Appeal decision

Appeal No. 2012-3397

Kanagawa, Japan	
Appellant	TOKYO OHKA KOGYO CO. LTD.
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The case of appeal against the examiner's decision of refusal of Japanese Patent

Application No. 2004-316960 entitled "POLYMER COMPOUND, PHOTORESIST COMPOSITION CONTAINING SUCH POLYMER COMPOUND, AND METHOD FOR FORMING RESIST PATTERN" (the application published on April 13, 2006 as Japanese Unexamined Patent Application Publication No. 2006-96965) 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 is an application filed on October 29, 2004 (priority claim: February 20, 2004, April 28, 2004, June 17, 2004, August 31, 2004). A reason for rejection dated March 22, 2011 was notified. In response, a written argument and a written amendment were submitted on May 18, 2011. Then, a reason for rejection (final) dated July 29, 2011 was notified, and in response a written argument and a written amendment were submitted on October 31, 2011. Then, a decision of refusal dated November 30, 2011 was issued with dismissal of the written amendment dated October 31, 2011. In response, an appeal against the examiner's decision of refusal was made on February 22, 2012, and a written amendment was submitted at the same time. During a reconsideration procedure by the examiner before appeal examination, a reason for rejection dated March 30, 2012 was issued. A written argument was submitted on May 30, 2012 but a reconsideration report dated June 18, 2012 was made. A questioning dated December 13, 2013.

No. 2 The Invention

The inventions according to Claims 1 to 9 of the present application should be 2/17

specified in the matters recited in Claims 1 to 9 of the Claims in view of the claims, specification, and drawings to which an amendment was made by the written argument submitted on February 22, 2012. The invention according to Claim 1 (hereinafter referred to as "the Invention") is set forth as below:

"A polymer compound with variable alkaline solubility by acid action, comprising a constituent unit (a1) derived from a compound represented by at least the following general formula (2):

[Chemical Formula 1]



(where R_1 is a cycloaliphatic group with a carbon number of 20 or less having an adamantane skeleton (with the proviso that a group having a carbonyl group is excluded), n is 0 or an integer of 1 to 5, and R_2 is hydrogen atom or lower alkyl group with a carbon number of 20 or less)."

No. 3 The summary of reasons for refusal in the reconsideration by examiner before appeal

The reasons for refusal during the reconsideration procedure by examiner before appeal examination dated March 30, 2012 may be summarized as below:

"The inventions according to Claims 1 to 5 and 7 to 9 of this application are identical to the inventions described in the specification, the claims, or drawings originally attached to the following patent application, which was filed before and published after the filing date of the present application. Further, the inventor of the present application is not identical to those who made an invention according to a prior patent application before filing the present application. Further, at the time of filing the present application, the Applicant thereof is not identical to those of the prior patent application. Thus, the inventions are not patentable under the provision of Article 29bis of the Patent Act.

Note

1. Japanese Patent Application No. 2004-28595 (Japanese Unexamined Patent Application Publication No. 2005-220059)"

No. 4 Judgment by the body

1. Prior application and the specification of the prior application

Japanese Patent Application No. 2004-28595 is an application filed on February 4, 2004 before the priority date of the present application, by the Applicant named Daicel Corporation with inventors of Koyama Yutaka, Inoue Keizo, and Iwahama Takahiro, which was published as Japanese Unexamined Patent Application Publication No. 2005-220059 on August 18, 2005 after the priority date of the present application.

The specification and the claims originally attached to the application of the prior application are collectively referred to as "the prior application."

2. The description of the specification of the prior application

The prior application discloses the following matters:

Note that the following summarized portion relies on a publication of the unexamined application corresponding to the prior application of Japanese Unexamined Patent Application Publication No. 2005-220059.

A. "An unsaturated carboxylic acid hemiacetal ester represented by the following formula (1):

[Chemical Formula 1]

$$\begin{array}{c} R^{a} \\ \hline \\ 0 \\ 0 \\ R^{b} \\ R^{c} \\ R^{d} \end{array}$$
 (1)

(where R^a is a hydrogen atom, a halogen atom, an alkyl group with a carbon number of 1 to 6 or a haloalkyl group with a carbon number of 1 to 6, R^b is a hydrocarbon group with a hydrogen atom at position 1, R^c is a hydrogen atom or a hydrocarbon group, and R^d is an organic group comprising cyclic skeleton)." (the Claims, Claim 1)

B. "The halogen atom in the above R^a includes ... Alkyl group with a carbon number of 1 to 6 may include for example, methyl ... Among them, ... particularly a methyl group is preferable. ..." (Paragraph 0017)

C. "The 'ring' constituting a cyclic skeleton in an organic group comprising cyclic skeleton of R^d may include a monocyclic or polycyclic non-aromatic or aromatic ring. The polycyclic non-aromatic ring may include, for example, an adamantane ring, etc. ..." (Paragraph 0020)

D. "Typical examples of the unsaturated carboxylic acid hemiacetal ester represented by formula (1) may include, but are not limited to, the following compounds.

[1-1] 1-(adamantan-1-yloxy)ethyl (meth)acrylate ..." (paragraph 0024)

E. "In addition to unsaturated carboxylic acid hemiacetal esters represented by formula (1), a compound where R^b and R^c are both hydrogen atoms in formula (1) is also useful as a monomer of a polymer compound for photoresist. The repeating unit corresponding to this compound exerts acid-leaving function or hydrophilic function in a polymer compound.

Such compound may include a compound corresponding to an example of an unsaturated carboxylic acid hemiacetal ester represented by the aforesaid formula (1) (a compound where $R^{b}=R^{c}=H$), etc.

The compound where both R^b and R^c are hydrogen atoms in formula (1) (the compound represented by formula (B)) may be produced, for example, by reacting the unsaturated carboxylic acid represented by formula (3) with the halomethyl ether represented by formula (A) in the presence of a base as shown in the following reaction scheme.

[Chemical Formula 5]



(where R^a and R^d are the same as above and Y is a halogen atom)

The halogen atom in Y includes chlorine, bromine, iodine, etc. The reaction is carried out in the presence or absence of a solvent. The aforementioned solvent may be used as a solvent. The base may include, for example, an organic base such as triethylamine or pyridine, or an inorganic base such as sodium hydroxide, sodium carbonate, or sodium hydrogencarbonate. The usage amount of unsaturated carboxylic acid represented by formula (3) may be, for example, 0.5 to 10 mole or so, preferably 0.8 to 2 mole or so on the basis of 1 mole of the halomethyl ether compound represented by formula (A). The usage amount of the base may be, for example, 1 to 5 mole or so on the basis of 1 mole of the halomethyl ether compound represented by formula (A). To suppress the polymerization of the halomethyl ether compound represented by formula (A) or a reaction product, a small amount of a polymerization inhibitor such as 4-methoxy phenol may be added in a system. The reaction temperature is normally -10°C to 100°C, preferably 0 to 60°C or so. After the completion of the reaction, the reaction product may be separated and purified by separating means such as acidity or alkalinity,

extraction, condensation, distillation, crystallization, recrystallization, or column chromatography.

The halomethyl ether represented by formula (A) may be produced, for example, by reacting a hydroxy compound represented by formula (C) with formaldehyde or its equivalents (paraformaldehyde, 1,3,5-trioxane, etc.) and a halogenated hydrogen represented by formula (D) as shown in the following reaction scheme.

[Chemical Formula 6]

$$\begin{array}{ccc} R^{d} - OH & \xrightarrow{HCHO} & Y - CH_{2} - 0 - R^{d} \\ (C) & (D) & (A) \end{array}$$

(where R^d and Y are the same as above)

The halogenated hydrogen represented by formula (D) may include, for example, hydrochloride, hydrobromide, etc. The reaction is carried out in the presence or absence of a solvent. The aforementioned solvent may be used as a solvent. The usage amount of formaldehyde or its equivalents may be, for example, 0.8 to 10 mole or so, preferably 1 to 1.5 mole or so in formaldehyde basis on the basis of the 1 mole of the hydroxy compound represented by formula (C). The usage amount of the halogenated hydrogen represented by formula (D) may be, for example, 1 to 5 mole or so on the basis of 1 mole of hydroxy compound represented by formula (C), or may be an excess amount. The reaction temperature is normally -10°C to 100°C, preferably 0 to 60°C or so. After the completion of the reaction, the reaction product may be separated and purified by separating means such as acidity or alkalinity, extraction, condensation, distillation, crystallization, recrystallization, or column chromatography." (Paragraphs 0032 to 0036)

F. "Production example 1

A mixture of 43.2 g of adamantane ethanol, 48.1 g of vinyl propionate, 15.3 g of 7/17

sodium carbonate, 120 ml of toluene, and 1.62 g of di-µ-chlorobis(1,5-cyclooctadiene) diiridium (I) was fed to a four-necked flask and stirred for four hours under a nitrogen atmosphere while being heated with 100°C. The precipitate in a reaction liquid was filtered and the filtrate was depressurized and condensed. The condensate was purified by depressurized distillation to obtain 34.8 g of 2-(adamantan-1-yl) ethyl vinyl ether represented by the following formula (8).

[Chemical Formula 9]

[Spectrum data of 2-(adamantan-1-yl)ethyl vinyl ether]

1H-NMR (CDCl3) δ: 1.46(t,2H), 1.53(d,6H), 1.62-1.72(m,6H), 1.95(m,3H), 3.73(t,2H), 3.96(m,1H), 4.16(m,1H), 6.46(m,1H)

Production example 2

A mixture of 32.8 g of 2-(adamantan-1-yl)ethyl vinyl ether, 68.4 g of methacrylic acid, 0.16 g of phosphoric acid, 0.164 g of 4-methoxy phenol, and 290 ml of toluene was fed to a four-necked flask and stirred for six hours at 20°C under a nitrogen atmosphere. After the completion of the reaction, a reaction liquid was washed twice with 500 ml of 10 weight% sodium carbonate water solution and once with 500 ml of 10 weight% saline water, and an organic layer was depressurized and condensed. The condensate was purified by silica gel chromatography to obtain 38.6 g of 1-[2-(adamantan-1yl)ethoxy]ethyl(meth)acrylate represented by formula (9).

[Chemical Formula 10]



[Spectrum data of 1-[2-(adamantan-1-yl)ethoxy]ethyl(meth)acrylate]

1H-NMR (CDCl3) δ:1.37-1.41(m,2H), 1.43(d,3H), 1.50(d,6H), 1.60-1.71(m,6H), 1.93(m,3H), 1.96(m,3H), 3.53(m,1H), 3.72(m,1H), 5.60(m,1H), 5.97(m,1H), 6.16(m,1H)" (paragraphs 0075 to 0076)

3. The invention described in the specification of the prior application

The specification of the prior application discloses in the summarized portions A and E that "in 'formula (1)':



a compound where R^b and R^c are both hydrogen atoms is also useful as a monomer of a polymer compound for photoresist. The repeating unit corresponding to this compound exerts acid-leaving function or hydrophilic function in a polymer compound." It further discloses that "Such compound may include a compound corresponding to an example of unsaturated carboxylic acid hemiacetal ester represented by the aforesaid formula (1) (a compound where $R^b=R^c=H$)" and "The compound where both R^b and R^c are hydrogen atoms in formula (1) (the compound represented by formula (B))."



Here, the summarized portions A and B disclose hydrogen atom, methyl group and

the like as "R^a," and further, the summarized portion C discloses "adamantane ring" and the like as "R^d" and the summarized portion D discloses a specific compound of "1- (adamantan-1-yloxy)ethyl (meth)acrylate" while it is a compound represented by the above "formula (1)." Furthermore, Examples disclose a specific compound of "1-[2-(adamantan-1-yl)ethoxy]ethyl(meth)acrylate" as in the summarized portion F, while it is a compound represented by the above "formula (1)." Thus, it specifically discloses a compound where "R^a" is a hydrogen atom or methyl group, and "R^d" is an "adamantane ring" (adamantyl group) or "adamantyl ethyl group."

Further, the summarized portion E clearly discloses that "In formula (B), R^a and R^d are the same as above." Therefore, it can be said that the specification of the prior application substantially discloses "(adamantan-1-yloxy)methyl (meth)acrylate" or "1-[2-(adamantan-1-yl)ethoxy]methyl (meth)acrylate," which corresponds to "a compound represented by formula (B)" where "R^a" is a hydrogen atom or methyl group, and "R^d" is an "adamantane ring" (adamantyl group) or "adamantyl ethyl group."

Consequently, the summarized portion E discloses that "a compound where R^b and R^c are both hydrogen atoms in 'formula (1)' is also useful as a monomer of a polymer compound for photoresist. The repeating unit corresponding to this compound exerts acid-leaving function or hydrophilic function in a polymer compound." Thus, it is recognized that the specification of the prior application discloses that "a monomer of (adamantan-1-yloxy)methyl (meth)acrylate or 1-[2-(adamantan-1-yl)ethoxy] methyl (meth)acrylate" may be used as "a monomer of a polymer compound for photoresist" and the repeating unit corresponding to said compound has an "acid-leaving function in a polymer compound."

Consequently, the specification of the prior application discloses obtaining "a polymer compound for photoresist" by polymerizing a monomer of "(adamantan-1-yloxy)methyl (meth)acrylate or 1-[2-(adamantan-1-yl)ethoxy] methyl (meth)acrylate," and the structure of "repeating unit" derived from said monomer incorporated into said polymer compound has an "acid-leaving function."

Accordingly, the specification of the prior application discloses the following

invention (hereinafter referred to as "the invention of the specification of the prior application").

"A polymer compound comprising a repeating unit with an acid-leaving function, the compound being derived from a monomer of (adamantan-1-yloxy)methyl (meth)acrylate or 1-[2-(adamantan-1-yl)ethoxy] methyl (meth)acrylate."

4. Comparison / Judgment

The Invention is compared to the invention of the specification of the prior application.

"(Adamantan-1-yloxy)methyl (meth)acrylate" in the invention of the specification of the prior application corresponds to "a compound (a monomer) represented by the following general formula (2):

[Chemical Formula 1]



where R_2 is a hydrogen atom or methyl group, n=0 and R_1 is an adamantane ring (adamantyl group)" of the Invention.

Further, "1-[2-(adamantan-1-yl)ethoxy] methyl (meth)acrylate" in the invention of the specification of the prior application corresponds to "a compound (a monomer) represented by the following general formula (2):

[Chemical Formula 1]



where R_2 is a hydrogen atom or methyl group, n=2 and R_1 is an adamantane ring (adamantyl group)" of the Invention.

Further, the fact that the polymer compound in the invention of the specification of the prior application "has an acid-leaving function" is construed as a function capable of changing alkaline solubility by generating a free carboxyl group in photoresist. Therefore, the function of the polymer compound of the invention of the specification of the prior application corresponds to the function of the variable solubility by acid action of the Invention.

Consequently, the polymer compound of the invention of the specification of the prior application corresponds to the polymer compound of the Invention.

Accordingly, the Invention is identical to the invention of the specification of the prior application.

No. 5 Appellant's allegation

The Appellant's arguments in the written argument submitted on May 30, 2012 are summarized as below.

"The compounds corresponding to Compounds [1-1] and [1-2], which are specifically raised as representative examples of unsaturated carboxylic acid hemiacetal ester represented by formula (1) (a compound where R^b=R^c=H), correspond to the compound represented by the general formula (2) of Claim 1 of the present application.

However, the specification of the prior application merely discloses the usefulness

of the compound represented by the above formula (B) as a monomer of a polymeric compound for photoresist. It actually fails to disclose any specific evidence (i.e., test result, etc.) showing the possibility to use the compound as a monomer of a polymeric compound for photoresist.

(2)

First of all, a patentable invention of chemical substances should have novelty and usefulness or industrial applicability as chemical substances. Therefore, the description of the specification is required to not only identify a chemical substance itself but also show its producibility and usefulness as a chemical substance. Further, in general, it is difficult to predict the usefulness of the invention of a chemical substance according to only the chemical structure. It is commonly known to those skilled in the art that actual tests are necessary to find such usefulness. In view of this, for saying that 'the invention according to a chemical substance is disclosed in the specification,' the specification is required to disclose actual test results of the chemical substance for usefulness in addition to identifying the chemical substance and showing its producibility so that those skilled in the art can recognize the usefulness.

However, the compound represented by the above formula (B) is unsaturated carboxylic acid acetal ester with an 'acetal structure,' not a 'hemiacetal structure.' Therefore, on the basis of only the test results of 1-[2-(adamantan-1yl)ethoxy]ethyl(meth)acrylate and 1-(bornyloxy)ethylmethacrylate, it cannot be said that the compounds represented by the above formula (B), which are totally different from compounds with a hemiacetal structure as a staple of the structure, are useful as photoresist.

(4)

Further, the specification of the prior application discloses a method of synthesizing a compound represented by the above formula (B) in paragraphs [0033] to [0036].

However, the properties of a compound will be unknown unless it is actually manufactured. Specifically, even if a compound represented by the above formula (B)

might be synthesized by a method described in the specification of the prior application, it is unknown as to whether a polymer compound might be synthesized from the synthesized compound unless it is actually tested... As seen above, whether or not a compound can be synthesized is totally different from whether or not a polymer compound can be produced from the synthesized compound. From also this viewpoint, since the specification of the prior application fails to disclose any test result showing that a polymer compound may actually be synthesized from the compound represented by the above formula (B), the specification fails to describe to the extent that those skilled in the art could recognize that a compound represented by the above formula (B) is useful for a monomer of a polymer compound.

(5)

As aforementioned, even those skilled in the art cannot recognize that the compounds represented by the above formula (B) are useful as a monomer of polymer compounds for photoresist according to the specification of the prior application without any test result of the compound represented by the above formula (B). Accordingly, the specification of the prior application fails to disclose the invention of 'a compound represented by the above formula (B)'."

The specification of the prior application specifically discloses in detail, however, in the summarized portion E,

reacting a hydroxy compound (C) and formaldehyde (or paraformaldehyde, etc.) and a halogenated hydrogen (D) by the following reaction scheme:

$$\begin{array}{ccc} R^{d} - OH & \xrightarrow{HCHO} & Y - CH_{2} - 0 - R^{d} \\ (C) & (D) & (A) \end{array}$$

to obtain a halomethyl ether compound (A), and then

reacting an unsaturated carboxylic acid (3) and the halomethyl ether compound (A) in the

presence of a base by the following reaction scheme:



to obtain "a compound represented by formula (B)" where "R^b and R^c are both hydrogen atoms."

Further, there is no particular reason or grounds that, when "R^d" is an "adamantane ring" (adamantyl group) or an "adamantyl ethyl group," a halomethyl ether compound (A) may not be produced from hydroxy compound (C). Further, there is no particular reason or grounds that "a compound represented by formula (B)" may not be produced or synthesized by the reaction of an unsaturated carboxylic acid (3) of acrylic acid or methacrylic acid and a halomethyl ether compound (A) where "R^d" is an "adamantane ring" (adamantyl group) or "adamantyl ethyl group."

Further, as aforementioned, "(adamantan-1-yloxy)methyl (meth)acrylate" or "1-[2-(adamantan-1-yl)ethoxy]methyl (meth)acrylate," which is "a compound represented by formula (B)" where "R^a" is a hydrogen atom or methyl group, "R^d" is an "adamantane ring" (adamantyl group) or "adamantyl ethyl group," is substantially disclosed in the specification of the prior application.

Further, "(adamantan-1-yloxy)methyl (meth)acrylate" or "1-[2-(adamantan-1yl)ethoxy]methyl (meth)acrylate" is a compound belonging to (meth)acrylate, and a vinyl monomer capable of producing a polymer compound by, e.g., radical polymerization with organic peroxides. Therefore, that is the monomer that provides the polymer compound described in the specification of the prior application. Specifically, it can be said that the specification of the prior application describes the polymer derived from the said monomer (a polymer compound).

Further, it discloses that a "repeating unit" corresponding to "a compound 15 / 17

represented by formula (B)" has an "acid-leaving function in a polymer compound." Therefore, there is no difference between "acetal structure" and "hemiacetal structure" in acid-leaving function or alkaline solubility. But the portion E discloses "a compound of formula (1) where R^b and R^c are both hydrogen atoms may be useful as a monomer of a polymer compound for photoresist. The repeating unit corresponding to this compound exerts an acid-leaving function or hydrophilic function in a polymer compound." Therefore, it cannot be said that an "acetal structure" derived from "a compound represented by formula (B)" may not have an acid-leaving function or alkaline solubility.

Further, as aforementioned, the specification of the prior application sufficiently specifically describes a method of synthesizing or producing "a compound represented by formula (B)," and that the compound is (meth)acrylate. Further there is a concrete ground for the embodiment where "R^d" is an "adamantane ring" (adamantyl group) or "adamantyl ethyl group." Therefore, it is not appropriate to say that the specification fails to disclose "(adamantan-1-yloxy)methyl (meth)acrylate" or "1-[2-(adamantan-1-yl)ethoxy]methyl (meth)acrylate" only for lack of examples in the prior specification.

Accordingly, the Appellant's argument is not acceptable.

Further, the Appellant provided details in the response letter submitted on February, 18, 2013, but no further points to be mentioned and rebutted may be found in addition to the above points.

No. 6 Closing

As seen above, the invention according to Claim 1 of the present application is identical to the invention described in the specification or the claims originally attached to the application of Japanese Patent Application No. 2004-28595 (Japanese Unexamined Patent Application Publication No. 2005-220059), that is, a patent application filed before the priority date of the present application and published after the priority date thereof.

Further, the inventors of the present application are not identical to those who made the aforesaid invention according to a patent application before the priority date of the present application. Further, the Applicant is not identical to the Applicant of the aforesaid patent application as of the filing date of the present application. Thus the inventions are not patentable under the provision of Article 29bis of the Patent Act.

Accordingly, the reason for refusal in the reconsideration by examiner before appeal is reasonable without making an examination of the other remaining claims. Thus, the present application should be rejected.

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

May 28, 2013

Chief administrative judge: ONODERA, Tsutomu

Administrative judge: KAGA, Naoto

Administrative judge: KURANO, Masaaki