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(54) **LUBRICANT ADDITIVE AND LUBRICANT COMPOSITION**

(57) A lubricating oil additive contains at least one steroid derivative selected from steroid saturated fatty acid ester, steroid carbonate ester and steroid ether. A lubricating oil composition added with the lubricating oil

additive exhibits a low friction coefficient and excellent wear resistance, heat resistance and oxidation resistance stability.

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**Description****Technical Field**

5 **[0001]** The present invention relates to a lubricating oil additive and lubricating oil composition.

**Background Art**

10 **[0002]** As the reduction in emission of carbon dioxide is being demanded in perspective of the global environment, more efficient utilization of energy is getting increasing attentions. For instance, machines including sliding portions can efficiently utilize the energy by reducing the friction energy with use of lubricating oil. In addition, prevention of wear, which is also one of the most important functions of the lubricating oil, leads to increase in lifetime of machines. Further, in perspective of waste reduction, the lubricating oil is demanded to have more long-drain capabilities, and to exhibit high oxidation stability and heat resistance.

15 As additives capable of friction reduction, a sulfur-containing molybdenum composition such as molybdenum dithiocarbamate (MoDTC) has been in use (see e.g., Patent Document 1).

**[0003]** Patent Document 1: JP-A-07-145187

**Disclosure of the Invention**

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**Problems to Be Solved by the Invention**

25 **[0004]** However, the metal-containing compounds disclosed in Patent Document 1 may be harmful to exhaust-purifying catalysts, so that ashless lubricating oil additives are being desired. Further, though sulfur and phosphorus are also capable of preventing wear, reduction of the use of them are demanded in view of their toxicity and harm to the exhaust-purifying catalysts.

Hence, an object of the invention is to provide a so-called ashless lubricating oil additive capable of providing sufficient lubrication without use of metals (e.g., Mo), sulfur or phosphorus, and to provide a lubricating oil composition added with the lubricating oil additive.

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**Means for Solving the Problems**

**[0005]** In order to solve the above-described problems, aspects of the invention provide the following lubricating oil additives and lubricating oil compositions:

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[1] a lubricating oil additive, containing at least one lanolin alcohol derivative selected from the group consisting of lanolin-alcohol saturated fatty acid ester, lanolin alcohol carbonate ester and lanolin alcohol ether;

[2] a lubricating oil additive, comprising at least one steroid derivative selected from steroid saturated fatty acid ester, steroid carbonate ester and steroid ether;

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[3] the lubricating oil additive according to [2], in which the steroid derivative is a cholesterol derivative;

[4] a lubricating oil composition, containing: lubricating base oil; and the lubricating oil additive according to any one of [1] to [3];

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[5] the lubricating oil composition according to [4], further containing at least one additive selected from the group consisting of viscosity index improvers, pour point depressants, antioxidants, ashless dispersants, friction modifiers, metal detergents, antiwear agents, rust inhibitors, metal deactivators, anti-emulsifiers and antifoaming agents; and

[6] the lubricating oil composition according to [4] or [5], in which the lubricating oil composition is used for engines, gears or industries.

50 **[0006]** The lubricating oil additive provided according to the aspect of the invention, although not containing a metal (e.g., Mo), sulfur or phosphorus, is capable of providing sufficient lubrication. Specifically, the lubricating oil composition obtained by adding the lubricating oil additive to suitable base oil exhibits a low friction coefficient and excellent wear resistance.

**Best Mode for Carrying Out the Invention**

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**[0007]** The lubricating oil additive according to the aspect of the invention contains at least one lanolin alcohol derivative selected from lanolin-alcohol saturated fatty acid ester, lanolin alcohol carbonate ester and lanolin alcohol ether.

The lanolin alcohol means a neutral alcohol component obtained by saponifying lanolin. Lanolin is a light-yellow waxen

substance obtained by refining "wool grease" (i.e., collected wash with which wool has been washed) through deacidification, decoloration and deodorization.

Lanolin alcohol contains: a steroid skeleton such as cholesterol, desmosterol, lanosterol, dihydrolanosterol and lanosterol; and a branched chain alcohol having 16 to 28 carbon atoms.

**[0008]** Such lanolin alcohol is available at Nippon Fine Chemical Co., Ltd.. By esterifying lanolin alcohol and fatty acid chloride under the presence of a base, lanolin alcohol fatty acid ester is obtainable.

Alternatively, lanolin alcohol fatty acid ester is also obtainable by reacting lanolin alcohol and saturated fatty acid with each other under the presence of a dehydration-condensation agent such as DCC (dicyclohexylcarbodiimide), DIC (diisopropylcarbodiimide) and EDC (1-ethyl-3-(3-dimethylaminopropyl)carbodiimide hydrochloride).

Further alternatively, lanolin alcohol fatty acid ester is also obtainable by dehydrating a sterol compound and saturated fatty acid under the presence of an acid catalyst.

**[0009]** Lanolin alcohol carbonate ester is obtainable by reacting lanolin alcohol and chlorocarbonate ester with each other under the presence of a base.

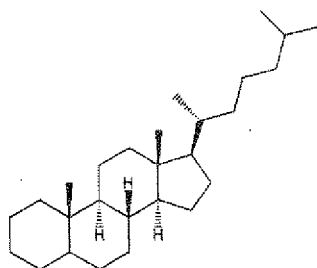
Lanolin alcohol alkyl ether is obtainable by reacting lanolin alcohol and alkyl bromide with each other under the presence of a base. Alternatively, lanolin alcohol alkyl ether is also obtainable by dehydrating and condensing lanolin alcohol and chain alcohol under the presence of an acid.

**[0010]** The lubricating oil additive according to the aspect of the invention contains at least one steroid derivative selected from steroid saturated fatty acid ester, steroid carbonate ester and steroid ether.

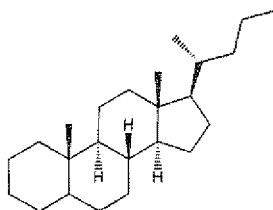
The steroid is a generic term for referring to compounds having cyclopenta[a]phenanthrene skeletons, of which structures are exemplarily represented by the following formulae (1) to (5).

**[0011]**

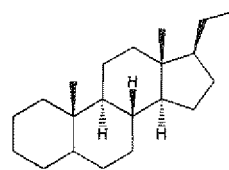
[Chemical Formula 1]



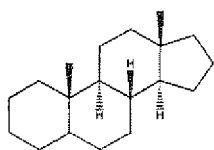
**(1) CHOLESTANE**



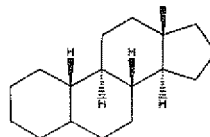
**(2) CHOLANE**



**(3) PREGNANE**



**(4) ANDROSTANE**



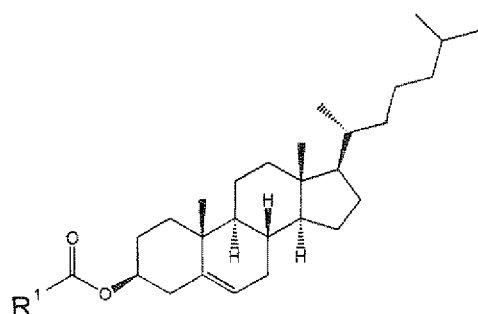
**(5) ESTRANE**

**[0012]** The steroid saturated fatty acid ester, steroid carbonate ester and steroid ether are compounds in which one or more saturated fatty acid ester group, carbonate ester group and ether group are bonded to the above-described skeletons.

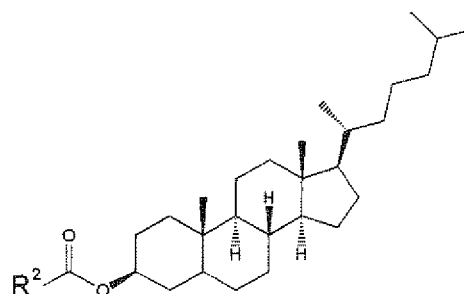
Examples of the steroid saturated fatty acid ester are cholesterol saturated fatty acid ester and cholestanol saturated fatty acid ester respectively represented by the following formulae (6) and (7).

**[0013]**

[Chemical Formula 2]



(6) CHOLESTEROL ESTER



(7) CHOLESTANOL ESTER

**[0014]** In the above formulae (6) and (7),  $R^1$  and  $R^2$  represent a branched or non-branched saturated hydrocarbon group. When  $R^1$  and  $R^2$  are of unsaturated structure, the obtained lubricating oil additive may exhibit lower wear resistance and deteriorated oxidation stability. In addition,  $R^1$  and  $R^2$  preferably have 1 to 30 carbon atoms, more preferably 9 to 24 carbon atoms. When  $R^1$  and  $R^2$  have more than 30 carbon atoms, availability may be deteriorated. Examples of carboxylic acids for providing esters having  $R^1$  and  $R^2$  as their basic skeletons are a pelargonic acid, capric acid, lauric acid, myristic acid, pentadecanoic acid, palmitic acid, margaric acid, stearic acid, nonadecane acid, eicosanoic acid, docosanoic acid and tetra docosanoic acid.

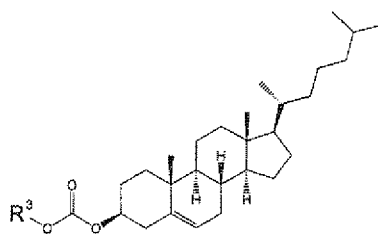
Further, the lubricating oil additive is preferably a cholesterol saturated fatty acid ester represented by the formula (6) in view of the availability and friction reduction capabilities.

**[0015]** Such steroid saturated fatty acid ester is readily available as a marketed product (for instance, cholesterol stearate by Wako Pure Chemical Industries, Ltd.). Alternatively, by esterifying a sterol compound and saturated fatty acid chloride under the presence of a base, sterol saturated fatty acid ester is obtainable. Further alternatively, sterol ester is obtainable by reacting a sterol compound and saturated fatty acid with each other under the presence of a dehydration-condensation agent such as DCC (dicyclohexylcarbodiimide), DIC (diisopropylcarbodiimide) and EDC (1-methyl-3-(3-dimethylaminopropyl)carbodiimide hydrochloride). Still further alternatively, sterol ester is also obtainable by dehydrating a sterol compound and saturated fatty acid under the presence of an acid catalyst.

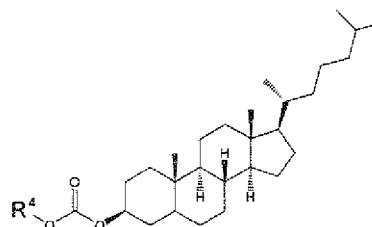
**[0016]** Examples of the steroid carbonate ester are cholesterol carbonate ester and cholesterol carbonate ester respectively represented by the following formulae (8) and (9).

**[0017]**

[Chemical Formula 3]



(8) CHOLESTEROL CARBONATE ESTER



(9) CHOLESTANOL CARBONATE ESTER

**[0018]**  $R^3$  and  $R^4$  represent a branched or non-branched hydrocarbon group.  $R^3$  and  $R^4$  preferably have 1 to 30 carbon atoms, more preferably 9 to 24 carbon atoms. When  $R^3$  and  $R^4$  have more than 30 carbon atoms, availability may be deteriorated. Examples of carboxylic acids for providing esters having  $R^3$  and  $R^4$  as their basic skeletons are a pelargonic acid, capric acid, lauric acid, myristic acid, pentadecanoic acid, palmitic acid, margaric acid, stearic acid, oleic acid,

linolic acid, linolenic acid, arachidonic acid, docosahexaenoic acid, eicosapentaenoic acid and erucic acid.

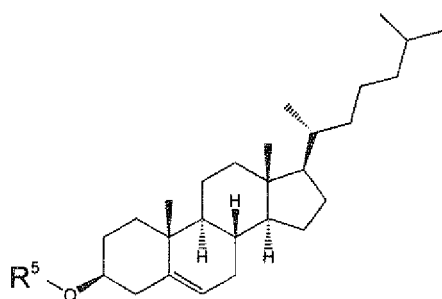
The lubricating oil additive is preferably a cholesterol carbonate ester represented by the formula (8) in view of the availability.

**[0019]** Such steroid carbonate ester is also readily available as a marketed product (for instance, cholesterol oleyl carbonate by Tokyo Chemical Industry Co., Ltd.). Alternatively, steroid carbonate ester is also obtainable by reacting a sterol compound and chloroformate with each other under the presence of a base.

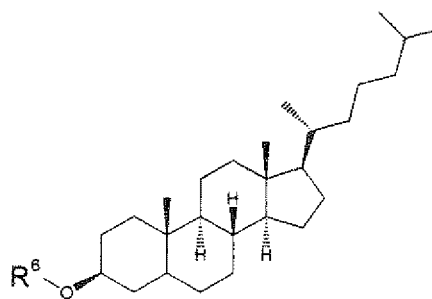
**[0020]** Examples of the steroid ether are cholesterol ether and cholestanol ether respectively represented by the following formulae (10) and (11).

**[0021]**

[Chemical Formula 4]



**(10) CHOLESTEROL ETHER**



**(11) CHOLESTANOL ETHER**

**[0022]**  $R^5$  and  $R^6$  represent a branched or non-branched hydrocarbon group. In addition,  $R^5$  and  $R^6$  preferably have 1 to 30 carbon atoms, more preferably 9 to 24 carbon atoms. When  $R^5$  and  $R^6$  have more than 30 carbon atoms, availability may be deteriorated. Examples of  $R^5$  and  $R^6$  are a nonyl group, decyl group, undecyl group, dodecyl group, tridecyl group, tetradecyl group, pentadecyl group, hexadecyl group, heptadecyl group, octadecyl group, nonadecyl group and eicosyl group.

The lubricating oil additive is preferably a cholesterol ether represented by the formula (10) in view of the availability.

**[0023]** Such steroid ether is obtainable by, for instance, reacting a sterol compound and alkyl bromide with each other under the presence of a base. Alternatively, steroid ether is also obtainable by reacting a sterol compound and alcohol under the presence of an acid catalyst.

**[0024]** The lubricating oil composition according to the aspect of the invention contains a lubricating base oil and the above-described lubricating oil additive. As the lubricating base oil, mineral oil and/or synthetic oil is used. The mineral oil or synthetic oil is not particularly limited, but may be suitably selected from any mineral oil and synthetic oil that have been conventionally used as base oil of the lubricating oil.

Examples of the mineral oil are mineral oil refined by processing lubricating oil fractions by at least one of solvent-deasphalting, solvent-extracting, hydrocracking, solvent-dewaxing, catalytic-dewaxing and hydrorefining (the lubricating oil fractions are obtained by vacuum-distilling atmospheric residual oil obtained by atmospherically distilling crude oil such as paraffin crude oil, naphthene crude oil and aromatic crude oil) and mineral oil manufactured by isomerizing wax and GTL WAX. Namely, the examples are spindle oil, 70 neutral oil, 100 neutral oil, 150 neutral oil, 500 neutral oil and bright stock.

**[0025]** On the other hand, examples of the synthetic oil are polybutene, polyolefin ( $\alpha$ -olefin homopolymer or copolymer such as ethylene- $\alpha$ -olefin copolymer), various esters (such as polyol ester, diacid ester and phosphoric ester), various ethers (such as polyphenylether), polyglycol, alkylbenzene and alkyl naphthalene. Among the above synthetic oil, poly- $\alpha$ -olefin copolymer is particularly preferable in view of its low friction coefficient. The poly- $\alpha$ -olefin copolymer preferably has a molecular weight of 250 to 5000, more preferably 300 to 3000. When the molecular weight is less than 250, oil-film breakdown is more likely to be invited due to the low viscosity of the oil, and thus wear resistance is unfavorably deteriorated. On the other hand, when the molecular weight is more than 5000, the viscosity is increased, and the friction coefficient is unfavorably increased due to increase of stirring resistance.

**[0026]** In the aspect of the invention, one of the above mineral oil and synthetic oil may be singularly used or a combination of two or more thereof may be used as the base oil.

In terms of the viscosity of the base oil, kinematic viscosity at 40 degrees C is preferably 2 to 2000 mm<sup>2</sup>/s, more preferably

10 to 1500 mm<sup>2</sup>/s. When the kinematic viscosity at 40 degrees C is less than 2 mm<sup>2</sup>/s, oil-film breakdown is unfavorably likely to be invited. On the other hand, when the kinematic viscosity at 40 degrees C is more than 2000 mm<sup>2</sup>/s, flow resistance is increased, and the friction coefficient is unfavorably increased.

A content of the above-described lubricating oil additive is 0.01 to 5 mass% of the total amount of the composition, preferably 0.1 to 2 mass%. When the content of the lubricating oil additive is less than 0.01 mass%, the friction coefficient may not be sufficiently reduced, and wear resistance may not be sufficiently enhanced. On the other hand, when the content of the lubricating oil additive is more than 5 mass%, no advantage comparable to the increased content is obtained.

**[0027]** As described above, the lubricating oil composition according to the aspect of the invention contains: the lubricating base oil; and the lubricating oil additive containing at least one steroid derivative selected from steroid saturated fatty acid ester, steroid carbonate ester and steroid ether. The lubricating oil composition can exhibit a low friction coefficient and excellent wear resistance without use of metals (e.g., Mo), sulfur or phosphorus.

In addition, the above-described steroid derivative is a so-called ashless compound containing none of metal, sulfur or phosphorus. Thus, when used in internal combustion engines, the steroid derivative can suppress the degradation of catalysts and pose less environmental load.

Accordingly, the lubricating oil composition according to the aspect of the invention is favorably usable in machines in which friction wear is to occur. For instance, in the form of engine lubricating oil, the lubricating oil composition is favorably usable in gasoline or diesel internal combustion engine for vehicles and ships. Further, in the form of gear lubricating oil, the lubricating oil composition is favorably usable in differential gears, mission gears, manual transmissions, automatic transmissions and continuously variable transmissions. Still further, in the form of industrial lubricating oil, the lubricating oil composition is favorably usable in compressor oil, cutting oil and plastic working oil. Examples of the compressor are refrigerators and vacuum pumps. The cutting oil is used in, for instance, cutting work. The plastic working oil is used in rolling work, extrusion work, pultrusion work, shear work, bending work, deep drawing work and forge work.

**[0028]** The lubricating oil composition according to the aspect of the invention may further contain suitable additives. Specifically, examples of such additives are viscosity index improvers, pour point depressants, antioxidants, ashless dispersants, friction modifiers, metal detergents, antiwear agents, rust inhibitors, metal deactivators, anti-emulsifiers and antifoaming agents.

**[0029]** Examples of the viscosity index improver are non-dispersed polymethacrylate, dispersed polymethacrylate, an olefin copolymer (such as an ethylene-propylene copolymer), a dispersed olefin copolymer and a styrene copolymer (such as a styrenediene hydrated copolymer). The weight-average molecular weight of the viscosity index improver is preferably 5000 to 1000000, more preferably 100000 to 800000, when, for instance, dispersed or non-dispersed polymethacrylate is used. When an olefin copolymer is used, the weight-average molecular weight is preferably 800 to 300000, more preferably 10000 to 200000. For use in the lubricating oil composition, one of the above viscosity index improvers may be singularly adopted or a plurality thereof may be adopted in combination. Typically, the content of the viscosity index improver is in a range of 0.1 to 20 mass% of the total amount of the lubricating oil composition.

**[0030]** Examples of the pour point depressant are an ethylene-vinyl acetate copolymer, condensation product of chlorinated paraffin and naphthalene, condensation product of chlorinated paraffin and phenol, polymethacrylate and polyalkyl styrene. In particular, polymethacrylate is preferable. The content of the pour point depressant is typically in a range of 0.01 to 5 mass% of the total amount of the lubricating oil composition.

**[0031]** Examples of the antioxidant are amine antioxidants such as alkylated diphenylamine, phenyl- $\alpha$ -naphthylamine and alkylated phenyl- $\alpha$ -naphthylamine, phenol antioxidants such as 2,6-di-*t*-butylphenol, 4,4'-methylenebis(2,6-di-*t*-butylphenol), isooctyl-3-(3,5-di-*t*-butyl-4-hydroxyphenyl) propionate and n-octadecyl-3-(3,5-di-*t*-butyl-4-hydroxyphenyl) propionate, sulfur antioxidants such as dilauryl-3,3'-thiodipropionate, phosphorous antioxidants such as phosphite, zinc dithiophosphate antioxidants and molybdenum antioxidants. For use in the lubricating oil composition, one of the above antioxidants may be singularly adopted, or a plurality of them may be adopted in combination. Typically, two or more of them are preferably used in combination, and the content of the antioxidant is preferably 0.01 to 5 mass% of the total amount of the lubricating oil composition, more preferably 0.2 to 3 mass%.

**[0032]** Examples of the ashless dispersant are polybutenyl succinimide, polybutenyl benzylamine and polybutenylamine that have polybutenyl groups having number average molecular weight of 900 to 3500, and their derivatives such as their borated products. For use in the lubricating oil composition, one of the above ashless dispersants may be singularly adopted or a plurality thereof may be adopted in combination. Typically, the content of the ashless dispersant is in a range of 0.1 to 20 mass% of the total amount of the lubricating oil composition.

**[0033]** Examples of the friction modifier are an organic molybdenum compound, fatty acid, higher alcohol, fatty acid ester, oils and fats, amine, amide, phosphate ester, phosphite ester, phosphate ester amine salt, sulfur-containing compound such as olefin sulfide, sulfurized fatty acid ester and dibenzyl disulfide and chloride compound such as chlorinated paraffin. For use in the lubricating oil composition, one of the above friction modifiers may be singularly adopted or a plurality thereof may be adopted in combination. Typically, the content of the friction modifier is in a range of 0.05 to 4 mass% of the total amount of the lubricating oil composition.

**[0034]** Examples of the metal detergent are sulfonate, phenate, salicylate and naphthenate of alkali metal (e.g., sodium

(Na), potassium (K)) or alkali earth metal (e.g., calcium (Ca), magnesium (Mg)). One of the above metal detergents may be singularly used or a plurality of them may be used in combination. The total base number and the additive amount of the metal detergent may be suitably determined in accordance with the desired performances of the lubricating oil. The total base number is typically 0 to 500 mg KOH/g by a perchloric acid method, preferably 20 to 400 mg KOH/g. The content is typically in a range of 0.1 to 10 mass% of the total amount of the lubricating oil composition.

**[0035]** Examples of the antiwear agent are metal dithiophosphate (e.g., Zn, Pb, Sb, Mo), metal dithiocarbamate (e.g., Zn, Pb, Sb, Mo), metal naphthenate (e.g., Pb), fatty acid metal salt (e.g., Pb), boron compound, phosphate ester, phosphite ester, alkyl hydrogen phosphite, phosphate ester amine salt, phosphate ester metal salt (e.g., Zn), disulfide, sulfurized oils and fats, olefin sulfide, dialkyl polysulfide, diaryl alkyl polysulfide, diaryl polysulfide and solid lubricating antiwear agent such as graphite, molybdenum disulfide, antimony sulfide and polytetrafluoroethylene. For use in the lubricating oil composition, one of the above antiwear agents may be singularly adopted or a plurality thereof may be adopted in combination. Typically, the content of the antiwear agent is in a range of 0.1 to 5 mass% of the total amount of the lubricating oil composition.

**[0036]** Examples of the rust inhibitor are a fatty acid, alkenyl succinic acid half ester, fatty acid soap, alkyl sulfonate, polyvalent alcohol fatty acid ester, fatty acid amine, paraffin oxide and alkyl polyoxyethylene ether. The content of the rust inhibitor is typically in a range of 0.01 to 3 mass% of the total amount of the lubricating oil composition.

**[0037]** Examples of the metal deactivator is benzotriazole, triazole derivative, benzotriazole derivative and thiadiazole derivative. The content of the metal deactivator is typically in a range of 0.01 to 3 mass% of the total amount of the lubricating oil composition.

As the antifoaming agent, a liquid silicone is suitable, and a methylsilicone, a fluorosilicone and a polyacrylate may be used. The content of the antifoaming agent is preferably 0.0005 to 0.01 mass% of the total amount of the composition.

**[0038]** Examples of the anti-emulsifier are ethers such as polyoxyethylene alkyl ether and polyoxyethylene alkyl phenyl ether and esters such as sorbitan fatty acid ester, polyoxyethylene sorbitan fatty acid ester and polyoxyethylene fatty acid ester. The content of the antifoaming agent is preferably 0.005 to 1 mass% of the total amount of the composition.

**[0039]** In order to better obtain the advantages of the invention, the composition is preferably so prepared as to contain, among the above additives, the metal-containing compounds, sulfur compounds and phosphorus compounds at as small contents as possible.

## Examples

**[0040]** Next, the invention will be described in a further detail with reference to Examples, which by no means limit the invention.

Specifically, lubricating oil compositions as determined below were prepared, and the lubricating properties thereof (friction coefficients and wear resistance) were evaluated.

**[0041]** The components used for preparing the lubricating oil compositions are as follows.

### (1) Base Oil:

(1-1) Base oil A: mineral oil of 500 neutral fraction (HG500)

(1-2) Base oil B: commercially-available oil containing antioxidants, antiwear agents and the like (general-purpose oil exemplarily used as gear oil)

### (2) Additives

(2-1) FM A: cholesterol stearate (manufactured by Wako Pure Chemical Industries, Ltd.)

(2-2) FM B: cholesterol n-caprylate (manufactured by Tokyo Chemical Industry Co., Ltd.)

(2-3) FM C: cholesterol acetate (manufactured by Tokyo Chemical Industry Co., Ltd.)

(2-4) FM D: cholesterol n-octyl carbonate (manufactured by Tokyo Chemical Industry Co., Ltd.)

(2-5) FM E: cholesterol isostearate (manufactured by Tokyo Chemical Industry Co., Ltd.)

(2-6) FM F: cholesterol oleyl carbonate (manufactured by Tokyo Chemical Industry Co., Ltd.)

(2-7) Molybdenum dithiocarbamate (MoDTC)

(2-8) FM G: lanolin alcohol stearate (see below)

**[0042]** 100 g of lanolin alcohol and 40 g of pyridine were dissolved in 700 ml of toluene, and 131 g of stearoyl chloride and solution of 300-ml toluene were added at 0 degree C. The mixture was reacted for one day and then added with water, and insoluble matters were filtrated. Subsequently, the organic layers were washed with aqueous solution of 5% hydrochloric acid and water. After being dried with magnesium sulfate, the organic layers were condensed, so that 202 g of light yellow solid was obtained.

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[Examples 1 to 12 and Comparative 1]

**[0043]** Lubricating oil compositions (sample oils) containing components shown in Table 1 were respectively prepared, which were then subjected to tests detailed below for evaluation of the lubricating properties. The results are also shown in Table 1. For reference, the same test was also conducted on the base oil A (HG500) itself and the base oil B (commercially-available oil).

(1) Block-on-Ring Test

With use of a block-on-ring tester (LFW-1), the test was conducted under conditions where: the rotation speed was 500 rpm; the load was 44.5 to 177.8 N (10 to 40 lbs); the oil temperature was 60 degrees C; and the friction time was 15 minutes.

Specifically, the amount of the sample oil was set such that the half of the ring was soaked therein, H-60 Test Block (manufactured by Falex Corporation) was used as the test block, and S-10 Test Ring (manufactured by Falex Corporation) was used as the test ring. Load was applied on the block, and the resistance caused when the ring was rotated was measured with a strain gauge, and the friction coefficient was obtained. In addition, width of the wear trace after the test was also measured.

(2) Soda Pendulum Test (JASO Method)

Soda pendulum test was conducted at the oil temperature of 60 degrees C, and the friction coefficient was obtained.

(3) Shell Four-Ball EP Test (Based on ASTM D 2783)

The test was conducted with the rotation speed of 1800 rpm at a room temperature, and the last non-seizure load (LNL) and last weld load (LWL) were measured, from which load wear index (LWI) was then obtained. The larger the value of the load wear index becomes, the more favorable the load resistance capacity is.

**[0044]**

Table 1

|                                                                                                         |                    | Example |         |         |         |         |         |         |         |         |         |         | Comparative | Reference |        |
|---------------------------------------------------------------------------------------------------------|--------------------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|-------------|-----------|--------|
|                                                                                                         |                    | 1       | 2       | 3       | 4       | 5       | 6       | 7       | 8       | 9       | 10      | 11      | 12          | 1         | 2      |
|                                                                                                         |                    | residue | residue | residue | residue | residue | residue | residue | residue | residue | residue | residue | residue     | residue   |        |
| Composition<br>(mass%)                                                                                  | Base Oil A (HG500) | -       | -       | -       | -       | -       | -       | -       | -       | -       | -       | -       | -           | -         | -      |
|                                                                                                         | Base Oil B 1)      | -       | -       | -       | -       | -       | -       | -       | -       | -       | -       | -       | -           | -         | 100    |
|                                                                                                         | FM A               | 0.01    | 01      | 1       | 0.1     | 1       | -       | -       | -       | -       | -       | -       | -           | -         | -      |
|                                                                                                         | FM B               | -       | -       | -       | -       | -       | 1       | -       | -       | -       | -       | -       | -           | -         | -      |
|                                                                                                         | FM C               | -       | -       | -       | -       | -       | -       | 1       | -       | -       | -       | -       | -           | -         | -      |
|                                                                                                         | FM D               | -       | -       | -       | -       | -       | -       | -       | 1       | -       | -       | -       | -           | -         | -      |
|                                                                                                         | FM E               | -       | -       | -       | -       | -       | -       | -       | -       | 1       | -       | -       | -           | -         | -      |
|                                                                                                         | FM F               | -       | -       | -       | -       | -       | -       | -       | -       | -       | 1       | -       | -           | -         | -      |
|                                                                                                         | FM G               | -       | -       | -       | -       | -       | -       | -       | -       | -       | -       | 1       | 2           | -         | -      |
|                                                                                                         | MoDTC              | -       | -       | -       | -       | -       | -       | -       | -       | -       | -       | -       | -           | 1         | -      |
| LFW-1Friction<br>Coefficient                                                                            | 44.5               | 0.078   | 0.076   | 0.063   | 0.063   | 0.069   | 0.095   | 0.093   | 0.086   | 0.08    | 0.084   | 0.065   | 0.061       | 0.067     | 0.097  |
|                                                                                                         | 88.9               | 0.118   | 0.109   | 0.092   | 0.105   | 0.110   | 0.124   | 0.127   | 0.123   | 0.122   | 0.117   | 0.089   | 0.085       | 0.097     | 0.126  |
|                                                                                                         | 133.3              | 0.136   | 0.125   | 0.107   | 0.123   | 0.130   | 0.138   | 0.142   | 0.135   | 0.137   | 0.13    | 0.103   | 0.101       | 0.113     | 0.142  |
|                                                                                                         | 177.8              | 0.141   | 0.134   | 0.122   | 0.135   | 0.138   | 0.143   | 0.145   | 0.142   | 0.142   | 0.136   | 0.108   | 0.105       | 0.116     | 0.146  |
| LFW-1 Wear Trace Width (mm)                                                                             |                    | 1.418   | 0.352   | 0.341   | -       | -       | 0.406   | 0.4922  | 0.488   | 0.4394  | 0.3315  | 0.442   | 0.485       | 0.3725    | 0.6205 |
| Friction Coefficient (Soda Pendulum Test)                                                               |                    | -       | 0.158   | 0.116   | -       | -       | -       | -       | -       | -       | -       | 0.086   | 0.086       | 0.151     | 0.350  |
| ShellFour-Ball<br>EP Test                                                                               | LNL                | -       | -       | 392     | -       | -       | -       | -       | -       | -       | -       | 490     | 490         | -         | 214    |
|                                                                                                         | LWL                | -       | -       | 1236    | -       | -       | -       | -       | -       | -       | -       | 1569    | 1569        | -         | 1236   |
|                                                                                                         | LWI                | -       | -       | 173     | -       | -       | -       | -       | -       | -       | -       | 240     | 260         | -         | 146    |
| 1) commercially-available oil containing antioxidant, antiwear agent and the like (general-purpose oil) |                    |         |         |         |         |         |         |         |         |         |         |         |             |           |        |

[Example 13]

**[0045]** 98.7 mass% of the base oil A (HG500), 0.3 mass% of a phenol antioxidant and 1 mass% of FM A were mixed together, and a thermal stability test (based on JIS K2540) was conducted at 120 degrees C for 168 hours. No precipitate was observed at all.

**[0046]** As understandable from the above results, the lubricating oil additive according to the aspect of the invention was considerably excellent in friction-coefficient reduction and wear resistance, even though the additive contained no metal such as Mo or Zn, sulfur or phosphorus. Particularly notably, the friction reduction by the lubricating oil additive according to the aspect of the invention was much more excellent than the friction reduction by MoDTC (Comparative 1), i.e., an additive typically used so far for providing considerably excellent friction reduction. As understandable also from Examples 4 and 5, when added to commercially-available general-purpose lubricating oil, the lubricating oil additive according to the aspect of the invention was able to further reduce the friction coefficient. In addition, a small additive amount was sufficiently effective. Further, as understandable from Example 13, the lubricating oil additive according to the aspect of the invention was also excellent in heat resistance and oxidation resistance stability. Accordingly, the long-drain capabilities required for the lubricating oil can be achieved, thereby contributing to waste reduction.

### Industrial Applicability

**[0047]** The lubricating oil additive and the lubricating oil composition containing the same according to the aspects of the invention are applicable to various lubricating oil.

### Claims

1. A lubricating oil additive, comprising at least one lanolin alcohol derivative selected from the group consisting of lanolin-alcohol saturated fatty acid ester, lanolin alcohol carbonate ester and lanolin alcohol ether.
2. A lubricating oil additive, comprising at least one steroid derivative selected from steroid saturated fatty acid ester, steroid carbonate ester and steroid ether.
3. The lubricating oil additive according to claim 2, wherein the steroid derivative is a cholesterol derivative.
4. A lubricating oil composition, comprising: lubricating base oil; and the lubricating oil additive according to any one of claims 1 to 3.
5. The lubricating oil composition according to claim 4, further comprising at least one additive selected from the group consisting of viscosity index improvers, pour point depressants, antioxidants, ashless dispersants, friction modifiers, metal detergents, antiwear agents, rust inhibitors, metal deactivators, anti-emulsifiers and antifoaming agents.
6. The lubricating oil composition according claim to 4 or 5, wherein the lubricating oil composition is used for engines, gears or industries.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2008/060416

| <b>A. CLASSIFICATION OF SUBJECT MATTER</b><br><i>C10M159/02(2006.01)i, C10M129/16(2006.01)i, C10M129/70(2006.01)i, C10M129/84(2006.01)i, C10M129/86(2006.01)i, C10M129/95(2006.01)i, C10N30/00(2006.01)n, C10N30/06(2006.01)n, C10N30/08(2006.01)n, C10N30/10(2006.01)n,</i>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       |                                                                                                                                                                                                                             |                                                                                     |
|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------|
| <b>B. FIELDS SEARCHED</b><br>Minimum documentation searched (classification system followed by classification symbols)<br><i>C10M159/02, C10M129/16, C10M129/70, C10M129/84, C10M129/86, C10M129/95, C10N30/00, C10N30/06, C10N30/08, C10N30/10, C10N40/04, C10N40/20, C10N40/25</i><br>Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched<br>Jitsuyo Shinan Koho 1922-1996 Jitsuyo Shinan Toroku Koho 1996-2008<br>Kokai Jitsuyo Shinan Koho 1971-2008 Toroku Jitsuyo Shinan Koho 1994-2008<br>Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)<br>CAplus (STN), REGISTRY (STN)                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |                                                                                                                                                                                                                             |                                                                                     |
| <b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      |                                                                                                                                                                                                                             |                                                                                     |
| Category*                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                          | Citation of document, with indication, where appropriate, of the relevant passages                                                                                                                                          | Relevant to claim No.                                                               |
| X<br>A                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                             | GGRIBAILO, A. P. et al., Effect of liquid crystals on the lubricating properties of mineral oils. Khimiya i Tekhnologiya Topliv i Masel, 1983, No.7, p.18-20, ISSN 0023-1169                                                | 2-6<br>1                                                                            |
| X<br>A                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                             | IVANOV, V. E. et al., Effect of lubrication by technical-grade media on drawing parameters. Trenie i Iznos, 1993, Vol.14, No.5, p.926-30, ISSN 0202-4977                                                                    | 2-6<br>1                                                                            |
| X<br>A                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                             | RODNENKOV, V. G., Potentiometric studies of steel friction surface characteristics subsequent to dynamic contact with liquid crystal additives in lubricants. Trenie i Iznos, 2000, Vol.21, No.5, p.551-557, ISSN 0202-4977 | 2-6<br>1                                                                            |
| <input checked="" type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/> See patent family annex.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |                                                                                                                                                                                                                             |                                                                                     |
| * Special categories of cited documents:<br>"A" document defining the general state of the art which is not considered to be of particular relevance<br>"E" earlier application or patent but published on or after the international filing date<br>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)<br>"O" document referring to an oral disclosure, use, exhibition or other means<br>"P" document published prior to the international filing date but later than the priority date claimed<br>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention<br>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone<br>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art<br>"&" document member of the same patent family |                                                                                                                                                                                                                             |                                                                                     |
| Date of the actual completion of the international search<br>25 August, 2008 (25.08.08)                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |                                                                                                                                                                                                                             | Date of mailing of the international search report<br>02 September, 2008 (02.09.08) |
| Name and mailing address of the ISA/<br>Japanese Patent Office                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |                                                                                                                                                                                                                             | Authorized officer                                                                  |
| Facsimile No.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      |                                                                                                                                                                                                                             | Telephone No.                                                                       |

Form PCT/ISA/210 (second sheet) (April 2007)

INTERNATIONAL SEARCH REPORT

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| C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT |                                                                                                                                                                                                  |                       |
|-------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------|
| Category*                                             | Citation of document, with indication, where appropriate, of the relevant passages                                                                                                               | Relevant to claim No. |
| A                                                     | JP 7-133488 A (Croda Japan Kabushiki Kaisha),<br>23 May, 1995 (23.05.95),<br>Claims; Par. Nos. [0001], [0010]<br>(Family: none)                                                                  | 1-6                   |
| A                                                     | JP 2005-98495 A (Nissan Motor Co., Ltd.),<br>14 April, 2005 (14.04.05),<br>Claims; Par. Nos. [0047] to [0062], [0079] to<br>[0082]<br>& WO 2005/014761 A2 & EP 1666573 A2<br>& US 2006/263604 A1 | 1-6                   |

Form PCT/ISA/210 (continuation of second sheet) (April 2007)

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2008/060416

Continuation of A. CLASSIFICATION OF SUBJECT MATTER  
(International Patent Classification (IPC))

C10N40/04(2006.01)n, C10N40/20(2006.01)n, C10N40/25(2006.01)n

(According to International Patent Classification (IPC) or to both national  
classification and IPC)

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2008/060416

**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.:  
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:
  
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:

The lubricant additive containing a lanolin alcohol derivative set forth in claim 1 and the lubricant additive containing a steroid derivative set forth in claim 2 do not have a common special technical feature.

Consequently, claims 1-6 are not so linked as to form a single general inventive concept.

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

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, 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.

Form PCT/ISA/210 (continuation of first sheet (2)) (April 2007)

**REFERENCES CITED IN THE DESCRIPTION**

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**Patent documents cited in the description**

- JP 7145187 A [0003]