably 10 or more, and may be, for example, 15 or more or 20 or more. The sum of a, b, c, d, e, and f is preferably 200 or less, is more preferably 100 or less, is even more preferably 60 or less, and may be, for example, 50 or less or 30 or less.

[0134] These repeating units may be linear or branched, and may contain a ring structure. For example, $-(OC_6F_{12})-$ may be $-(OCF_2CF_2CF_2CF_2CF_2CF_2)-$, $-(OCF(CF_3)CF_2CF_2CF_2CF_2)-$, $-(OCF_2CF(CF_3)CF_2CF_2CF_2)-$, $-(OCF_2CF_2CF(CF_3)CF_2CF_2)-$, $(OCF_2CF_2CF_2CF(CF_3)CF_2)-$, $-(OCF_2CF_2CF_2CF_2CF(CF_3))-$, or the like. $-(OC_5F_{10})-$ may be $-(OCF_2CF_2CF_2CF_2CF_2)-$, $-(OCF(CF_3)CF_2CF_2CF_2)-$, $-(OCF_2CF(CF_3)CF_2CF_2)-$, $-(OCF_2CF_2CF(CF_3)CF_2)-$, $-(OCF_2CF_2CF_2CF(CF_3))-$, or the like. $-(OC_4F_8)-$ may be any of $-(OCF_2CF_2CF_2CF_2)-$, $-(OCF(CF_3)CF_2CF_2)-$, $-(OCF_2CF(CF_3)CF_2)-$, $-(OCF_2CF_2CF(CF_3))-$, $-(OC(CF_3)_2CF_2)-$, $-(OCF_2C

$(CF_3)_2)$—, —$(OCF(CF_3)CF(CF_3))$—, —$(OCF(C_2F_5)$ $CF_2)$—, and —$(OCF_2CF(C_2F_5))$—. —(OC_3F_6)— (that is, in the above formula, R^{Fa} is a fluorine atom) may be any of —$(OCF_2CF_2CF_2)$—, —$(OCF(CF_3)CF_2)$—, and $(OCF_2CF$ $(CF_3))$—. —(OC_2F_4)— may be any of —(OCF_2CF_2)— and —$(OCF(CF_3))$—.

[0135] The ring structure may be any of the following 3-membered ring, 4-membered rings, 5-membered rings, and 6-membered rings:
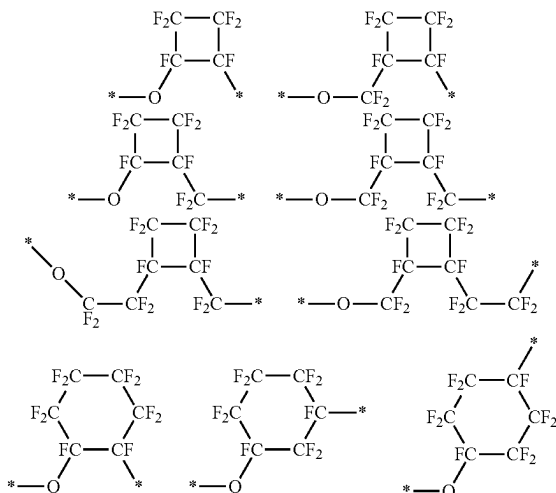
[Formula 14]
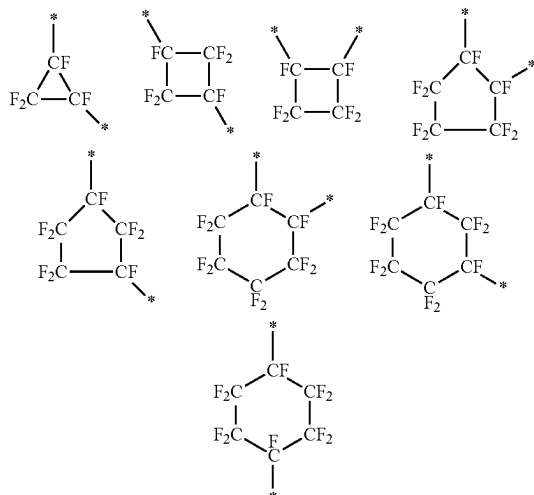
wherein * represents a bond position.

[0136] The ring structure may be preferably a 4-membered ring, a 5-membered ring, or a 6-membered ring, and more preferably a 4-membered ring or a 6-membered ring.

[0137] The repeating units having a ring structure may be preferably any of the following units:

[Formula 15]


wherein * represents a bond position.

[0138] In one embodiment, the above repeating units are linear. When the above repeating units are linear, the surface lubricity, friction durability, and the like of the surface-treating layer can be increased.

[0139] In one embodiment, the repeating units are branched. When the repeating units are branched, the dynamic friction coefficient of the surface-treating layer can be increased.

[0140] In one embodiment, R^F is each independently a group represented by any of the following formulae (f1) to (f6):

$$—(OC_3F_6)_d—(OC_2F_4)_e— \qquad (f1)$$

wherein d is an integer of 1 to 200, and e is 0 or 1;

$$—(OC_4F_8)_c—(OC_3F_6)_d—(OC_2F_4)_e—(OCF_2)_f— \qquad (f2)$$
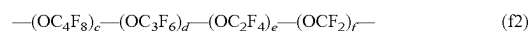
wherein c and d are each independently an integer of 0 or more and 30 or less, and e and f are each independently an integer of 1 or more and 200 or less,

[0141] the sum of c, d, e, and f is 2 or more, and

[0142] the occurrence order of the respective repeating units enclosed in parentheses provided with the subscript c, d, e, or f is not limited in the formula;

$$—(R^6—R^7)_g— \qquad (f3)$$

wherein R^6 is OCF_2 or OC_2F_4,

[0143] R^7 is a group selected from OC_2F_4, OC_3F_6, OC_4F_8, OC_5F_{10}, and OC_6F_{12}, or is a combination of two or three groups independently selected from these groups, and

[0144] g is an integer of 2 to 100;

$$—(R^6—R^7)_g—R^r—(R^{7'}—R^{6'})_{g'}— \qquad (f4)$$

wherein R^6 is OCF_2 or OC_2F_4,

[0145] R^7 is a group selected from OC_2F_4, OC_3F_6, OC_4F_8, OC_5F_{10}, and OC_6F_{12}, or is a combination of two or three groups independently selected from these groups,

[0146] $R^{6'}$ is OCF_2 or OC_2F_4,

[0147] $R^{7'}$ is a group selected from OC_2F_4, OC_3F_6, OC_4F_8, OC_5F_{10}, and OC_6F_{12}, or is a combination of two or three groups independently selected from these groups,
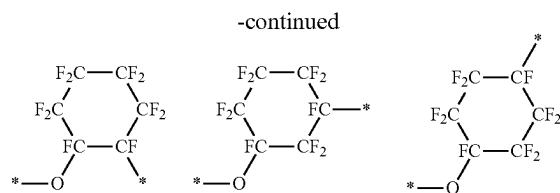
[0148] g is an integer of 2 to 100,
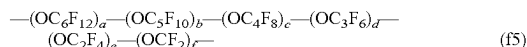
[0149] g' is an integer of 2 to 100, and

[0150] R^r is any of:

[Formula 16]
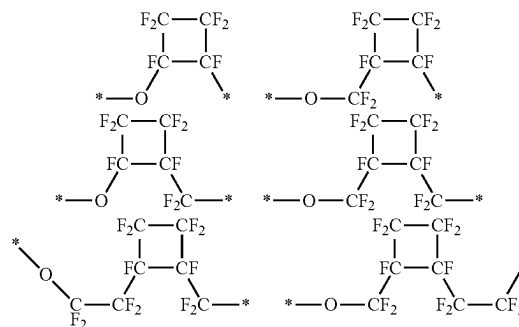
-continued

wherein * represents a bond position;

$$—(OC_6F_{12})_a—(OC_5F_{10})_b—(OC_4F_8)_c—(OC_3F_6)_d—(OC_2F_4)_e—(OCF_2)_f— \quad (f5)$$

wherein e is an integer of 1 or more and 200 or less; a, b, c, d, and f are each independently an integer of 0 or more and 200 or less; and the occurrence order of the respective repeating units enclosed in parentheses provided with a, b, c, d, e, or f is not limited in the formula; and

$$—(OC_6F_{12})_a—(OC_5F_{10})_b—(OC_4F_8)_c—(OC_3F_6)_d—(OC_2F_4)_e—(OCF_2)_f— \quad (f6)$$
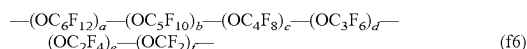
wherein f is an integer of 1 or more and 200 or less; a, b, c, d, and e are each independently an integer of 0 or more and 200 or less; and the occurrence order of the respective repeating units enclosed in parentheses provided with a, b, c, d, e, or f is not limited in the formula.

[0151] In the formula (f1), d is an integer of preferably 5 to 200, more preferably 10 to 100, and even more preferably 15 to 50, such as 25 to 35. In one embodiment, e is 1. In another embodiment, e is 0. In the above formula (f1), (OC_3F_6) is preferably a group represented by $(OCF_2CF_2CF_2)$, $(OCF_2CF(CF_3))$, or $(OCF(CF_3)CF_2)$, and is more preferably a group represented by $—(OCF_2CF_2CF_2)_d—$. In the above formula (f1), (OC_2F_4) is preferably a group represented by (OCF_2CF_2) or $(OCF(CF_3))$, and is more preferably a group represented by (OCF_2CF_2).
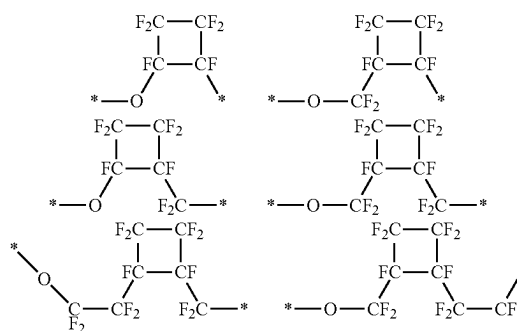
[0152] In the above formula (f2), e and f are each independently an integer of preferably 5 to 200, and more preferably 10 to 200. The sum of c, d, e, and f is preferably 5 or more, is more preferably 10 or more, and may be, for example, 15 or more or 20 or more. In one embodiment, the above formula (f2) is preferably a group represented by $—(OCF_2CF_2CF_2CF_2)_c—(OCF_2CF_2CF_2)_d—(OCF_2CF_2)_e—(OCF_2)_f—$. In another embodiment, the formula (f2) may be a group represented by $—(OC_2F_4)_e—(OCF_2)_f—$.

[0153] In the above formula (f3), R^6 is preferably OC_2F_4. In the above (f3), R^7 is preferably a group selected from OC_2F_4, OC_3F_6, and OC_4F_8, or a combination of two or three groups independently selected from these groups, and more preferably a group selected from OC_3F_6 and OC_4F_8. The combination of two or three groups independently selected from OC_2F_4, OC_3F_6, and OC_4F_8 is not limited, and examples include $—OC_2F_4OC_3F_6—$, $—OC_2F_4OC_4F_8—$, $—OC_3F_6OC_2F_4—$, $—OC_3F_6OC_3F_6—$, $—OC_3F_6OC_4F_8—$, $—OC_4F_8OC_4F_8—$, $—OC_4F_8OC_3F_6—$, $—OC_4F_8OC_2F_4—$, $—OC_2F_4OC_2F_4OC_3F_6—$, $—OC_2F_4OC_2F_4OC_4F_8—$, $—OC_2F_4OC_3F_6OC_2F_4—$, $—OC_2F_4OC_3F_6OC_3F_6—$, $—OC_2F_4OC_4F_8OC_2F_4—$, $—OC_3F_6OC_2F_4OC_2F_4—$, $—OC_3F_6OC_2F_4OC_3F_6—$, $—OC_3F_6OC_3F_6OC_2F_4—$, and $—OC_4F_8OC_2F_4OC_2F_4—$. In the above formula (f3), g is an integer of preferably 3 or more, and more preferably 5 or more. g is preferably an integer of 50 or less. In the above formula (f3), OC_2F_4, OC_3F_6, OC_4F_8, OC_5F_{10}, and OC_6F_{12} may be either linear or branched, and are preferably linear.

In this embodiment, the above formula (f3) is preferably $—(OC_2F_4—OC_3F_6)_g—$, or $—(OC_2F_4—OC_4F_8)_g—$.

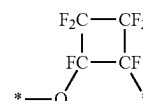[0154] In the formula (f4), R^6, R^7, and g have the same meanings as those described for the above formula (f3), and have the same embodiments. $R^{6'}$, $R^{7'}$, and g' have the same meanings as those of R^6, R^7, and g set forth in the above formula (f3), respectively, and have the same embodiments. R^r is preferably:

[Formula 17]

wherein * represents a bond position, and more preferably

[Formula 18]

wherein * represents a bond position.

[0155] In the above formula (f5), e is an integer of preferably 1 or more and 100 or less, and more preferably 5 or more and 100 or less. The sum of a, b, c, d, e, and f is preferably 5 or more, is more preferably 10 or more, and is, for example, 10 or more and 100 or less.

[0156] In the above formula (f6), f is an integer of preferably 1 or more and 100 or less, and more preferably 5 or more and 100 or less. The sum of a, b, c, d, e, and f is preferably 5 or more, is more preferably 10 or more, and is, for example, 10 or more and 100 or less.

[0157] In one embodiment, R^F is a group represented by the above formula (f1) or (f2).

[0158] In one embodiment, R^F is a group represented by the above formula (f1).

[0159] In one embodiment, R^F is a group represented by the above formula (f2).

[0160] In one embodiment, R^F is a group represented by the above formula (f3) or (f4).

[0161] In one embodiment, R^F is a group represented by the above formula (f3).

[0162] In one embodiment, R^F is a group represented by the above formula (f4).

[0163] In one embodiment, R^F is a group represented by the above formula (f5).

[0164] In one embodiment, R^F is a group represented by the above formula (f6).

[0165] In R^F, the ratio of e to f (hereinafter referred to as the "e/f ratio") is 0.1 to 10, preferably 0.2 to 5, more preferably 0.2 to 2, even more preferably 0.2 to 1.5, and yet more preferably 0.2 to 0.85. With an e/f ratio of 10 or less, the lubricity, friction durability, and chemical resistance (such as durability against artificial sweat) of the surface-treating layer obtained from this compound are more increased. The lower the e/f ratio is, the more increased the lubricity and the friction durability of the surface-treating layer are. On the other hand, with an e/f ratio of 0.1 or more, the stability of the compound can be more enhanced. The higher the e/f ratio is, the more increased the stability of the compound is.

[0166] In one embodiment, the e/f ratio is preferably 0.2 to 0.95, and more preferably 0.2 to 0.9.

[0167] In one embodiment, from the viewpoint of heat resistance, the e/f ratio is preferably 1.0 or more, and more preferably 1.0 to 2.0.

[0168] In the fluorine-containing silane compound, the number average molecular weight of the R^{F1} and R^{F2} moieties is not limited, and is, for example, 500 to 30,000, preferably 1,500 to 30,000, and more preferably 2,000 to 10,000. Herein, the number average molecular weight of R^{F1} and R^{F2} is a value obtained by ^{19}F-NMR measurement.

[0169] In another embodiment, the number average molecular weight of the R^{F1} and R^{F2} moieties is 500 to 30,000, preferably 1,000 to 20,000, more preferably 2,000 to 15,000, and even more preferably 2,000 to 10,000, and may be, for example, 3,000 to 6,000.
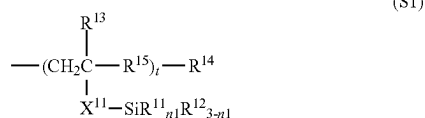
[0170] In another embodiment, the number average molecular weight of the R^{F1} and R^{F2} moieties may be 4,000 to 30,000, preferably 5,000 to 10,000, and more preferably 6,000 to 10,000.

[0171] In the above formulae (A1) and (A2), R^{Si} is each independently a monovalent group containing a Si atom to which a hydroxyl group, a hydrolyzable group, a hydrogen atom, or a monovalent organic group is bonded, and at least one R^{Si} is a monovalent group containing a Si atom to which a hydroxyl group or a hydrolyzable group is bonded.

[0172] In a preferable embodiment, R^{Si} is a monovalent group containing a Si atom to which a hydroxyl group or a hydrolyzable group is bonded, and more preferably a monovalent group containing a Si atom to which a hydrolyzable group is bonded.

[0173] In a preferable embodiment, R^{Si} is a group represented by the following formula (S1), (S2), (S3), (S4), or (S5):

[Formula 19]

$$\underset{\underset{X^{11}-SiR^{11}{}_{n1}R^{12}{}_{3-n1}}{\overset{\overset{R^{13}}{|}}{|}}{-(CH_2C-R^{15}){}_t-R^{14}} \qquad (S1)$$

$$-SiR^{11}{}_{n1}R^{12}{}_{3-n1} \qquad (S2)$$

$$-SiR^{a1}{}_{k1}R^{b1}{}_{l1}R^{c1}{}_{m1} \qquad (S3)$$

$$-CR^{d1}{}_{k2}R^{e1}{}_{l2}R^{f1}{}_{m2} \qquad (S4)$$

$$-NR^{g1}R^{h1} \qquad (S5)$$

[0174] In the above formulae, R^{11} is each independently a hydroxyl group or a hydrolyzable group.

[0175] R^{11} is preferably each independently a hydrolyzable group.

[0176] R^{11} is preferably each independently $-OR^j$, $-OCOR^j$, $-O-N=CR^j{}_2$, $-NR^j{}_2$, $-NHR^j$, $-NCO$, or a halogen (in these formulae, R^j represents a substituted or unsubstituted C_{1-4} alkyl group), and more preferably $-OR^j$ (that is, an alkoxy group). Examples of R^j include unsubstituted alkyl groups such as a methyl group, an ethyl group, a propyl group, an isopropyl group, a n-butyl group, and an isobutyl group; and substituted alkyl groups such as a chloromethyl group. Among such groups, an alkyl group, in particular an unsubstituted alkyl group, is preferable, and a methyl group or an ethyl group is more preferable. In one embodiment, R^j is a methyl group, and in another embodiment, R^j is an ethyl group.

[0177] In the above formulae, R^{12} is each independently a hydrogen atom or a monovalent organic group. Such a monovalent organic group is a monovalent organic group excluding the above hydrolyzable group.

[0178] In R^{12}, the monovalent organic group is preferably a C_{1-20} alkyl group, more preferably a C_{1-6} alkyl group, and even more preferably a methyl group.

[0179] In the above formulae, n1 is each independently an integer of 0 to 3 for each $(SiR^{11}{}_{n1}R^{12}{}_{3-n1})$ unit. However, when R^{Si} is a group represented by the formula (S1) or (S2), the R^{Si} moiety at the terminal of the formula (A1) and the formula (A2) (hereinafter, also referred to simply as the "terminal moiety" of the formula (A1) and the formula (A2)) has at least one $(SiR^{11}{}_{n1}R^{12}{}_{3-n1})$ unit wherein n1 is 1 to 3. That is, in such terminal moieties, not all n1 are 0 at the same time. In other words, in the terminal moieties of the formula (A1) and the formula (A2), at least one Si atom to which a hydroxyl group or a hydrolyzable group is bonded is present.

[0180] n1 is each independently an integer of preferably 1 to 3, more preferably 2 to 3, and even more preferably 3 for each $(SiR^{11}{}_{n1}R^{12}{}_{3-n1})$ unit.

[0181] In the above formulae, X^{11} is each independently a single bond or a divalent organic group. Such a divalent organic group is preferably $-R^{28}-O_x-R^{29}-$ (wherein R^{28} and R^{29} are each independently a single bond or a C_{1-20} alkylene group, and x is 0 or 1). Such a C_{1-20} alkylene group may be linear or may be branched, and is preferably linear. Such a C_{1-20} alkylene group is preferably a C_{1-10} alkylene group, more preferably a C_{1-6} alkylene group, and even more preferably a C_{1-3} alkylene group.

[0182] In one embodiment, X^{11} is each independently $-C_{1-6}$ alkylene-$O-C_{1-6}$ alkylene- or $-O-C_{1-6}$ alkylene-.

[0183] In a preferable embodiment, X^{11} is each independently a single bond or a linear C_{1-6} alkylene group, preferably a single bond or a linear C_{1-3} alkylene group, more preferably a single bond or a linear C_{1-2} alkylene group, and even more preferably a linear C_{1-2} alkylene group.

[0184] In the above formulae, R^{13} is each independently a hydrogen atom or a monovalent organic group. Such a monovalent organic group is preferably a C_{1-20} alkyl group. Such a C_{1-20} alkyl group may be linear or branched, and is preferably linear.

[0185] In a preferable embodiment, R^{13} is each independently a hydrogen atom or a linear C_{1-6} alkyl group, preferably a hydrogen atom or a linear C_{1-3} alkyl group, and preferably a hydrogen atom or a methyl group.

[0186] In the above formulae, t is each independently an integer of 2 or more.

[0187] In a preferable embodiment, t is each independently an integer of 2 to 10, and preferably an integer of 2 to 6.

[0188] In the above formulae, R^{14} is each independently a hydrogen atom, a halogen atom, or $-X^{11}-SiR^{11}_{n1}R^{12}_{3-n1}$. Such a halogen atom is preferably an iodine atom, a chlorine atom, or a fluorine atom, and more preferably a fluorine atom. In a preferable embodiment, R^{14} is a hydrogen atom.
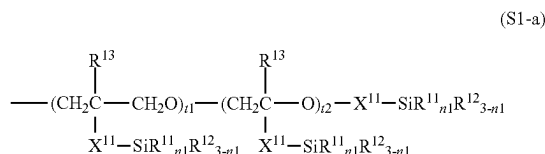
[0189] In the above formulae, R^{15} is each independently at each occurrence a single bond, an oxygen atom, an alkylene group having 1 to 6 carbon atoms, or an alkyleneoxy group having 1 to 6 carbon atoms.

[0190] In one embodiment, R^{15} is each independently at each occurrence an oxygen atom, an alkylene group having 1 to 6 carbon atoms, or an alkyleneoxy group having 1 to 6 carbon atoms.

[0191] In a preferable embodiment, R^{15} is a single bond.

[0192] In one embodiment, the formula (S1) is the following formula (S1-a):

[Formula 20]

(S1-a)

$$-(CH_2C-CH_2O)_{t1}-(CH_2C-O)_{t2}-X^{11}-SiR^{11}_{n1}R^{12}_{3-n1}$$

with R^{13} above each C, and $X^{11}-SiR^{11}_{n1}R^{12}_{3-n1}$ below each C.

wherein

[0193] R^{11}, R^{12}, R^{13}, X^{11}, and n1 have the same meanings as those described for the above formula (S1),

[0194] t1 and t2 are each independently an integer of 1 or more, preferably an integer of 1 to 10, and more preferably an integer of 2 to 10, such as an integer of 1 to 5 or an integer of 2 to 5, and

[0195] the occurrence order of the respective repeating units enclosed in parentheses provided with t1 and t2 is not limited in the formula.

[0196] In a preferable embodiment, the formula (S1) is the following formula (S1-b):

[Formula 21]

(S1-b)

$$-(CH_2C)_t-H$$

with R^{13} above C, and $X^{11}-SiR^{11}_{n1}R^{12}_{3-n1}$ below C.
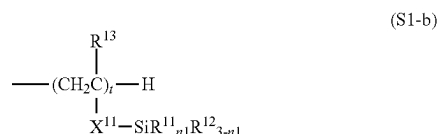
wherein R^{11}, R^{12}, R^{13}, X^{11}, n1, and t have the same meanings as those described for the above formula (S1).

[0197] In the above formula, R^{a1} is each independently $-Z^1-SiR^{21}_{p1}R^{22}_{q1}R^{23}_{r1}$.

[0198] Z^1 is each independently an oxygen atom or a divalent organic group. Below, the structure denoted as Z^1 is bonded to $(SiR^{21}_{p1}R^{22}_{q1}R^{23}_{r1})$ on the right side.

[0199] In a preferable embodiment, Z^1 is a divalent organic group.

[0200] In a preferable embodiment, Z^1 does not include a group that forms a siloxane bond with the Si atom to which Z^1 is bonded. Preferably, in the formula (S3), (Si-Z^1-Si) does not contain a siloxane bond.

[0201] Z^1 is preferably a C_{1-6} alkylene group, $-(CH_2)_{z1}-O-(CH_2)_{z2}-$ (wherein z1 is an integer of 0 to 6 such as an integer of 1 to 6, and z2 is an integer of 0 to 6 such as an integer of 1 to 6), or $-(CH_2)_{z3}$-phenylene-$(CH_2)_{z4}-$ (wherein z3 is an integer of 0 to 6 such as an integer of 1 to 6, and z4 is an integer of 0 to 6 such as an integer of 1 to 6). Such a C_{1-6} alkylene group may be linear or may be branched, and is preferably linear. These groups may be substituted with, for example, one or more substituents selected from a fluorine atom, a C_{1-6} alkyl group, a C_{2-6} alkenyl group, and a C_{2-6} alkynyl group, and are preferably unsubstituted.

[0202] In a preferable embodiment, Z^1 is a C_{1-6} alkylene group or $-(CH_2)_{z3}$-phenylene-$(CH_2)_{z4}-$, and preferably -phenylene-$(CH_2)_{z4}-$. When Z^1 is such a group, light resistance, in particular ultraviolet resistance, can be further enhanced.

[0203] In another preferable embodiment, Z^1 is a C_{1-3} alkylene group. In one embodiment, Z^1 may be $-CH_2CH_2CH_2-$. In another embodiment, Z^1 may be $-CH_2CH_2-$.

[0204] R^{21} is each independently $-Z^{1'}-SiR^{21'}_{p1'}R^{22'}_{q1'}R^{23'}_{r1'}$.

[0205] $Z^{1'}$ is each independently an oxygen atom or a divalent organic group. Below, the structure denoted as $Z^{1'}$ is bonded to $(SiR^{21'}_{p1'}R^{22'}_{q1'}R^{23'}_{r1'})$ on the right side.

[0206] In a preferable embodiment, $Z^{1'}$ is a divalent organic group.

[0207] In a preferable embodiment, $Z^{1'}$ does not include a group that forms a siloxane bond with the Si atom to which $Z^{1'}$ is bonded. Preferably, in the formula (S3), (Si-$Z^{1'}$-Si) does not contain a siloxane bond.

[0208] $Z^{1'}$ is preferably a C_{1-6} alkylene group, $-(CH_2)_{z1'}-O-(CH_2)_{z2'}-$ (wherein z1' is an integer of 0 to 6 such as an integer of 1 to 6, and z2' is an integer of 0 to 6 such as an integer of 1 to 6), or $-(CH_2)_{z3'}$-phenylene-$(CH_2)_{z4'}-$ (wherein z3' is an integer of 0 to 6 such as an integer of 1 to 6, and z4' is an integer of 0 to 6 such as an integer of 1 to 6). Such a C_{1-6} alkylene group may be linear or may be branched, and is preferably linear. These groups may be substituted with, for example, one or more substituents selected from a fluorine atom, a C_{1-6} alkyl group, a C_{2-6} alkenyl group, and a C_{2-6} alkynyl group, and are preferably unsubstituted.

[0209] In a preferable embodiment, $Z^{1'}$ is a C_{1-6} alkylene group or $-(CH_2)_{z3'}$-phenylene-$(CH_2)_{z4'}-$, and preferably -phenylene-$(CH_2)_{z4'}-$. When $Z^{1'}$ is such a group, light resistance, in particular ultraviolet resistance, can be further enhanced.

[0210] In another preferable embodiment, $Z^{1'}$ is a C_{1-3} alkylene group. In one embodiment, $Z^{1'}$ may be $-CH_2CH_2CH_2-$. In another embodiment, $Z^{1'}$ may be $-CH_2CH_2-$.

[0211] $R^{21'}$ is each independently $-Z^{1''}-SiR^{22''}_{q1'}-R^{23''}_{r1''}$.

[0212] $Z^{1''}$ is each independently an oxygen atom or a divalent organic group. Below, the structure denoted as $Z^{1''}$ is bonded to $(SiR^{22''}_{q1'}R^{23''}_{r1''})$ on the right side.

[0213] In a preferable embodiment, $Z^{1''}$ is a divalent organic group.

[0214] In a preferable embodiment, $Z^{1''}$ does not include a group that forms a siloxane bond with the Si atom to which $Z^{1''}$ is bonded. Preferably, in the formula (S3), (Si-$Z^{1''}$-Si) does not contain a siloxane bond.

[0215] $Z^{1''}$ is preferably a C_{1-6} alkylene group, —$(CH_2)_{z1''}$—O—$(CH_2)_{z2''}$— (wherein $z1''$ is an integer of 0 to 6 such as an integer of 1 to 6, and $z2''$ is an integer of 0 to 6 such as an integer of 1 to 6), or —$(CH_2)_{z3''}$-phenylene-$(CH_2)_{z4''}$— (wherein $z3''$ is an integer of 0 to 6 such as an integer of 1 to 6, and $z4''$ is an integer of 0 to 6 such as an integer of 1 to 6). Such a C_{1-6} alkylene group may be linear or may be branched, and is preferably linear. These groups may be substituted with, for example, one or more substituents selected from a fluorine atom, a C_{1-6} alkyl group, a C_{2-6} alkenyl group, and a C_{2-6} alkynyl group, and are preferably unsubstituted.

[0216] In a preferable embodiment, $Z^{1''}$ is a C_{1-6} alkylene group or —$(CH_2)_{z3''}$-phenylene-$(CH_2)_{z4''}$—, and preferably -phenylene-$(CH_2)_{z4''}$—. When $Z^{1''}$ is such a group, light resistance, in particular ultraviolet resistance, can be further enhanced.

[0217] In another preferable embodiment, $Z^{1''}$ is a C_{1-3} alkylene group. In one embodiment, $Z^{1''}$ may be —$CH_2CH_2CH_2$—. In another embodiment, $Z^{1''}$ may be —CH_2CH_2—.

[0218] $R^{22''}$ is each independently a hydroxyl group or a hydrolyzable group.

[0219] $R^{22'}$ is preferably each independently a hydrolyzable group.

[0220] $R^{22''}$ is preferably each independently —OR^j, —$OCOR^j$, —O—N=CR^j_2, —NR^j_2, —NHR^j, —NCO, or a halogen (in these formulae, R^j represents a substituted or unsubstituted C_{1-4} alkyl group), and more preferably —OR^j (that is, an alkoxy group). Examples of R^j include unsubstituted alkyl groups such as a methyl group, an ethyl group, a propyl group, an isopropyl group, a n-butyl group, and an isobutyl group; and substituted alkyl groups such as a chloromethyl group. Among such groups, an alkyl group, in particular an unsubstituted alkyl group, is preferable, and a methyl group or an ethyl group is more preferable. In one embodiment, R^j is a methyl group, and in another embodiment, R^j is an ethyl group.

[0221] $R^{23''}$ is each independently a hydrogen atom or a monovalent organic group. Such a monovalent organic group is a monovalent organic group excluding the above hydrolyzable group.

[0222] In one embodiment, the monovalent organic group is a C_{1-20} alkyl group, more preferably a C_{1-6} alkyl group, and even more preferably a methyl group.

[0223] $q1''$ is each independently an integer of 0 to 3, and $r1''$ is each independently an integer of 0 to 3. The sum of $q1''$ and $r1''$ is 3 in the (Si$R^{22''}_{q1''}R^{23''}_{r1''}$) unit.

[0224] $q1''$ is each independently an integer of preferably 1 to 3, more preferably 2 to 3, and even more preferably 3 for each (Si$R^{22''}_{q1''}R^{23''}_{r1''}$) unit.

[0225] $R^{22'}$ is each independently a hydroxyl group or a hydrolyzable group.

[0226] $R^{22'}$ is preferably each independently a hydrolyzable group.

[0227] $R^{22'}$ is preferably each independently —OR^j, —$OCOR^j$, —O—N=CR^j_2, —NR^j_2, —NHR^j, —NCO, or a halogen (in these formulae, R^j represents a substituted or unsubstituted C_{1-4} alkyl group), and more preferably —OR^j (that is, an alkoxy group). Examples of R^j include unsubstituted alkyl groups such as a methyl group, an ethyl group, a propyl group, an isopropyl group, a n-butyl group, and an isobutyl group; and substituted alkyl groups such as a chloromethyl group. Among such groups, an alkyl group, in particular an unsubstituted alkyl group, is preferable, and a methyl group or an ethyl group is more preferable. In one embodiment, R^j is a methyl group, and in another embodiment, R^j is an ethyl group.

[0228] $R^{23'}$ is each independently a hydrogen atom or a monovalent organic group. Such a monovalent organic group is a monovalent organic group excluding the hydrolyzable group.

[0229] In one embodiment, the monovalent organic group is a C_{1-20} alkyl group, more preferably a C_{1-6} alkyl group, and even more preferably a methyl group.

[0230] $p1'$ is each independently an integer 0 to 3, $q1'$ is each independently an integer of 0 to 3, and $r1'$ is each independently an integer of 0 to 3. The sum of p', $q1'$, and $r1'$ is 3 in the (Si$R^{21'}_{p1'}R^{22'}_{q1'}R^{23'}_{r1'}$) unit.

[0231] In one embodiment, $p1'$ is 0.

[0232] In one embodiment, $p1'$ may be each independently an integer of 1 to 3, an integer of 2 to 3, or 3 for each (Si$R^{21'}_{p1'}R^{22'}_{q1'}R^{23'}_{r1'}$) unit. In a preferable embodiment, $p1'$ is 3.

[0233] In one embodiment, $q1'$ is each independently an integer of 1 to 3, preferably an integer of 2 to 3, and more preferably 3 for each (Si$R^{21'}_{p1'}R^{22'}_{q1'}R^{23'}_{r1'}$) unit.

[0234] In one embodiment, $p1'$ is 0, and $q1'$ is each independently an integer of 1 to 3, preferably an integer of 2 to 3, and even more preferably 3 for each (Si$R^{21'}_{p1'}R^{22'}_{q1'}R^{23'}_{r1'}$) unit.

[0235] R^{22} is each independently a hydroxyl group or a hydrolyzable group.

[0236] R^{22} is preferably each independently a hydrolyzable group.

[0237] R^{22} is preferably each independently —OR^j, —$OCOR^j$, —O—N=CR^j_2, —NR^j_2, —NHR^j, —NCO, or a halogen (in these formulae, R^j represents a substituted or unsubstituted C_{1-4} alkyl group), and more preferably —OR^j (that is, an alkoxy group). Examples of R^j include unsubstituted alkyl groups such as a methyl group, an ethyl group, a propyl group, an isopropyl group, a n-butyl group, and an isobutyl group; and substituted alkyl groups such as a chloromethyl group. Among such groups, an alkyl group, in particular an unsubstituted alkyl group, is preferable, and a methyl group or an ethyl group is more preferable. In one embodiment, R^j is a methyl group, and in another embodiment, R^j is an ethyl group.

[0238] R^{23} is each independently a hydrogen atom or a monovalent organic group. Such a monovalent organic group is a monovalent organic group excluding the hydrolyzable group.

[0239] In one embodiment, the monovalent organic group is a C_{1-20} alkyl group, more preferably a C_{1-6} alkyl group, and even more preferably a methyl group.

[0240] $p1$ is each independently an integer 0 to 3, $q1$ is each independently an integer of 0 to 3, and $r1$ is each independently an integer of 0 to 3. The sum of $p1$, $q1$, and $r1$ is 3 in the (Si$R^{21}_{p1}R^{22}_{q1}R^{23}_{r1}$) unit.

[0241] In one embodiment, $p1$ is 0.

[0242] In one embodiment, $p1$ may be each independently an integer of 1 to 3, an integer of 2 to 3, or 3 for each (Si$R^{21}_{p1}R^{22}_{q1}R^{23}_{r1}$) unit. In a preferable embodiment, $p1$ is 3.

[0243] In one embodiment, q1 is each independently an integer of 1 to 3, preferably an integer of 2 to 3, and more preferably 3 for each $(SiR^{21}_{p1}R^{22}_{q1}R^{23}_{r1})$ unit.

[0244] In one embodiment, p1 is 0, and q1 is each independently an integer of 1 to 3, preferably an integer of 2 to 3, and even more preferably 3 for each $(SiR^{21}_{p1}R^{22}_{q1}R^{23}_{r1})$ unit.

[0245] In the above formula, R^{b1} is each independently a hydroxyl group or a hydrolyzable group.

[0246] R^{b1} is preferably each independently a hydrolyzable group.

[0247] R^{b1} is preferably each independently —OR, —OCOR^j, —O—N═CR$^j_2$, —NR$^j_2$, —NHR^j, —NCO, or a halogen (in these formulae, R^j represents a substituted or unsubstituted C_{1-4} alkyl group), and more preferably —OR^j (that is, an alkoxy group). Examples of R^j include unsubstituted alkyl groups such as a methyl group, an ethyl group, a propyl group, an isopropyl group, a n-butyl group, and an isobutyl group; and substituted alkyl groups such as a chloromethyl group. Among such groups, an alkyl group, in particular an unsubstituted alkyl group, is preferable, and a methyl group or an ethyl group is more preferable. In one embodiment, R^j is a methyl group, and in another embodiment, R^j is an ethyl group.

[0248] In the above formula, R^{c1} is each independently a hydrogen atom or a monovalent organic group. Such a monovalent organic group is a monovalent organic group excluding the above hydrolyzable group.

[0249] In one embodiment, the monovalent organic group is a C_{1-20} alkyl group, more preferably a C_{1-6} alkyl group, and even more preferably a methyl group.

[0250] k1 is each independently an integer 0 to 3, l1 is each independently an integer of 0 to 3, and m1 is each independently an integer of 0 to 3. The sum of k1, l1, and m1 is 3 in the $(SiR^{a1}_{k1}R^{b1}_{l1}R^{c1}_{m1})$ unit.

[0251] In one embodiment, k1 is each independently an integer of 1 to 3, preferably 2 or 3, and more preferably 3 for each $(SiR^{a1}_{k1}R^{b1}_{l1}R^{c1}_{m1})$ unit. In a preferable embodiment, k1 is 3.

[0252] In the above formulae (A1) and (A2), when R^{Si} is a group represented by the formula (S3), preferably, at least two Si atoms to which a hydroxyl group or a hydrolyzable group is bonded are present in the terminal moieties of the formula (A1) and the formula (A2).

[0253] In a preferable embodiment, the group represented by the formula (S3) has any one of -Z^1-SiR$^{22}_{q1}$R$^{23}_{r1}$ (wherein q1 is an integer of 1 to 3, preferably 2 or 3, and more preferably 3, and r1 is an integer of 0 to 2), -Z$^{1'}$-SiR$^{22'}_{q1'}$R$^{23'}_{r1'}$ (wherein q1' is an integer of 1 to 3, preferably 2 or 3, and more preferably 3, and r1' is an integer of 0 to 2), and -Z$^{1''}$-SiR$^{22''}_{q1''}$R$^{23''}_{r1''}$ (wherein q1'' is an integer of 1 to 3, preferably 2 or 3, and more preferably 3, and r1'' is an integer of 0 to 2). Z^1, Z$^{1'}$, Z$^{1''}$, R^{22}, R^{23}, R$^{22'}$, R$^{23'}$, R$^{22''}$, and R$^{23''}$ have the same meanings as defined above.

[0254] In a preferable embodiment, when R$^{21'}$ is present in the formula (S3), q1'' is an integer of 1 to 3, preferably 2 or 3, and more preferably 3 in at least one and preferably all R$^{21'}$ groups.

[0255] In a preferable embodiment, when R^{21} is present in the formula (S3), p1' is 0, and q1' is an integer of 1 to 3, preferably 2 or 3, and more preferably 3 in at least one and preferably all R^{21} groups.

[0256] In a preferable embodiment, when R^{a1} is present in the formula (S3), p1 is 0, and q1 is an integer of 1 to 3, preferably 2 or 3, and more preferably 3 in at least one and preferably all R^{a1} groups.

[0257] In a preferable embodiment, in the formula (S3), k1 is 2 or 3 and preferably 3, p1 is 0, q1 is 2 or 3 and preferably 3.

[0258] R^{d1} is each independently -Z^2-CR$^{31}_{p2}$R$^{32}_{q2}$R$^{33}_{r2}$.

[0259] Z^2 is each independently a single bond, an oxygen atom, or a divalent organic group. Below, the structure denoted as Z^2 is bonded to $(CR^{31}_{p2}R^{32}_{q2}R^{33}_{r2})$ on the right side.

[0260] In a preferable embodiment, Z^2 is a divalent organic group.

[0261] Z^2 is preferably a C_{1-6} alkylene group, —(CH$_2$)$_{z5}$—O—(CH$_2$)$_{z6}$— (wherein z5 is an integer of 0 to 6 such as an integer of 1 to 6, and z6 is an integer of 0 to 6 such as an integer of 1 to 6), or —(CH$_2$)$_{z7}$-phenylene-(CH$_2$)$_{z8}$— (wherein z7 is an integer of 0 to 6 such as an integer of 1 to 6, and z8 is an integer of 0 to 6 such as an integer of 1 to 6). Such a C_{1-6} alkylene group may be linear or may be branched, and is preferably linear. These groups may be substituted with, for example, one or more substituents selected from a fluorine atom, a C_{1-6} alkyl group, a C_{2-6} alkenyl group, and a C_{2-6} alkynyl group, and are preferably unsubstituted.

[0262] In a preferable embodiment, Z^2 is a C_{1-6} alkylene group or —(CH$_2$)$_{z7}$-phenylene-(CH$_2$)$_{z8}$—, and preferably -phenylene-(CH$_2$)$_{z8}$—. When Z^2 is such a group, light resistance, in particular ultraviolet resistance, can be further enhanced.

[0263] In another preferable embodiment, Z^2 is a C_{1-3} alkylene group. In one embodiment, Z^2 may be —CH$_2$CH$_2$CH$_2$—. In another embodiment, Z^2 may be —CH$_2$CH$_2$—.

[0264] R^{31} is each independently -Z$^{2'}$-CR$^{32'}_{q2'}$R$^{33'}_{r2'}$.

[0265] $Z^{2'}$ is each independently a single bond, an oxygen atom, or a divalent organic group. Below, the structure denoted as $Z^{2'}$ is bonded to $(CR^{32'}_{q2'}R^{33'}_{r2'})$ on the right side.

[0266] $Z^{2'}$ is preferably a C_{1-6} alkylene group, —(CH$_2$)$_{z5'}$—O—(CH$_2$)$_{z6'}$— (wherein z5' is an integer of 0 to 6 such as an integer of 1 to 6, and z6' is an integer of 0 to 6 such as an integer of 1 to 6), or —(CH$_2$)$_{z7'}$-phenylene-(CH$_2$)$_{z8'}$— (wherein z7' is an integer of 0 to 6 such as an integer of 1 to 6, and z8' is an integer of 0 to 6 such as an integer of 1 to 6). Such a C_{1-6} alkylene group may be linear or may be branched, and is preferably linear. These groups may be substituted with, for example, one or more substituents selected from a fluorine atom, a C_{1-6} alkyl group, a C_{2-6} alkenyl group, and a C_{2-6} alkynyl group, and are preferably unsubstituted.

[0267] In a preferable embodiment, $Z^{2'}$ is a C_{1-6} alkylene group or —(CH$_2$)$_{z7'}$-phenylene-(CH$_2$)$_{z8'}$—, and preferably -phenylene-(CH$_2$)$_{z8'}$—. When $Z^{2'}$ is such a group, light resistance, in particular ultraviolet resistance, can be further enhanced.

[0268] In another preferable embodiment, $Z^{2'}$ is a C_{1-3} alkylene group. In one embodiment, $Z^{2'}$ may be —CH$_2$CH$_2$CH$_2$—. In another embodiment, $Z^{2'}$ may be —CH$_2$CH$_2$—.

[0269] $R^{32'}$ is each independently -Z^3-SiR$^{34}_{n2}$R$^{35}_{3-n2}$.

[0270] Z^3 is each independently a single bond, an oxygen atom, or a divalent organic group. Below, the structure denoted as Z^3 is bonded to $(SiR^{34}_{n2}R^{35}_{3-n2})$ on the right side.

[0271] In one embodiment, Z^3 is an oxygen atom.

[0272] In one embodiment, Z^3 is a divalent organic group.

[0273] Z^3 is preferably a C_{1-6} alkylene group, $—(CH_2)_{z5''}—O—(CH_2)_{z6''}—$ (wherein z5" is an integer of 0 to 6 such as an integer of 1 to 6, and z6" is an integer of 0 to 6 such as an integer of 1 to 6), or $—(CH_2)_{z7''}$-phenylene-$(CH_2)_{z8''}—$ (wherein z7" is an integer of 0 to 6 such as an integer of 1 to 6, and z8" is an integer of 0 to 6 such as an integer of 1 to 6). Such a C_{1-6} alkylene group may be linear or may be branched, and is preferably linear. These groups may be substituted with, for example, one or more substituents selected from a fluorine atom, a C_{1-6} alkyl group, a C_{2-6} alkenyl group, and a C_{2-6} alkynyl group, and are preferably unsubstituted.

[0274] In a preferable embodiment, Z^3 is a C_{1-6} alkylene group or $—(CH_2)_{z7''}$-phenylene-$(CH_2)_{z8''}—$, preferably -phenylene-$(CH_2)_{z8''}—$. When Z^3 is such a group, light resistance, in particular ultraviolet resistance, can be further enhanced.

[0275] In another preferable embodiment, Z^3 is a C_{1-3} alkylene group. In one embodiment, Z^3 may be $—CH_2CH_2CH_2—$. In another embodiment, Z^3 may be $—CH_2CH_2—$.

[0276] R^{34} is each independently a hydroxyl group or a hydrolyzable group.

[0277] R^{34} is preferably each independently a hydrolyzable group.

[0278] R^{34} is preferably each independently $—OR^j$, $—OCOR^j$, $—O—N=CR^j_2$, $—NR^j_2$, $—NHR^j$, $—NCO$, or a halogen (in these formulae, R^j represents a substituted or unsubstituted C_{1-4} alkyl group), and more preferably $—OR^j$ (that is, an alkoxy group). Examples of R^j include unsubstituted alkyl groups such as a methyl group, an ethyl group, a propyl group, an isopropyl group, a n-butyl group, and an isobutyl group; and substituted alkyl groups such as a chloromethyl group. Among such groups, an alkyl group, in particular an unsubstituted alkyl group, is preferable, and a methyl group or an ethyl group is more preferable. In one embodiment, R^j is a methyl group, and in another embodiment, R^j is an ethyl group.

[0279] R^{35} is each independently a hydrogen atom or a monovalent organic group. Such a monovalent organic group is a monovalent organic group excluding the above hydrolyzable group.

[0280] In R^{35}, the monovalent organic group is preferably a C_{1-20} alkyl group, more preferably a C_{1-6} alkyl group, and even more preferably a methyl group.

[0281] In the above formula, n2 is each independently an integer of 0 to 3 for each $(SiR^{34}_{n2}R^{35}_{3-n2})$ unit. However, when R^{Si} is a group represented by the formula (S4), the terminal moieties of the formula (A1) and the formula (A2) have at least one $(SiR^{34}_{n2}R^{35}_{3-n2})$ unit wherein n2 is 1 to 3. That is, in such terminal moieties, not all n2 are 0 at the same time. In other words, in the terminal moieties of the formula (A1) and the formula (A2), at least one Si atom to which a hydroxyl group or a hydrolyzable group is bonded is present.

[0282] n2 is each independently an integer of preferably 1 to 3, more preferably 2 to 3, and even more preferably 3 for each $(SiR^{34}_{n2}R^{35}_{3-n2})$ unit.

[0283] $R^{33'}$ is each independently a hydrogen atom, a hydroxyl group, or a monovalent organic group. Such a monovalent organic group is a monovalent organic group excluding the hydrolyzable group.

[0284] In $R^{33'}$, the monovalent organic group may preferably be a C_{1-20} alkyl group or a group represented by $R^{41'}—(OR^{42'})_{y1'}—O_{y2'}—$, wherein $R^{41'}$ is a hydrogen atom or a C_{1-6} alkyl group, $R^{42'}$ is each independently a C_{1-6} alkylene group, y1' is an integer of 1 to 30, and y2' is 0 or 1.

[0285] In one embodiment, $R^{41'}$ is a hydrogen atom.

[0286] In another embodiment, $R^{41'}$ is a C_{1-6} alkyl group and preferably a C_{1-3} alkyl group.

[0287] $R^{42'}$ is each independently a C_{1-6} alkylene group, preferably a C_{1-3} alkylene group, and more preferably a C_{2-3} alkylene group.

[0288] y1' is an integer of 1 to 30, preferably an integer of 1 to 20, more preferably an integer of 1 to 10, even more preferably an integer of 2 to 10, such as an integer of 2 to 6, an integer of 2 to 4, an integer of 3 to 6, or an integer of 4 to 6.

[0289] In one embodiment, $R^{33'}$ is a hydroxyl group.

[0290] In another embodiment, $R^{33'}$ is a C_{1-20} alkyl group and more preferably a C_{1-6} alkyl group.

[0291] In another embodiment, $R^{33'}$ is $R^{41'}—(OR^{42'})_{y1'}—O_{y2'}—$.

[0292] q2' is each independently an integer of 0 to 3, and r2' is each independently an integer of 0 to 3. The sum of q2' and r2' is 3 in the $(CR^{32'}_{q2'}R^{33'}_{r2'})$ unit.

[0293] q2' is each independently an integer of preferably 1 to 3, more preferably 2 to 3, and even more preferably 3 for each $(CR^{32'}_{q2'}R^{33'}_{r2'})$ unit.

[0294] R^{32} is each independently $-Z^3-SiR^{34}_{n2}R^{35}_{3-n2}$. Such $-Z^3-SiR^{34}_{n2}R^{35}_{3-n2}$ has the same meanings as those described for $R^{32'}$.

[0295] R^{33} is each independently a hydrogen atom, a hydroxyl group, or a monovalent organic group. Such a monovalent organic group is a monovalent organic group excluding the hydrolyzable group.

[0296] In R^{33}, the monovalent organic group may preferably be a C_{1-20} alkyl group or a group represented by $R^{41}—(OR^{42})_{y1}—O_{y2}—$, wherein R^{41} is a hydrogen atom or a C_{1-6} alkyl group, R^{42} is each independently a C_{1-6} alkylene group, y1 is an integer of 1 to 30, and y2 is 0 or 1.

[0297] In one embodiment, R^{41} is a hydrogen atom.

[0298] In another embodiment, R^{41} is a C_{1-6} alkyl group and preferably a C_{1-3} alkyl group.

[0299] R^{42} is each independently a C_{1-6} alkylene group, preferably a C_{1-3} alkylene group, and more preferably a C_{2-3} alkylene group.

[0300] y1 is an integer of 1 to 30, preferably an integer of 1 to 20, more preferably an integer of 1 to 10, even more preferably an integer of 2 to 10, such as an integer of 2 to 6, an integer of 2 to 4, an integer of 3 to 6, or an integer of 4 to 6.

[0301] In one embodiment, R^{33} is a hydroxyl group.

[0302] In another embodiment, R^{33} is a C_{1-20} alkyl group and more preferably a C_{1-6} alkyl group.

[0303] In another embodiment, R^{33} is $R^{41}—(OR^{42})_{y1}—O_{y2}—$.

[0304] p2 is each independently an integer of 0 to 3, q2 is each independently an integer of 0 to 3, and r2 is each independently an integer of 0 to 3. The sum of p2, q2, and r2 is 3 in the $(CR^{31}_{p2}R^{32}_{q2}R^{33}_{r2})$ unit.

[0305] In one embodiment, p2 is 0.

[0306] In one embodiment, p2 may be each independently an integer of 1 to 3, an integer of 2 to 3, or 3 for each $(CR^{31}{}_{p2}R^{32}{}_{q2}R^{33}{}_{r2})$ unit. In a preferable embodiment, p2 is 3.

[0307] In one embodiment, q2 is each independently an integer of 1 to 3, preferably an integer of 2 to 3, and more preferably 3 for each $(CR^{31}{}_{p2}R^{32}{}_{q2}R^{33}{}_{r2})$ unit.

[0308] In one embodiment, p2 is 0, and q2 is each independently an integer of 1 to 3, preferably an integer of 2 to 3, and even more preferably 3 for each $(CR^{31}{}_{p2}R^{32}{}_{q2}R^{33}{}_{r2})$ unit.

[0309] R^{e1} is each independently $-Z^3-SiR^{34}{}_{n2}R^{35}{}_{3-n2}$. Such $-Z^3-SiR^{34}{}_{n2}R^{35}{}_{3-n2}$ has the same meanings as those described for $R^{32'}$.

[0310] R^{f1} is each independently a hydrogen atom, a hydroxyl group, or a monovalent organic group. Such a monovalent organic group is a monovalent organic group excluding the hydrolyzable group.

[0311] In R^{f1}, the monovalent organic group may preferably be a C_{1-20} alkyl group or a group represented by $R^{43}-(OR^{44})_{y3}-O_{y4}-$, wherein R^{43} is a hydrogen atom or a C_{1-6} alkyl group, R^{44} is each independently a C_{1-6} alkylene group, y3 is an integer of 1 to 30, and y4 is 0 or 1.

[0312] In one embodiment, R^{43} is a hydrogen atom.

[0313] In another embodiment, R^{43} is a C_{1-6} alkyl group and preferably a C_{1-3} alkyl group.

[0314] R^{44} is each independently a C_{1-6} alkylene group, preferably a C_{1-3} alkylene group, and more preferably a C_{2-3} alkylene group.

[0315] y3 is an integer of 1 to 30, preferably an integer of 1 to 20, more preferably an integer of 1 to 10, even more preferably an integer of 2 to 10, such as an integer of 2 to 6, an integer of 2 to 4, an integer of 3 to 6, or an integer of 4 to 6.

[0316] In one embodiment, R^{f1} is a hydroxyl group.

[0317] In another embodiment, R^{f1} is a C_{1-20} alkyl group and more preferably a C_{1-6} alkyl group.

[0318] In another embodiment, R^{f1} is $R^{43}-(OR^{44})_{y3}-O_{y4}-$.

[0319] k2 is each independently an integer 0 to 3, l2 is each independently an integer of 0 to 3, and m2 is each independently an integer of 0 to 3. The sum of k2, l2, and m2 is 3 in the $(CR^{d1}{}_{k2}R^{e1}{}_{l2}R^{f1}{}_{m2})$ unit.

[0320] In the above formulae (A1) and (A2), when R^{Si} is a group represented by the formula (S4), preferably, at least two Si atoms to which a hydroxyl group or a hydrolyzable group is bonded are present in the terminal moieties of the formula (A1) and the formula (A2).

[0321] In one embodiment, when R^{Si} is a group represented by the formula (S4), 2 or more, for example, 2 to 27, preferably 2 to 9, more preferably 2 to 6, even more preferably 2 to 3, and particularly preferably 3 $(SiR^{34}{}_{n2}R^{35}{}_{3-n2})$ units wherein n2 is 1 to 3, preferably 2 or 3, and more preferably 3 are present in each terminal moiety of the formula (A1) and the formula (A2).

[0322] In a preferable embodiment, when $R^{32'}$ is present in the formula (S4), n2 is an integer of 1 to 3, preferably 2 or 3, and more preferably 3 in at least one and preferably all $R^{32'}$ groups.

[0323] In a preferable embodiment, when R^{32} is present in the formula (S4), n2 is an integer of 1 to 3, preferably 2 or 3, and more preferably 3 in at least one and preferably all R^{32} groups.

[0324] In a preferable embodiment, when R^{e1} is present in the formula (S4), n2 is an integer of 1 to 3, preferably 2 or 3, and more preferably 3 in at least one and preferably all R^{a1} groups.

[0325] In a preferable embodiment, k2 is 0, l2 is 2 or 3 and preferably 3, and n2 is 2 or 3 and preferably 3 in the formula (S4).

[0326] R^{g1} and R^{h1} are each independently $-Z^4-SiR^{11}{}_{n1}R^{12}{}_{3-n1}$, $-Z^4-SiR^{a1}{}_{k1}R^{b1}{}_{l1}R^{c1}{}_{m1}$, or $-Z^4-CR^{d1}{}_{k2}R^{e1}{}_{l2}R^{f1}{}_{m2}$. Here, R^1, R^{12}, R^{a1}, R^{b2}, R^{c1}, R^{d1}, R^{e1}, R^{f1}, n1, k1, l1, m1, k2, l2, and m2 are as defined above.

[0327] In a preferable embodiment, R^{g1} and R^{h1} are each independently $-Z^4-SiR^{11}{}_{n1}R^{12}{}_{3-n1}$.

[0328] Z^4 is each independently a single bond, an oxygen atom, or a divalent organic group. Below, the structure denoted as Z^4 is bonded to $(SiR^{11}{}_{n1}R^{12}{}_{3-n1})$ on the right side.

[0329] In one embodiment, Z^4 is an oxygen atom.

[0330] In one embodiment, Z^4 is a divalent organic group.

[0331] Z^4 is preferably a C_{1-6} alkylene group, $-(CH_2)_{z5''}-O-(CH_2)_{z6''}-$ (wherein z5'' is an integer of 0 to 6 such as an integer of 1 to 6, and z6'' is an integer of 0 to 6 such as an integer of 1 to 6), or $-(CH_2)_{z7''}$-phenylene-$(CH_2)_{z8''}-$ (wherein z7'' is an integer of 0 to 6 such as an integer of 1 to 6, and z8'' is an integer of 0 to 6 such as an integer of 1 to 6). Such a C_{1-6} alkylene group may be linear or may be branched, and is preferably linear. These groups may be substituted with, for example, one or more substituents selected from a fluorine atom, a C_{1-6} alkyl group, a C_{2-6} alkenyl group, and a C_{2-6} alkynyl group, and are preferably unsubstituted.

[0332] In a preferred embodiment, Z^4 is a C_{1-6} alkylene group or $-(CH_2)_{z7''}$-phenylene-$(CH_2)_{z8''}-$, and preferably -phenylene-$(CH_2)_{z8''}-$. When Z^3 is such a group, light resistance, in particular ultraviolet resistance, can be further enhanced.

[0333] In another preferable embodiment, Z^4 is a C_{1-3} alkylene group. In one embodiment, Z^4 may be $-CH_2CH_2CH_2-$. In another embodiment, Z^4 may be $-CH_2CH_2-$.

[0334] In one embodiment, R^{Si} is a group represented by the formula (S2), (S3), (S4), or (S5). These compounds are capable of forming a surface-treating layer having high surface lubricity.

[0335] In one embodiment, R^{Si} is a group represented by the formula (S1), (S3), (S4), or (S5). These compounds have a plurality of hydrolyzable groups at one terminal, and are therefore capable of forming a surface-treating layer that firmly adheres to a substrate and that has high friction durability.

[0336] In one embodiment, R^{Si} is a group represented by the formula (S1), (S3), or (S4). These compounds have a plurality of hydrolyzable groups at one terminal, and are therefore capable of forming a surface-treating layer that firmly adheres to a substrate and that has high friction durability.

[0337] In one embodiment, R^{Si} is a group represented by the formula (S3) or (S4). These compounds can have a plurality of hydrolyzable groups branched from one Si atom or C atom at one terminal, and are therefore capable of forming a surface-treating layer that has higher friction durability.

[0338] In one embodiment, R^{Si} is a group represented by the formula (S1).

[0339] In one embodiment, R^{Si} is a group represented by the formula (S2).

[0340] In one embodiment, R^{Si} is a group represented by the formula (S3).

[0341] In one embodiment, R^{Si} is a group represented by the formula (S4).

[0342] In one embodiment, R^{Si} is a group represented by the formula (S5).

[0343] In the formulae (A1) and (A2), X^A is interpreted as a linker connecting fluoropolyether moieties (R^{F1} and R^{F2}), which mainly provide water-repellency, surface lubricity and the like, to a moiety (R^{Si}) providing the ability of binding to a substrate. Accordingly, such X^A may be a single bond or any group as long as the compounds represented by the formulae (A1) and (A2) can stably exist.

[0344] In the above formula (A1), $\alpha 1$ is an integer of 1 to 9, and $\beta 1$ is an integer of 1 to 9. These $\alpha 1$ and $\beta 1$ may vary depending on the valence of X^A. The sum of $\alpha 1$ and $\beta 1$ is the same as the valence of X^A. For example, when X^A is a decavalent organic group, the sum of $\alpha 1$ and $\beta 1$ is 10, e.g., $\alpha 1$ may be 9 and β may be 1, $\alpha 1$ may be 5 and $\beta 1$ may be 5, or $\alpha 1$ may be 1 and $\beta 1$ may be 9. When X^A is a divalent organic group, $\alpha 1$ and $\beta 1$ are 1.

[0345] In the above formula (A2), γ is an integer of 1 to 9. γ may vary depending on the valence of X^A. That is, γ is a value obtained by subtracting 1 from the valence of X^A.

[0346] X^A is each independently a single bond or a di- to decavalent organic group.

[0347] In one embodiment, X^A does not include a siloxane bond (—Si—O—Si—).

[0348] The di- to decavalent organic group in X^A is preferably a di- to octavalent organic group. In one embodiment, such a di- to decavalent organic group is preferably a di- to tetravalent organic group, and more preferably a divalent organic group. In another embodiment, such a di- to decavalent organic group is preferably a tri- to octavalent organic group, and more preferably a tri- to hexavalent organic group.

[0349] In one embodiment, X^A is a single bond or a divalent organic group, $\alpha 1$ is 1, and $\beta 1$ is 1.

[0350] In one embodiment, X^A is a single bond or a divalent organic group, γ is 1.

[0351] In one embodiment, X^A is a tri- to hexavalent organic group, $\alpha 1$ is 1, and $\beta 1$ is 2 to 5.

[0352] In one embodiment, X^A is a tri- to hexavalent organic group, and γ is 2 to 5.

[0353] In one embodiment, X^A is a trivalent organic group, $\alpha 1$ is 1, and $\beta 1$ is 2.

[0354] In one embodiment, X^A is a trivalent organic group, and γ is 2.

[0355] When X^A is a single bond or a divalent organic group, the formulae (A1) and (A2) are represented by the following formulae (A1') and (A2').
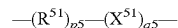
[Formula 22]

$$R^{F1}—X^A—R^{Si} \tag{A1'}$$

$$R^{Si}—X^A—R^{F2}—X^A—R^{Si} \tag{A2'}$$

[0356] In one embodiment, X^A is a single bond.

[0357] In another embodiment, X^A is a divalent organic group.

[0358] In one embodiment, examples of X^A include a single bond or a divalent organic group represented by the following formula:

$$—(R^{51})_{p5}—(X^{51})_{q5}—$$

wherein

[0359] R^{51} represents a single bond, —$(CH_2)_{s5}$—, or an o-, m- or p-phenylene group, and is preferably —$(CH_2)_{s5}$—,

[0360] s5 is an integer of 1 to 20, preferably 1 to 6, more preferably 1 to 3, and even more preferably 1 or 2,

[0361] X^{51} represents —$(X^{52})_{l5}$—,

[0362] X^{52} each independently represents a group selected from the group consisting of —O—, —S—, —CO—, —NR^{54}—, —$(CH_2)_{n5}$—, an o-, m- or p-phenylene group, —COO—, —OCO—, —$CONR^{54}$—, —$NR^{54}CO$—, —O—$CONR^{54}$—, —$NR^{54}CO$—O—, —$Si(R^{53})_2$—, and —$(Si(R^{53})_2O)_{m5}$—$Si(R^{53})_2$—,

[0363] R^{53} each independently represents a phenyl group, a C_{1-6} alkyl group, or a C_{1-6} alkoxy group, is preferably a phenyl group or a C_{1-6} alkyl group, and is more preferably a methyl group,

[0364] R^{54} each independently represents a hydrogen atom, a phenyl group, or a C_{1-6} alkyl group (preferably a methyl group),

[0365] m5 is each independently at each occurrence an integer of 1 to 100, and preferably an integer of 1 to 20,

[0366] n5 is each independently at each occurrence an integer of 1 to 20, preferably an integer of 1 to 6, and more preferably an integer of 1 to 3,

[0367] l5 is an integer of 1 to 10, preferably an integer of 1 to 5, and more preferably an integer of 1 to 3,

[0368] p5 is 0 or 1, and

[0369] q5 is 0 or 1,

[0370] provided that at least one of p5 and q5 is 1, the occurrence order of the respective repeating units enclosed within parentheses provided with p5 or q5 is not limited, and the right side is bonded to R^{Si}.

[0371] In a preferable embodiment, X^A is

[0372] X^A is a single bond or a divalent organic group represented by the following formula:

$$(R^{51})_{p5}—(X^{51})_{q5}—$$
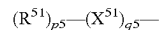
wherein

[0373] R^{51} is a single bond or —$(CH_2)_{s5}$—,

[0374] s5 is an integer of 1 to 20,

[0375] X^{51} is —$(X^{52})_{l5}$—,

[0376] X^{52} each independently represents a group selected from the group consisting of —O—, —S—, —CO—, —NR^{54}—, —$(CH_2)_{n5}$—, —COO—, —OCO—, —$CONR^{54}$—, —$NR^{54}CO$—, —O—$CONR^{54}$—, —$NR^{54}CO$—O—, —$Si(R^{53})_2$—, and —$(Si(R^{53})_2O)_{m5}$—$Si(R^{53})_2$—,

[0377] R^{53} is each independently a phenyl group, a C_{1-6} alkyl group, or a C_{1-6} alkoxy group,

[0378] R^{54} is each independently a hydrogen atom, a phenyl group, or a C_{1-6} alkyl group,

[0379] m5 is each independently an integer of 1 to 100,

[0380] n5 is each independently an integer of 1 to 20,

[0381] l5 is an integer of 1 to 10,

[0382] p5 is 0 or 1, and

[0383] q5 is 0 or 1,

[0384] provided that at least one of p5 and q5 is 1, the occurrence order of the respective repeating units enclosed within parentheses provided with p5 or q5 is not limited, and

[0385] the right side is bonded to R^{Si}.

[0386] Preferably, X^A may be each independently

[0387] a single bond,

[0388] a C_{1-20} alkylene group,

[0389] $-R^{51}-X^{53}-R^{52}-$, or

[0390] $-X^{54}-R^5-$

[0391] wherein R^{51} and R^{52} are as defined above,

[0392] X^{53} is

[0393] $-O-$,

[0394] $-S-$,

[0395] $-C(O)O-$,

[0396] $-CONR^{54}-$,

[0397] $-O-CONR^{54}-$,

[0398] $-Si(R^{53})_2-$,

[0399] $-(Si(R^{53})_2O)_{m5}-Si(R^{53})_2-$,

[0400] $-O-(CH_2)_{u5}-(Si(R^{53})_2O)_{m5}-Si(R^{53})_2-$,

[0401] $-O-(CH_2)_{u5}-Si(R^{53})_2-O-Si(R^{53})_2-CH_2CH_2-Si(R^{53})_2-O-Si(R^{53})_2-$,

[0402] $-O-(CH_2)_{u5}-Si(OCH_3)_2OSi(OCH_3)_2-$,

[0403] $-CONR^{54}-(CH_2)_{u5}-(Si(R^{53})_2O)_{m5}-Si(R^{53})_2-$, or

[0404] $-CONR^{54}-(CH_2)_{u5}-N(R^{54})-$,

[0405] (wherein R^{53}, R^{54}, and m5 are as defined above, and

[0406] u5 is an integer of 1 to 20, preferably an integer of 2 to 6, and more preferably an integer of 2 to 3), and

[0407] X^{54} represents

[0408] $-S-$,

[0409] $-C(O)O-$,

[0410] $-CONR^{54}-$,

[0411] $-O-CONR^{54}-$,

[0412] $-CONR^{54}-(CH_2)_{u5}-(Si(R^{54})_2O)_{m5}-Si(R^{54})_2-$, or

[0413] $-CONR^{54}-(CH_2)_{u5}-N(R^{54})-$,

[0414] wherein each symbol is as defined above.

[0415] More preferably, X^A is each independently

[0416] a single bond,

[0417] a C_{1-20} alkylene group,

[0418] $-(CH_2)_{s5}-X^{53}-$,

[0419] $-(CH_2)_{s5}-X^{53}-(CH_2)_{t5}-$,

[0420] $-X^{54}-$, or

[0421] $-X^{54}-(CH_2)_{t5}-$,

[0422] wherein X^{53}, X^{54}, s5, and t5 are as defined above.

[0423] More preferably, X^A is each independently

[0424] a single bond,

[0425] a C_{1-20} alkylene group,

[0426] $-(CH_2)_{s5}-X^{53}-(CH_2)_{t5}-$, or

[0427] $-X^{54}-(CH_2)_{t5}-$,

[0428] wherein each symbol is as defined above.

[0429] In a preferable embodiment, X^A may be each independently

[0430] a single bond,

[0431] a C_{1-20} alkylene group,

[0432] $-(CH_2)_{s5}-X^{53}-$, or

[0433] $-(CH_2)_{s5}-X^{53}-(CH_2)_{t5}-$,

[0434] wherein

[0435] X^{53} is $-O-$, $-CONR^{54}-$, or $-O-CONR^{54}-$,

[0436] R^{54} each independently represents a hydrogen atom, a phenyl group, or a C_{1-6} alkyl group,

[0437] s5 is an integer of 1 to 20, and

[0438] t5 is an integer of 1 to 20.

[0439] In a preferable embodiment, X^A may each independently be

[0440] $-(CH_2)_{s5}-O-(CH_2)_{t5}-$ or

[0441] $-CONR^{54}-(CH_2)_{t5}-$,

[0442] wherein

[0443] R 54 each independently represents a hydrogen atom, a phenyl group, or a C_{1-6} alkyl group,

[0444] s5 is an integer of 1 to 20, and

[0445] t5 is an integer of 1 to 20.

[0446] In one embodiment, X^A may each independently be

[0447] a single bond,

[0448] a C_{1-20} alkylene group,

[0449] $-(CH_2)_{s5}-O-(CH_2)_{t5}-$,

[0450] $-(CH_2)_{s5}-(Si(R^{53})_2O)_{m5}-Si(R^{53})_2-(CH_2)_{t5}-$,

[0451] $-(CH_2)_{s5}-O-(CH_2)_{u5}-(Si(R^{53})_2O)_{m5}-Si(R^{53})_2-(CH_2)_{t5}-$, or

[0452] $-(CH_2)_{s5}-O-(CH_2)_{t5}-Si(R^{53})_2-(CH_2)_{u5}-Si(R^{53})_2-(C_vH_{2v})-$,

[0453] wherein R^{53}, m5, s5, t5, and u5 are as defined above, and v5 is an integer of 1 to 20, preferably an integer of 2 to 6, and more preferably an integer of 2 to 3.

[0454] In the above formula, $-(C_vH_{2v})-$ may be linear or branched and may be, for example, $-CH_2CH_2-$, $-CH_2CH_2CH_2-$, $-CH(CH_3)-$, or $-CH(CH_3)CH_2-$.

[0455] X^A (typically the hydrogen atom of X^A) each independently is optionally substituted with one or more substituents selected from a fluorine atom, a C_{1-3} alkyl group, and a C_{1-3} fluoroalkyl group (preferably a C_{1-3} perfluoroalkyl group). In a preferable embodiment, X^A is unsubstituted.

[0456] In one embodiment, X^A may each independently be a group other than an $-O-C_{1-6}$ alkylene group.

[0457] In another embodiment, examples of X^A include the following groups:

[Formula 23]

[Formula 24]

-continued
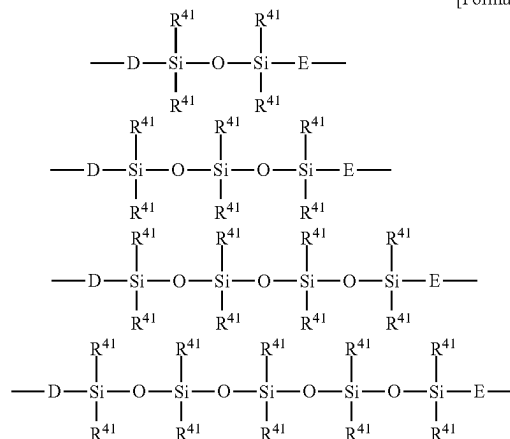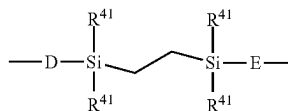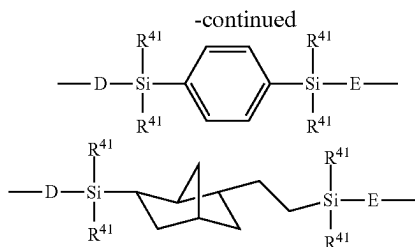
wherein R^{41} is each independently a hydrogen atom, a phenyl group, an alkyl group having 1 to 6 carbon atoms, or a C_{1-6} alkoxy group, and preferably a methyl group;

[0458]   D is a group selected from:

[0459] $-CH_2O(CH_2)_2-$,

[0460] $-CH_2O(CH_2)_3-$,

[0461] $-CF_2O(CH_2)_3-$,

[0462] $-(CH_2)_2-$,

[0463] $-(CH_2)_3-$,

[0464] $-(CH_2)_4-$,

[0465] $-CONH-(CH_2)_3-$,

[0466] $-CON(CH_3)-(CH_2)_3-$,

[0467] $-CON(Ph)-(CH_2)_3-$ (wherein Ph means phenyl), and

[Formula 25]
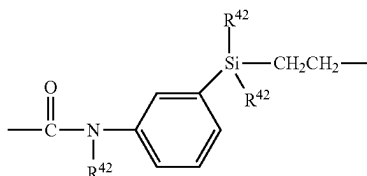
[0468]   (wherein R^{42} each independently represents a hydrogen atom, a C_{1-6} alkyl group, or a C_{1-6} alkoxy group, preferably a methyl group or a methoxy group, and more preferably a methyl group);

[0469]   E is $-(CH_2)_n-$ (n is an integer of 2 to 6); and

[0470]   D is bonded to R^{F1} or R^{F2} of the molecular backbone, and E is bonded to R^{Si}.

[0471]   Specific examples of X^4 include:

[0472]   a single bond,

[0473] $-CH_2OCH_2-$,

[0474] $-CH_2O(CH_2)_2-$,

[0475] $-CH_2O(CH_2)_3-$,

[0476] $-CH_2O(CH_2)_4-$,

[0477] $-CH_2O(CH_2)_5-$,

[0478] $-CH_2O(CH_2)_6-$,

[0479] $-CH_2O(CH_2)_3Si(CH_3)_2OSi(CH_3)_2(CH_2)_2-$,

[0480] $-CH_2O(CH_2)_3Si(CH_3)_2OSi(CH_3)_2OSi(CH_3)_2(CH_2)_2-$,

[0481] $-CH_2O(CH_2)_3Si(CH_3)_2O(Si(CH_3)_2O)_2Si(CH_3)_2(CH_2)_2-$,

[0482] $-CH_2O(CH_2)_3Si(CH_3)_2O(Si(CH_3)_2O)_3Si(CH_3)_2(CH_2)_2-$,

[0483] $-CH_2O(CH_2)_3Si(CH_3)_2O(Si(CH_3)_2O)_{10}Si(CH_3)_2(CH_2)_2-$,

[0484] $-CH_2O(CH_2)_3Si(CH_3)_2O(Si(CH_3)_2O)_{20}Si(CH_3)_2(CH_2)_2-$,

[0485] $-CH_2OCF_2CHFOCF_2-$,

[0486] $-CH_2OCF_2CHFOCF_2CF_2-$,

[0487] $-CH_2OCF_2CHFOCF_2CF_2CF_2-$,

[0488] $-CH_2OCH_2CF_2CF_2OCF_2-$,

[0489] $-CH_2OCH_2CF_2CF_2OCF_2CF_2-$

[0490] $-CH_2OCH_2CF_2CF_2OCF_2CF_2CF_2-$,

[0491] $-CH_2OCH_2CF_2CF_2OCF(CF_3)CF_2OCF_2-$,

[0492] $-CH_2OCH_2CF_2CF_2OCF(CF_3)CF_2OCF_2CF_2-$,

[0493] $-CH_2OCH_2CF_2CF_2OCF(CF_3)CF_2OCF_2CF_2CF_2-$,

[0494] $-CH_2OCH_2CHFCF_2OCF_2-$, $-CH_2OCH_2CHFCF_2OCF_2CF_2-$,

[0495] $-CH_2OCH_2CHFCF_2OCF_2CF_2CF_2-$,

[0496] $-CH_2OCH_2CHFCF_2OCF(CF_3)CF_2OCF_2-$,

[0497] $-CH_2OCH_2CHFCF_2OCF(CF_3)CF_2OCF_2CF_2-$,

[0498] $-CH_2OCH_2CHFCF_2OCF(CF_3)CF_2OCF_2CF_2CF_2-$,

[0499] $-CH_2OCF_2CHFOCF_2CF_2CF_2-C(O)NH-CH_2-$,

[0500] $-CH_2OCH_2(CH_2)_7CH_2Si(OCH_3)_2OSi(OCH_3)_2(CH_2)_2Si(OCH_3)_2OSi(OCH_3)_2(CH_2)_2-$,

[0501] $-CH_2OCH_2CH_2CH_2Si(OCH_3)_2OSi(OCH_3)_2(CH_2)_3-$,

[0502] $-CH_2OCH_2CH_2CH_2Si(OCH_2CH_3)_2OSi(OCH_2CH_3)_2(CH_2)_3-$,

[0503] $-CH_2OCH_2CH_2CH_2Si(OCH_3)_2OSi(OCH_3)_2(CH_2)_2-$,

[0504] $-CH_2OCH_2CH_2CH_2Si(OCH_2CH_3)_2OSi(OCH_2CH_3)_2(CH_2)_2-$,

[0505] $-(CH_2)_2-Si(CH_3)_2-(CH_2)_2-$,

[0506] $-CH_2-$,

[0507] $-(CH_2)_2-$,

[0508] $-(CH_2)_3-$,

[0509] $-(CH_2)_4-$,

[0510] $-(CH_2)_5-$,

[0511] $-(CH_2)_6-$,

[0512] $-CO-$,

[0513] $-CONH-$,

[0514] $-CONH-CH_2-$,

[0515] $-CONH-(CH_2)_2-$,

[0516] $-CONH-(CH_2)_3-$,

[0517] $-CONH-(CH_2)_4-$,

[0518] $-CONH-(CH_2)_5-$,

[0519] $-CONH-(CH_2)_6-$,

[0520] $-CON(CH_3)-CH_2-$,

[0521] $-CON(CH_3)-(CH_2)_2-$,

[0522] $-CON(CH_3)-(CH_2)_3-$,

[0523] $-CON(CH_3)-(CH_2)_4-$,

[0524] $-CON(CH_3)-(CH_2)_5-$,

[0525] $-CON(CH_3)-(CH_2)_6-$,

[0526] $-CONH-(CH_2)_2NH(CH_2)_3-$,

[0527] $-CONH-(CH_2)_6NH(CH_2)_3-$,

[0528] $-CH_2O-CONH-(CH_2)_3-$,

[0529] $-CH_2O-CONH-(CH_2)_6-$,

[0530] $-S-(CH_2)_3-$,

[0531] $-(CH_2)_2S(CH_2)_3-$,

[0532] $-CONH-(CH_2)_3Si(CH_3)_2OSi(CH_3)_2(CH_2)_2-$,

[0533] $-CONH-(CH_2)_3Si(CH_3)_2OSi(CH_3)_2OSi(CH_3)_2(CH_2)_2-$,

[0534] $-CONH-(CH_2)_3Si(CH_3)_2O(Si(CH_3)_2O)_2Si(CH_3)_2(CH_2)_2-$,

[0535] $-CONH-(CH_2)_3Si(CH_3)_2O(Si(CH_3)_2O)_3Si(CH_3)_2(CH_2)_2-$,

[0536] —CONH—(CH₂)₃Si(CH₃)₂O(Si(CH₃)₂O)₁₀Si(CH₃)₂(CH₂)₂—,

[0537] —CONH—(CH₂)₃Si(CH₃)₂O(Si(CH₃)₂O)₂₀Si(CH₃)₂(CH₂)₂—,

[0538] —C(O)O—(CH₂)₃—,

[0539] —C(O)O—(CH₂)₆—,

[0540] —CH₂—O—(CH₂)₃—Si(CH₃)₂—(CH₂)₂—Si(CH₃)₂—(CH₂)₂—,

[0541] —CH₂—O—(CH₂)₃—Si(CH₃)₂—(CH₂)₂—Si(CH₃)₂—CH(CH₃)—,

[0542] —CH₂—O—(CH₂)₃—Si(CH₃)₂—(CH₂)₂—Si(CH₃)₂—(CH₂)₃—,

[0543] —CH₂—O—(CH₂)₃—Si(CH₃)₂—(CH₂)₂—Si(CH₃)₂—CH(CH₃)—CH₂—,

[0544] —OCH₂—, —O(CH₂)₃—,

[0545] —OCFHCF₂—,

[Formula 26]
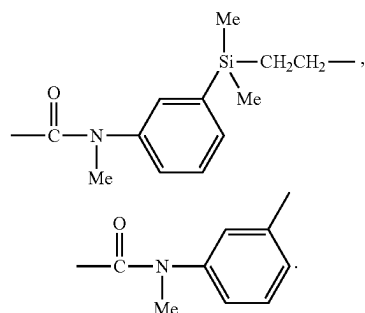
[0546] In yet another embodiment, X^A is each independently a group represented by the formula: —$(R^{16})_{x1}$—$(CFR^{17})_{y1}$—$(CH_2)_{z1}$—. In the formula, x1, y1, and z1 are each independently an integer of 0 to 10, the sum of x1, y1, and z1 is 1 or more, and the occurrence order of the respective repeating units enclosed in parentheses is not limited in the formula.

[0547] In the above formula, R^{16} is each independently an oxygen atom, phenylene, carbazolylene, —NR^{18}— (wherein R^{18} represents a hydrogen atom or an organic group), or a divalent organic group. Preferably, R^{18} is an oxygen atom or a divalent polar group.

[0548] Examples of the "divalent polar group" include, but are not limited to, —C(O)—, —$C(=NR^{19})$— and —$C(O)NR^{19}$— (wherein R^{19} represents a hydrogen atom or a lower alkyl group). The "lower alkyl group" is, for example, an alkyl group having 1 to 6 carbon atoms, such as methyl, ethyl, and n-propyl, that is optionally substituted with one or more fluorine atoms.

[0549] In the above formula, R^{17} is each independently a hydrogen atom, a fluorine atom, or a lower fluoroalkyl group, and is preferably a fluorine atom. The "lower fluoroalkyl group" is, for example, a fluoroalkyl group having 1 to 6 carbon atoms and preferably 1 to 3 carbon atoms, preferably a perfluoroalkyl group having 1 to 3 carbon atoms, more preferably a trifluoromethyl group or a pentafluoroethyl group, and even more preferably a trifluoromethyl group.

[0550] X^A is bonded to R^{F1} or R^{F2} on the left side, and bonded to R^{Si} on the right side.

[0551] In yet another embodiment, examples of X^A include the following groups:

[Formula 27]
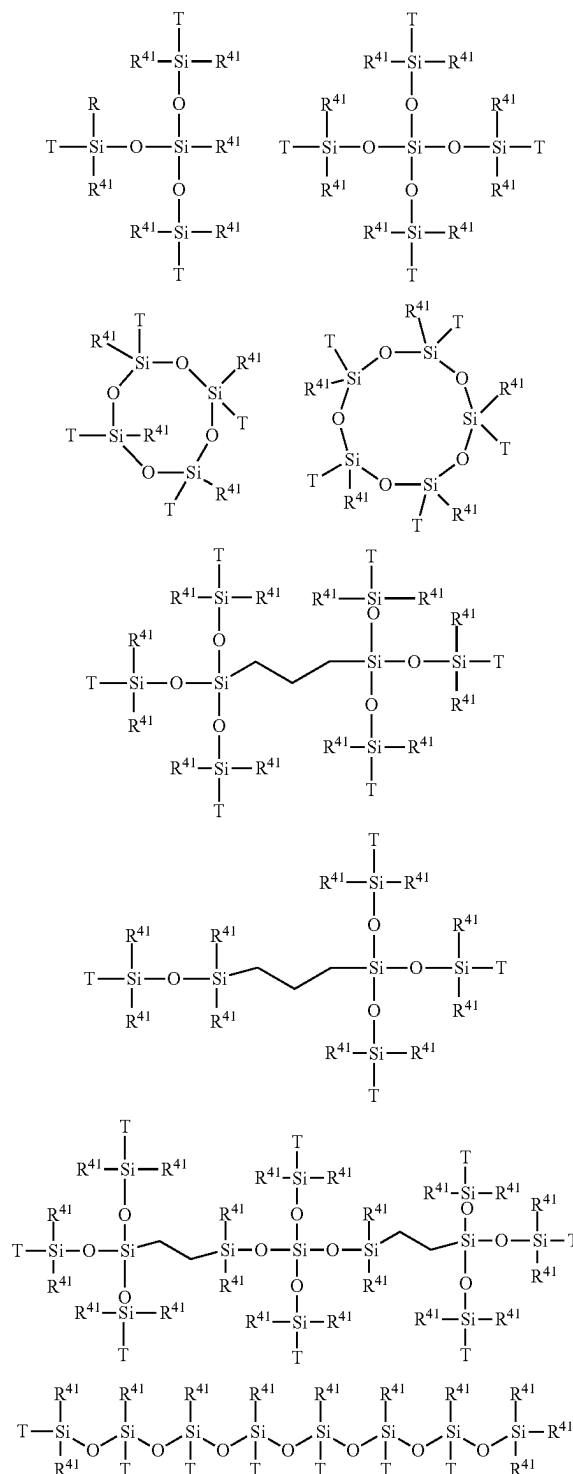
wherein

[0552] R^{41} is each independently a hydrogen atom, a phenyl group, an alkyl group having 1 to 6 carbon atoms, or a C_{1-6} alkoxy group, and is preferably a methyl group;

[0553] some of the groups of T in each X^A group are the following groups bonded to R^{F1} or R^{F2} of the molecular backbone:

[0554] —CH$_2$O(CH$_2$)$_2$—,

[0555] —CH$_2$O(CH$_2$)$_3$—,

[0556] —CF$_2$O(CH$_2$)$_3$—,

[0557] —(CH$_2$)$_2$—,

[0558] —(CH$_2$)$_3$—,

[0559] —(CH$_2$)$_4$—,

[0560] —CONH—(CH$_2$)$_3$—,

[0561] —CON(CH$_3$)—(CH$_2$)$_3$—,

[0562] —CON(Ph)—(CH$_2$)$_3$— (wherein Ph means phenyl), or

[Formula 28]
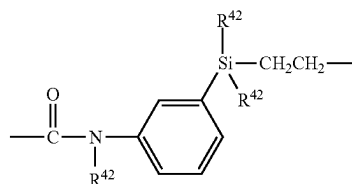
(wherein R^{42} each independently represents a hydrogen atom, a C_{1-6} alkyl group, or a C_{1-6} alkoxy group, preferably a methyl group or a methoxy group, and more preferably a methyl group); and

some other groups of T are bonded to R^{Si} of the molecular backbone, and the remaining groups of T, if present, are each independently a methyl group, a phenyl group, a C_{1-6} alkoxy group, or a radical scavenging group or a UV absorbing group.

[0563] The radical scavenging group is not limited as long as it can scavenge a radical generated by light irradiation, and examples include residues of benzophenones, benzotriazoles, benzoates, phenyl salicylates, crotonic acids, malonates, organoacrylates, hindered amines, hindered phenols, or triazines.

[0564] The UV absorbing group is not limited as long as it can absorb ultraviolet rays, and examples include residues of benzotriazoles, hydroxybenzophenones, esters of substituted and unsubstituted benzoic acid or salicylic acid compounds, acrylates or alkoxy cinnamates, oxamides, oxanilides, benzoxazinones, and benzoxazoles.

[0565] In a preferable embodiment, examples of preferable radical scavenging groups or UV absorbing groups include:

[Formula 29]
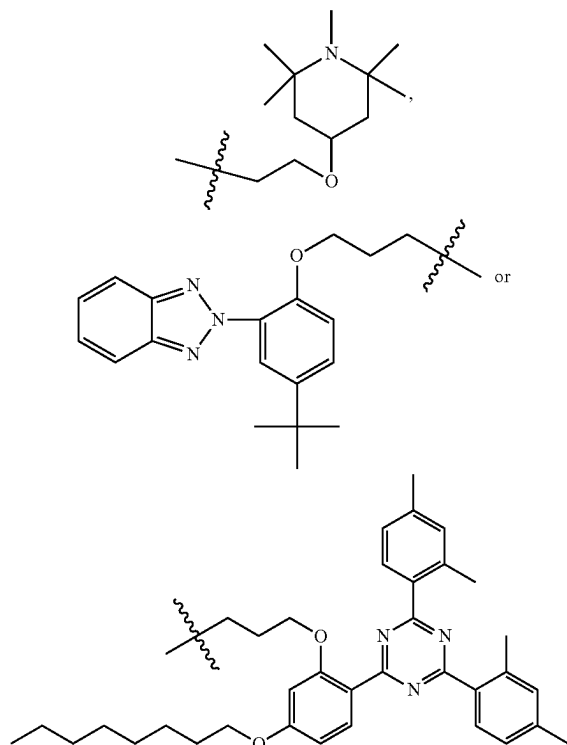
[0566] In this embodiment, X^A may be each independently a tri- to decavalent organic group.

[0567] In yet another embodiment, examples of X^A include the following groups:

[Formula 30]

$$—R^{25}—N—R^{26}—$$
$$\underset{\displaystyle R^{27}—}{|}$$
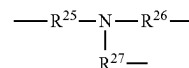
wherein R^{25}, R^{26}, and R^{27} are each independently a di- to hexavalent organic group; and

[0568] R^{25} is bonded to at least one R^{F1}, and R^{26} and R^{27} are each bonded to at least one R^{Si}.

[0569] In one embodiment, R^{25} is a single bond, a C_{1-20} alkylene group, a C_{3-20} cycloalkylene group, a C_{5-20} arylene group, —R^{57}—X^{58}—R^{59}—, —X^{58}—R^{59}—, or —R^{57}—X^{58}—. R^{57} and R^{59} are each independently a single bond, a C_{1-20} alkylene group, a C_{3-20} cycloalkylene group, or a C_{5-20} arylene group. X^{58} is —O—, —S—, —CO—, —O—CO—, or —COO—.

[0570] In one embodiment, R^{26} and R^{27} are each independently a hydrocarbon, or a group having at least one atom selected from N, O, and S at a terminal or in the main chain of a hydrocarbon, and preferable examples include a C_{1-6} alkyl group, —R^{36}—R^{37}—R^{36}, and —R^{36}—CHR^{38}$_2$—. Here, R^{36} is each independently a single bond or an alkyl group having 1 to 6 carbon atoms, and preferably an alkyl group having 1 to 6 carbon atoms. R^{37} is N, O, or S, and preferably N or O. R^{38} is —R^{45}—R^{46}—R^{45}—, —R^{46}—

R^{45}—, or —R^{45}—R^{46}—. Here, R^{45} is each independently an alkyl group having 1 to 6 carbon atoms. R^{46} is N, O, or S, and preferably O.

[0571] In this embodiment, X^A may be each independently a tri- to decavalent organic group.

[0572] In yet another embodiment, X^A is a group represented by:

[Formula 31]
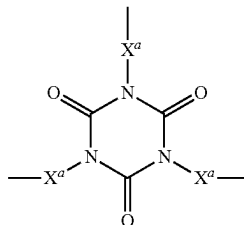
wherein X^a is a single bond or a divalent organic group.

[0573] X^a is a single bond or divalent linking group directly bonded to the isocyanuric ring. X^a is preferably a single bond, an alkylene group, or a divalent group containing at least one bond selected from the group consisting of an ether bond, an ester bond, an amide bond, and a sulfide bond, and more preferably a single bond, an alkylene group having 1 to 10 carbon atoms, or a divalent hydrocarbon group having 1 to 10 carbon atoms and containing at least one bond selected from the group consisting of an ether bond, an ester bond, an amide bond, and a sulfide bond.

[0574] X^a is even more preferably a group represented by the following formula:

$$—(CX^{121}X^{122})_{x1}—(X^{a1})_{y1}—(CX^{123}X^{124})_{z1}—$$
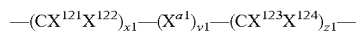
wherein X^{121} to X^{124} are each independently H, F, OH, or —$OSi(OR^{121})_3$ (wherein three R^{121} are each independently an alkyl group having 1 to 4 carbon atoms),

[0575] X^{a1} is —C(=O) NH—, —NHC(=O)—, —O—, —C(=O)O—, —OC(=O)—, —OC(=O) O—, or —NHC(=O)NH— (each bond is bonded to $CX^{121}X^{122}$ on the left side), and

[0576] x1 is an integer of 0 to 10, y1 is 0 or 1, and z1 is an integer of 1 to 10.

[0577] X^{a1} is preferably —O— or —C(=O)O—.

[0578] X^a is particularly preferably a group represented by the following formula:

$$—(CF_2)_{m11}—(CH_2)_{m12}—O—(CH_2)_{m13}—$$
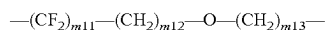
wherein m11 is an integer of 1 to 3, m12 is an integer of 1 to 3, and m13 is an integer of 1 to 3; a group represented by the following formula:

$$—(CF_2)_{m14}—(CH_2)_{m15}—O—CH_2CH(OH)—(CH_2)_{m16}—$$
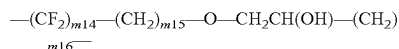
wherein m14 is an integer of 1 to 3, m15 is an integer of 1 to 3, and m16 is an integer of 1 to 3; a group represented by the following formula:

$$—(CF_2)_{m17}—(CH_2)_{m18}—$$
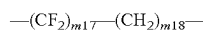
wherein m17 is an integer of 1 to 3, and m18 is an integer of 1 to 3; a group represented by the following formula:

$$—(CF_2)_{m19}—(CH_2)_{m20}—O—CH_2CH(OSi(OCH_3)_3)—(CH_2)_{m21}—$$
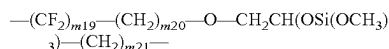
wherein m19 is an integer of 1 to 3, m20 is an integer of 1 to 3, and m21 is an integer of 1 to 3; or a group represented by the following formula:

$$—(CH_2)_{m22}—$$
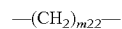
wherein m22 is an integer of 1 to 3.

[0579] X^a is not limited, and specific examples include —CH_2—, —C_2H_4—, —C_3H_6—, —C_4H_8—, —C_4H_8—O—CH_2—, —CO—O—CH_2—CH(OH)—CH_2—, —$(CF_2)_{n5}$— (n5 is an integer of 0 to 4), —$(CF_2)_{n5}$—$(CH_2)_{m5}$— (n5 and m5 are each independently an integer of 0 to 4), —$CF_2CF_2CH_2OCH_2CH(OH)CH_2$—, and —$CF_2CF_2CH_2OCH_2CH(OSi(OCH_3)_3)CH_2$—.

[0580] In this embodiment, X^A may be each independently a di- or trivalent organic group.

[0581] In one embodiment, the fluorine-containing silane compound represented by the formula (A1) or the formula (A2) does not contain a siloxane bond (—Si—O—Si—).

[0582] The number average molecular weight of the fluorine-containing silane compound represented by the above formula (A1) or (A2) is not limited, and may be $5×10^2$ to $1×10^5$. In particular, the average molecular weight is preferably 2,000 to 32,000 and more preferably 2,500 to 12,000 from the viewpoint of friction durability. The "average molecular weight" refers to a number average molecular weight, and the "average molecular weight" is a value obtained by ^{19}F-NMR measurement.

[0583] The surface-treating layer can be formed by treating the substrate surface with the surface-treating agent containing a fluorine-containing silane compound. For example, the surface-treating layer can be obtained by applying the surface-treating agent to the substrate to form a film of the surface-treating agent, and leaving the film to stand still or subjecting the film to a heat treatment to cure the film of the surface-treating agent.

[0584] In one embodiment, the fluorine-containing silane compound in the surface-treating agent is a compound represented by the formula (A1).

[0585] In another embodiment, the fluorine-containing silane compound in the surface-treating agent is a compound represented by the formula (A2).

[0586] In another embodiment, the fluorine-containing silane compound in the surface-treating agent is a compound represented by the formula (A1) and a compound represented by the formula (A2).

[0587] In the surface-treating agent, the compound represented by the formula (A2) is preferably 0.1 mol % or more and 35 mol % or less based on the total of the compound represented by the formula (A1) and the compound represented by the formula (A2). The lower limit of the content of the compound represented by the formula (A2) may be preferably 0.1 mol %, more preferably 0.2 mol %, even more preferably 0.5 mol %, yet more preferably 1 mol %, particularly preferably 2 mol %, and especially 5 mol %, based on the total of the compound represented by the formula (A1) and the compound represented by the formula (A2). The upper limit of the content of the compound represented by the formula (A2) may be preferably 35 mol %, more preferably 30 mol %, even more preferably 20 mol %, and yet more preferably 15 mol % or 10 mol %, based on the total of the compound represented by the formula (A1) and the compound represented by the formula (A2). The compound represented by the formula (A2) is preferably 0.1 mol % or more and 30 mol % or less, more preferably 0.1 mol

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% or more and 20 mol % or less, even more preferably 0.2 mol % or more and 10 mol % or less, yet more preferably 0.5 mol % or more and 10 mol % or less, and particularly preferably 1 mol % or more and 10 mol % or less, such as 2 mol % or more and 10 mol % or less or 5 mol % or more and 10 mol % or less, based on the total of the compound represented by the formula (A1) and the compound represented by the formula (A2). With the compound represented by the formula (A2) being within such a range, friction durability can be more increased.

[0588]   The component (A) may be a mixture of the compound represented by the formula (A1) or the formula (A2) with a compound obtained by condensing a portion of these compounds.

[0589]   The surface-treating agent may contain a solvent, an alcohol, an (unreactive) fluoropolyether compound that can be understood as a fluorine-containing oil or preferably a perfluoropolyether compound (hereinafter, collectively referred to as a "fluorine-containing oil"), an (unreactive) silicone compound that can be understood as a silicone oil (hereinafter, referred to as a "silicone oil"), a compatibilizer, a catalyst, a surfactant, a polymerization inhibitor, a sensitizer, and the like.

[0590]   Examples of the solvent include aliphatic hydrocarbons such as hexane, cyclohexane, heptane, octane, nonane, decane, undecane, dodecane, and mineral spirits; aromatic hydrocarbons such as benzene, toluene, xylene, naphthalene, and solvent naphtha; esters such as methyl acetate, ethyl acetate, propyl acetate, n-butyl acetate, iso-propyl acetate, isobutyl acetate, cellosolve acetate, propylene glycol methyl ether acetate, carbitol acetate, diethyl oxalate, ethyl pyruvate, ethyl 2-hydroxybutyrate, ethyl acetoacetate, amyl acetate, methyl lactate, ethyl lactate, methyl 3-methoxypropionate, ethyl 3-methoxypropionate, methyl 2-hydroxyisobutyrate, and ethyl 2-hydroxyisobutyrate; ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone, 2-hexanone, cyclohexanone, methyl amino ketone, and 2-heptanone; glycol ethers such as ethyl cellosolve, methyl cellosolve, methyl cellosolve acetate, ethyl cellosolve acetate, propylene glycol monomethyl ether, propylene glycol monoethyl ether, propylene glycol monobutyl ether, propylene glycol monomethyl ether acetate, propylene glycol monoethyl ether acetate, propylene glycol monobutyl ether acetate, dipropylene glycol dimethyl ether, and ethylene glycol monoalkyl ether; alcohols such as methanol, ethanol, iso-propanol, n-butanol, isobutanol, tert-butanol, sec-butanol, 3-pentanol, octyl alcohol, 3-methyl-3-methoxybutanol, and tert-amyl alcohol; glycols such as ethylene glycol and propylene glycol; cyclic ethers such as tetrahydrofuran, tetrahydropyran, and dioxane; amides such as N, N-dimethylformamide and N, N-dimethylacetamide; ether alcohols such as methyl cellosolve, cellosolve, isopropyl cellosolve, butyl cellosolve, and diethylene glycol monomethyl ether; diethylene glycol monoethyl ether acetate; and fluorine-containing solvents such as 1,1,2-trichloro-1,2,2-trifluoroethane, 1,2-dichloro-1,1,2,2-tetrafluoroethane, dimethyl sulfoxide, 1,1-dichloro-1,2,2,3,3-pentafluoropropane (HCFC 225), 1,1,2,2,3,3,4-heptafluorocyclopentane (e.g., Zeorora H), perfluorobutyl methyl ether ($C_4F_9OCH_3$) (e.g., HFE 7100), perfluorobutyl ethyl ether ($C_4F_9OC_2H_5$) (e.g., HFE 7200), perfluorohexyl methyl ether ($C_2F_5CF(OCH_3)C_3F_7$) (e.g., HFE 7300),

CF_3CH_2OH, $CF_3CF_2CH_2OH$, and $(CF_3)_2CHOH$. Alternatively, the solvent may be a mixed solvent of two or more of such solvents.

[0591]   Examples of the alcohol include alcohols having 1 to 6 carbon atoms and optionally substituted with one or more fluorine atoms, such as methanol, ethanol, iso-propanol, tert-butanol, CF_3CH_2OH, $CF_3CF_2CH_2OH$, $(CF_3)_2CHOH$, $H(CF_2CF_2)CH_2OH$, $H(CF_2CF_2)_2CH_2OH$, and $H(CF_2CF_2)_3CH_2OH$. Such an alcohol added to the surface-treating agent increases the stability of the surface-treating agent and improves the miscibility between the component (A) and the component (B).

[0592]   In a preferable embodiment, the surface-treating agent contains a solvent and an alcohol.

[0593]   The alcohol content is preferably 0.1 to 50% by mass, more preferably 0.5 to 30% by mass, even more preferably 1.0 to 20% by mass, and yet more preferably 2.0 to 10% by mass, based on the total of the solvent and the alcohol.

[0594]   The fluorine-containing oil is not limited, and examples include compounds (perfluoropolyether compounds) represented by the following general formula (3):

$$Rf^5—(OC_4F_8)_{a'}—(OC_3F_6)_{b'}—(OC_2F_4)_{c'}—(OCF_2)_{d'}—Rf^6 \qquad (3)$$
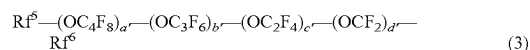
[0595]   wherein Rf^5 represents a C_{1-16} alkyl group optionally substituted with one or more fluorine atoms (preferably, a C_{1-16} perfluoroalkyl group), Rf^6 represents a C_{1-16} alkyl group optionally substituted with one or more fluorine atoms (preferably, a C_{1-16} perfluoroalkyl group), a fluorine atom, or a hydrogen atom, and Rf^5 and Rf^6 are more preferably each independently a C_{1-3} perfluoroalkyl group;

[0596]   a', b', c' and d' respectively represent the numbers of repeating units of four perfluoropolyethers constituting the main backbone of the polymer and are mutually independently an integer of 0 or more and 300 or less, and the sum of a', b', c', and d' is at least 1, preferably 1 to 300, and more preferably 20 to 300; and the occurrence order of the respective repeating units in parentheses provided with the subscript a', b', c', or d' is not limited in the formula. Among these repeating units, $—(OC_4F_8)—$ may be any of $—(OCF_2CF_2CF_2CF_2)—$, $—(OCF(CF_3)CF_2CF_2)—$, $—(OCF_2CF(CF_3)CF_2)—$, $—(OCF_2CF_2CF(CF_3))—$, $—(OC(CF_3)_2CF_2)—$, $—(OCF_2C(CF_3)_2)—$, $—(OCF(CF_3)CF(CF_3))—$, $—(OCF(C_2F_5)CF_2)—$, and $(OCF_2CF(C_2F_5))—$, and is preferably $—(OCF_2CF_2CF_2CF_2)—$. $—(OC_3F_6)—$ may be any of $—(OCF_2CF_2CF_2)—$, $—(OCF(CF_3)CF_2)—$, and $(OCF_2CF(CF_3))—$, and is preferably $—(OCF_2CF_2CF_2)—$. $—(OC_2F_4)—$ may be any of $—(OCF_2CF_2)—$ and $(OCF(CF_3))—$, and is preferably $—(OCF_2CF_2)—$.

[0597]   Examples of the perfluoropolyether compound represented by the above general formula (3) include a compound represented by any of the following general formulae (3a) and (3b) (which may be used singly or as a mixture of two or more).

$$Rf^5—(OCF_2CF_2CF_2)_{b''}—Rf^6 \qquad (3a)$$

$$Rf^5—(OCF_2CF_2CF_2CF_2)_{a''}—(OCF_2CF_2CF_2)_{b''}—(OCF_2CF_2)_{c''}—(OCF_2)_{d''}—Rf^6 \qquad (3b)$$
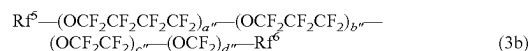
[0598] wherein Rf^5 and Rf^6 are as described above; in the formula (3a), b" is an integer of 1 or more and 100 or less; and in the formula (3b), a" and b" are each independently an integer of 0 or more and 30 or less, and c" and d" are each independently an integer of 1 or more and 300 or less. The occurrence order of the respective repeating units enclosed in parentheses provided with the subscript a", b", c", or d" is not limited in the formulae.

[0599] From another viewpoint, the fluorine-containing oil may be a compound represented by the general formula Rf^3—F, wherein Rf^3 is a C_{5-16} perfluoroalkyl group. The fluorine-containing oil may be a chlorotrifluoroethylene oligomer.

[0600] The fluorine-containing oil may have an average molecular weight of 500 to 10,000. The molecular weight of the fluorine-containing oil may be measured using GPC.

[0601] The fluorine-containing oil may be contained in an amount of, for example, 0 to 50 mass %, preferably 0 to 30 mass %, and more preferably 0 to 5 mass %, based on the surface-treating agent. In one embodiment, the surface-treating agent is substantially free of the fluorine-containing oil. Being substantially free of the fluorine-containing oil means that the fluorine-containing oil is not contained at all, or an extremely small amount of the fluorine-containing oil may be contained.

[0602] In one embodiment, the average molecular weight of the fluorine-containing oil may be greater than the average molecular weight of the fluorine-containing silane compound. With such average molecular weights, better friction durability and surface lubricity can be obtained particularly when a surface-treating layer is formed by a vacuum deposition method.

[0603] In one embodiment, the average molecular weight of the fluorine-containing oil may be smaller than the average molecular weight of the fluorine-containing silane compound. With such average molecular weights, a cured product having high friction durability and high surface lubricity can be formed while suppressing deterioration of the transparency of the surface-treating layer obtained from the compound.

[0604] The fluorine-containing oil contributes to increasing surface lubricity of the layer formed by the surface-treating agent.

[0605] For example, a linear or cyclic silicone oil having 2,000 or fewer siloxane bonds can be used as the silicone oil. The linear silicone oil may be a so-called straight silicone oil or modified silicone oil. Examples of the straight silicone oil include dimethyl silicone oil, methyl phenyl silicone oil, and methyl hydrogen silicone oil. Examples of the modified silicone oil include those obtained by modifying a straight silicone oil with alkyl, aralkyl, polyether, higher fatty acid ester, fluoroalkyl, amino, epoxy, carboxyl, alcohol, or the like. Examples of the cyclic silicone oil include cyclic dimethylsiloxane oil.

[0606] Such a surface-treating agent can include, for example, 0 to 300 parts by mass and preferably 50 to 200 parts by mass of such silicone oil based on a total of 100 parts by mass of the fluorine-containing silane compound of the present disclosure (in the case of two or more, the total thereof, and the same also applies below).

[0607] The silicone oil contributes to increasing the surface lubricity of the surface-treating layer.

[0608] Examples of the compatibilizing agent include a fluorine-substituted alcohol such as 2,2,2-trifluoroethanol, 2,2,3,3,3-pentafluoro-1-propanol, or 2,2,3,3,4,4,5,5-octafluoro-1-pentanol, and preferably a fluorine-substituted alcohol having a CF_2H terminal, as well as s a fluorine-substituted aryl such as 1,3-bis(trifluoromethyl)benzene and preferably a fluorine-substituted benzene.

[0609] Examples of the catalyst include acids (such as acetic acid and trifluoroacetic acid), bases (such as ammonia, triethylamine, and diethylamine), and transition metals (such as Ti, Ni, and Sn).

[0610] The catalyst promotes hydrolysis and dehydration condensation of the fluorine-containing silane compound of the present disclosure, and promotes formation of the layer to be formed by the surface-treating agent.

[0611] Examples of other components include, in addition to those described above, tetraethoxysilane, methyltrimethoxysilane, 3-aminopropyltrimethoxysilane, 3-glycidoxypropyltrimethoxysilane, and methyltriacetoxysilane.

[0612] The surface-treating agent can be formed into pellets by impregnating a porous material such as a porous ceramic material or a metal fiber such as a fiber obtained by, for example, solidifying steel wool in a cotton-like form therewith. Such pellets can be used in, for example, vacuum deposition.

[0613] The surface-treating agent may contain, in addition to the components described above, trace amounts of Pt, Rh, Ru, 1,3-divinyltetramethyldisiloxane, triphenylphosphine, NaCl, KCl, silane condensation product, and the like as impurities.

[0614] The layer of the surface-treating agent can be formed by applying the surface-treating agent to the underlayer. The coating method is not limited. For example, a wet coating method and a dry coating method can be used.

[0615] Examples of the wet coating method include dip coating, spin coating, flow coating, spray coating, roll coating, gravure coating, and similar methods.

[0616] Examples of the dry coating method include deposition (usually, vacuum deposition), sputtering, CVD, and similar methods. Specific examples of the deposition method (usually, a vacuum deposition method) include resistive heating, high-frequency heating using electron beam, microwave, or the like, ion beam, and similar methods. Specific examples of the CVD method include plasma-CVD, optical CVD, thermal CVD, and similar methods.

[0617] Furthermore, coating by an atmospheric pressure plasma method can be performed.

[0618] When using the wet coating method, the surface-treating agent can be applied to the substrate surface after being diluted with a solvent. From the viewpoint of the stability of the composition of the present disclosure and the volatility of solvents, the following solvents are preferably used: perfluoroaliphatic hydrocarbons having 5 to 12 carbon atoms (such as perfluorohexane, perfluoromethylcyclohexane, and perfluoro-1,3-dimethylcyclohexane); polyfluoroaromatic hydrocarbons (such as bis(trifluoromethyl)benzene); polyfluoroaliphatic hydrocarbons (such as $C_6F_{13}CH_2CH_3$ (such as Asahiklin (registered trademark) AC-6000 manufactured by Asahi Glass Co., Ltd.), and 1,1,2,2,3,3,4-heptafluorocyclopentane (such as Zeorora (registered trademark) H manufactured by Zeon Corporation)); alkyl perfluoroalkyl ethers (the perfluoroalkyl group and the alkyl group may be linear or branched) such as hydrofluoroether (HFE) (such as perfluoropropyl methyl

ether ($C_3F_7OCH_3$) (such as Novec (trademark) 7000 manufactured by Sumitomo 3M Limited), perfluorobutyl methyl ether ($C_4F_9OCH_3$) (such as Novec (trademark) 7100 manufactured by Sumitomo 3M Limited), perfluorobutyl ethyl ether ($C_4F_9OC_2H_5$) (such as Novec (trademark) 7200 manufactured by Sumitomo 3M Limited), and perfluorohexyl methyl ether ($C_2F_5CF(OCH_3)C_3F_7$) (such as Novec (trademark) 7300 manufactured by Sumitomo 3M Limited), or $CF_3CH_2OCF_2CHF_2$ (such as Asahiklin (registered trademark) AE-3000 manufactured by Asahi Glass Co., Ltd.)). One of these solvents may be used singly, or two or more may be used as a mixture. In particular, hydrofluoroether is preferable, and perfluorobutyl methyl ether ($C_4F_9OCH_3$) and/or perfluorobutyl ethyl ether ($C_4F_9OCH_5$) is particularly preferable.

[0619] When using the dry coating method, the surface-treating agent may be directly subjected to the dry coating method, or may be diluted with the above solvent before being subjected to the dry coating method.

[0620] A layer of the surface-treating agent is preferably formed such that the surface-treating agent coexists in the layer with a catalyst for hydrolysis and dehydrative condensation. Conveniently, in the case of a wet coating method, the surface-treating agent is diluted with a solvent, and then, immediately before application to the substrate surface, a catalyst may be added to the diluted solution of the surface-treating agent. In the case of a dry coating method, the surface-treating agent to which a catalyst has been added is directly used in a deposition (usually vacuum deposition) treatment, or a pellet-like material may be used in a deposition (usually vacuum deposition) treatment, wherein the pellets are obtained by impregnating a porous body of metal such as iron or copper with the surface-treating agent to which the catalyst has been added.

[0621] The catalyst may be any suitable acid or base. Acid catalyst may be, for example, acetic acid, formic acid, or trifluoroacetic acid. The base catalyst may be, for example, ammonia or an organic amine.

[0622] The thickness of the surface-treating layer is not limited. Preferably, the thickness of the surface-treating layer in the case of an optical member is in the range of 1 to 200 nm, preferably 1 to 100 nm, and more preferably 1 to 50 nm, such as 1 to 30 nm, from the viewpoint of optical performance, friction durability, and antifouling properties.

(Article)

[0623] The present disclosure provides an article comprising the laminate of the present disclosure.

[0624] In a preferable embodiment, the article of the present disclosure may be glass or mirror glass for vehicles, marine vessels, and aircrafts, such as automobile glass, automobile door mirror or automobile fender mirror, or glass for use in on-board cameras. In addition, the article may be glass used in security cameras and outdoor glass such as roadside mirror.

[0625] Moreover, the article of the present disclosure may be an optical member. Examples of the optical member include lenses of glasses or the like; displays such as PDPs and LCDs; touch panel sheets for devices such as cell phones and portable information terminals; optical discs such as Blu-ray® discs, DVD discs, CD-Rs, and MOs; optical fibers; and display surfaces of watches and clocks.

[0626] The article of the present disclosure may be medical equipment or a medical material.

[0627] So far, the laminate and the article of the present disclosure have been described in detail. The laminate, the article, and the like of the present disclosure are not limited to those exemplified above.

[0628] The present disclosure includes the following embodiments.

[1] A laminate comprising a substrate, an underlayer disposed on the substrate, and a surface-treating layer disposed on the underlayer, wherein

[0629] the underlayer is a layer formed of:

(A) polysilazane represented by the following formula (P):

[Formula 1]

$$\left[\begin{array}{c} R^{61} \\ | \\ Si-N \\ | \quad | \\ R^{62} \quad R^{63} \end{array} \right]_n$$
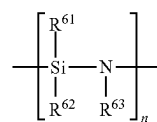
wherein

[0630] R^{61} is a hydrogen atom or a C_{1-6} alkyl group,

[0631] R^{62} is a hydrogen atom, a C_{1-6} alkyl group, or a group having a functional group,

[0632] R^{63} is a hydrogen atom, a C_{1-6} alkyl group, or a group having a functional group, and

[0633] n is an integer of 2 to 100; or

(B) aminosilane represented by the following formula (Q):

$$N(R^{71}-SiR^{72}{}_3)_m R^{73}{}_{3-m} \qquad \text{[Formula 2]}$$

wherein

[0634] R^{71} is each independently a C_{1-6} alkylene group,

[0635] R^{72} is each independently a hydrolyzable group,

[0636] R^{73} is a hydrogen atom or a C_{1-6} alkyl group, and

[0637] m is 2 or 3; and

[0638] the surface-treating layer is a layer formed of a fluorine-containing silane compound.

[2] The laminate according to [1], wherein the fluorine-containing silane compound is a compound represented by the following formula (A1) or (A2):

[Formula 3]

$$R^{F1}{}_{\alpha 1}\text{-}X^4\text{---}R^{Si}{}_{\beta 1} \qquad (A1)$$

$$R^{Si}{}_{\gamma 1}\text{---}X^4\text{---}R^{F2}\text{---}X^4\text{---}R^{Si}{}_{\gamma 1} \qquad (A2)$$
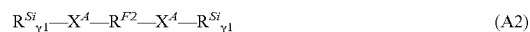
wherein

[0639] R^{F1} is each independently Rf^1— or Rf^1—R^F—O_q—,

[0640] R^{F2} is —R^{f2}— or —$Rf^2{}_p$—R^F—O_q—,

[0641] R^F is each independently a divalent fluoropolyether group,

[0642] Rf^1 is each independently a C_{1-16} alkyl group optionally substituted with one or more fluorine atoms,

[0643] Rf^2 is a C_{1-6} alkylene group optionally substituted with one or more fluorine atoms,

[0644] p is 0 or 1,

[0645] q is each independently 0 or 1,

[0646] R^{Si} is each independently a monovalent group containing a Si atom to which a hydroxyl group, a hydrolyzable group, a hydrogen atom, or a monovalent organic group is bonded,

[0647] at least one R^{Si} is a monovalent group containing a Si atom to which a hydroxyl group or a hydrolyzable group is bonded,

[0648] X^A is each independently a single bond or a di- to decavalent organic group,

[0649] $\alpha 1$ is an integer of 1 to 9,

[0650] $\beta 1$ is an integer of 1 to 9, and

[0651] $\gamma 1$ is each independently an integer of 1 to 9.

[3] The laminate according to [2], wherein

[0652] Rf^1 is each independently a C_{1-16} perfluoroalkyl group, and

[0653] Rf^2 is each independently a C_{1-6} perfluoroalkylene group.

[4] The laminate according to [2] or [3], wherein

[0654] R^{F1} is each independently $Rf^1-R^F-O_q-$, and

[0655] R^{F2} is $-Rf^2_p-R^F-O_q-$.

[5] The laminate according to any one of [2] to [4], wherein

[0656] R^F is each independently a group represented by the following formula:

$$-(OC_6F_{12})_a-(OC_5F_{10})_b-(OC_4F_8)_c-(OC_3R^{Fa}_6)_d-(OC_2F_4)_e-(OCF_2)_f-$$

wherein

[0657] R^{Fa} is each independently a hydrogen atom, a fluorine atom, or a chlorine atom; and

[0658] a, b, c, d, e, and f are each independently an integer of 0 to 200, the sum of a, b, c, d, e, and f is 1 or more, and the occurrence order of the respective repeating units enclosed in parentheses provided with a, b, c, d, e, or f is not limited in the formula, provided that when all R^{Fa} groups are hydrogen atoms or chlorine atoms, at least one of a, b, c, e, and f is 1 or more.

[6] The laminate according to [5], wherein R^{Fa} is a fluorine atom.

[7] The laminate according to any one of [2] to [6], wherein

[0659] R^F is each independently a group represented by the following formula (f1), (f2), (f3), (f4), (f5), or (f6):

$$(OC_3F_6)_d-(OC_2F_4)_e-\qquad\text{(f1)}$$

wherein d is an integer of 1 to 200, and e is 0 or 1;

$$-(OC_4F_8)_c-(OC_3F_6)_d-(OC_2F_4)_e-(OCF_2)_f-\qquad\text{(f2)}$$

wherein c and d are each independently an integer of 0 to 30,

[0660] e and f are each independently an integer of 1 to 200,

[0661] the sum of c, d, e, and f is an integer of 10 to 200, and

[0662] the occurrence order of the respective repeating units enclosed in parentheses provided with the subscript c, d, e, or f is not limited in the formula;

$$-(R^6-R^7)_g-\qquad\text{(f3)}$$

wherein R^6 is OCF_2 or OC_2F_4,

[0663] R^7 is a group selected from OC_2F_4, OC_3F_6, OC_4F_8, OC_5F_{10}, and OC_6F_{12}, or is a combination of two or three groups selected from these groups, and

[0664] g is an integer of 2 to 100;

$$-(R^6-R^7)_g-R^r-(R^7-R^6)_{g'}-\qquad\text{(f4)}$$

wherein R^6 is OCF_2 or OC_2F_4,

[0665] R^7 is a group selected from OC_2F_4, OC_3F_6, OC_4F_8, OC_5F_{10}, and $OC\,F_{12}$, or is a combination of two or three groups independently selected from these groups,

[0666] $R^{6'}$ is OCF_2 or OC_2F_4,

[0667] $R^{7'}$ is a group selected from OC_2F_4, OC_3F_6, OC_4F_8, OC_5F_{10}, and $OC\,F_{12}$, or is a combination of two or three groups independently selected from these groups,

[0668] g is an integer of 2 to 100,

[0669] g' is an integer of 2 to 100, and

[0670] R^r is any of:

[Formula 4]

wherein * represents a bond position;

$$-(OC_6F_{12})_a-(OC_5F_{10})_b-(OC_4F_8)_c-(OC_3F_6)_d-(OC_2F_4)_e-(OCF_2)_f-\qquad\text{(f5)}$$

wherein e is an integer of 1 or more and 200 or less, a, b, c, d, and f are each independently an integer of 0 or more and 200 or less, and the occurrence order of the respective repeating units enclosed in parentheses provided with a, b, c, d, e or f is not limited in the formula; and

$$(OC_6F_{12})_a-(OC_5F_{10})_b-(OC_4F_8)_c-(OC_3F_6)_d-(OC_2F_4)_e-(OCF_2)_f-\qquad\text{(f6)}$$

wherein f is an integer of 1 or more and 200 or less, a, b, c, d, and e are each independently an integer of 0 or more and 200 or less, and the occurrence order of the respective repeating units enclosed in parentheses provided with a, b, c, d, e or f is not limited in the formula.

[8] The laminate according to any one of [2] to [7], wherein

[0671] R^{Si} is a group represented by the following formula (S1), (S2), (S3), (S4), or (S5):

[Formula 5]

$$-(CH_2C\overset{\textstyle R^{13}}{\underset{\textstyle X^{11}-SiR^{11}_{n1}R^{12}_{3-n1}}{|}}-R^{15})_t-R^{14}\qquad\text{(S1)}$$

$$-SiR^{11}_{n1}R^{12}_{3-n1}\qquad\text{(S2)}$$

$$-SiR^{a1}_{k1}R^{b1}_{l1}R^{c1}_{m1}\qquad\text{(S3)}$$

$$-CR^{d1}_{k2}R^{e1}_{l2}R^{f1}_{m2} \quad (S4)$$

$$-NR^{g1}R^{h1} \quad (S5)$$

wherein

[0672] R^{11} is each independently a hydroxyl group or a hydrolyzable group,

[0673] R^{12} is each independently a hydrogen atom or a monovalent organic group,

[0674] n1 is each independently an integer of 0 to 3 for each $(SiR^{11}_{n1}R^{12}_{3-n1})$ unit,

[0675] X^{11} is each independently a single bond or a divalent organic group,

[0676] R^{13} is each independently a hydrogen atom or a monovalent organic group,

[0677] t is each independently an integer of 2 or more,

[0678] R^{14} is each independently a hydrogen atom, a halogen atom, or $-X^{11}-SiR^{11}_{n1}R^{12}_{3-n1}$,

[0679] R^{15} is each independently at each occurrence a single bond, an oxygen atom, an alkylene group having 1 to 6 carbon atoms, or an alkyleneoxy group having 1 to 6 carbon atoms,

[0680] R^{a1} is each independently $-Z^{1}-SiR^{21}_{p1}R^{22}_{q1}R^{23}_{r1}$,

[0681] Z^{1} is each independently an oxygen atom or a divalent organic group,

[0682] R^{21} is each independently $-Z^{1'}-SiR^{21'}_{p1}R^{22'}_{q1}R^{23'}_{r1'}$,

[0683] R^{22} is each independently a hydroxyl group or a hydrolyzable group,

[0684] R^{23} is each independently a hydrogen atom or a monovalent organic group,

[0685] p1 is each independently an integer of 0 to 3,

[0686] q1 is each independently an integer of 0 to 3,

[0687] r1 is each independently an integer of 0 to 3,

[0688] the sum of p1, q1, and r1 is 3 in the $(SiR^{21}_{p1}R^{22}_{q1}R^{23}_{r1})$ unit,

[0689] $Z^{1'}$ is each independently an oxygen atom or a divalent organic group,

[0690] $R^{21'}$ is each independently $-Z^{1''}-SiR^{22''}_{q1''}R^{23''}_{r1''}$,

[0691] $R^{22'}$ is each independently a hydroxyl group or a hydrolyzable group,

[0692] $R^{23'}$ is each independently a hydrogen atom or a monovalent organic group,

[0693] p1' is each independently an integer of 0 to 3,

[0694] q1' is each independently an integer of 0 to 3,

[0695] r1' is each independently an integer of 0 to 3,

[0696] the sum of p1', q1', and r1' is 3 in the $(SiR^{21'}_{p1}R^{22'}_{q1}R^{23'}_{r1'})$ unit,

[0697] $Z^{1''}$ is each independently an oxygen atom or a divalent organic group,

[0698] $R^{22''}$ is each independently a hydroxyl group or a hydrolyzable group,

[0699] $R^{23''}$ is each independently a hydrogen atom or a monovalent organic group,

[0700] q1" is each independently an integer of 0 to 3,

[0701] r1" is each independently an integer of 0 to 3,

[0702] the sum of q1" and r1" is 3 in the $(SiR^{22''}_{q1''}R^{23''}_{r1''})$ unit,

[0703] R^{b1} is each independently a hydroxyl group or a hydrolyzable group,

[0704] R^{c1} is each independently a hydrogen atom or a monovalent organic group,

[0705] k1 is each independently an integer of 0 to 3,

[0706] l1 is each independently an integer of 0 to 3,

[0707] m1 is each independently an integer of 0 to 3,

[0708] the sum of k1, l1, and m1 is 3 in the $(SiR^{a1}_{k1}R^{b1}_{l1}R^{c1}_{m1})$ unit,

[0709] R^{d1} is each independently $-Z^{2}-CR^{31}_{p2}R^{32}_{q2}R^{33}_{r2}$,

[0710] Z^{2} is each independently a single bond, an oxygen atom, or a divalent organic group,

[0711] R^{31} is each independently $-Z^{2'}-CR^{32'}_{92}R^{33'}_{21}$,

[0712] R^{32} is each independently $-Z^{3}-SiR^{34}_{n2}R^{35}_{3-n2}$,

[0713] R^{33} is each independently a hydrogen atom, a hydroxyl group, or a monovalent organic group,

[0714] p2 is each independently an integer of 0 to 3,

[0715] q2 is each independently an integer of 0 to 3,

[0716] r2 is each independently an integer of 0 to 3,

[0717] the sum of p2, q2, and r2 is 3 in the $(CR^{31}_{p2}R^{32}_{q2}R^{33}_{r2})$ unit,

[0718] $Z^{2'}$ is each independently a single bond, an oxygen atom, or a divalent organic group,

[0719] $R^{32'}$ is each independently $-Z^{3}-SiR^{34}_{n2}R^{35}_{3-n2}$,

[0720] $R^{33'}$ is each independently a hydrogen atom, a hydroxyl group, or a monovalent organic group,

[0721] q2' is each independently an integer of 0 to 3,

[0722] r2' is each independently an integer of 0 to 3,

[0723] the sum of q2' and r2' is 3 in the $(CR^{32'}_{q2}R^{33'}_{r2'})$ unit,

[0724] Z^{3} is each independently a single bond, an oxygen atom, or a divalent organic group,

[0725] R^{34} is each independently a hydroxyl group or a hydrolyzable group,

[0726] R^{35} is each independently a hydrogen atom or a monovalent organic group,

[0727] n2 is each independently an integer of 0 to 3,

[0728] R^{e1} is each independently $-Z^{3}-SiR^{34}_{n2}R^{35}_{3-n2}$,

[0729] R^{f1} is each independently a hydrogen atom, a hydroxyl group, or a monovalent organic group,

[0730] k2 is each independently an integer of 0 to 3,

[0731] l2 is each independently an integer of 0 to 3,

[0732] m2 is each independently an integer of 0 to 3,

[0733] the sum of k2, l2, and m2 is 3 in the $(CR^{d1}_{k2}R^{e1}_{l2}R^{f1}_{m2})$ unit,

[0734] R^{g1} and R^{h1} are each independently $-Z^{4}-SiR^{11}_{n1}R^{12}_{3-n1}$, $-Z^{4}-SiR^{a1}_{k1}R^{b1}_{l1}R^{c1}_{m1}$, or $-Z^{4}-CR^{d1}_{k2}R^{e1}_{l2}R^{f1}_{m2}$, and

[0735] Z^{4} is each independently a single bond, an oxygen atom, or a divalent organic group,

[0736] provided that in the formulae (S1), (S2), (S3), (S4), and (S5), at least one Si atom to which a hydroxyl group or a hydrolyzable group is bonded is present.

[9] The laminate according to any one of [2] to [8], wherein α1, β1, and γ1 are each 1.

[10] The laminate according to any one of [2] to [9], wherein X^{4} is a single bond or a divalent organic group represented by the following formula:

$$-(R^{51})_{p5}-(X^{51})_{q5}-$$

wherein

[0737] R^{51} is a single bond or $-(CH_2)_{s5}-$,

[0738] s5 is an integer of 1 to 20,

[0739] X^{51} is $-(X^{52})_{l5}-$,

[0740] X^{52} each independently represents a group selected from the group consisting of $-O-$, $-S-$, $-CO-$, $-NR^{54}-$, $-(CH_2)_{n5}-$, $-COO-$, $-OCO-$, $-CONR^{54}-$, $-NR^{54}CO-$,

—O—CONR^{54}—, —NR^{54}CO—O—, —Si(R^{53})$_2$—, and —(Si(R^{53})$_2$O)$_{m5}$—Si(R^{53})$_2$—,

[0741] R^{53} is each independently a phenyl group, a C$_{1-6}$ alkyl group, or a C$_{1-6}$ alkoxy group,

[0742] R^{54} is each independently a hydrogen atom, a phenyl group, or a C$_{1-6}$ alkyl group,

[0743] m5 is each independently an integer of 1 to 100,

[0744] n5 is each independently an integer of 1 to 20,

[0745] 15 is an integer of 1 to 10,

[0746] p5 is 0 or 1, and

[0747] q5 is 0 or 1,

[0748] provided that at least one of p5 and q5 is 1, the occurrence order of the respective repeating units enclosed in parentheses provided with p5 or q5 is not limited, and

[0749] the right side is bonded to R^{Si}.

[11] The laminate according to any one of [2] to [8], wherein X^4 is a group represented by the following formula:
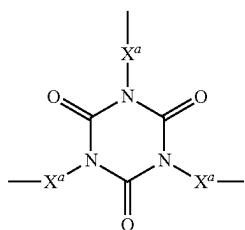
[Formula 6]

wherein X^a is a single bond or a divalent organic group.

[12] The laminate according to any one of [1] to [11], wherein the polysilazane is represented by formula (P) wherein R^{61} is a hydrogen atom, R^{62} is a hydrogen atom, and R^{63} is a hydrogen atom.

[13] The laminate according to any one of [1] to [12], wherein the polysilazane has a number average molecular weight of 1,000 to 10,000.

[14] The laminate according to any one of [1] to [13], wherein the aminosilane is represented by the following formula (Q-1):

HN(R^{71}—SiR$^{72}_3$)$_2$ [Formula 7]

wherein

[0750] R^{71} is each independently a C$_{1-6}$ alkylene group, and

[0751] R^{72} is each independently a hydrolyzable group.

[15] The laminate according to any one of [1] to [14], wherein the substrate is glass, metal, or ceramic.

[16] An article comprising the laminate according to any one of [1] to.

[17] The article according to [16], which is automobile glass, outdoor glass, or a display.

EXAMPLES

[0752] Hereinafter, the laminate of the present disclosure will now be described in the Examples, but the present disclosure is not limited to the following Examples. In the Examples, the occurrence order of units constituting repeating fluoropolyether is not limited, and the chemical formulae shown below indicate average compositions.

[0753] Chemical solutions (A) to (F) were prepared as compositions for forming an underlayer.

(Polysilazane-Containing Composition)

[0754] (A): Polysilazane (Durazane 2800, manufactured by Merck) was dissolved in dibutyl ether (manufactured by Tokyo Chemical Industry Co., Ltd.) so as to have a concentration of 0.2% by mass, and the mixture was regarded as a chemical solution (A).

[0755] (B): Polysilazane (Durazane 2800, manufactured by Merck) was dissolved in dibutyl ether (manufactured by Tokyo Chemical Industry Co., Ltd.) so as to have a concentration of 0.5% by mass, and the mixture was regarded as a chemical solution (B).

[0756] (C): Polysilazane (Durazane 2800, manufactured by Merck) was dissolved in dibutyl ether (manufactured by Tokyo Chemical Industry Co., Ltd.) so as to have a concentration of 5.0% by mass, and the mixture was regarded as a chemical solution (C).

(Aminosilane-Containing Composition)

[0757] (D): Bis-3-trimethoxysilylpropylamine (manufactured by Tokyo Chemical Industry Co., Ltd.) was dissolved in hydrofluoroether (Novec HFE 7200, manufactured by 3M) so as to have a concentration of 0.2% by mass, and the mixture was regarded as a chemical solution (D).

[0758] (E): Bis-3-trimethoxysilylpropylamine (manufactured by Tokyo Chemical Industry Co., Ltd.) was dissolved in hydrofluoroether (Novec HFE 7200, manufactured by 3M) so as to have a concentration of 0.5% by mass, and the mixture was regarded as a chemical solution (E).

[0759] (F): Bis-3-trimethoxysilylpropylamine (manufactured by Tokyo Chemical Industry Co., Ltd.) was dissolved in hydrofluoroether (Novec HFE 7200, manufactured by 3M) so as to have a concentration of 5.0% by mass, and the mixture was regarded as a chemical solution (F).

[0760] The following water-repelling solutions (1) to (6) were prepared as compositions for forming a surface-treating layer.

[0761] (1): The following compound (1) was dissolved in hydrofluoroether (Novec HFE 7200, manufactured by 3M) so as to have a concentration of 0.18 by mass, and the mixture was regarded as a water-repelling solution (1).

[0762] (2): The following compound (2) was dissolved in hydrofluoroether (Novec HFE 7200, manufactured by 3M) so as to have a concentration of 0.1% by mass, and the mixture was regarded as a water-repelling solution (2).

[0763] (3): The following compound (3) was dissolved in hydrofluoroether (Novec HFE 7200, manufactured by 3M) so as to have a concentration of 0.18 by mass, and the mixture was regarded as a water-repelling solution (3).

[0764] (4): The following compound (4) was dissolved in hydrofluoroether (Novec HFE 7200, manufactured by 3M) so as to have a concentration of 0.1% by mass, and the mixture was regarded as a water-repelling solution (4).

[0765] (5): The following compound (5) was dissolved in hydrofluoroether (Novec HFE 7200, manufactured

by 3M) so as to have a concentration of 0.1% by mass, and the mixture was regarded as a water-repelling solution (5).

[0766]   (6): The following (6) compound was dissolved in hydrofluoroether (Novec HFE 7200, manufactured by 3M) so as to have a concentration of 0.1% by mass, and the mixture was regarded as a water-repelling solution (6).
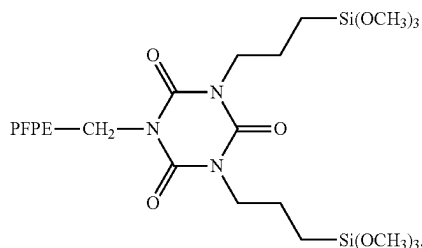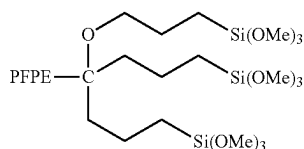
Compound (1):

[Formula 32]

wherein PFPE is $CF_3CF_2CF_2$—$(OCF_2CF_2CF_2)_{25}$—OCF_2CF_2—.

Compound (2):

[Formula 33]

wherein PFPE is CF_3—$(OCF_2CF_2)_{22}(OCF_2)_{20}$—.

Compound (3):

[Formula 34]
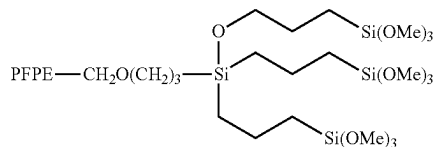
wherein PFPE is CF_3—$(OCF_2CF_2)_{22}(OCF_2)_{20}$—.

Compound (4):

[0767]

$$CF_3CF_2CF_2\text{—}(OCF_2CF_2CF_2)_{25}\text{—}OCF_2CF_2\text{—}$$
$$CH_2OCH_2CH_2CH_2Si(OCH_3)_3$$

Compound (5):

[0768]

$$C_6F_{13}\text{-ph-}Si(OMe)_3$$

Compound (6):

[0769]

$$C_6F_{13}\text{—}CH_2CH_2\text{—}Si(OMe)_3$$
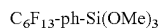
wherein Me is a methyl group, and ph is a phenylene group.

(Substrate)

[0770]   The top surface of a float glass (clear) substrate (planar dimension 70 mm×150 mm) having a thickness of 2.0 mm was washed with water and alcohol and then dried to give a glass substrate.

[0771]   Using an atmospheric pressure plasma discharge apparatus (Si-PlasmaR, manufactured by Creating Nano Tech., Inc.), an atmospheric pressure plasma treatment was performed on the top surface of the glass substrate under the following conditions for washing and activation.

<Gas Conditions>

[0772]   Discharge gas: Argon gas (Ar), Reactive gas: Oxygen gas (O_2)

<Plasma Irradiation Conditions>

[0773]   Plasma power: 744 W (output controlled in the range of 740 to 750 W)

[0774]   Ar flow: 34 L/min (flow controlled at 30 to 40 L/min)

[0775]   O2 flow: 34 L/min (flow controlled at 30 to 40 L/min)

[0776]   PCW (cooling water): 2.6 L/min (flow controlled at 2.0 to 3.0 L/min)

[0777]   Plasma irradiation was performed, with the distance between the plasma irradiated surface and the top surface of the substrate being within the range of 0.5 to 3.0 mm.

(Formation of Underlayer)

[0778]   Using the chemical solutions (A) to (F) prepared above, the underlayer was formed on the glass substrate. Specifically, 300 mL each of the chemical solutions (A) to (F) prepared above was added to a separate vessel. Then, the glass substrate was immersed in the composition in each vessel for 20 seconds. Thereafter, the substrate was raised at 20 mm/see, the treated substrate was allowed to stand still at room temperature for 3 hours, and thus underlayer-treated substrates (A) to (F) were formed.

(Formation of Surface-Treating Layer)

Example 1

[0779]   Using a commercially available spray coater equipped with two fluid nozzles, the water-repelling solution (1) prepared above was uniformly sprayed onto the surface of the underlayer-treated substrate (A) at a head speed of 70 mm/sec. The amount of the water-repelling solution (1) applied was 60 g per unit m2 of the substrate. Then, the substrate was removed and left to stand still in air at room temperature for 24 hours, and thus a water-repelling substrate (1A) was obtained.

Example 2

[0780] A water-repelling substrate (1B) was obtained in the same manner as in Example 1 except that the underlayer-treated substrate (B) was used in place of the underlayer-treated substrate (A).

Example 3

[0781] A water-repelling substrate (1C) was obtained in the same manner as in Example 1 except that the underlayer-treated substrate (C) was used in place of the underlayer-treated substrate (A).

Example 4

[0782] A water-repelling substrate (1D) was obtained in the same manner as in Example 1 except that the underlayer-treated substrate (D) was used in place of the underlayer-treated substrate (A).

Example 5

[0783] A water-repelling substrate (1E) was obtained in the same manner as in Example 1 except that the underlayer-treated substrate (E) was used in place of the underlayer-treated substrate (A).

Example 6

[0784] A water-repelling substrate (1F) was obtained in the same manner as in Example 1 except that the underlayer-treated substrate (F) was used in place of the underlayer-treated substrate (A).

Example 7

[0785] Using a commercially available spray coater equipped with two fluid nozzles, the water-repelling solution (2) prepared above was uniformly sprayed onto the surface of the underlayer-treated substrate (B) at a head speed of 70 mm/sec. The amount of the water-repelling solution (2) applied was 60 g per unit m2 of the substrate. Then, the substrate was removed and left to stand still in air at room temperature for 24 hours, and thus a water-repelling substrate (2B) was obtained.

Example 8

[0786] A water-repelling substrate (2E) was obtained in the same manner as in Example 6 except that the underlayer-treated substrate (E) was used in place of the underlayer-treated substrate (B).

Example 9

[0787] Using a commercially available spray coater equipped with two fluid nozzles, the water-repelling solution (3) prepared above was uniformly sprayed the surface of the underlayer-treated substrate (B) at a head speed of 70 mm/sec. The amount of the water-repelling solution (3) applied was 60 g per unit m2 of the substrate. Then, the substrate was removed and left to stand still in air at room temperature for 24 hours, and thus water-repelling substrate (3B) was obtained.

Comparative Example 1

[0788] Using a commercially available spray coater equipped with two fluid nozzles, the water-repelling solution (1) prepared above was uniformly sprayed onto the surface of a glass substrate not treated with the underlayer at a head speed of 70 mm/sec. The amount of the water-repelling solution (1) applied was 50 g per unit m2 of the substrate. Then, the substrate was removed and left to stand still in air at room temperature for 24 hours, and thus water-repelling substrate (1) was obtained.

Comparative Example 2

[0789] A water-repelling substrate (2) was obtained in the same manner as in Comparative Example 1 except that the water-repelling solution (2) was used in place of the water-repelling solution (1).

Comparative Example 3

[0790] A water-repelling substrate (3) was obtained in the same manner as in Comparative Example 1 except that the water-repelling solution (3) was used in place of the water-repelling solution (1).

Comparative Example 4

[0791] A water-repelling substrate (4) was obtained in the same manner as in Comparative Example 1 except that the water-repelling solution (4) was used in place of the water-repelling solution (1).

Comparative Example 5

[0792] A water-repelling substrate (5) was obtained in the same manner as in Comparative Example 1 except that the water-repelling solution (5) was used in place of the water-repelling solution (1).

Comparative Example 6

[0793] A water-repelling substrate (6) was obtained in the same manner as in Comparative Example 1 except that the water-repelling solution (6) was used in place of the water-repelling solution (1).

(Evaluation)

[0794] The water-repelling substrates 1A to 1F, 2B, 2E, 3B, and 1 to 6 obtained above were subjected to the following evaluations.

Measurement of Static Water Contact Angle

[0795] The static water contact angle was measured using a fully automatic contact angle meter DropMaster 700 (manufactured by Kyowa Interface Science Co., Ltd.). Specifically, the static water contact angle was obtained by dripping 2 μL of water from a microsyringe onto a horizontally placed substrate and taking a still image with a video microscope 1 second after the dripping. Concerning the measured value of the static water contact angle, five different points on the surface-treating layer of the substrate were measured, and the average value thereof was calculated and used.

Evaluation of Accelerated Weather Resistance

[0796] An accelerated weathering resistance test was performed on the water-repelling substrates 1A to 1F, 2B, 2E, 3B, and 1 to 6 obtained above. The steps of (1) UV irradiation for 24 hours, (2) irradiation with ion-exchanged water for 5 minutes, and (3) leaving the substrate in the dark for 1 hour were repeated three time, which were thus regarded as one cycle (75 hours), and the static contact angle of water was measured after each cycle. For UV irradiation, a UVB-313 lamp (manufactured by Q-Lab, irradiance of 0.63 W/m² at 310 nm) was used, the distance between the lamp and the surface-treating layer of the substrate was 5 cm, and the temperature of the plate on which the substrate was placed was 63° C. The substrate was temporarily removed when measuring the static water contact angle, and the surface-treating layer was wiped back and forth five times with Kimwipe (trade name, manufactured by Jujo Kimberly Co., Ltd.). Immediately thereafter, the static water contact angle was measured. The static water contact angle after a given number of cycles is shown in Table 1 wherein the result of Test Example 1 is denoted as the number of cycles being 0.

TABLE 1

Example	Comparative Example	Substrate	Underlayer chemical solution	Water-repelling solution	0	1	2	3	4	5	6	7	8
1		Water-repelling substrate 1A	A	1	114	116	115	115	114	113	114	113	113
2		Water-repelling substrate 1B	B	1	114	115	115	115	116	114	115	114	113
3		Water-repelling substrate 1C	C	1	114	115	114	115	114	114	115	116	115
4		Water-repelling substrate 1D	D	1	114	115	114	114	114	115	115	114	114
5		Water-repelling substrate 1E	E	1	114	115	114	114	115	115	116	115	114
6		Water-repelling substrate 1F	F	1	116	115	115	114	114	113	112	108	106
7		Water-repelling substrate 2B	B	2	114	114	114	115	113	110	102	102	99
8		Water-repelling substrate 2E	E	2	115	114	114	114	113	113	111	109	109
9		Water-repelling substrate 3B	B	3	114	111	106	87	82	72	70		
	1	Glass substrate	—	1	114	115	114	113	114	114	114	111	108
	2	Glass substrate	—	2	114	113	113	108	104	96	85	79	73
	3	Glass substrate	—	3	115	110	92	79	69				
	4	Glass substrate	—	4	115	99	62						
	5	Glass substrate	—	5	107	102	93						
	6	Glass substrate	—	6	107	105	103	90					

Example	Comparative Example	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23
1		112	111	108	105	96	93	84	84	68						
2		113	111	109	109	107	104	98	94	90	90	84	89			
3		112	110	110	109	108	108	108	101	98	92	91	85	84	73	
4		113	112	110	110	108	105	102	98	88	86	86	85	75		
5		113	113	109	109	108	107	102	103	102	103	95	89	89	82	78
6		97														
7		95	85	82	80											
8		108	106	103	95											
9																
	1	104	93	94	79	76										
	2															
	3															
	4															
	5															
	6															

## INDUSTRIAL APPLICABILITY

[0797] The laminate of the present disclosure can be suitably used in various applications, especially an article for outdoor applications.

1. A laminate comprising a substrate, an underlayer disposed on the substrate, and a surface-treating layer disposed on the underlayer, wherein

the underlayer is a layer formed of:

(A) polysilazane represented by the following formula (P):

[Formula 1]

$$\left[\begin{array}{c} R^{61} \\ | \\ -Si-N- \\ | \quad | \\ R^{62} \quad R^{63} \end{array}\right]_{n}$$
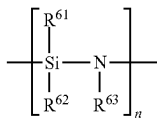
wherein

R^{61} is a hydrogen atom or a C_{1-6} alkyl group,

R^{62} is a hydrogen atom, a C_{1-6} alkyl group, or a group having a functional group,

R^{63} is a hydrogen atom, a C_{1-6} alkyl group, or a group having a functional group, and

n is an integer of 2 to 100; or

(B) aminosilane represented by the following formula (Q):

$$N(R^{71}-SiR^{72}{}_3)_m R^{73}{}_{3-m} \qquad [Formula\ 2]$$

wherein

R^{71} is each independently a C_{1-6} alkylene group,

R^{72} is each independently a hydrolyzable group,

R^{73} is a hydrogen atom or a C_{1-6} alkyl group, and

m is 2 or 3; and

the surface-treating layer is a layer formed of a fluorine-containing silane compound.

2. The laminate according to claim 1, wherein the fluorine-containing silane compound is a compound represented by the following formula (A1) or (A2):

[Formula 3]

$$R^{F1}{}_{\alpha 1}-X^4-R^{Si}{}_{\beta 1} \qquad (A1)$$

$$R^{Si}{}_{\gamma 1}-X^4-R^{F2}-X^4-R^{Si}{}_{\gamma 1} \qquad (A2)$$

wherein

R^{F1} is each independently Rf^1- or $Rf^1-R^F-O_q-$,

R^{F2} is $-Rf^2-$ or $-Rf^2{}_p-R^F-O_q-$,

R^F is each independently a divalent fluoropolyether group,

Rf^1 is each independently a C_{1-16} alkyl group optionally substituted with one or more fluorine atoms,

Rf^2 is a C_{1-6} alkylene group optionally substituted with one or more fluorine atoms,

p is 0 or 1,

q is each independently 0 or 1,

R^{Si} is each independently a monovalent group containing a Si atom to which a hydroxyl group, a hydrolyzable group, a hydrogen atom, or a monovalent organic group is bonded,

at least one R^{Si} is a monovalent group containing a Si atom to which a hydroxyl group or a hydrolyzable group is bonded,

X^4 is each independently a single bond or a di- to decavalent organic group,

$\alpha 1$ is an integer of 1 to 9,

$\beta 1$ is an integer of 1 to 9, and

$\gamma 1$ is each independently an integer of 1 to 9.

3. The laminate according to claim 2, wherein

Rf^1 is each independently a C_{1-16} perfluoroalkyl group, and
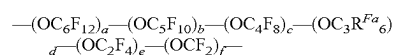
Rf^2 is each independently a C_{1-6} perfluoroalkylene group.

4. The laminate according to claim 2, wherein

R^{F1} is each independently $Rf^1-R^F-O_q-$, and

R^{F2} is $-Rf^2{}_p-R^F-O_q-$.

5. The laminate according to claim 2, wherein

R^F is each independently a group represented by the following formula:

$$-(OC_6F_{12})_a-(OC_5F_{10})_b-(OC_4F_8)_c-(OC_3R^{Fa}{}_6)_d-(OC_2F_4)_e-(OCF_2)_f-$$

wherein

R^{Fa} is each independently a hydrogen atom, a fluorine atom, or a chlorine atom; and

a, b, c, d, e, and f are each independently an integer of 0 to 200, the sum of a, b, c, d, e, and f is 1 or more, and the occurrence order of the respective repeating units enclosed in parentheses provided with a, b, c, d, e, or f is not limited in the formula, provided that when all R^{Fa} groups are hydrogen atoms or chlorine atoms, at least one of a, b, c, e, and f is 1 or more.
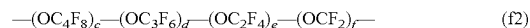
6. The laminate according to claim 5, wherein R^{Fa} is a fluorine atom.

7. The laminate according to claim 2, wherein

R^F is each independently a group represented by the following formula (f1), (f2), (f3), (f4), (f5), or (f6):

$$-(OC_3F_6)_d-(OC_2F_4)_e- \qquad (f1)$$

wherein d is an integer of 1 to 200, and e is 0 or 1;

$$-(OC_4F_8)_c-(OC_3F_6)_d-(OC_2F_4)_e-(OCF_2)_f- \qquad (f2)$$

wherein c and d are each independently an integer of 0 to 30,

e and f are each independently an integer of 1 to 200,

the sum of c, d, e, and f is an integer of 10 to 200, and

the occurrence order of the respective repeating units enclosed in parentheses provided with the subscript c, d, e, or f is not limited in the formula;

$$-(R^6-R^7)_g- \qquad (f3)$$

wherein R^6 is OCF_2 or OC_2F_4,

R^7 is a group selected from OC_2F_4, OC_3F_6, OC_4F_8, OC_5F_{10}, and OC_6F_{12}, or is a combination of two or three groups selected from these groups, and

g is an integer of 2 to 100;

$$-(R^6-R^7)_g-R^r-(R^7-R^6)_{g'}- \qquad (f4)$$
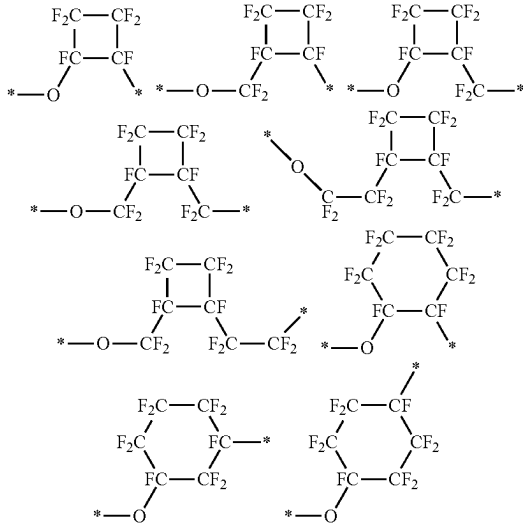
wherein R^6 is OCF_2 or OC_2F_4,

R^7 is a group selected from OC_2F_4, OC_3F_6, OC_4F_8, OC_5F_{10}, and OC_6F_{12}, or is a combination of two or three groups independently selected from these groups,

$R^{6'}$ is OCF_2 or OC_2F_4,

$R^{7'}$ is a group selected from OC_2F_4, OC_3F_6, OC_4F_8, OC_5F_{10}, and OC_6F_{12}, or is a combination of two or three groups independently selected from these groups,
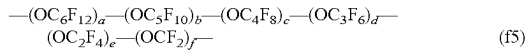
g is an integer of 2 to 100,

g' is an integer of 2 to 100, and

R^r is any of:

[Formula 4]

wherein * represents a bond position;

$$-(OC_6F_{12})_a-(OC_5F_{10})_b-(OC_4F_8)_c-(OC_3F_6)_d-$$
$$(OC_2F_4)_e-(OCF_2)_f- \quad (f5)$$

wherein e is an integer of 1 or more and 200 or less, a, b, c, d, and f are each independently an integer of 0 or more and 200 or less, and the occurrence order of the respective repeating units enclosed in parentheses provided with a, b, c, d, e or f is not limited in the formula; and

$$-(OC_6F_{12})_a-(OC_5F_{10})_b-(OC_4F_8)_c-(OC_3F_6)_d-$$
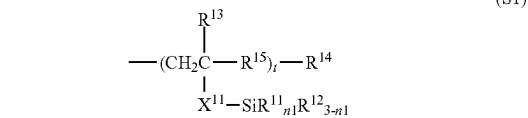$$(OC_2F_4)_e-(OCF_2)_f- \quad (f6)$$
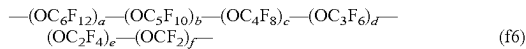
wherein f is an integer of 1 or more and 200 or less, a, b, c, d, and e are each independently an integer of 0 or more and 200 or less, and the occurrence order of the respective repeating units enclosed in parentheses provided with a, b, c, d, e or f is not limited in the formula.

8. The laminate according to claim **2**, wherein

R^{Si} is a group represented by the following formula (S1), (S2), (S3), (S4), or (S5):

[Formula 5]

$$-(CH_2C-R^{15})_t-R^{14} \quad (S1)$$

with R^{13} above and $X^{11}-SiR^{11}{}_{n1}R^{12}{}_{3-n1}$ below the $(CH_2C-R^{15})_t$ chain

$$-SiR^{11}{}_{n1}R^{12}{}_{3-n1} \quad (S2)$$

$$-SiR^{a1}{}_{k1}R^{b1}{}_{l1}R^{c1}{}_{m1} \quad (S3)$$

$$-CR^{d1}{}_{k2}R^{e1}{}_{l2}R^{f1}{}_{m2} \quad (S4)$$

$$-NR^{g1}R^{h1} \quad (S5)$$

wherein

R^{11} is each independently a hydroxyl group or a hydrolyzable group,

R^{12} is each independently a hydrogen atom or a monovalent organic group,

n1 is each independently an integer of 0 to 3 for each $(SiR^{11}{}_{n1}R^{12}{}_{3-n1})$ unit,

X^{11} is each independently a single bond or a divalent organic group,

R^{13} is each independently a hydrogen atom or a monovalent organic group,

t is each independently an integer of 2 or more,

R^{14} is each independently a hydrogen atom, a halogen atom, or $-X^{11}-SiR^{11}{}_{n1}R^{12}{}_{3-n1}$,

R^{15} is each independently at each occurrence a single bond, an oxygen atom, an alkylene group having 1 to 6 carbon atoms, or an alkyleneoxy group having 1 to 6 carbon atoms,

R^{a1} is each independently $-Z^1-SiR^{21}{}_{p1}R^{22}{}_{q1}R^{23}{}_{r1}$,

Z^1 is each independently an oxygen atom or a divalent organic group,

R^{21} is each independently $-Z^{1'}-SiR^{21'}{}_{p1'}R^{22'}{}_{q1'}R^{23'}{}_{r1'}$,

R^{22} is each independently a hydroxyl group or a hydrolyzable group,

R^{23} is each independently a hydrogen atom or a monovalent organic group,

p1 is each independently an integer of 0 to 3,

q1 is each independently an integer of 0 to 3,

r1 is each independently an integer of 0 to 3,

the sum of p1, q1, and r1 is 3 in the $(SiR^{21}{}_{p1}R^{22}{}_{q1}R^{23}{}_{r1})$ unit,

$Z^{1'}$ is each independently an oxygen atom or a divalent organic group,

$R^{21'}$ is each independently $-Z^{1''}-SiR^{22''}{}_{q1''}R^{23''}{}_{r1''}$,

$R^{22'}$ is each independently a hydroxyl group or a hydrolyzable group,

$R^{23'}$ is each independently a hydrogen atom or a monovalent organic group,

p1' is each independently an integer of 0 to 3,

q1' is each independently an integer of 0 to 3,

r1' is each independently an integer of 0 to 3,

the sum of p1', q1', and r1' is 3 in the $(SiR^{21'}{}_{p1'}R^{22'}{}_{q1'}R^{23'}{}_{r1'})$ unit,

$Z^{1''}$ is each independently an oxygen atom or a divalent organic group,

$R^{22''}$ is each independently a hydroxyl group or a hydrolyzable group,

$R^{23''}$ is each independently a hydrogen atom or a monovalent organic group,

q1" is each independently an integer of 0 to 3,

r1" is each independently an integer of 0 to 3,

the sum of q1" and r1" is 3 in the $(SiR^{22''}q1''R^{23''}-1'')$ unit,

R^{b1} is each independently a hydroxyl group or a hydrolyzable group,

R^{c1} is each independently a hydrogen atom or a monovalent organic group,

k1 is each independently an integer of 0 to 3,

l1 is each independently an integer of 0 to 3,

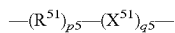m1 is each independently an integer of 0 to 3,

the sum of k1, l1, and m1 is 3 in the $(SiR^{a1}{}_{k1}R^{b1}{}_{l1}R^{c1}{}_{m1})$ unit,

R^{d1} is each independently $-Z^2-CR^{31}{}_{p2}R^{32}{}_{q2}R^{33}{}_{r2}$,

Z^2 is each independently a single bond, an oxygen atom, or a divalent organic group,

R^{31} is each independently $-Z^{2'}-CR^{32'}{}_{q2'}R^{33'}{}_{r2'}$,

R^{32} is each independently -Z^3-SiR$^{34}_{n2}$R$^{35}_{3-n2}$,

R^{33} is each independently a hydrogen atom, a hydroxyl group, or a monovalent organic group,

p2 is each independently an integer of 0 to 3,

q2 is each independently an integer of 0 to 3,

r2 is each independently an integer of 0 to 3,

the sum of p2, q2, and r2 is 3 in the (CR$^{31}_{p2}$R$^{32}_{q2}$R$^{33}_{r2}$) unit,

$Z^{2'}$ is each independently a single bond, an oxygen atom, or a divalent organic group,

$R^{32'}$ is each independently -Z^3-SiR$^{34}_{n2}$R$^{35}_{3-n2}$,

$R^{33'}$ is each independently a hydrogen atom, a hydroxyl group, or a monovalent organic group,

q2' is each independently an integer of 0 to 3,

r2' is each independently an integer of 0 to 3,

the sum of q2' and r2' is 3 in the (CR$^{32'}_{q2'}$R$^{33'}_{r2'}$) unit,

Z^3 is each independently a single bond, an oxygen atom, or a divalent organic group,

R^{34} is each a independently hydroxyl group or a hydrolyzable group,

R^{35} is each independently a hydrogen atom or a monovalent organic group,

n2 is each independently an integer of 0 to 3,

R^{e1} is each independently -Z^3-SiR$^{34}_{n2}$R$^{35}_{3-n2}$,

R^{f1} is each independently a hydrogen atom, a hydroxyl group, or a monovalent organic group,

k2 is each independently an integer of 0 to 3,

l2 is each independently an integer of 0 to 3,

m2 is each independently an integer of 0 to 3,

the sum of k2, l2, and m2 is 3 in the (CR$^{d1}_{k2}$R$^{e1}_{l2}$R$^{f1}_{m2}$) unit,

R^{g1} and R^{h1} are each independently -Z^4-SiR$^{11}_{n1}$R$^{12}_{3-n1}$, -Z^4-SiR$^{a1}_{k1}$R$^{b1}_{l1}$R$^{c1}_{m1}$, or -Z^4-CR$^{d1}_{k2}$R$^{e1}_{l2}$R$^{f1}_{m2}$, and

Z^4 is each independently a single bond, an oxygen atom, or a divalent organic group,

provided that in the formulae (S1), (S2), (S3), (S4), and (S5), at least one Si atom to which a hydroxyl group or a hydrolyzable group is bonded is present.

9. The laminate according to claim **2**, wherein α1, β1, and γ1 are each 1.

10. The laminate according to claim **2**, wherein X^4 is a single bond or a divalent organic group represented by the following formula:

$$—(R^{51})_{p5}—(X^{51})_{q5}—$$

wherein

R^{51} is a single bond or —(CH$_2$)$_{s5}$—,

s5 is an integer of 1 to 20,

X^{51} is —(X^{52})$_{l5}$—,

X^{52} each independently represents a group selected from the group consisting of —O—, —S—, —CO—, —NR^{54}—, —(CH$_2$)$_{n5}$—, —COO—, —OCO—,

—CONR^{54}—, —NR^{54}CO—, —O—CONR^{54}—, —NR^{54}CO—O—, —Si(R^{53})$_2$—, and —(Si(R^{53})$_2$O)$_{m5}$—Si(R^{53})$_2$—,

R^{53} is each independently a phenyl group, a C$_{1-6}$ alkyl group, or a C$_{1-6}$ alkoxy group,

R^{54} is each independently a hydrogen atom, a phenyl group, or a C$_{1-6}$ alkyl group,

m5 is each independently an integer of 1 to 100,

n5 is each independently an integer of 1 to 20,

l5 is an integer of 1 to 10,

p5 is 0 or 1, and

q5 is 0 or 1,

provided that at least one of p5 and q5 is 1, the occurrence order of the respective repeating units enclosed in parentheses provided with p5 or q5 is not limited, and the right side is bonded to R^{Si}.

11. The laminate according to claim **2**, wherein X^4 is a group represented by the following formula:
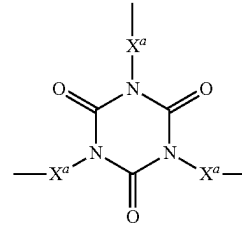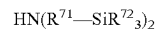
[Formula 6]

wherein X^a is a single bond or a divalent organic group.

12. A laminate according to claim **1**, wherein the polysilazane is represented by formula (P) wherein R^{61} is a hydrogen atom, R^{62} is a hydrogen atom, and R^{63} is a hydrogen atom.

13. The laminate according to claim **1**, wherein the polysilazane has a number average molecular weight of 1,000 to 10,000.

14. The laminate according to claim **1**, wherein the aminosilane is represented by the following formula (Q-1):

HN(R^{71}—SiR$^{72}_3$)$_2$ [Formula 7]

wherein

R^{71} is each independently a C$_{1-6}$ alkylene group, and

R^{72} is each independently a hydrolyzable group.

15. The laminate according to claim **1**, wherein the substrate is glass, metal, or ceramic.

16. An article comprising the laminate according to claim **1**.

17. The article according to claim **16**, which is automobile glass, outdoor glass, or a display.

* * * * *