

## Appeal decision

Appeal No. 2015- 6131

USA

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The case of appeal against the examiner's decision of refusal of Japanese Patent Application No. 2012-554984, entitled "ANTIREFLECTIVE HARDMASK COMPOSITION AND A METHOD OF PREPARING A PATTERNED MATERIAL USING SAME" (September 1, 2011 international publication, International Publication No. WO2011/106040, June 13, 2013, National Publication, National Publication of International Patent Application No. 2013-522654) has resulted in the following appeal decision.

### Conclusion

The appeal of the case was groundless.

### Reason

#### No. 1 History of the procedures

The present application was filed on October 22, 2010 (priority claim under the Paris Convention received by the foreign office: February 24, 2010 (US)) as an international filing date, a notification of reasons for refusal was issued on June 30, 2014, and a written argument and a written amendment were submitted on October 15, 2014. A decision for refusal was issued on December 3, 2014, an appeal against an examiner's decision of refusal was filed on April 2, 2015, and a written amendment was submitted on the same day.

#### No. 2 Decision to dismiss amendment

Decision to dismiss amendment by the written amendment submitted on April 2, 2015 (hereinafter referred to as the "Amendment")

#### [Conclusion of Decision to Dismiss Amendment]

The Amendment shall be dismissed.

#### [Reason]

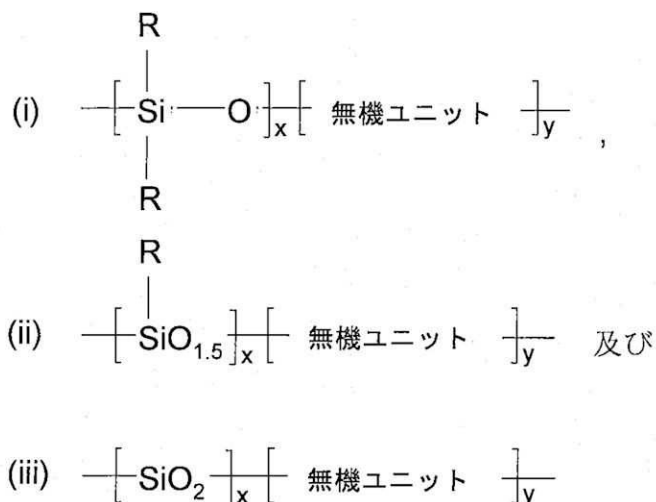
##### 1 Details of the Amendment

The Amendment is to amend the scope of claims amended by the written amendment submitted on October 15, 2014 (hereinafter referred to as "before the Amendment") and the amendment of claim 1 is as follows (underlines, which were added by the body, indicate words related to the amendment.).

(1) Claims 1 and 2 before the Amendment

"1. An antireflective hardmask composition comprising a repeating structural unit selected from the group illustrated below:

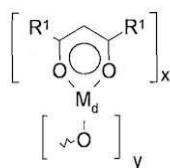
[Formula 1]



無機ユニット            Inorganic Unit  
及び and

wherein x is at least 1; y is at least 1; R is an organic functional unit selected from one or more chromophore moieties, transparent moieties, and crosslinking components; and said inorganic unit has the following formula:

[Formula 2]



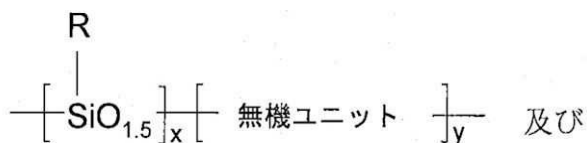
wherein x and y have the meanings given above; M<sub>d</sub> is selected from Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, and lanthanide rare earth metals; and R<sup>1</sup> is the same or different and is selected from C<sub>1</sub>-C<sub>6</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkoxy, phenyl, and phenoxy."

"2. A composition in accordance with claim 1 wherein said polymer further comprises an acid generator."

(2) Claim 1 after the Amendment

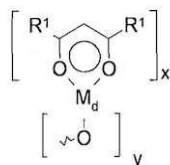
"1. An antireflective hardmask composition comprising a polymer composed of a repeating structural unit selected from the group illustrated below and an acid

generator:  
[Formula 1]



無機ユニット            Inorganic Unit  
及び    and

wherein x is at least 1; y is at least 1; R is an organic functional unit selected from one or more chromophore moieties, transparent moieties, and crosslinking components; and said inorganic unit has the following formula:  
[Formula 2]



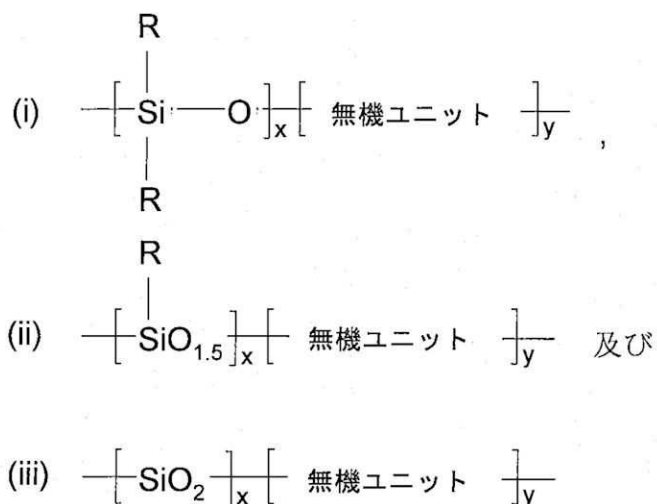
wherein x and y have the meanings given above; M<sub>d</sub> is Ti; and R<sup>1</sup> is the same or different and is selected from C<sub>1</sub>-C<sub>6</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkoxy, phenyl, and phenoxy."

2. As to whether amendment adds a new matter or not and purpose of amendment  
(1) Amended matter

The Amendment of claim 1 consists of the following amended matters.

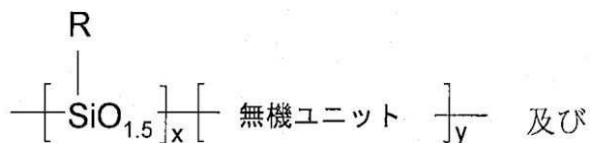
A. Claim 1 is deleted and claim 2 is changed to an independent claim to be the new claim 1.

B. From a matter necessary to specify the invention according to claim 1 before the Amendment, "a repeating structural unit selected from the group illustrated below:  
[Formula 1]



無機ユニット            Inorganic Unit  
及び and

"a part of the choices is deleted and the matter is limited to "a repeating structural unit selected from the group illustrated below:  
[Formula 1]



無機ユニット            Inorganic Unit  
及び and

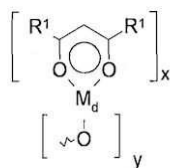
."

In the above amended matter the presence of "and," which is to be deleted, is apparently an editorial error and the above amended matter will be examined without the "and."

C. A matter necessary to specify the invention according to claim 1 before the Amendment, "a repeating structural unit" is amended to "a polymer composed of a repeating structural unit."

D. From a matter necessary to specify the invention according to claim 1 before the Amendment, "inorganic unit" the statement "has the following formula:" is amended to "has the following formula;"

E. On a matter necessary to specify the invention according to claim 1 before the Amendment, "inorganic unit" having "the following formula:  
[Formula 2]



"a part of the choices of " M<sub>d</sub> " which "is selected from Ti, Zr, Hf, Sc, Y, La, V, Nb, Ta, and lanthanide rare earth metals" is deleted and " M<sub>d</sub> " is limited to " M<sub>d</sub> " which "is Ti".

F. On a matter necessary to specify the invention according to claim 2 before the Amendment, the relation between "polymer" and "acid generator," the statement "said polymer further comprises an acid generator" is amended to "comprising a polymer" "and an acid generator."

(2) Purpose, etc. of amendment

A. The amended matter (1) A above falls under the purpose of the deletion of a claim or claims in Article 17-2(5)(i) of the Patent Act.

B. The amended matters (1) B and E above delete a part of the choices of "repeating structural unit" and " M<sub>d</sub> " in "inorganic unit," respectively, and restrict a matter necessary to specify the invention according to claim 1 before the Amendment. Since the field of industrial application and the problems to be solved of the claimed invention before and after this amendment are found to be the same, the amended matters comply with the requirement as provided in Article 17-2(3) of the Patent Act and fall under the purpose of restriction of the scope of claims in Article 17-2(5)(ii) of the Patent Act.

C. The amended matters (1) C, D, and F above fall under the purpose of the correction of errors in Article 17-2(5)(iii) of the Patent Act and, since they do not add any new matter, comply with the requirement as provided in Article 17-2(3) of the Patent Act.

3. Consideration on independent requirements for patentability

Since the Amendment of claim 1 includes amended matters having the purpose of restriction of the scope of claims in Article 17-2(5)(ii) of the Patent Act, whether the invention according to Claim 1 after the Amendment (hereinafter, referred to as "Amended Invention of the Application") is independently patentable at the time of the patent application (complies with the provisions of Article 126(7) of the Patent Act which is applied mutatis mutandis pursuant to the provisions of Article 17-2(6) of the Patent Act) will be examined.

(1) Amended Invention of the Application

Amended Invention of the Application is as described in 1 (2) above.

(2) As to Article 36(6)(i) of the Patent Act (requirements for support)

A. Description of the Application includes the following statement. (Underlines were added by the body. The same shall apply hereinafter.)

(A) "Background Art

[0002]

The need to remain cost and performance competitive in the production of semiconductor devices has driven the industry to a continuing increase in device density with a concomitant decrease in device geometry. To facilitate the shrinking device dimensions, new lithographic materials, processes, and tools are being considered. Typical lithographic processes involve formation of a patterned resist layer by patternwise exposing the radiation-sensitive resist to an imaging radiation. The image is subsequently developed by contacting the exposed resist layer with a material (typically an aqueous alkaline developer) to selectively remove portions of the resist layer to reveal the desired pattern. The pattern is subsequently transferred to an underlying material by etching the material in openings of the patterned resist layer. After the transfer is complete, the remaining resist layer is then removed. Currently, 248 nm and 193 nm lithography are being pursued to print sub-200 nm features.

[0003]

To do this, tools with higher numerical aperture (NA) are emerging. The higher NA allows for improved resolution but reduces the depth of focus of aerial images projected onto the resist. Because of the reduced depth of focus, a thinner resist will be required. As the thickness of the resist is decreased, the resist becomes less effective as a mask for subsequent dry etch image transfer to the underlying substrate. Without significant improvement in the etch resistance exhibited by current single layer resists, these systems cannot provide the necessary lithography and etch properties for high-resolution lithography.

[0004]

Another problem with single layer resist systems is critical dimension (CD) control. Substrate reflections at ultraviolet (UV) and deep ultraviolet (DUV) wavelengths are notorious for producing standing wave effects and resist notching, which severely limit CD control of single layer resists. Notching results from substrate topography and non-uniform substrate reflectivity, which causes local variations in exposure energy on the resist. Standing waves are thin film interference (TFI) or periodic variations of light intensity through the resist thickness. These light variations are introduced because planarization of the resist presents a different thickness through the underlying topography. Thin film interference plays a dominant role in CD control of single layer photoresist processes, causing large changes in the effective exposure dose due to a tiny change in the optical phase. Thin film interference effects are described in Non Patent Document 1, the teaching of which is incorporated herein by reference.

[0005]

Bottom antireflective coatings or BARCs have been used with single layer resists to reduce thin film interference. However, these thin absorbing BARCs have fundamental limitations. For some lithographic imaging processes, the resist used does not provide sufficient resistance to subsequent etching steps to enable effective transfer of the desired pattern to a layer underlying the resist. The resist typically gets consumed after transferring the pattern into the underlying BARC and substrates. In addition, the migration to smaller sub-90 nm node feature sizes requires the use of an ultrathin resist (>200 nm) in order to avoid image collapse. In many instances where a substantial etching depth is required, and/or where it is desired to use certain etchants for a given

underlying material, the resist thickness is insufficient to complete the etch process. In addition, the radiation-sensitive resist material employed does not provide sufficient resistance to subsequent etching steps to enable effective transfer of the desired pattern to the layer underlying the radiation-sensitive resist and anti-reflective coating (ARC).

[0006]

In many cases, where the underlying material layer to be etched is thick, where a substantial etching depth is required, where it is desirable to use certain etchants for a given underlying layer, or any combination of the above, it would be desirable to employ an antireflective hardmask. The antireflective hardmask layer could serve as an intermediate layer between the patterned radiation-sensitive resist material and the underlying material layer to be patterned. The antireflective hardmask layer receives the pattern from the patterned radiation-sensitive resist material layer by reactive ion etching (RIE) followed by the transfer of the pattern to the underlying material layer. The antireflective hardmask layer should be able to withstand the etching processes required to transfer the pattern onto the underlying material layer. Furthermore, a thin antireflective hardmask layer is desirable to receive the pattern by RIE from the resist layer, especially if a thin resist is used. While many materials useful as ARC compositions are known, there is a need for improved antireflective hardmask compositions with high etch selectivity to the radiation-sensitive resist material and to the underlying material layer. Further, many of the known antireflective hardmasks are difficult to apply to the substrate, e.g., applying these ARCs may require the use of chemical vapor deposition (Patent Document 1; Patent Document 2). It would be advantageous to apply the antireflective hardmask material by spin-on techniques like conventional organic BARC currently used in manufacturing.

[0007]

In addition, antireflective hardmask materials are difficult to remove after pattern transfer. Typically organic BARC are removed by a wet or dry ashing process. CVD deposited hardmask layers are difficult to remove without damaging the underlying dielectric substrate. Ideally, the antireflective hardmask materials can be removed easily by a wet strip with high selectivity to the underlying substrates.

[0008]

Thus, it would be desirable to be able to perform lithographic techniques with high etch selectivity yet sufficient resistance to multiple etchings. Such lithographic techniques would enable production of highly detailed semiconductor devices.

... (omission) ...

Technical Problem

[0011]

The present invention provides antireflective hardmask compositions and techniques for the use of antireflective hardmask compositions for processing of semiconductor devices. In one aspect of the invention, an antireflective hardmask layer for lithography is provided."

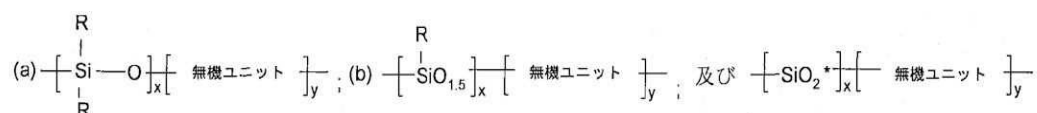
(B) "Description of Embodiments

[0015]

A lithographic structure comprises an antireflective hardmask composition (hereinafter "the composition") disclosed herein. In one aspect of the invention the

composition comprises a polymer with an Si-O backbone containing a non-silicon inorganic moiety incorporated into the backbone comprising at least one chromophore moiety, at least one transparent moiety, and a crosslinking component. The polymer may comprise any combination of an acid generator and an additional crosslinking component. The composition comprises a polymer having an Si-O backbone and a non-silicon based inorganic unit incorporated into the backbone and having a repeating structural formula selected from the group consisting of the following structural formulas:

[Formula 1]

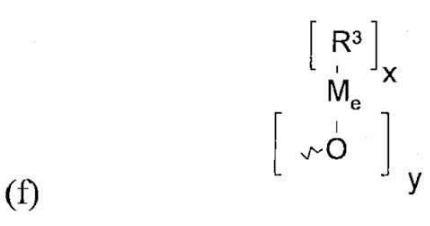
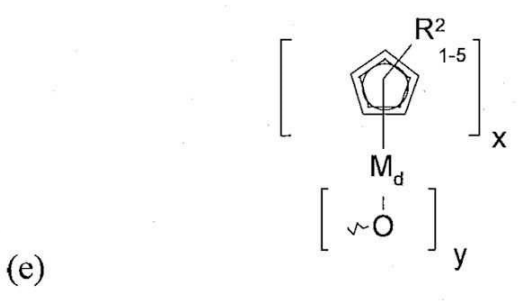
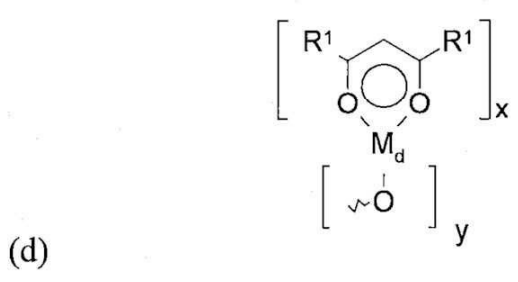
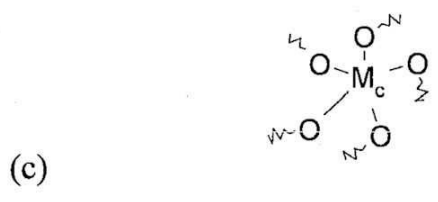
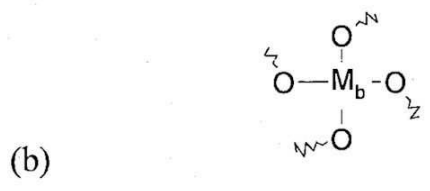
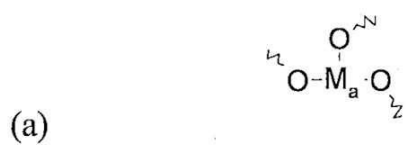


無機ユニット                      Inorganic Unit  
及び and

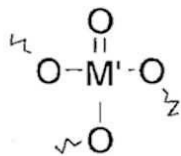
wherein x is at least 1; y is at least 1; R is a chromophore, at least one transparent moiety, or at least one transparent moiety and a crosslinking component with the proviso that the inorganic unit contains no silicon; and

[Formula 2]





wherein x and y have the meanings above;  $M_a$  is Sc, Y, La, B, Al, Ga, In, or a lanthanide metal;  $M_b$  is Ti, Zr, Hf, Ge, or Sn;  $M_c$  is V, Nb, Ta, P, or [Formula 3]



$M_d$  is Ti, Zr, Hf, Sc, Y, La, a lanthanide metal, V, Nb, or Ta;  $M_e$  is Ge or Sn;  $M'$  is V or P;  $R^1$  is the same or different and is  $C_1$ - $C_6$  alkyl,  $C_1$ - $C_6$  alkoxy, phenyl, or phenoxy;  $R^2$  is the same or different and is  $C_1$ - $C_6$  alkyl or  $C_1$ - $C_6$  alkoxy;  $R^3$  is the same or different and is  $C_1$ - $C_6$  alkyl or phenyl, with the proviso that  $M_d$  may be another transition metal or a metal of Groups 2 to 5 of the Periodic Table.

[0016]

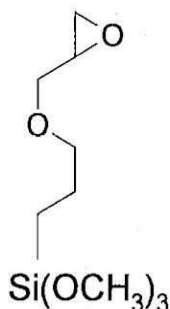
In an exemplary embodiment wherein the polymer contains Si-O in the backbone and an inorganic unit which includes no Si, the composition has at least one or more Si-O units and at least one or more inorganic non-silicon containing units. The presence of the inorganic unit within the polymer backbone ensures hardmask properties of the composition (especially when any one of the underlayers comprises a silicon oxide, nitride, and/or dielectric containing Si and C) and stripability. The Si-O-containing units may be a siloxane, silsesquioxane, or silicon oxide (sol gel) building block. The antireflective hardmask layer should have solution and film-forming characteristics conducive to layer formation by conventional spin coating."

(C) " Example 1:

[0058]

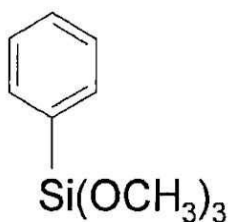
#### Preparation and Analysis of an Antireflective Hardmask Layer

A sample vial was charged with 4 g of Dowanol (Trademark) PM solution, obtained from Aldrich, and a 1 g mixture of sol gel precursors. The Dowanol (Trademark) PM solution was prepared by combining Dowanol (Trademark) PM (100 g) and FC-4430 surfactant (0.5 g). The 1 g mixture of sol gel precursor comprised 0.381 g Silane Precursor A (0.45 mol %); 0.069 g Silane Precursor B (0.1 mol %); and 0.549 g  $Ti(OC_4H_9)_4$ , where Silane Precursor A was a compound having the following structured formula:



and Silane Precursor B was a compound having the following structural formula:

[Formula 5]



[0059]

The contents of the vial were stirred for 5 minutes at room temperature, after which 1 g of 1N HCl was added disperse under vigorous stirring. The resulting clear solution was stirred at room temperature for 1 hour and then filtered through a 0.2 micron membrane filter. The filtrate was directly applied to a silicon wafer and spun at 3,000 rpm for 60 seconds followed by exposure to a temperature of 215°C for 2 minutes.

[0060]

The thus prepared antireflective hardmask layer was analyzed to determine its thickness, and its index optical constants: refraction (n) and extraction coefficient (k), measured at 193 nm, utilizing an n&k Analyzer (Trademark), manufactured by n&k Technology, Inc. In addition, a Rutherford Back Scattering (RBS) analysis of the hardmask layer was conducted. The results of these analyses are summarized in Table 1.

Example 2:

[0061]

Preparation and Analysis of an Antireflective Hardmask Layer

A second antireflective hardmask layer was prepared and analyzed in accordance with the procedure set forth in Example 1 but for the constituency of the 1 gram of silane precursor introduced into the sample vial.

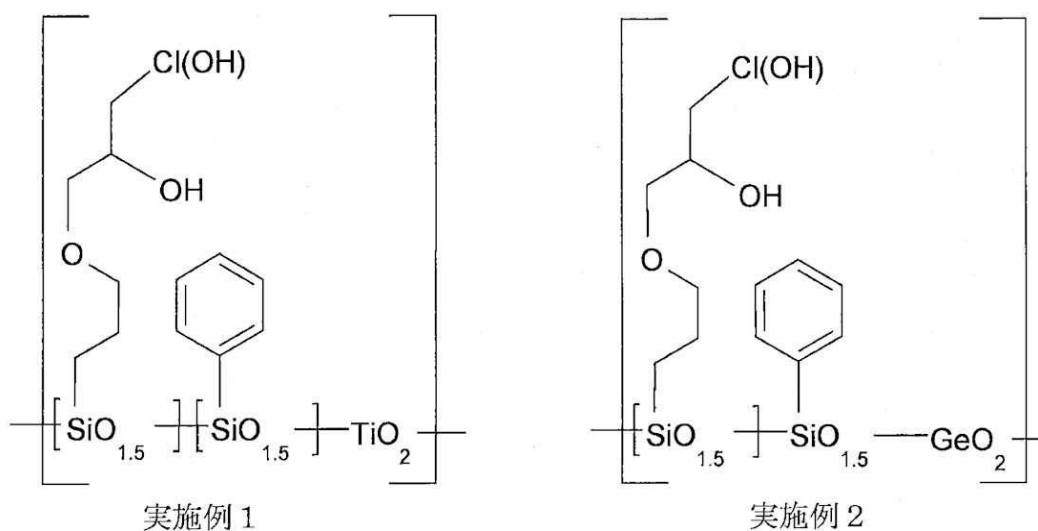
[0062]

In this example Silane Precursor A was utilized in a weight of 0.444 g, Silane Precursor B was used in a weight of 0.081 g and the third constituent was  $\text{Ge}(\text{OC}_2\text{H}_5)_4$ , present in an amount of 0.475 g.

[0063]

The repeating structural units of the antireflective hardmask composition layers of Examples 1 and 2 are as follows:

[Formula 6]



実施例 1      Example 1  
 実施例 2      Example 2

[0064]

A summary of the antireflective hardmask composition layers of Examples 1 and 2 is provided in Table 1.

[Table 1]

表 1						
反射防止ハードマスク層						
実施例 No.	シラン前駆体 A, g	シラン前駆体 B, g	第 3 成分, g	層の厚さ, Å	n (193nm)	k (193nm)
1	0.381	0.069	Ti(OC <sub>4</sub> H <sub>9</sub> ) <sub>4</sub> , .549	2332	1.735	0.301
2	0.444	0.081	Ge(OC <sub>2</sub> H <sub>5</sub> ) <sub>4</sub> , .475	3073	1.725	0.294

原子濃度, %							
実施例 No.	[S]	[O]	[C]	[H]	[Cl]	[Ti]	[Ge]
1	6.1 ± 2	19.3 ± 2	30.1 ± 2	37.3 ± 2	2.9 ± 0	4.3 ± 0.2	なし
2	6.1 ± 2	18.9 ± 2	27.0 ± 2	40.9 ± 2	2.9 ± 0	なし	4.2 ± 0.2

表 1    Table 1

反射防止ハードマスク層    Antireflective Hardmask Layer

実施例 No.                    Example No.

シラン前駆体 A, g            Silane Precursor A, g

シラン前駆体 B, g            Silane Precursor B, g

第 3 成分, g                  Third Constituent, g

層の厚さ, Å                  Layer Thickness, Ang

原子濃度, %	Atomic Concentration, %
実施例 No.	Example No.
なし	None

Example 3:

[0065]

193 nm Lithographic Layer over Antireflective Hardmask Layer

An approximately 250 nm thick layer of acrylic-based photoresist was spun-coated over the cured antireflective hardmask layer of Example 1. This radiation-sensitive photoresist imaging layer was heated at 130°C for 60 seconds. The heated imaging layer was then imaged using a 0.75 NA 193 nm ASML Stepper with conventional and annular illumination using an AFSM reticle.

[0066]

After patternwise exposure, the radiation sensitive photoresist imaging layer was heated at 130°C for 60 seconds. The image was developed using a conventional developer (0.26 M TMAH). The resulting pattern had 120 nm contact holes, 113.75 nm lines, and 104 nm space patterns.

Example 4:

[0067]

Pattern Transfer by Reactive Ion Etching

The contact holes, lines, and space patterns formed on the antireflective hardmask layer in Example 3 were transferred onto silicon oxide by a 20 second fluorocarbon based etch using a LAM RIE tool.

[0068]

The observed etch selectivity of the antireflective hardmask layer to the photoresist was approximately 2:1. The etch rate of the blanket films of Example 1 in a trifluoromethane/methane plasma was 613 Ang/min.; that in a chlorine plasma 813 Ang/min.; and that in a nitrogen/hydrogen plasma 62 Ang/min. The silicon oxide etch rate was 2000 Ang/min.

Example 5:

[0069]

Stripping the Antireflective Hardmask Composition Layer

The antireflective hardmask composition layers of Examples 1 and 2 were removed from the silicon wafers upon which they were coated by dipping the wafers in a liquid mixture of hydrogen fluoride/ethylene glycol, prepared by mixing 2 ml of 49% HF in 250 ml ethylene glycol at 60°C, followed by dipping the thus treated wafers in AZ400T (Trademark) (a commercially available strip formulation) produced by Clariant at 80°C, each dip being conducted for 5 minutes."

B. From A. (A) above, the problems of the Amended Invention of the Application are recognized to be the following.

"to provide an antireflective hardmask layer for lithography with high etch selectivity yet sufficient resistance to multiple etchings."

C. As stated in A. (B), [0015] in the description of the Application describes a polymer with an Si-O backbone and a non-silicon inorganic unit incorporated into the backbone and the polymer has choices (a) to (c) for the repeating structural formula, the inorganic unit contained in the repeating structural formula has choices (a) to (f), there are many choices of metals for " M<sub>a</sub> " to " M<sub>e</sub> " contained in the inorganic unit, and therefore very many kinds of polymers are included. The polymer of Amended Invention of the Application corresponds to a polymer in which the repeating structural formula "(b)" is selected for the repeating structural formula of the polymer, the inorganic unit "(d)" is selected for the inorganic unit, and "Ti" is selected for " M<sub>d</sub> " in the formula "(d)."

D. As stated in A. (C) above, the polymers actually synthesized and confirmed for the effect of antireflective hardmask compositions containing the polymers stated in the description of the Application are, however, only the polymers of Examples 1 and 2, in which the repeating structural formula "(b)" is selected for the repeating structural formula and "TiO<sub>2</sub>" or "GeO<sub>2</sub>" is selected for the inorganic unit.

E. Although it is common general knowledge for those skilled in the art that physical properties of a polymer largely vary depend on the structure of the repeating unit constituting the polymer, no concrete ground is found to be stated even referring to the whole description of the Application, nor it is recognized to be obvious that the polymers of Amended Invention of the Application, more specifically, polymers in which an inorganic unit of a polymer of Example 1 or 2 is replaced with an inorganic unit (d) wherein M<sub>d</sub> is Ti, have properties similar to those of the polymers of Example 1 or 2 for etching selectivity and etching resistance.

F. Accordingly, it is not admitted that the detailed explanation of the invention is described so that a person skilled in the art can understand that Amended Invention of the Application can solve the problem of Amended Invention of the Application that is "to provide an antireflective hardmask layer for lithography with high etch selectivity yet sufficient resistance to multiple etchings."

G. Thus, it cannot be said that Amended Invention of the Application is described in the detailed description of the invention. Therefore, the statement of the scope of claims of the Application does not comply with the requirement as provided in 36(6)(i) of the Patent Act and Amended Invention of the Application is not independently patentable at the time of the patent application.

(3) As to inventive step (Article 29(2) of the Patent Act)

A. Described matters in Cited Documents

(A) U.S. Patent Application Publication No. 2007/0015083 (hereinafter, referred to as "Citation 1"), a publication distributed before the date of the priority claim of the Application (hereinafter, referred to as "Priority Date"), cited as Cited Document 1 in reasons for refusal of the examiner's decision states the following on "ANTIREFLECTIVE COMPOSITION AND PROCESS OF MAKING ALITHOGRAPHIC STRUCTURE."

a "FIELD OF INVENTION

[0001] The invention relates to an antireflective composition used in lithography, and to a process of making a lithographic structure from the antireflective composition."

b "BACKGROUND OF THE INVENTION

[0002] In the process of making semiconductor devices, photoresists and antireflective materials are applied to a substrate. Photoresists are photosensitive films used to transfer an image to a substrate. A photoresist is formed on a substrate and then exposed to a radiation source through a photomask (reticle). Exposure to the radiation provides a photochemical transformation of the photoresist, thus transferring the pattern of the photomask to the photoresist. The photoresist is then developed to provide a relief image that permits selective processing of the substrate.

[0003] Photoresists are typically used in lithographic structures to create features such as vias, trenches, or a combination of the two, in a dielectric material. In such a process, the reflection of radiation during exposure of the photoresist can limit the resolution of the image patterned in the photoresist due to reflections from the material beneath the photoresist. Reflection of radiation from the substrate/photoresist interface can also produce variations in the radiation intensity during exposure, resulting in non-uniform linewidths. Also, unwanted scattering of radiation exposes regions of the photoresist not intended, which again results in linewidth variation. The amount of scattering and reflection will vary from one region of the substrate to another, resulting in further linewidth variation.

[0004] With recent trends towards high-density semiconductor devices, there is a movement in the industry to use low wavelength radiation sources into the deep ultraviolet light (300 nm or less) for imaging a photoresist, e.g., KrF excimer laser light (248 nm), ArF excimer laser light (193 nm), excimer laser light (157 nm), electron beams, and soft x-rays. However, the use of low wavelength radiation often results in increased reflections from the upper resist surface as well as the surface of the underlying substrate.

[0005] Substrate reflections at ultraviolet and deep ultraviolet wavelengths are notorious for producing standing wave effects and resist notching, which severely limit critical dimension (CD) control. Notching results from substrate topography and non-uniform substrate reflectivity which causes local variations in exposure energy on the resist. Standing waves are thin film interference or periodic variations of light intensity through the resist thickness. These light variations are introduced because planarization of the resist presents a different thickness through the underlying topography. Thin film interference plays a dominant role in CD control of single material photoresist processes, causing large changes in the effective exposure dose due to a tiny change in the optical phase. Thin film interference effects are described in "Optimization of optical properties of resist processes" (T. Brunner, SPIE 10 Proceedings Vol. 1466, 1991, 297).

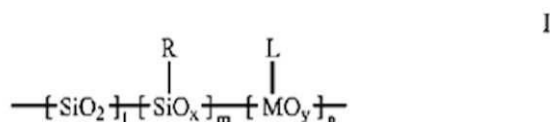
[0006] Bottom anti-reflective coatings (BARCs) have been used with photoresists to reduce thin film interference with some success. However, these relatively thin absorbing BARCs have fundamental limitations. At times, the photoresist does not provide sufficient resistance to subsequent etching steps to enable effective transfer of the desired pattern to a material, e.g., a dielectric, beneath the photoresist. The photoresist is consumed after transferring the pattern into the underlying BARC and substrates. In addition, the trend to smaller sub-90 nm node feature sizes requires the

use of relatively thin photoresists (>200 nm) to avoid image collapse. If a substantial etching depth is required, or if it is desired to use certain etchants for a given underlying material, the photoresist thickness is now insufficient to complete the etch process. Consequently, the photoresist does not effectively transfer the desired pattern into the underlying substrate or antireflective material.

[0007] The present trend to 248 nm, 193 nm, and 157 nm lithography and the demand for sub-200 nm features requires development of new processing schemes. To accomplish this, tools with higher numerical aperture (NA) are emerging. The higher NA allows for improved resolution but reduces the depth of focus of aerial images projected onto the photoresist. Because of the reduced depth of focus, a thinner photoresist is required. However, as the thickness of the photoresist is decreased, the photoresist becomes less effective as a mask for subsequent dry etch image transfer to the underlying substrate. Without significant improvement in the etch resistance exhibited by current single material photoresists, these systems cannot provide the necessary etch characteristics for high resolution lithography."

c "SUMMARY OF THE INVENTION

[0008] The invention is directed to a composition comprising a polymer of the following formula I,



[0009] wherein  $1 \leq x \leq 2$ ;  $1 \leq y \leq 5$ ;  $l \geq 0$ ;  $m > 0$ ;  $n > 0$ ; R is a chromophore, M is a metal selected from Group IIIB to Group VIB, lanthanides, Group IIIA, Group IVA except silicon; and L is an optional ligand.

[0010] The composition is typically used to form an antireflective material used in lithography. In particular, the antireflective material is used to pattern an underlying substrate, particularly, in high resolution lithography using low wavelength radiation sources into the deep ultraviolet to soft x-rays."

d "[0016] The - MO<sub>y</sub> - polymeric unit can be any one of many different metal-oxide forms. An exemplary list of such metal-oxide forms for a particular metal is as follows:

[0017] 1. - MO<sub>3</sub>-; wherein M is Sc, Y, lanthanide, and Group IIIA; B, Al, Ga, or In.

[0018] 2. - MO<sub>4</sub>-; wherein M is Group IVB; Ti, Zr or Hf, and Group IIIA; Sn or Ge.

[0019] 3. - MO<sub>5</sub>-; wherein M is Group IVB; V, Nb or Ta; or P. The Group VB metals are also known to form stable metal oxo forms, LMO<sub>3</sub>, wherein L is an oxo.

[0020] 4. -LMO-; many of the listed metals form stable acetoacetato-metal complexes.

[0021] 5. -LMO-; many of the listed metals form stable cyclopentadienyl-metal complexes.

[0022] 6. -LMO-; wherein L is an alkoxy ligand; M is Sc, Y, or lanthanide, Group IVB, and Group VB

[0023] 7. -LMO-; wherein L is an alkyl or phenyl ligand; M is Group IIIA or Group IVA."

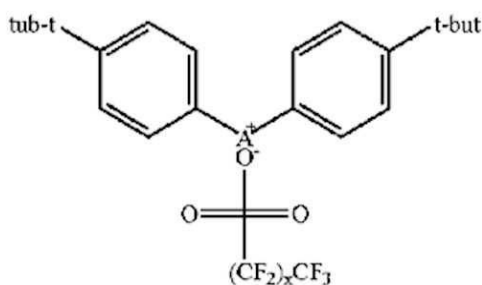
e "[0040] The antireflective compositions will likely contain an acid generator, which is



used to catalyze the crosslinking of the polymer. The acid generator can be a compound that liberates acid upon thermal treatment. A listing of known thermal acid generators includes 2,4,4,6-tetrabromocyclohexadienone, benzoin tosylate, 2-nitrobenzyl tosylate, and other alkyl esters of organic sulfonic acids. Compounds that generate a sulfonic acid upon activation are generally suitable. Other suitable thermally activated acid generators are described in U.S. Pat. Nos. 5,886,102 and 5,939,236; the disclosures of these two patents as related to the thermally activated, acid generating compounds are incorporated herein by reference.

[0041] If desired, a radiation-sensitive acid generator can be used as an alternative to a thermally activated acid generator or in combination with a thermally activated acid generator. Examples of suitable radiation-sensitive acid generators are described in U.S. Pat. Nos. 5,886,102 and 5,939,236, the disclosures of these two patents as related to radiation sensitive, acid generating compounds are incorporated herein by reference. Other radiation-sensitive acid generators known in the resist art can be used so long as they are compatible with the other components of the antireflective composition.

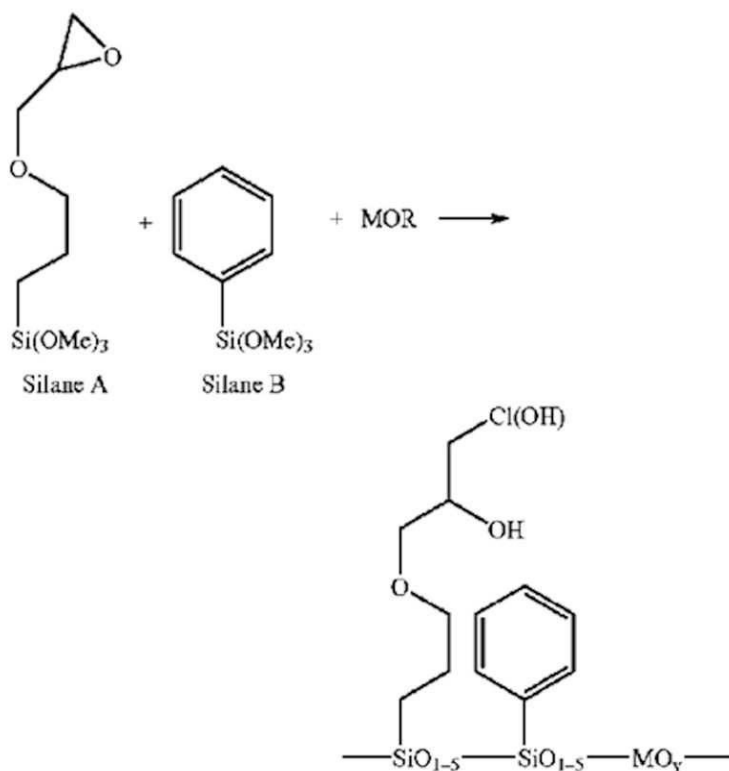
[0042] An acid generator of the formula below provides an antireflective material with optimal characteristics.



[0043] wherein A is S or I, and x is 0 to 7."

f "EXAMPLE 1

[0071]



#### EXAMPLE 1A

[0072] Silane A (0.381 g, 0.45 mol %), Silane B (0.069 g, 0.1 mol %), and  $\text{Ti}(\text{On-Bu})_4$  (0.549 g, 0.45 mol %) were added to a reaction vial with Dowanol PM (Aldrich, 4 g). The solution mixture was stirred at room temperature for about 5 min, followed by the dropwise addition of 1N HCl (1 g). The resulting clear solution was stirred at room temperature for 1 hr, then filtered through a 0.2 micron membrane. The filtered solution was applied to a silicon wafer spun at 3000 rpm for 60 sec. The deposited composition was heated at 215°C for 2 min.

[0073] The optical constants of the resulting silicon-metal oxide, antireflective material were measured using an n&k analyzer. Index of refraction (n)=1.735 and extinction coefficient (k)=0.301 at 193 nm. The material had a thickness of 233 nm.

....

#### EXAMPLE 2

[0078] Lithography (193 nm):

[0079] The silicon-metal oxide, antireflective material described in Example 1A was used for lithography. A material of acrylic-based photoresist, a product of JSR microelectronics, was spun-coated over the silicon-metal oxide, antireflective material to a thickness of about 250 nm. The radiation-sensitive imaging material was baked at 130°C. for 60 seconds. The image was then developed using a commercial developer (0.26M TMAH). The resulting pattern showed 120 nm contact holes and 113.75 nm lines and 104 nm space patterns.

[0080] Pattern Transfer by Reactive Ion Etching:

[0081] The contact hole and lines and space pattern formed on the silicon-metal oxide, antireflective material were transferred into silicon oxide by a 20 second fluorocarbon-

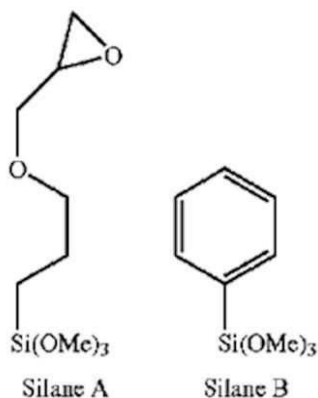
based etch using a LAM RIE tool. The observed etch selectivity of antireflective hardmask composition to the resist was approximately 2:1. The etch rate of blanket films from example 1A in CHF<sub>3</sub>/CH<sub>4</sub> plasma was 613 Å/min (silicon oxide etch rate 2000 Å/min), in chlorine 813 Å/min, and in N<sub>2</sub>/H<sub>2</sub> 62 Å/min.

[0082] Stripping the Silicon-Metal Oxide, Antireflective Materials by a Wet Strip:

[0083] The silicon-metal oxide, antireflective materials described in Examples 1A and 1B were stripped in HF/ethylene glycol (2 ml 49% HF in 250 mL ethylene glycol) at 60°C and AZ400T, which is commercially available from Clariant, at 80°C. in 5 min dip."

g. From a to f above, Citation 1 is recognized to describe the following invention.

"An antireflective hardmask composition comprising a silicon-metal oxide, obtained by adding the following Silane A (0.381 g, 0.45 mol %), the following Silane B (0.069 g, 0.1 mol %), and Ti(On-Bu)<sub>4</sub> (0.549 g, 0.45 mol %) to a reaction vial with Dowanol PM (Aldrich, 4 g), stirring the solution mixture at room temperature for about 5 min., followed by the dropwise addition of 1N HCl (1 g), stirring the resulting clear solution at room temperature for 1 hr, and then filtering the solution through a 0.2 micron membrane.



" (hereinafter, referred to as "Cited Invention 1.")

(B) Japanese Unexamined Patent Application Publication No. 2009-126940 (hereinafter, referred to as "Citation 2."), a publication distributed in Japan before the Priority Date of the Application, cited as Cited Document 2 in reasons for refusal of the examiner's decision states the following on "Metal oxide-containing film-forming composition, metal oxide-containing film, metal oxide-containing film-bearing substrate, and patterning method using the same" (title of invention).

a "Technical Field

[0001]

This invention relates to a metal oxide-containing film-forming composition suitable for forming a metal oxide-containing film for use as an intermediate layer in a multilayer resist process which is used in micropatterning in the manufacturing process of semiconductor devices and the like, especially for forming such a metal oxide-containing film by spin coating; a metal oxide-containing film formed therefrom; a metal oxide-containing film-bearing substrate; and a patterning method using the same.

## Background Art

[0002]

In the drive for higher integration and operating speeds in LSI devices, the pattern feature size is made drastically finer. Under the miniaturizing trend, lithography has achieved formation of finer patterns by using a light source with a shorter wavelength and by a choice of a proper resist composition for the shorter wavelength. Predominant among others are positive photoresist compositions which are used as a single layer. These single-layer positive photoresist compositions are based on resist resins possessing a framework having etching resistance to dry etching with chlorine or fluorine gas plasma and provided with a resist mechanism such that exposed areas become dissolvable. The resist composition is coated on a substrate to be processed and exposed to a pattern of light, after which the exposed areas of the resist coating are dissolved to form a pattern. Then, the substrate can be processed by dry etching with the remaining resist pattern serving as an etching mask.

[0003]

In an attempt to achieve a finer feature size; i.e., to reduce the pattern width with the thickness of a photoresist coating kept unchanged, the photoresist coating becomes low in resolution performance. If the photoresist coating is developed with a liquid developer to form a pattern, the so-called "aspect ratio" becomes too high, resulting in pattern collapse. For this reason, the miniaturization is accompanied by a thickness reduction of the photoresist coating.

[0004]

On the other hand, a method commonly used for the processing of a processable substrate is by processing a substrate by dry etching with the patterned photoresist film made an etching mask. Since a dry etching method capable of establishing a full etching selectivity between the photoresist film and the processable substrate is not available in practice, the resist film is also damaged during substrate processing. That is, the resist film breaks down during substrate processing, failing to transfer the resist pattern to the processable substrate faithfully. As the pattern feature size is reduced, resist materials are required to have higher resistance to dry etching.

With the progress of the exposure wavelength toward a shorter wavelength, the resin to be used for resist compositions is required to have less light absorption at the exposure wavelength. In response to changes from i-line to KrF and to ArF, the resin has made a transition to novolac resins, polyhydroxystyrene, and aliphatic polycyclic skeleton resins. Actually, the etching rate under the above-indicated dry etching conditions has been accelerated. Advanced photoresist compositions featuring a high resolution tend to be rather low in etching resistance.

This suggests the inevitability that a processable substrate is dry etched through a thinner photoresist coating having weaker etching resistance. It is urgently required to have a material and process suited for this processing stage.

[0005]

One solution to these problems is a multilayer resist process. The process involves forming an intermediate film on a processable substrate, and forming a photoresist film (resist overcoat film) thereon, wherein the intermediate film with etching selectivity different from that of the resist overcoat film intervenes between the resist overcoat film and the processable substrate, patterning the resist overcoat film,

then dry etching the intermediate film through the overcoat resist pattern as a dry etching mask for thereby transferring the pattern to the intermediate film, and dry etching the processable substrate through the intermediate film pattern as a dry etching mask for thereby transfer the pattern to the processable substrate.

... (omission) ...

[0008]

Also included in the multilayer resist process is a tri-layer resist process which can use general resist compositions as used in the single-layer resist process. In the tri-layer resist process, for example, an organic film of novolac resin or the like is formed on a processable substrate as a resist undercoat film, a silicon-containing film is formed thereon as a resist intermediate film, and an ordinary organic photoresist film is formed thereon as a resist overcoat film. On dry etching with a fluorine gas plasma, the resist overcoat film of organic nature provides a satisfactory etching selectivity ratio relative to the silicon-containing resist intermediate film. Then, the resist pattern is transferred to the silicon-containing resist intermediate film by dry etching with a fluorine gas plasma. With this process, even on use of a resist composition which is difficult to form a pattern having a sufficient thickness to allow for direct processing of a processable substrate, or a resist composition which has insufficient dry etching resistance to allow for substrate processing, a pattern of novolac film having sufficient dry etching resistance to allow for substrate processing is obtainable like the bilayer resist process, so long as the pattern can be transferred to the silicon-containing film.

... (omission) ...

[0011]

Such silicon-containing films conventionally used in the multilayer resist process suffer from several problems. For example, as is well known in the art, where an attempt is made to form a resist pattern by photolithography, exposure light is reflected by the substrate and interferes with the incident light, incurring the problem of so-called standing waves. To produce a microscopic pattern of a resist film without edge roughness, an antireflective coating must be provided as an intermediate layer. Reflection control is essential particularly under high-NA exposure conditions of the advanced lithography.

[0012]

In the multilayer resist process, especially the process of forming a silicon-containing film as an intermediate layer by CVD, it becomes necessary for reflection control purposes to provide an organic antireflective coating between the resist overcoat film and the silicon-containing intermediate film. However, the provision of the organic antireflective coating entails the necessity that the organic antireflective coating be patterned by using the resist overcoat film as a dry etching mask. That is, the antireflective coating is dry etched by using the resist overcoat film as a mask, before which the process proceeds to processing of the silicon-containing intermediate layer. Then the overcoat photoresist must bear an additional load of dry etching corresponding to the processing of the antireflective coating. While photoresist films used in the advanced lithography become thinner, this dry etching load is not negligible. Therefore, greater attention is paid to the tri-layer resist process in which a light-absorbing silicon-containing film not creating such an etching load is applied as an intermediate film.

[0013]

Known light-absorbing silicon-containing intermediate films include light-

absorbing silicon-containing films of spin coating type. For example, Patent Document 8 (Japanese Unexamined Patent Application Publication No. 2005-15779) discloses the provision of an aromatic structure as the light-absorbing structure. Since the aromatic ring structure capable of effective light absorption acts to reduce the rate of dry etching with a fluorine gas plasma, this approach is disadvantageous for the purpose of dry etching the intermediate film without an additional load to the photoresist film. Since it is thus undesirable to incorporate a large amount of such light-absorbing substituent groups, the amount of incorporation must be limited to the minimum.

[0014]

Further, the dry etching rate of the resist undercoat film during reactive dry etching with an oxygen gas plasma as commonly used in the processing of the resist undercoat film by using the intermediate film as a dry etching mask is preferably low so as to increase the etching selectivity ratio between the intermediate film and the undercoat film. To this end, the intermediate film is desired to have a higher content of silicon which is highly reactive with fluorine etchant gas. The requirement arising from the conditions of processing both the overcoat photoresist film and the undercoat organic film gives preference to an intermediate film having a higher content of silicon which is highly reactive with fluorine gas.

[0015]

In actual silicon-containing intermediate film-forming compositions of spin coating type, however, organic substituent groups are incorporated into the silicon-containing compounds so that the silicon-containing compounds may be dissolvable in organic solvents. Of the silicon-containing intermediate films known in the art, an SOG film-forming composition adapted for KrF excimer laser lithography is disclosed in Non-Patent Document 1 (J. Appl. Polym. Sci., Vol. 88, 636-640 (2003)). However, since light-absorbing groups in this composition are described nowhere, it is believed that this composition forms a silicon-containing film without an antireflective function. This film fails to suppress reflection during exposure by the lithography using the advanced high-NA exposure system. It would be impossible to produce microscopic pattern features.

[0016]

In the advanced semiconductor process using such a high-NA exposure system, the photoresist film has seen a more outstanding reduction in thickness. Then in etching the intermediate film using a photoresist as an etching mask, an attempt to increase the silicon content while possessing an antireflection function, as such, is expected to encounter difficulty in facilitating pattern transfer to the silicon-containing intermediate film. There is a demand for a material having a higher etching rate.

[0017]

In addition to the requirements for the dry etching properties and antireflection effect, the composition for forming an intermediate film with a high silicon content suffers from several problems, of which shelf stability is the most outstanding. Shelf stability relates to the phenomenon that a silicon-containing compound in the composition changes its molecular weight during shelf storage as a result of condensation of silanol groups on the silicon-containing compound. Such molecular weight changes show up as film thickness variations and lithography performance variations. In particular, the lithography performance is sensitive, and so, even when the condensation of silanol groups within the molecule takes place merely to such an

extent that it does not show up as a film thickness buildup or molecular weight change, it can be observed as variations of microscopic pattern features.

[0018]

As is known in the art, such highly reactive silanol groups can be rendered relatively stable if they are kept in acidic conditions. See Non-Patent Document 2 (C. J. Brinker and G. W. Scherer, "Sol-Gel Science: The Physics and Chemistry of Sol-Gel Processing," Academic Press, San Diego (1990)). Further, addition of water improves shelf stability as disclosed in Non-Patent Document 1 (J. Appl. Polym. Sci., Vol. 88, 636-640 (2003)), Patent Document 9 (Japanese Unexamined Patent Application Publication No. 2004-157469) and Patent Document 10 (Japanese Unexamined Patent Application Publication No. 2004-191386). However, practically, the condensation reaction of silanol groups is not inhibited completely in the silicon-containing compounds prepared by the methods of these patent publications even when such measures are taken. The silicon-containing compound in the composition slowly alters over time, and a silicon-containing film formed from such an altered composition changes in nature. Then the composition must be held in a refrigerated or frozen state until just before use, and on use, must be brought to a use temperature (typically 23°C) and consumed quickly.

... (omission) ...

#### Disclosure of the Invention

##### Problems to be Solved by the Invention

[0019]

Objects of the present invention are to provide a metal oxide-containing film-forming composition in which (1) the metal oxide-containing film has a light-absorbing capability to allow for satisfactory pattern formation even under high-NA exposure conditions in case of disposing a metal oxide-containing film on an organic film, disposing a photoresist film thereon, and forming a resist pattern, (2) the metal oxide-containing film serves as a satisfactory dry etching mask between the overlying photoresist film and the underlying organic film of the metal oxide-containing film, and (3) the composition is fully shelf stable; a metal oxide-containing film formed from the composition; a substrate having the metal oxide-containing film disposed thereon; and a patterning method."

##### b. " Means for Solving the Problems

[0020]

The inventors have studied the lithographic properties and stability of a metal oxide-containing intermediate film-forming composition and found that combining a metal oxide-containing compound obtained through hydrolytic condensation of hydrolyzable silicon compounds with components (B), (C), and (D) defined below provides

- (1) a metal oxide-containing film that can suppress reflection under high-NA exposure conditions of either dry or immersion lithography technique when light-absorbing groups described below are incorporated;
- (2) the metal oxide-containing film having a sufficient etching selectivity ratio to serve as a dry etching mask; and
- (3) a metal oxide-containing film-forming composition that is fully shelf stable so that

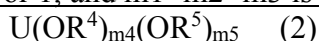
its lithography performance undergoes little or no change over time, thereby completing the present invention.

[0021]

Accordingly, the present invention provides a heat curable metal oxide-containing film-forming composition comprising: (A) a metal oxide-containing compound obtained through hydrolytic condensation between one or more hydrolyzable silicon compounds represented by the general formula (1), and one or more hydrolyzable metal compounds represented by the general formula (2):



wherein R is an alkyl having 1 to 6 carbon atoms, R<sup>1</sup>, R<sup>2</sup> and R<sup>3</sup> are each a hydrogen atom or a monovalent organic group having 1 to 30 carbon atoms, m<sub>1</sub>, m<sub>2</sub> and m<sub>3</sub> are each 0 or 1, and m<sub>1</sub>+m<sub>2</sub>+m<sub>3</sub> is an integer of 0 to 3;



wherein R<sup>4</sup> and R<sup>5</sup> are each an organic group having 1 to 30 carbon atoms; m<sub>4</sub>+m<sub>5</sub> is the valence dependent on U; m<sub>4</sub> and m<sub>5</sub> are each an integer equal to or greater than 0; and U is an element selected from the elements of Groups III, IV, and V in the Periodic Table, excluding silicon;

(B) at least one compound represented by the general formula (3) or (4):



wherein L is lithium, sodium, potassium, rubidium, or cesium, X is a hydroxyl group or a one or more valent organic acid group having 1 to 30 carbon atoms, "a" is an integer equal to or greater than 1, "b" is 0 or an integer equal to or greater than 1, and a + b is equal to the valence of hydroxyl group or organic acid group;



wherein M is sulfonium, iodonium, or ammonium, A is X described above or a non-nucleophilic counter ion, "a" is an integer equal to or greater than 1, "b" is 0 or an integer equal to or greater than 1, and a + b is equal to the valence of hydroxyl group or organic acid group;

(C) a mono or multi valent organic acid having 1 to 30 carbon atoms, and

(D) an organic solvent (Claim 1)."

c. "[0088]

Examples of compounds represented by the formula (2) in which U is titanium include titanium methoxide, titanium ethoxide, titanium propoxide, titanium butoxide, titanium amyloxy, titanium hexyloxy, titanium cyclopentoxide, titanium cyclohexyloxy, titanium allyloxy, titanium phenoxide, titanium methoxyethoxide, titanium ethoxyethoxide, titanium dipropoxyethylacetoacetate, titanium dibutoxybisethylacetoacetate, titanium dipropoxide bis(2,4-pantandionate), titanium dibutoxy bis(2,4-pentanedionate), and the like as monomers."

d. "[0160]

In the present invention, a photoacid generator may be used. Examples of the photoacid generator which can be used in the present invention include:

(A-I) onium salts of the formula (P1a-1), (P1a-2) or (P1b) below,

(A-II) diazomethane derivatives of the formula (P2) below,

(A-III) glyoxime derivatives of the formula (P3) below,

(A-IV) bissulfone derivatives of the formula (P4) below,



(A-V) sulfonic acid esters of N-hydroxyimide compounds of the formula (P5) below,  
(A-VI)  $\beta$ -ketosulfonic acid derivatives,  
(A-VII) disulfone derivatives,  
(A-VIII) nitrobenzylsulfonate derivatives, and  
(A-IX) sulfonate derivatives.  
... (omission) ...

[0178]

Among these, in particular, there are preferably used onium salts such as triphenylsulfonium trifluoromethanesulfonate, (p-tert-butoxyphenyl)diphenylsulfonium trifluoromethanesulfonate, tris(p-tert-butoxyphenyl)sulfonium trifluoromethanesulfonate, triphenylsulfonium p-toluenesulfonate, (p-tert-butoxyphenyl)diphenylsulfonium p-toluenesulfonate, tris(p-tert-butoxyphenyl)sulfonium p-toluenesulfonate, trinaphthylsulfonium trifluoromethanesulfonate, cyclohexylmethyl(2-oxocyclohexyl)sulfonium trifluoromethanesulfonate, (2-norbornyl)methyl(2-oxocyclohexyl)sulfonium trifluoroethanesulfonate, and 1,2'-naphthylcarbonylmethyltetrahydrothiophenium triflate; diazomethane derivatives such as bis(benzenesulfonyl)diazomethane, bis(p-toluenesulfonyl)diazomethane, bis(cyclohexylsulfonyl)diazomethane, bis(n-butylsulfonyl)diazomethane, bis(isobutylsulfonyl)diazomethane, bis(sec-butylsulfonyl)diazomethane, bis(n-propylsulfonyl)diazomethane, bis(isopropylsulfonyl)diazomethane, and bis(tert-butylsulfonyl)diazomethane; glyoxime derivatives such as bis-O-(p-toluenesulfonyl)- $\alpha$ -dimethylglyoxime and bis-O-(n-butanefulfonyl)- $\alpha$ -dimethylglyoxime; bissulfone derivatives such as bisnaphthylsulfonylmethane; and sulfonic acid ester derivatives of N-hydroxyimide compounds such as N-hydroxysuccinimide methanesulfonate, N-hydroxysuccinimide trifluoromethanesulfonate, N-hydroxysuccinimide 1-propanesulfonate, N-hydroxysuccinimide 2-propanesulfonate, N-hydroxysuccinimide 1-pentanesulfonate, N-hydroxysuccinimide p-toluenesulfonate, N-hydroxynaphthalimide methanesulfonate, and N-hydroxynaphthalimide benzenesulfonate."

e. " Examples

[0200]

Synthesis Examples and Examples are given below together with Comparative Examples for further illustrating the invention, although the invention is not limited by such descriptions. All percents are by weight. The molecular weight (Mw) is determined by gel permeation chromatography (GPC).

... (omission) ...

[0202]

Synthetic Example 2

A mixture of 10 g of phenyltrimethoxysilane, 20 g of 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane, 10 g of titanium tetrabutoxide, 20 g of 2,4-pentanedione, and 35 g of propylene glycol methyl ether was added to a mixture of 40 g of propylene glycol methyl ether, 1 g of methanesulfonic acid, and 50 g of deionized water. The mixture was held at 30°C for 12 hours while hydrolytic condensation took place. Then the by-product alcohol was distilled off under a reduced pressure. To the remaining solution, 200 ml of propylene glycol methyl ether was added. Further concentration under a reduced pressure yielded 120 g of a propylene glycol methyl ether

solution of metal oxide-containing compound 2 (polymer concentration 20%). The molecular weight was determined versus polystyrene standards and the product was found to have a Mw of 8,000.

... (omission) ...

[0210]

#### Examples and Comparative Examples

Metal oxide-containing film-forming composition solutions were respectively prepared by dissolving metal oxide-containing compounds 1 to 6, silicon-containing compounds 7 and 8, acids, thermal crosslink accelerators, and additives in a solvent at ratios shown in Table 1, and filtering through a fluoroplastic filter having a pore size of 0.1  $\mu\text{m}$ . These solutions are designated as Sol. 1 to 18 respectively.

[0211]

[Table 1]

金属酸化物含有膜形成用組成物

	No.	金属酸化物含有化合物、ケイ素含有化合物 (質量部)	熱架橋促進剤 (質量部)	酸 (質量部)	溶剤 (質量部)	水/安定剤 (質量部)	その他添加物 (質量部)
実施例 1	Sol.1	化合物1(4.0)	TPSOAc (0.04)	マレイン酸 (0.04)	プロピレングリコールメチルエーテル (100)	なし	なし
実施例 2	Sol.2	化合物1(4.0)	TPSOH (0.04)	シュウ酸 (0.04)	プロピレングリコールメチルエーテル (100)	水(10)	なし
実施例 3	Sol.3	化合物1(4.0)	TPSCI (0.04) TPSOAc (0.003)	マレイン酸 (0.04)	プロピレングリコールメチルエーテル (100)	水(10)	TPSNf (0.02)
実施例 4	Sol.4	化合物1(4.0)	TPSMA (0.04) TMAOAc (0.003)	マレイン酸 (0.04) シュウ酸 (0.04)	プロピレングリコールメチルエーテル (100)	水(10) 安定剤1(5)	なし
実施例 5	Sol.5	化合物1(4.0) 化合物7(0.4)	TPSN (0.04)	マレイン酸 (0.04) シュウ酸 (0.04)	プロピレングリコールメチルエーテル (100)	水(10) 安定剤1(5)	なし
実施例 6	Sol.6	化合物1(4.0) 化合物8(0.4)	TPSMA (0.04)	マレイン酸 (0.04)	プロピレングリコールメチルエーテル (100)	安定剤1(5)	なし
実施例 7	Sol.7	化合物1(3.2) 化合物7(0.4) 化合物8(0.4)	TPSOAc (0.04)	フマル酸 (0.04)	プロピレングリコールメチルエーテル (100)	安定剤1(5)	なし
実施例 8	Sol.8	化合物2(4.0)	TPSOAc (0.04)	マレイン酸 (0.04)	プロピレングリコールメチルエーテル (100)	水(10) 安定剤2(5)	なし
実施例 9	Sol.9	化合物2(4.0)	TPSMA (0.04)	マレイン酸 (0.04)	プロピレングリコールメチルエーテル (100)	水(10) 安定剤2(5)	なし
実施例 10	Sol.10	化合物3 (4.0)	TPSOAc (0.04)	マレイン酸 (0.04)	プロピレングリコールエチルエーテル (100)	水(10) 安定剤2(5)	なし
実施例 11	Sol.11	化合物3(4.0)	TPSOH (0.04)	マレイン酸 (0.04)	プロピレングリコールエチルエーテル (100)	水(10) 安定剤3(5)	なし
実施例 12	Sol.12	化合物4(4.0)	TPSMA (0.04)	マレイン酸 (0.04)	プロピレングリコールプロピルエーテル (100)	水(10) 安定剤3(5)	なし
実施例 13	Sol.13	化合物4(4.0)	TPSOAc (0.04)	マレイン酸 (0.04)	プロピレングリコールプロピルエーテル (100)	水(10) 安定剤4(5)	なし
実施例 14	Sol.14	化合物5(4.0)	TPSMA (0.04)	マレイン酸 (0.04)	プロピレングリコールプロピルエーテル (100)	水(10) 安定剤4(5)	なし
実施例 15	Sol.15	化合物5(4.0)	TPSN (0.04)	マレイン酸 (0.04)	プロピレングリコールプロピルエーテル (100)	水(10) 安定剤5(5)	なし
比較例 1	Sol.16	化合物6(4.0)	TPSMA (0.04)	マレイン酸 (0.04)	プロピレングリコールプロピルエーテル (100)	水(10) 安定剤5(5)	なし
比較例 2	Sol.17	化合物1(4.0)	なし	マレイン酸 (0.04)	プロピレングリコールプロピルエーテル (100)	水(10) 安定剤5(5)	なし
比較例 3	Sol.18	化合物1(4.0)	TPSMA (0.04)	なし	プロピレングリコールプロピルエーテル (100)	水(10) 安定剤5(5)	なし

金属酸化物含有膜形成用組成物 Metal oxide-containing film-forming composition  
 金属酸化物含有化合物、ケイ素含有化合物 (質量部) Metal oxide-containing compound, Si-containing compound (pbw)  
 熱架橋促進剤 (質量部) Thermal crosslink accelerator (pbw)

酸 (質量部)	Acid (pbw)
溶剤 (質量部)	Solvent (pbw)
水/安定剤 (質量部)	Water/Stabilizer (pbw)
その他添加物 (質量部)	Other additives (pbw)

実施例	Example
比較例	Comparative Example
化合物	Compound
なし	None
マレイン酸	maleic acid
シュウ酸	oxalic acid
フマル酸	fumaric acid
プロピレングリコールメチルエーテル	Propylene glycol methyl ether
水	water
安定剤	Stabilizer

[0212]

TPSOAc: triphenylsulfonium acetate (pho-degradable thermal crosslink accelerator)

TPSOH: triphenylsulfonium hydroxide (pho-degradable thermal crosslink accelerator)

TPSCl: triphenylsulfonium chloride (pho-degradable thermal crosslink accelerator)

TPSMA: mono(triphenylsulfonium) maleate (pho-degradable thermal crosslink accelerator)

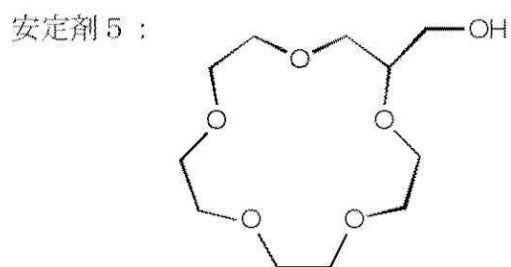
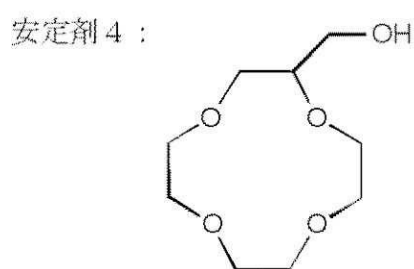
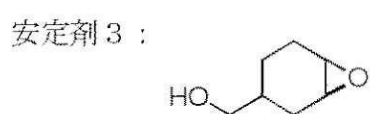
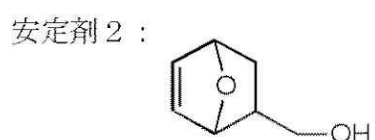
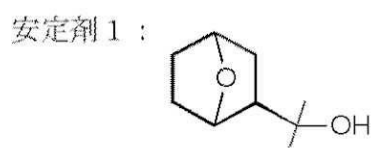
TPSN: triphenylsulfonium nitrate (pho-degradable thermal crosslink accelerator)

TMAOAc: tetramethylammonium acetate (non-pho-degradable thermal crosslink accelerator)

TPSNf: triphenylsulfonium nonafluorobutanesulfonate (photo acid generator)

[0213]

[Formula 17]



"

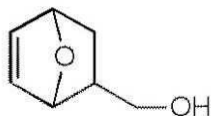
安定剂 1	Stabilizer 1
安定剂 2	Stabilizer 2
安定剂 3	Stabilizer 3
安定剂 4	Stabilizer 4
安定剂 5	Stabilizer 5

"

f. From [Table 1] above in Citation 2, it is found that Sol. 8, a solution of a metal oxide-containing film-forming composition, comprises metal oxide-containing compound 2, TPSOAc (triphenylsulfonium acetate), maleic acid, propylene glycol

methyl ether, water, and Stabilizer 2 represented by the following formula.

安定剂 2 :

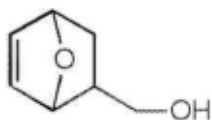


安定剂 2      Stabilizer 2

g. From a to f above, Citation 2 is found to describe the following invention.

"A solution of a metal oxide-containing film-forming composition comprising metal oxide-containing compound 2, TPSOAc (triphenylsulfonium acetate), maleic acid, propylene glycol methyl ether, and water, wherein a solution of the metal oxide-containing compound 2 in propylene glycol methyl ether is obtained by adding a mixture of 10 g of phenyltrimethoxysilane, 20 g of 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane, 10 g of titanium tetrabutoxide, 20 g of 2,4-pentanedione, and 35 g of propylene glycol methyl ether to a mixture of 40 g of propylene glycol methyl ether, 1 g of methanesulfonic acid, and 50 g of deionized water, holding the mixture at 30°C for 12 hours to allow hydrolytic condensation, then distilling the by-product alcohol off under a reduced pressure, adding 200 ml of propylene glycol methyl ether to the remaining solution, and, furthermore, concentrating it under reduced pressure.

Stabilizer 2:



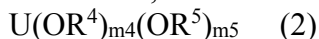
" (hereinafter, referred to as "Cited Invention 2.")

h. Moreover, from a to f above, Citation 2 is found to state the following technical matter:

"in a thermocurable metal oxide-containing film-forming composition comprising a metal oxide-containing compound obtained through hydrolytic condensation between one or more hydrolyzable silicon compounds represented by the general formula (1) and one or more hydrolyzable metal compounds represented by the general formula (2):



wherein R is an alkyl having 1 to 6 carbon atoms, R<sup>1</sup>, R<sup>2</sup> and R<sup>3</sup> are each a hydrogen atom or a monovalent organic group having 1 to 30 carbon atoms, m<sub>1</sub>, m<sub>2</sub>, and m<sub>3</sub> are each 0 or 1, and m<sub>1</sub>+m<sub>2</sub>+m<sub>3</sub> is an integer of 0 to 3;



wherein R<sup>4</sup> and R<sup>5</sup> are each an organic group having 1 to 30 carbon atoms; m<sub>4</sub>+m<sub>5</sub> is the valence dependent on U; m<sub>4</sub> and m<sub>5</sub> are each an integer equal to or greater than 0; and U is an element selected from the elements of Group III, IV and V in the Periodic Table, excluding silicon, examples of compounds represented by the formula (2) in which U is titanium include titanium butoxide, titanium dibutoxybisethylacetoacetate,

etc. as monomers."

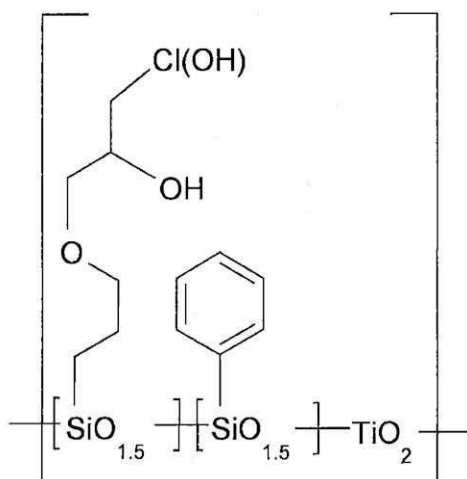
## B. Examination when the main Cited Invention is Cited Invention 1

### (A) Comparison

Cited Invention 1 is compared with Amended Invention of the Application.

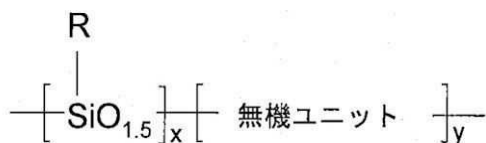
a. "An antireflective hardmask composition" in Cited Invention 1 corresponds to "an antireflective hardmask composition" in Amended Invention of the Application.

b. Since "a silicon-metal oxide" in Cited Invention 1 is made from the same materials of the same composition in the same method as the repeating structural unit of the antireflective hardmask composition layer of Example 1 described in [0058], [0059], [0063], and [0064] in the description of the Application, it is found to be "a polymer composed of the following repeating structural unit



" which is identical with the aforementioned repeating structural unit of the antireflective hardmask composition layer of Example 1. Referring to the statement "Suitable chromophore moieties include, but are not limited to, phenyl, chrysenes, pyrenes, fluoranthrenes, anthrones, benzophenones, thioxanthenes, and anthracenes" in [0018] and the statement "Examples of the transparent moiety, chromophore moiety, and crosslinking component attached to the Si-O unit suitable for this invention are further described in Patent Document 4 and Japanese Patent Application No. 2004-158639, filing date 5/28/2004; priority Japanese Patent Application No. 2003-157808 with priority date 6/3/2003 and Japanese Patent Application No. 2004-172222, filing date 6/10/2004 and references cited therein, the disclosures of which are incorporated herein by reference." in [0027] in the description of the Application, and taking it into consideration that a silicon containing compound composed of the repeating unit listed first from the left in the aforementioned repeating structural units is described as an example of silicon containing compounds containing a cross-linking monovalent organic group in [0023], [0024], and [0029] to [0034] in Japanese Unexamined Patent Application Publication No. 2005-352104, which is an unexamined patent application publication of Japanese Patent Application No. 2004-172222 cited in the description of the Application, "a silicon-metal oxide" in Cited Invention 1 is identical with "a

polymer composed of a repeating structural unit selected from the following group:  
[Formula 1]



無機ユニット            Inorganic Unit

" in terms that "wherein x is at least 1; y is at least 1; R is an organic functional unit selected from one or more chromophore moieties, transparent moieties, and crosslinking components."

c. From a and b above, Amended Invention of the Application and Cited Invention 1 are identical in terms that

"An antireflective hardmask composition comprising a polymer composed of a repeating structural unit selected from the following group:  
[Formula 1]



無機ユニット            Inorganic Unit

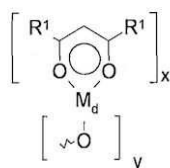
wherein x is at least 1; y is at least 1; R is an organic functional unit selected from one or more chromophore moieties, transparent moieties, and crosslinking components" and are different in the following points.

The different feature 1:

While the antireflective hardmask composition in Amended Invention of the Application comprises an acid generator, the antireflective hardmask composition in Cited Invention 1 does not comprise an acid generator.

The different feature 2:

While the "inorganic unit" in the formula of the aforementioned "repeating structural unit" in Amended Invention of the Application "has the following formula:  
[Formula 2]



wherein x and y have the meanings given above; M<sub>d</sub> is Ti; and R<sup>1</sup> is the same or



different and is selected from C<sub>1</sub>-C<sub>6</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkoxy, phenyl, and phenoxy," that in Cited Invention 1 is "TiO<sub>2</sub>."

(B) Judgment of difference

The aforementioned different feature 1 and different feature 2 will be judged.

a. The different feature 1

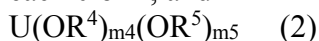
Since [0040] to [0043] in Citation 1 states that the antireflective compositions "will likely contain an acid generator, which is used to catalyze the cross-linking of the polymer," incorporating an acid generator into an antireflective hardmask composition in Cited Invention 1 is an idea that a person skilled in the art could conceive easily.

b. The different feature 2

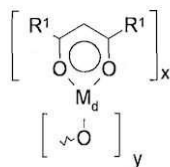
Citation 2 states, as stated in A. (B) h. above, the following technical matter: "in a thermocurable metal oxide-containing film-forming composition comprising a metal oxide-containing compound obtained through hydrolytic condensation between one or more hydrolyzable silicon compounds represented by the general formula (1) and one or more hydrolyzable metal compounds represented by the general formula (2):



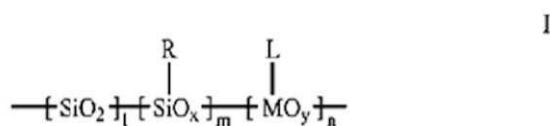
wherein R is an alkyl having 1 to 6 carbon atoms, R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> are each a hydrogen atom or a monovalent organic group having 1 to 30 carbon atoms, m<sub>1</sub>, m<sub>2</sub>, and m<sub>3</sub> are each 0 or 1, and m<sub>1</sub>+m<sub>2</sub>+m<sub>3</sub> is an integer of 0 to 3;



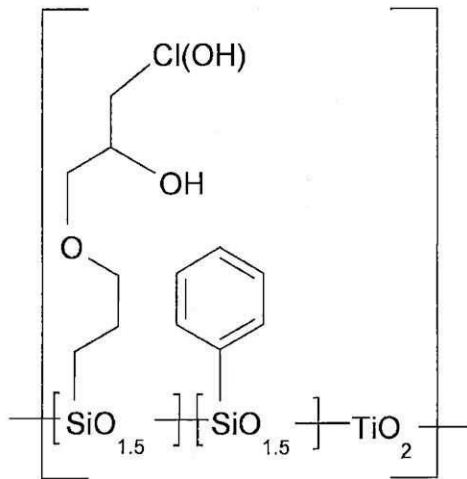
wherein R<sup>4</sup> and R<sup>5</sup> are each an organic group having 1 to 30 carbon atoms; m<sub>4</sub>+m<sub>5</sub> is the valence dependent on U; m<sub>4</sub> and m<sub>5</sub> are each an integer equal to or greater than 0; and U is an element selected from the elements of Groups III, IV, and V in the Periodic Table, excluding silicon, examples of compounds represented by the formula (2) in which U is titanium include titanium butoxide, titanium dibutoxybisethylacetoacetate, etc. as monomers." If "titanium butoxide" is used as the hydrolyzable metal compound represented by the aforementioned formula (2), then the metal oxide contained in the metal oxide-containing compound is "TiO<sub>2</sub>" and if "titanium dibutoxybisethylacetoacetate" is used as the hydrolyzable metal compound illustrated in the aforementioned formula (2), then the metal oxide contained in the metal oxide-containing compound is the "inorganic unit" "represented by the following formula:



wherein x is at least 1, y is at least 1, M<sub>d</sub> is Ti, and R<sup>1</sup> is different and is C<sub>1</sub> alkyl or C<sub>2</sub> alkoxy" in Amended Invention of the Application. [0008], [0009], [0016], and [0020] in Citation 1 states that in "a polymer of the following formula I



wherein  $1 \leq x \leq 2$ ,  $1 \leq y \leq 5$ ,  $l \geq 0$ ,  $m > 0$ ;  $n > 0$ ; R is a chromophore, M is a metal selected from Groups IIIB to Group VIB, lanthanides, Group IIIA, Group IVA except silicon; and L is an optional ligand" comprised in a composition, - MO<sub>y</sub> - polymer unit can be -LMO- and many metals stated form stable acetoacetate-metal complexes. As stated in B. (A) b above, the "silicon-metal oxide" in Cited Invention 1 is found to be "a polymer composed of the following repeating structural unit



" and correspond to "a polymer of formula I" stated in Citation 1 and also to the aforementioned "metal oxide-containing compound obtained through hydrolytic condensation between one or more hydrolyzable silicon compounds represented by the general formula (1) and one or more hydrolyzable metal compounds represented by the general formula (2)" stated in Citation 2. Therefore, applying the aforementioned technical matter stated in Citation 2 and using "titanium dibutoxybisethylacetoacetate" instead of "Ti(On-Bu)<sub>4</sub> (titanium butoxide)" to produce the configuration of Amended Invention of the Application relating to the aforementioned different feature 2 when synthesizing silicon-metal oxide in Cited Invention 1 is a matter that the statement in [0008], [0009], [0016], and [0020] in Citation 1 suggests and an idea that a person skilled in the art could conceive easily.

The polymer contained in the antireflective hardmask composition of Amended Invention of the Application is not actually synthesized in the description of the Application, and effects of Amended Invention of the Application are unknown. Amended Invention of the Application cannot be recognized to have an advantageous function and effect that a person skilled in the art cannot foresee.

### (C) Summary

As described above, Amended Invention of the Application could be easily made by a person skilled in the art based on the described matters in Cited Invention 1 and Citation 2. Thus, the appellant should not be granted a patent for it independently at the time of patent application under the provisions of Article 29(2) of the Patent Act.

### C. Examination when the main Cited Invention is Cited Invention 2

#### (A) Comparison

Cited Invention 2 is compared with Amended Invention of the Application.

a. [0020] in Citation 2 states "The inventors have studied the lithographic properties and stability of a metal oxide-containing intermediate film-forming composition and found that combining a metal oxide-containing compound obtained through hydrolytic condensation of hydrolyzable silicon compounds with components (B), (C), and (D) defined below provides

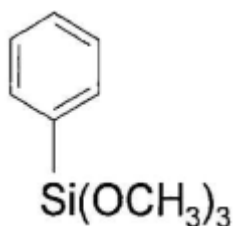
(1) a metal oxide-containing film that can suppress reflection under high-NA exposure conditions of either dry or immersion lithography technique when light-absorbing groups described below are incorporated;

(2) the metal oxide-containing film having a sufficient etching selectivity ratio to serve as a dry etching mask; and

... (omission) ...

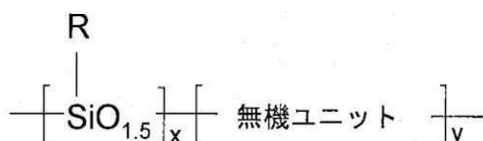
thereby completing the present invention." and the metal oxide-containing membrane formed from "a solution of a metal oxide-containing film-forming composition" in Cited Invention 2 is found to function as an antireflective hardmask. Therefore, "a solution of a metal oxide-containing film-forming composition" in Cited Invention 2 corresponds to "an antireflective hardmask composition" in Amended Invention of the Application.

b. "A propylene glycol methyl ether solution of metal oxide-containing compound 2" in Cited Invention 2 is obtained by "adding a mixture of 10 g of phenyltrimethoxysilane, 20 g of 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane, 10 g of titanium tetrabutoxide, 20 g of 2,4-pentanedione, and 35 g of propylene glycol methyl ether to a mixture of 40 g of propylene glycol methyl ether, 1 g of methanesulfonic acid, and 50 g of deionized water, holding the mixture at 30°C for 12 hours to allow hydrolytic condensation, then distilling the by-product alcohol off under reduced pressure, adding 200 ml of propylene glycol methyl ether to the remaining solution, and, furthermore, concentrating it under reduced pressure" and the "metal oxide-containing compound 2" is found to be a compound obtained through hydrolytic condensation of "phenyltrimethoxysilane," which is identical with "silane precursor B" stated in [0058] in the description of the Application, which is a compound having the following structural formula:



," "2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane," and "tetrabutoxytitanium," which is identical with "Ti (OC<sub>4</sub>H<sub>9</sub>)<sub>4</sub>" stated in [0058] in the description of the Application. Referring to the statement "Suitable chromophore moieties include, but are not limited to, phenyl, chrysenes, pyrenes, fluoranthrenes, anthracenes, benzophenones, thioxanthenes, and anthracenes" in [0018] and the statement "Examples of the transparent moiety, chromophore moiety, and crosslinking component attached to the Si-O unit suitable for this invention are further described in Patent Document 4 and Japanese Patent Application No. 2004-158639, filing date 5/28/2004; priority Japanese Patent Application No. 2003-157808 with priority date 6/3/2003 and Japanese Patent

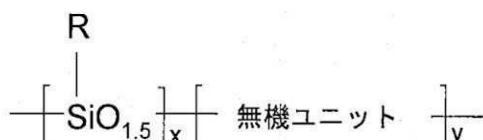
Application No. 2004-172222, filing date 6/10/2004 and references cited therein, the disclosures of which are incorporated herein by reference." in [0027] in the description of the Application, and taking it into consideration that "2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane" is described as an example of silicon containing compounds containing a cross-linking monovalent organic group in [0023], [0024], and [0029] to [0034] in Japanese Unexamined Patent Application Publication No. 2005-352104, which is an unexamined patent application publication of Japanese Patent Application No. 2004-172222 cited in the description of the Application, "metal oxide-containing compound 2" in Cited Invention 2 is identical with "a polymer composed of a repeating structural unit selected from the following group [Formula 1]



無機ユニット            Inorganic Unit

" in Amended Invention of the Application in terms that "wherein x is at least 1; y is at least 1; R is an organic functional unit selected from one or more chromophore moieties, transparent moieties, and crosslinking components."

c. From a and b above, Amended Invention of the Application and Cited Invention 2 are identical in terms of "an antireflective hardmask composition comprising a polymer composed of a repeating structural unit selected from the following group. [Formula 1]



無機ユニット            Inorganic Unit

wherein x is at least 1; y is at least 1; R is an organic functional unit selected from one or more chromophore moieties, transparent moieties, and crosslinking components" and differ in the following points.

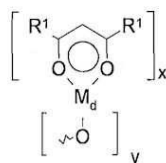
The different feature 1:

While the antireflective hardmask composition in Amended Invention of the Application comprises an acid generator, the antireflective hardmask composition in Cited Invention 2 does not comprise an acid generator.

The different feature 2:

While the "inorganic unit" in the formula of the aforementioned "repeating

structural unit" in Amended Invention of the Application "has the following formula:  
[Formula 2]



wherein x and y have the meanings given above;  $M_d$  is Ti; and  $R^1$  is the same or different and is selected from  $C_1$ - $C_6$  alkyl,  $C_1$ - $C_6$  alkoxy, phenyl, and phenoxy," there is no such specification in Cited Invention 2.

(B) Judgment of difference

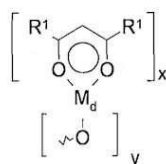
The aforementioned different feature 1 and different feature 2 will be judged.

a. The different feature 1

Since [0160] to [0178] in Citation 2 states "In the present invention, a photoacid generator may be used" and it is apparent that "acid generators" include "photo acid generators," incorporating a photoacid generator into a solution of a metal oxide-containing film-forming composition in Cited Invention 2 is an idea that a person skilled in the art could conceive easily.

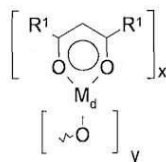
b. The different feature 2

As stated in C. (A) b above, "metal oxide-containing compound 2" in Cited Invention 2 is found to be a compound obtained through hydrolytic condensation between "phenyltrimethoxysilane," "2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane," and "tetrabutoxytitanium," besides the aforementioned "phenyltrimethoxysilane," "2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane," and "tetrabutoxy titanium," "2, 4-pentanedione" which are added upon hydrolytic condensation. [0020] to [0036] in International Publication No. WO2013/125562 states that alkoxy titanium compounds such as "tetrabutoxytitanium" react with di-carbonyl compounds such as "2,4-pentanedione" to form titanium complexes and up to 2n alkoxy group in an alkoxy titanium compound having 2n + 2 alkoxy groups are substituted by dicarbonylate under normal reaction conditions. "Tetrabutoxytitanium" and "2,4-pentanedione" in "metal oxide-containing compound 2" of Cited Invention 2 are found to form "titanium complexes" by substituting up to 2 butoxy groups in the for butoxy groups of "tetrabutoxytitanium" with "2,4-pentanedionate." In "metal oxide-containing compound 2" obtained through hydrolytic condensation of the "titanium complex" with "phenyltrimethoxysilane" and "2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane" the aforementioned "titanium complex" is the "inorganic unit" that "has the following formula;



wherein x is at least 1; y is at least 1, M<sub>d</sub> is Ti; and R<sup>1</sup> is the same and is C<sub>1</sub> alkyl" in Amended Invention of the Application.

Even if the "inorganic unit" in "metal oxide-containing compound 2" in Cited Invention 2 is not a compound of "the following formula



wherein x is at least 1, y is at least 1, M<sub>d</sub> is Ti, and R<sup>1</sup> is the same and is C<sub>1</sub> alkyl" Citation 2 states, as stated in A. (B) h. above, the following technical matter: "in a thermocurable metal oxide-containing film-forming composition comprising a metal oxide-containing compound obtained through hydrolytic condensation between one or more hydrolyzable silicon compounds represented by the general formula (1) and one or more hydrolyzable metal compounds represented by the general formula (2):



wherein R is an alkyl having 1 to 6 carbon atoms, R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> are each a hydrogen atom or a monovalent organic group having 1 to 30 carbon atoms, m<sub>1</sub>, m<sub>2</sub>, and m<sub>3</sub> are each 0 or 1, and m<sub>1</sub>+m<sub>2</sub>+m<sub>3</sub> is an integer of 0 to 3;



wherein R<sup>4</sup> and R<sup>5</sup> are each an organic group having 1 to 30 carbon atoms; m<sub>4</sub>+m<sub>5</sub> is the valence dependent on U; m<sub>4</sub> and m<sub>5</sub> are each an integer equal to or greater than 0; and U is an element selected from the elements of Groups III, IV, and V in the Periodic Table, excluding silicon, examples of compounds represented by the formula (2) in which U is titanium include titanium butoxide, titanium dibutoxybisethylacetoacetate, etc. as monomers." and if "titanium dibutoxybisethylacetoacetate" is used as the hydrolyzable metal compound illustrated in the aforementioned formula (2), then the metal oxide contained in the metal oxide-containing compound is found to be the "inorganic unit" "represented by the following formula" "wherein x is at least 1; y is at least 1, M<sub>d</sub> is Ti; and R<sup>1</sup> is different and is C<sub>1</sub> alkyl or C<sub>2</sub> alkoxy" in Amended Invention of the Application. Therefore, applying the aforementioned technical matter stated in Citation 2 and using "titanium dibutoxybisethylacetoacetate" instead of "tetrabutoxytitanium" to produce the configuration of Amended Invention of the Application relating to the aforementioned different feature 2 when synthesizing "metal oxide-containing compound 2" in Cited Invention 2 is an idea that a person skilled in the art could conceive easily.

Therefore, the aforementioned different feature 2 is a feature not substantially different, or is an idea that a person skilled in the art can easily conceive based on Cited Invention 2 and the described matters in Citation 2.

The polymer contained in the antireflective hardmask composition of Amended Invention of the Application is not actually synthesized in the description of the Application and effects of Amended Invention of the Application are unknown. Amended Invention of the Application cannot be recognized to have an advantageous function and effect that a person skilled in the art cannot foresee.

(C) Summary

As described above, Amended Invention of the Application could be easily made by a person skilled in the art based on Cited Invention 2 and the described matters in Citation 2. Thus, the appellant should not be granted a patent for it independently at the time of patent application under the provisions of Article 29(2) of the Patent Act.

#### 4 Closing

Therefore, the Amendment violates the provisions of Article 126(7) of the Patent Act which is applied mutatis mutandis pursuant to the provisions of Article 17-2(6) of the Patent Act. Thus, the Amendment should be dismissed in accordance with the provisions of Article 53(1) of the Patent Act to be applied mutatis mutandis in Article 159(1) of the Patent Act.

#### No. 3 Regarding the invention of the case

##### 1 The Invention

As in Conclusion of Decision to Dismiss Amendment in "No. 2" above, the Amendment is dismissed. Therefore, the claimed invention of the Application is an invention stated in claims 1-17 before the Amendment, in which the invention according to claim 2 (herein after, referred to as "the Invention") is the invention as described in No. 2 1 (1) above.

##### 2. Described matters in Cited Documents

The described matters in Cited Documents are as stated in No. 2 3. (3) A above.

##### 3 Judgment

As examined in No. 2 2. (1) above, Amended Invention of the Application corresponds to an invention in which the matters specifying the invention of the Invention are limited.

As examined in No. 2, 3 above, Amended Invention of the Application that includes all the constitution feature of the Invention and limits the Invention further more could be easily made by a person skilled in the art based on Cited Invention 1 and the described matters in Citation 2 or based on Cited Invention 2 and the described matters in Citation 2. Thus, the Invention could be easily made by a person skilled in the art for the same reason.

Also, the Invention includes Amended Invention of the Application as its subordinate concept. Since, as examined in No. 2, 3 above, Amended Invention of the Application is not considered to be described in the detailed explanation of the invention, the Invention is not considered to be described in the detailed explanation of the invention for the same reason.

##### No. 4 Closing

As described above, the Invention is not considered to be described in the detailed explanation of the invention. Thus, the appellant should not be granted a patent for the Invention in accordance with the provisions of Article 36(6)(i) of the Patent Act. Moreover, the Invention could be easily made by a person skilled in the art for the same reason based on Cited Invention 1 and the described matters in Citation 2 or based on Cited Invention 2 and the described matters in Citation 2. Thus, the appellant should not be granted a patent for the Invention in accordance with the provisions of Article

29(2) of the Patent Act.

Therefore, the Application should be rejected without examining other claims.

Therefore, the appeal decision shall be made as described in the conclusion.

March 14, 2016

Chief administrative judge: FUJIWARA, Keishi

Administrative judge: HONDA, Hiroyuki

Administrative judge: HIGUCHI, Nobuhiro