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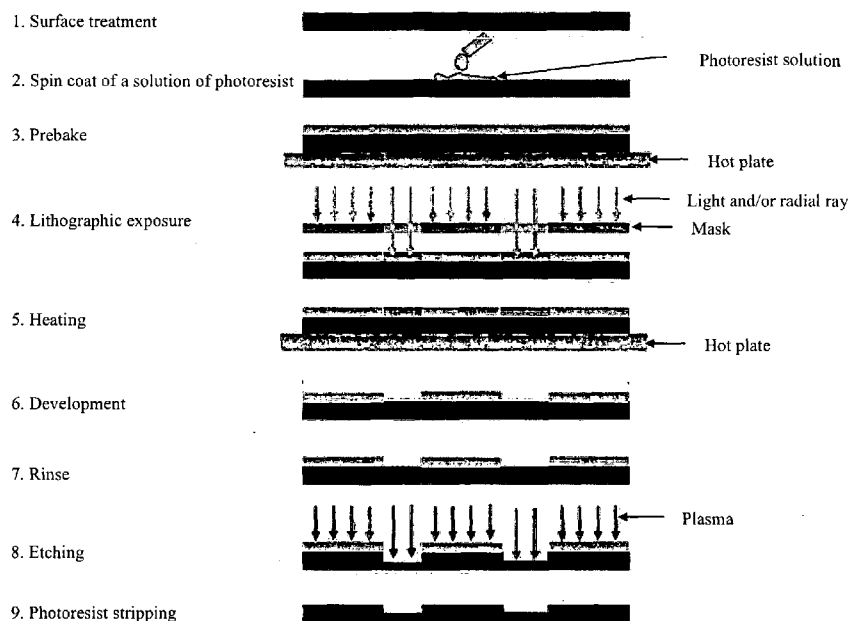
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(54) Title: REAGENT AND COMPOSITION OF RESIST

[Fig. 1]



(57) **Abstract:** Described is a reagent that enhances acid generation of a photoacid generator and composition containing such reagent.

Description

Title of Invention: REAGENT AND COMPOSITION OF RESIST

CROSS REFERENCE TO RELATED APPLICATION

[0001] This application claims the benefit under 35 U.S.C. section 119(e) of U.S. Provisional Patent Application Serial No. 61/822,603 filed on May 13, 2013, the disclosure of which is hereby incorporated herein in its entirety by this reference.

Technical Field

[0002] Several aspects of the disclosure relate to the fields of chemistry and reagents that enhance acid generation and also to the compositions of resists.

Background

[0003] Current high-resolution lithographic processes are based on chemically amplified resists (CARs) and are used to pattern features with dimensions less than 100 nm. As feature dimensions shrink to below 50 nm.

[0004] Method for forming pattern features with dimensions less than 100 nm is disclosed in US 7851252 (filed on February 17, 2009), the contents of the entirety of which are incorporated herein by this reference.

Summary of Invention

[0005] Described are reagents that enhance acid generation of an acid generator and compositions of photoresists. Typically, such a reagent assists the generation of a Bronsted acid. Furthermore, such a reagent can apply to the generation of a Lewis acid.

Typically, such a reagent generates an intermediate having a reducing character.

[0006] Examples of such reagents are bisaryl ketones, arylalkyl ketone, bisarylmethyl halides, benzoin, carbazoles, alkoxy (or aryloxy) benzophenone, alkoxy (or aryloxy) methylnaphthalene, and reagents containing at least one benzyl group.

[0007] In certain embodiments, the reagent is characterized in that: when energy is directly or indirectly applied to an acceptor other than the reagent; the reagent generates an intermediate by receiving energy from the excited acceptor or reacting with the excited acceptor or reactive chemical species generated from the acceptor; and the intermediate enhances generation of acid from a precursor.

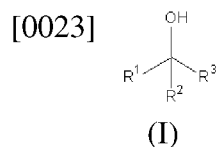
[0008] In certain embodiments, the reagent is characterized in that: when energy is directly or indirectly applied to the reagent; an intermediate is generated from the reagent; and the intermediate enhances generation of acid from a precursor.

[0009] In certain embodiments, the intermediate may have a reducing character. In certain embodiments, the intermediate may be a radical, such as a ketyl radical.

[0010] In certain embodiments, the intermediate discharges at least one of a hydrogen atom and a hydrogen ion that has reducing characteristics.

- [0011] In certain embodiments, the energy includes irradiation with a light, such as an extreme ultraviolet light or a light having a wavelength less than or equal to 15 nm. The energy may include exposure to particle ray such as electron beam.
- [0012] As an example, the composition of photoresist can be applied to the manufacture of electronic devices, such as, semiconductor devices and electro optical devices.
- [0013] A reagent in certain embodiments relating to an aspect of the present invention is characterized in that: a feed of energy to the reagent or to an acceptor receiving the energy generates an intermediate from the reagent; and the intermediate enhances a generation of chemical species acid from a precursor such as acid and base.
- [0014] It is preferred that the intermediate has a reducing character or an electron donor character. A photoacid generator may easily generate acid or base by receiving an electron from the intermediate.
- [0015] It is preferred that the intermediate is a reactive intermediate such as radical.
- [0016] It is preferred that the intermediate discharges at least one of a hydrogen atom and hydrogen ion that have reducing characteristics.
- [0017] It is preferred that the intermediate is a ketyl radical. Such ketyl radical can be stabilized aryl group and the reagent having at least one aryl group can easily form such ketyl radical.
- [0018] It is preferred that the feed of the energy is carried out by irradiation with a light or exposure to particle ray.
- [0019] It is preferred that the feed of the energy is carried out by an irradiation of the reagent with at least one of a light of which the wavelength is equal to or shorter than 15 nm and an electron beam. The feed of energy carried out by the irradiation with such light or electron beam enables formation of pattern with ultrafine structure.
- [0020] A composition in certain embodiments relating to an aspect of the present invention includes: any one of the reagent above; and a first compound that functions as a generation source of acid.
- [0021] It is preferred that the composition further includes: a second compound that has a bond cleavable by chemical species such as acid and base.
- [0022] A composition in certain embodiments relating to an aspect of the present invention includes: a reagent represented by formula (I); and a first compound that functions as a generation source of acid.

Chem.1



- [0024] It is preferred that R¹ is a hydrogen atom; R² is a hydrogen atom, an alkyl carbonyl

group, an aryl carbonyl group, an alkyl group, an alkenyl group, an aralkyl group, an alkynyl group, an alkyl group containing a cyclic or poly cyclic moiety, or a substituent containing at least one atom other than carbon atom and hydrogen atom; and R³ is a hydrogen atom, an alkyl carbonyl group, an aryl carbonyl group, an alkyl group, an alkenyl group, an aralkyl group, an alkynyl group, an alkyl group containing a cyclic or poly cyclic moiety, or a substituent containing at least one atom other than carbon atom and hydrogen atom.

[0025] A composition in certain embodiments relating to an aspect of the present invention includes: a reagent; and a first compound that generates acid by accepting energy, accepting an electron or at least one hydrogen atom.

[0026] With regard to the composition, it is preferred that the reagent includes: a hydroxyl group; and a first cyclic moiety that contains a carbon atom bonded to the hydroxyl group and a hydrogen atom.

[0027] With regard to the composition, it is preferred that the reagent further includes a second cyclic moiety, and the first cyclic moiety containing at least two atoms which are also contained in the second cyclic moiety.

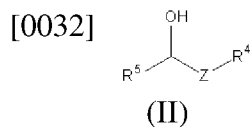
[0028] With regard to the composition, it is preferred that the reagent further includes a third cyclic moiety, and the first cyclic moiety contains at least two atoms that are also contained in the third cyclic moiety.

[0029] With regard to the composition, it is preferred that the first cyclic moiety is either a six membered ring or a five membered ring.

[0030] With regard to the composition, it is preferred that the second cyclic moiety is an aromatic group.

[0031] A composition in certain embodiments relating to an aspect of the present invention includes: a reagent represented by formula (II); and a first compound that generates acid by accepting energy, accepting an electron or at least one hydrogen atom.

Chem.2



With regard to composition, it is preferred that: Z is a carbonyl group, a methylene group, an alkoxymethylene, an aryloxymethylene, or a hydroxymethylene; R⁴ is an aryl group or an aryl group containing an aromatic group and a substituent on the aromatic group containing at least one atom other than carbon atom and hydrogen atom; and R⁵ is a hydrogen atom, an alkyl carbonyl group, an aryl carbonyl group, an alkyl group, an alkenyl group, an aralkyl group, an alkynyl group, an alkyl group containing a cyclic or poly cyclic moiety, or a substituent containing at least one atom other than carbon atom and hydrogen atom.

- [0033] With regard to the composition, it is preferred that the first compound is an organic salt containing an iodonium ion or a sulfonium ion.
- [0034] With regard to the reagent, it is preferred that the immediate is generated by abstracting a hydrogen atom from the reagent.
- [0035] With regard to the reagent, it is preferred that a difference between at least one of a first oxidation potential of a ground state and a second oxidation potential of an excited state of the immediate and at least one of a first reduction potential of the ground state and a second reduction potential of the excited state of the precursor is equal to or greater than 0.10 eV.
- [0036] With regard to the reagent, it is preferred that the first reduction potential is lower than at least one of the first oxidation potential and the second oxidation potential.
- [0037] With regard to the reagent, it is preferred that the intermediate has reducing character.
- [0038] With regard to the reagent, it is preferred that the intermediate is a radical.
- [0039] With regard to the reagent, it is preferred that the intermediate discharge at least one of a hydrogen atom and hydrogen ion that have reducing characteristics.
- [0040] With regard to the reagent, it is preferred that the intermediate is a ketyl radical.
- [0041] With regard to the reagent, it is preferred that the feed of the energy is carried out by irradiation of the reagent with a light.
- [0042] With regard to the reagent, it is preferred that the feed of the energy is carried out by an irradiation of the reagent with at least one of a light of which the wavelength is equal to or less than 15 nm and an electron beam.
- [0043] With regard to the reagent, it is preferred that the intermediate is generated by having a hydrogen atom of the reagent abstracted.
- [0044] A method for manufacturing a device in certain embodiments relating to an aspect of the present invention the method includes: applying a solution of any one of the composition to a substrate such that a film including the composition is formed on the substrate; and irradiating the film with at least one of an electromagnetic ray and a particle ray such that a first portion of the film is irradiated with the at least one of the electromagnetic ray and the particle ray while a second portion of the film is not irradiated with the at least one of the electromagnetic ray and the particle ray.
- [0045] With regard to the method, it is preferred that the method further includes: removing the first portion.
- [0046] With regard to the method, it is preferred that the method further includes: etching the substrate such that a third portion of the substrate on which the first portion has been present is etched.
- [0047] With regard to the method, it is preferred that the irradiating of the film is carried out using at least one of an EUV light and an electron beam.

- [0048] A reagent in certain embodiments relating to an aspect of the present invention is characterized in that: when energy is directly or indirectly applied to the reagent, an intermediate is generated from the reagent; and the intermediate enhances generation of acid from a precursor.
- [0049] With regard to the reagent, it is preferred that the intermediate has a reducing character.
- [0050] With regard to the reagent, it is preferred that the intermediate is a radical.
- [0051] With regard to the reagent, it is preferred that the intermediate is a ketyl radical.
- [0052] With regard to the reagent, it is preferred that the intermediate discharges at least one of a hydrogen atom and a hydrogen ion that has reducing characteristics.
- [0053] With regard to the reagent, it is preferred that the energy includes irradiation with light.
- [0054] With regard to the reagent, it is preferred that the light has a wavelength less than or equal to 15 nm.

Brief Description of Drawings

- [0055] In the drawings, which illustrate what is currently considered to be the best mode for carrying out the invention:
- [0056] [Fig. 1]FIG. 1 shows fabrication processes of a device, such as, an integrated circuit (IC) utilizing a photoresist including an acid generation enhancer.

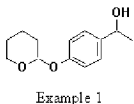
DETAILED DESCRIPTION

- [0057] The disclosure is further described with the aid of the following illustrative Examples.
- Experimental Procedures:
- [0058] Synthesis of 4-hydropyranylacetophenone.
- [0059] 10.0 g of 4-hydroxyacetophenone and 9.89 g of 2H-dihydropyran are dissolved in 80.0 g of methylene chloride. 0.74 g of pyridinium p toluenesulfonate is added to the methylene chloride solution containing 4-hydroxyacetophenone and 2H-dihydropyran. The mixture is stirred at 25 degrees Celsius for 3 hours. Afterwards, the mixture is further stirred after addition of 1% aqueous solution of sodium hydroxide. The organic phase is collected through separation by liquid extraction. 14.4 g of 4-hydropyranylacetophenone is obtained by evaporating solvents from the collected organic phase.
- [0060] Synthesis of 1-(4-tetrahydropyranylphenyl)ethanol (Example 1).
- [0061] 5.0 g of 4-hydropyranylacetophenone and 0.10 g of potassium hydroxide are dissolved in ethanol. 1.04 g of sodium borohydride is added to the ethanol solution containing 4-hydropyranylacetophenone and potassium hydroxide. The mixture is added at 25 degrees Celsius for 3 hours. Afterwards, alkali in the mixture is neutralized

by 10% aqueous solution of hydrochloric acid. The organic phase is collected through separation by liquid extraction using 100 g of methylene chloride. 4.52 g of 1-(4-tetrahydropyranylphenyl)ethanol is obtained by evaporating solvents from the organic phase.

Chem.3

[0062]



[0063]

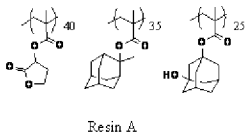
Synthesis of resin A.

[0064]

A solution containing 5.0 g of alpha-methacryloyloxy-gamma-butyrolactone, 6.03 g of 2-methyladamantane-2-methacrylate, and 4.34 g of 3-hydroxyadamantane-1-methacrylate, 0.51 g of dimethyl-2,2'-azobis(2-methylpropionate), and 26.1 g of tetrahydrofuran is prepared. The prepared solution is added for 4 hours to 20.0 g of tetrahydrofuran placed in flask with stirring and boiling. After the addition of the prepared solution, the mixture is heated to reflux for 2 hours and cooled to room temperature. Addition of the mixture drop-wise to a mixed liquid containing 160 g of hexane and 18 g of tetrahydrofuran (with vigorously stirring) precipitates the copolymer. The copolymer is isolated by filtration. Purification of the copolymer is carried out by vacuum drying following twice washings with 70 g of hexane. Thereby, 8.5 g of white powder of the copolymer is obtained.

Chem.4

[0065]



[0066]

Preparation of samples for evaluation.

[0067]

Sample 1 is prepared by dissolving 300 mg of resin A, 36.7 mg of 4,4'-di-(t-butylphenyl)iodonium nonafluorobutanesulfonate as a photoacid generator, and 15.0 mg of coumarin 6 as an indicator in 2000 mg of cyclohexanone.

[0068]

Sample 2 is prepared by dissolving 6.0 mg of 1-(4-tetrahydropyranylphenyl)ethanol, 300 mg of resin A, 36.7 mg of 4,4'-di-(t-butylphenyl)iodonium nonafluorobutanesulfonate as a photoacid generator, and 15.0 mg of coumarin 6 as an indicator in 2000 mg of cyclohexane.

[0069]

Evaluation of efficiency of acid generation.

[0070]

Films are formed on 4 inch quartz wafers by spin coating of Samples 1 and 2. Each of the films is irradiated with electron beams of which volumes are 0, 10, 20, 30, and 40 mC/cm² output by an electron beam lithography apparatus. Subsequent to the electron beam exposure, the efficiencies for the films are obtained by plotting ab-

sorbances at 534 nm, which of each of the films are assigned to quantities of protonated coumarin 6 generated by the respective volumes of electron beams.

[0071] Table 1 shows the relative acid generation efficiencies for the Samples 1 and 2. In Table 1, the acid generation efficiency for the Sample 1 is used as a benchmark. The results shown in Table 1 indicate that the acid generation efficiency is improved by the reduction of the photoacid generator by ketyl radical formed from 1-(4-tetrahydropyranylphenyl)ethanol. In other words, 1-(4-tetrahydropyranylphenyl)ethanol functions as an Acid Generation Enhancer (AGE).

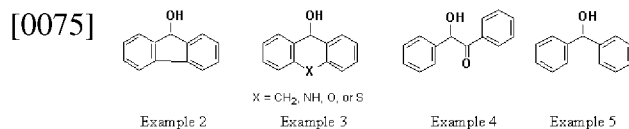
[0072] [Table 1]The relative acid generation efficiencies for Samples 1 and 2.

	Relative acid-generation efficiency
Sample 1	1.0
Sample 2	1.2

[0073] Based upon the results, a reactive intermediate having reducing character is considered to enhance the efficiency of acid generation.

[0074] Each of Examples 2, 3, 4, and 5 can also be used as an AGE.

Chem.5



[0076] It is preferred that a carbon atom which is bonded to hydroxy group or will be the radical center is bonded to at least one aryl group because such aryl group can stabilize generated radical.

[0077] FIG. 1 shows fabrication processes of a device, such as, integrated circuit (IC) using a photoresist including the acid generation enhancer (AGE) obtained by the processes by the above procedures.

[0078] A silicon wafer is provided. The surface of silicon wafer is oxidized by heating the silicon wafer in the presence of oxygen gas.

[0079] A solution containing AGE, resin A, and a photoacid generator is applied to the surface of a silicon wafer by spin coating. A film containing AGE, resin A, and the photoacid generator is formed on the surface of the silicon wafer.

[0080] An irradiation of the film with an EUV light through a mask is carried out after a prebake of the silicon wafer. The deprotection reaction of resin A is induced by acid-generation by photoreaction of the photoacid generator assisted by AGE.

[0081] Development of the irradiated film is performed after the prebake.

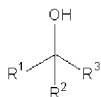
[0082] The irradiated film and the silicon wafer are exposed to a plasma. After that, the remaining film is removed.

[0083] An electronic device, such as, an integrated circuit is fabricated utilizing the processes shown in FIG. 1. The deterioration of the device due to the irradiation with a

light is suppressed compared to existing photoresists since times for irradiation of the film are shortened.

Claims

- [Claim 1] A reagent characterized in that:
a feed of an energy to the reagent or to an acceptor receiving the energy generates an intermediate from the reagent; and
the intermediate enhances a generation of acid from a precursor.
- [Claim 2] The reagent of claim 1, wherein the intermediate has a reducing character.
- [Claim 3] The reagent of claim 1, wherein the intermediate is a radical.
- [Claim 4] The reagent of claim 1, wherein the intermediate discharges at least one of a hydrogen atom and hydrogen ion that have reducing characteristics.
- [Claim 5] The reagent of claim 1, wherein the intermediate is a ketyl radical.
- [Claim 6] The reagent of claim 1, wherein the feed of the energy is carried out by irradiation with a light.
- [Claim 7] The reagent of claim 1, wherein the feed of the energy is carried out by an irradiation of the reagent with at least one of a light of which the wavelength is equal to or shorter than 15 nm and an electron beam.
- [Claim 8] A composition, comprising:
the reagent of claim 1; and
a first compound that functions as a generation source of acid.
- [Claim 9] The composition of claim 8, further comprising:
a second compound that has a bond cleavable by acid.
- [Claim 10] A composition comprising:
a reagent represented by formula (I) ; and
a first compound that functions as a generation source of acid.



(I)

wherein:

R¹ is a hydrogen atom;

R² is a hydrogen atom, an alkyl carbonyl group, an aryl carbonyl group, an alkyl group, an alkenyl group, an aralkyl group, an alkynyl group, an alkyl group containing a cyclic or poly cyclic moiety, or a substituent containing at least one atom other than carbon atom and hydrogen atom; and

R³ is a hydrogen atom, an alkyl carbonyl group, an aryl carbonyl group, an alkyl group, an alkenyl group, an aralkyl group, an alkynyl group, an

alkyl group containing a cyclic or poly cyclic moiety, or a substituent containing at least one atom other than carbon atom and hydrogen atom.

[Claim 11]

A composition, comprising:

a reagent; and

a first compound that generates acid by accepting energy or at least one hydrogen atom, the reagent including:

a hydroxyl group; and

a first cyclic moiety that contains a carbon atom bonded to the hydroxyl group and a hydrogen atom.

[Claim 12]

The composition of claim 11, wherein:

the reagent further includes a second cyclic moiety, and

the first cyclic moiety contains at least two atoms which are also contained in the second cyclic moiety.

[Claim 13]

The composition of claim 12, wherein:

the reagent further includes a second cyclic moiety, and

the first cyclic moiety contains at least two atoms that are also contained in the third cyclic moiety

[Claim 14]

The composition of claim 11, wherein the first cyclic moiety is either a six membered ring or a five membered ring.

[Claim 15]

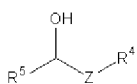
The composition of claim 11, wherein the second cyclic moiety is an aromatic group.

[Claim 16]

A composition, comprising:

a reagent represented by formula (II); and

a first compound that generates acid by accepting an energy or at least one hydrogen atom,



(II)

wherein:

Z is a carbonyl group, a methylene group, an alkoxymethylene, an aryloxymethylene, or a hydroxymethylene;

R⁴ is an aryl group or an aryl group containing an aromatic group and a substituent on the aromatic group containing at least one atom other than carbon atom and hydrogen atom; and

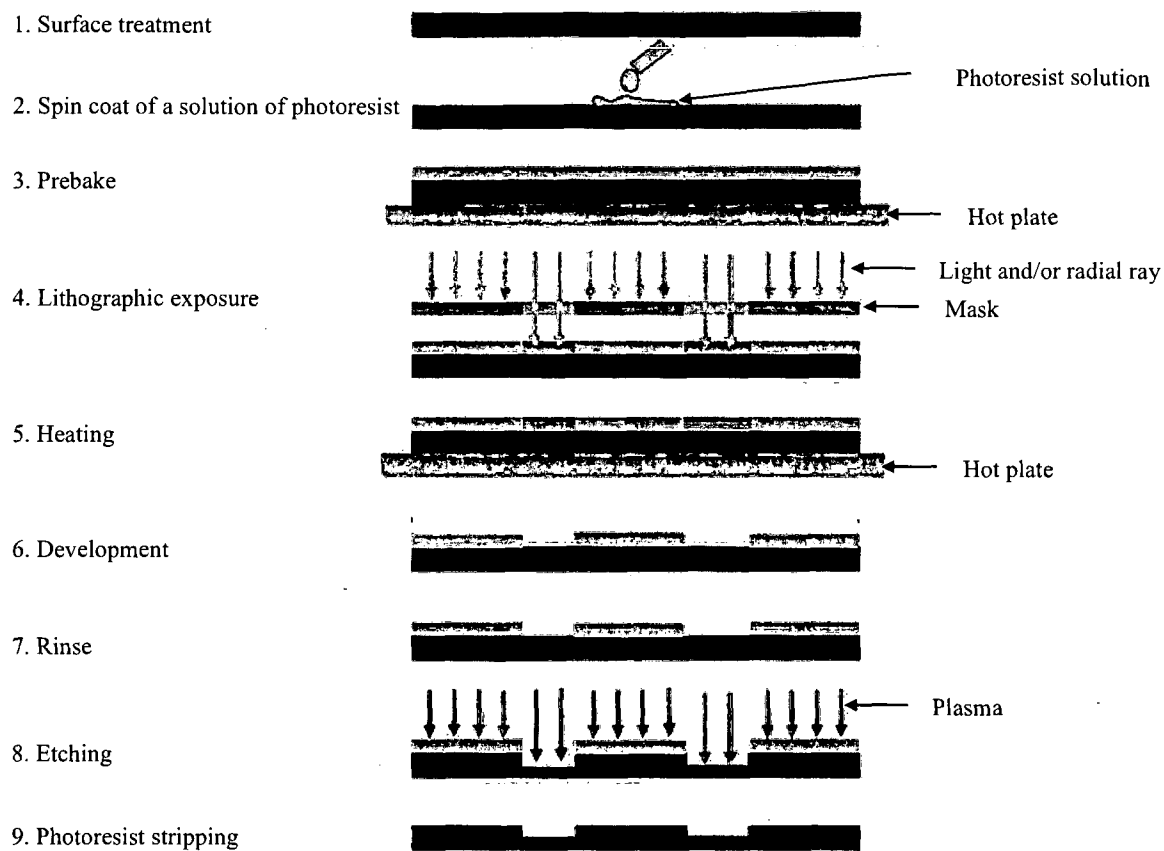
R⁵ is a hydrogen atom, an alkyl carbonyl group, an aryl carbonyl group, an alkyl group, an alkenyl group, an aralkyl group, an alkynyl group, an alkyl group containing a cyclic or poly cyclic moiety, or a substituent

- containing at least one atom other than carbon atom and hydrogen atom.
- [Claim 17] The composition of claim 10, wherein the first compound is an organic salt containing an iodonium ion or a sulfonium ion.
- [Claim 18] The reagent of claim 1, wherein the immediate is generated by abstracting a hydrogen atom from the reagent.
- [Claim 19] The reagent of claim 1, wherein a difference between at least one of a first oxidation potential of a ground state and a second oxidation potential of an excited state of the immediate and at least one of a first reduction potential of the ground state and a second reduction potential of the excited state of the precursor is equal to or greater than 0.10 eV.
- [Claim 20] The reagent of claim 19, wherein the first reduction potential is lower than at least one of the first oxidation potential and the second oxidation potential.
- [Claim 21] The reagent of claim 1, wherein the intermediate has reducing character.
- [Claim 22] The reagent of claim 1, wherein the intermediate is a radical.
- [Claim 23] The reagent of claim 1, wherein the intermediate discharge is at least one of a hydrogen atom and hydrogen ion that have reducing characteristics.
- [Claim 24] The reagent of claim 1, wherein the intermediate is a ketyl radical.
- [Claim 25] The reagent of claim 1, wherein the feed of the energy is carried out by irradiation of the reagent with a light.
- [Claim 26] The reagent of claim 1, wherein the feed of the energy is carried out by an irradiation of the reagent with at least one of a light of which the wavelength is equal to or less than 15 nm and an electron beam.
- [Claim 27] The reagent of claim 1, wherein the intermediate is generated by having a hydrogen atom of the reagent abstracted.
- [Claim 28] A method for manufacturing a device, the method comprising: applying a solution of the composition of claim 9 to a substrate such that a film including the composition is formed on the substrate; and irradiating the film with at least one of an electromagnetic ray and a particle ray such that a first portion of the film is irradiated with the at least one of the electromagnetic ray and the particle ray while a second portion of the film is not irradiated with the at least one of the electromagnetic ray and the particle ray.
- [Claim 29] The method of claim 28, further comprising: removing the first portion.

- [Claim 30] The method of claim 29, further comprising:
etching the substrate such that a third portion of the substrate on which the first portion has been present is etched.
- [Claim 31] The method of claim 28, wherein irradiating the film is carried out using at least one of an EUV light and an electron beam.
- [Claim 32] Every novel feature, compound or process disclosed herein.
- [Claim 33] A reagent characterized in that:
when energy is directly or indirectly applied to the reagent, an intermediate is generated from the reagent; wherein the intermediate enhances generation of acid from a precursor.
- [Claim 34] The reagent of claim 1, wherein the intermediate has a reducing character.
- [Claim 35] The reagent of claim 1, wherein the intermediate is a radical.
- [Claim 36] The reagent of claim 35, wherein the intermediate is a ketyl radical.
- [Claim 37] The reagent of claim 1, wherein the intermediate discharges at least one of a hydrogen atom and a hydrogen ion that has reducing characteristics.
- [Claim 38] The reagent of claim 1, wherein the energy comprises irradiation with light.
- [Claim 39] The reagent of claim 36, wherein the light has a wavelength less than or equal to 15 nm.

1/1

[Fig. 1]



International application No.
PCT/JP2014/002531

Form PCT/ISA/210 (second sheet) (July 2009)

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2014/002531

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☒ Claims Nos.: 32
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

The term 'Every novel feature, compound or process disclosed herein' used in Claim 32 is totally unclear such that it is impossible to determine the subject matter that may be reasonably expected to be claimed.

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of additional fees.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- ☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- ☐ No protest accompanied the payment of additional search fees.