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(54) LUBRICANT OIL COMPOSITION

(57) A lubricant oil composition of the present invention that is prepared by blending a boron-containing compound therein, the boron-containing compound having a ratio of an integrated value of peaks in the 5 to 25 ppm chemical shift to an integrated value of peaks in the -10

ppm to 25 ppm chemical shift, as determined by 11 B-NMR measurement in deuterochloroform using BF_3 ·OEt₂ as an external standard (0 ppm), of 0.5 or higher and 1.0 or lower.

Description

Technical Field

[0001] The present invention relates to a lubricant oil composition, and, more particularly, to a lubricant oil composition suitable for internal combustion engines such as gasoline engines, diesel engines or engines using dimethyl ether as fuel, and gas engines.

Background Art

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[0002] In recent years, improvement of fuel-saving properties of automobiles is being considered in order to reduce energy loss and emission of carbon dioxide during automobile driving. As one measure to improve the fuel-saving properties of automobiles, weight reduction of automobile bodies is being promoted. However, lubricant oils are also required to contribute to the fuel-saving properties, and reduction of viscosity of lubricant oils and further improvement of friction reducing properties provided by lubricant oils are considered accordingly.

[0003] Conventionally, the use of a friction reducing agent is considered to achieve further improvement of friction reducing properties provided by lubricant oils. Known friction reducing agents include ashless-type friction modifiers such as fatty acid amides, fatty acid esters and aliphatic ethers, and molybdenum-based friction reducing agents such as molybdenum dithiocarbamate (refer to Patent Document 1, for example). Molybdenum-based friction reducing agents have a higher friction reducing effect than ashless-type friction modifiers, especially in a boundary lubrication area, and are therefore blended into many engine oils.

[0004] Boric acid-based lubricating additives are also conventionally known as additives having antiwear properties and lubricating properties. For example, Patent Document 2 describes that an organic boric acid ester may be blended together with a 1,3,4-thiadiazole compound, a dithiocarbamate and a non-sulfur molybdenum compound into a lubricant oil composition and that a synergistic wear-resistant effect in conjunction with these compounds is thereby obtained. The organic boric acid ester used in Patent Document 2 is a reaction product obtained by a reaction of a fatty oil and a diethanolamine followed by a reaction with boric acid.

Citation List

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Patent Literature

[0005]

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Patent Document 1: JP 2011-12213A Patent Document 2: JP 2006-502287A

Summary of Invention

40 Technical Problem

[0006] In recent years, the demand for reduction of fuel consumption is further increasing and lubricant oils are also required to have a lower viscosity and higher friction reducing properties accordingly. However, the friction coefficient of a lubricant oil cannot be sufficiently reduced by simply blending a molybdenum-based friction reducing agent into the lubricant oil, and therefore, a lubricating additive or a combination of additives that helps to further reduce friction is in demand.

[0007] On the other hand, conventional boron-based compounds, such as one disclosed in Patent Document 2, have relatively high antiware properties but is hardly effective in reducing the friction coefficient. Thus, the addition of a conventional boron-based compound hardly contributes to the ability of lubricant oils to reduce friction.

[0008] The present invention has been made in view of the above problems, and it is, therefore, an object of the present invention to provide a lubricant oil composition having improved friction reducing performance that helps to reduce friction and having high fuel-saving properties.

Solution to Problem

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[0009] The inventors of the present invention have conducted intensive studies to solve the above-mentioned problem. As a result, the inventors have surprisingly found that the blend of a specific boron-containing compound into a lubricant oil improves the ability of the lubricant oil to reduce friction, and have accomplished the present invention.

[0010] In other words, the present invention provides the following (1) to (14).

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- (1) A lubricant oil composition that is prepared by blending a boron-containing compound therein, the boron-containing compound having a ratio of an integrated value of peaks in the 5 to 25 ppm chemical shift to an integrated value of peaks in the -10 ppm to 25 ppm chemical shift, as determined by ^{11}B -NMR measurement in deuterochloroform using BF $_3$ ·OE $_2$ as an external standard (0 ppm), of 0.5 or higher and 1.0 or lower.
- (2) The lubricant oil composition according to (1) above, in which the boron-containing compound has a ratio of a boron atom content to a nitrogen atom content of 0.6 or higher on a mass basis.
- (3) The lubricant oil composition according to (1) or (2) above, in which the boron-containing compound is obtained by acting a boric acid or a boric acid derivative on an organic compound having a hydroxyl group or an amino group.
- (4) The lubricant oil composition according to (3) above, in which the organic compound is at least one compound selected from amine compounds (A), amide compounds having a hydroxyl group (B), imide compounds having an amino group (C), ester compounds having a hydroxyl group (D), and alcohol compounds having a hydroxyl group (E).
- (5) The lubricant oil composition according to any one of (1) to (4) above, in which the boron-containing compound is blended therein in an amount of 0.01 to 30% by weight.
- (6) The lubricant oil composition according to any one of (1) to (5) above, prepared by blending, in addition to the boron-containing compound, at least one of a viscosity index improver, a molybdenum compound, a zinc dialkyldithi-ophosphate, an antioxidant, a metal-based detergent and an ashless-type dispersant into a base oil composed of a mineral oil and/or a synthetic oil.
- (7) The lubricant oil composition according to any one of (1) to (6) above, being a lubricant oil composition for internal combustion engines.
- (8) A lubricant oil composition that is prepared by blending a boron-containing compound therein, the boron-containing compound obtained by heating and stirring an organic compound having a hydroxyl group or an amino group and a boric acid or a boric acid derivative at a temperature of 100°C or higher and 150°C or lower.
- (9) A lubricant oil composition, containing a boron-containing compound having a ratio of an integrated value of peaks in the 5 to 25 ppm chemical shift to an integrated value of peaks in the -10 ppm to 25 ppm chemical shift, as determined by ¹¹B-NMR measurement in deuterochloroform using BF₃·OEt₂ as an external standard (0 ppm), of 0.5 or higher and 1.0 or lower.
- (10) A lubricant oil composition, containing a boron-containing compound obtained by heating and stirring an organic compound having a hydroxyl group or an amino group and a boric acid or a boric acid derivative at a temperature of 100°C or higher and 150°C or lower.
- (11) The lubricant oil composition according to (9) or (10) above, further containing a base oil composed of a mineral oil and/or a synthetic oil, and at least one of a viscosity index improver, a molybdenum compound, a zinc dialkyldithiophosphate, an antioxidant, a metal-based detergent and an ashless-type dispersant.
- (12) A method for producing a lubricant oil composition, comprising: blending a boron-containing compound having a ratio of an integrated value of peaks in the 5 to 25 ppm chemical shift to an integrated value of peaks in the -10 ppm to 25 ppm chemical shift, as determined by 11 B-NMR measurement in deuterochloroform using BF $_3$ ·OEt $_2$ as an external standard (0 ppm), of 0.5 or higher and 1.0 or lower into a base oil composed of a mineral oil and/or a synthetic oil to produce the lubricant oil composition.
- (13) A method for producing a lubricant oil composition, comprising: obtaining a boron-containing compound by heating and stirring an organic compound having a hydroxyl group or an amino group and a boric acid or a boric acid derivative at a temperature of 100°C or higher and 150°C or lower; and blending the boron-containing compound into a base oil composed of mineral oil and/or a synthetic oil to produce a lubricant oil composition.
- (14) The method for producing a lubricant oil composition according to (12) or (13) above, further comprising: blending at least one of a viscosity index improver, a molybdenum compound, a zinc dialkyldithiophosphate, an antioxidant, a metal-based detergent and an ashless-type dispersant into the base oil. Advantageous Effect of Invention
- **[0011]** The lubricant oil composition of the present invention, which is reduced in friction coefficient and thereby improved in the ability to reduce friction by the blend of a specific boron-containing compound, can improve the fuel-saving properties of internal combustion engines and so on.

Description of Embodiments

55 [0012] A preferred embodiment of the present invention will be hereinafter described in detail.

[0013] A lubricant oil composition of the present invention is prepared by blending a boron-containing compound into a lubricant base oil (hereinafter occasionally referred simply to as "base oil").

[Boron-containing compound]

[0014] The boron-containing compound of the present invention is a boron-based lubricating additive obtained by acting a boric acid or a boric acid derivative on an organic compound having a hydroxyl group or an amino group, and, specifically, is a boron-containing compound obtained by heating and stirring an organic compound having a hydroxyl group or an amino group and boric acid or a boric acid derivative at 100°C or higher and 150°C or lower.

[0015] The boron-containing compound of the present invention improve the ability to reduce friction while improving the antiwear properties of the lubricant oil composition by decreasing the friction coefficient of the lubricant oil composition. [0016] The boron-containing compound of the present invention has a ratio of an integrated value of peaks in the 5 to 25 ppm chemical shift to an integrated value of peaks in the -10 to 25 ppm chemical shift, as determined by 11 B-NMR measurement in deuterochloroform using BF $_3$ ·OEt $_2$ as an external standard (0 ppm), of 0.50 or higher and 1.0 or lower. [0017] It is known that, in an 11 B-NMR measurement using BF $_3$ ·OEt $_2$ as a standard (0 ppm), peaks of three-coordinate boron-based compounds appear in the 5 to 25 ppm chemical shift. Thus, the integrated value ratio described above being 0.50 or higher means that the proportion of a three-coordinate boron-containing compound is high.

[0018] It is also known that peaks of four-coordinate boron-containing compounds appear in the -10 to 5 ppm range. Thus, the boron-containing compound of the present invention contains a three-coordinate boron-containing compound at a molar ratio of 0.50 or higher based on the total amount of three- and four-coordinate boron-containing compounds. [0019] The three-coordinate boron-based compound is a component that contributes to the reduction of friction. Thus, when the integrated value ratio or the molar ratio described above is lower than 0.50, the lubricant oil composition has a high friction coefficient, which means the lubricant oil composition cannot be improved in the ability to reduce friction. [0020] From the standpoint of the ability to further reduce the friction coefficient, the integrated value ratio described above (or the molar ratio described above) is preferably 0.55 or higher.

[0021] When the boron-containing compound of the present invention contains nitrogen atoms, the ratio of a boron atom content to a nitrogen atom content in the compound (B/N ratio) is preferably 0.6 or higher, more preferably 0.7 or higher, on a mass basis. Although not specifically limited, the B/N ratio is preferably 2.0 or lower, more preferably 1.5 or lower. In the present invention, when the B/N ratio is 0.6 or higher, the lubricant oil composition is suitably improved in the ability to reduce friction.

[0022] The boron-containing compound preferably has a boron atom content of 0.1 to 3% by mass, more preferably 1 to 3% by mass.

[0023] The boron-containing compound of the present invention is usually blended in the amount of 0.01 to 30% by mass, preferably 0.1 to 15% by mass, more preferably 0.5 to 5% by mass, of the lubricant oil composition.

[0024] In the present invention, when the amount of the boron-containing compound is equal to or greater than the above-mentioned lower limit value, the boron-containing compound can suitably exhibit the function of reducing the friction coefficient.

[0025] Examples of the boric acid derivative include orthoboric acid, metaboric acid, tetraboric acid, boron oxide, halogenated borons such as boron trifluoride, boron tribromide and boron trichloride, and borate esters such as trimethyl borate, triethyl borate, tributyl borate, triisopropyl borate and tributyl borate.

[0026] In the present invention, as the organic compound having a hydroxyl group or an amino group, there are mentioned at least one compound selected from amine compounds (A), amide compounds having a hydroxyl group (B), imide compounds having an amino group (C), ester compounds having a hydroxyl group (D), and alcohol compounds having a hydroxyl group (E), for example.

[0027] The compounds (A) to (E) are described in detail below.

[Amine compounds (A)]

[0028] The amine compounds (A) include amine compounds having one or more hydroxyl groups and one or more amino groups (a1), amine compounds having two or more amino groups (a2), amine compounds having one amino group only (a3), or the like.

[0029] More specifically, the amine compounds (a1) include the compounds represented by the general formulae (A1) and (A2) below, and the amine compounds (a1) or amine compounds (a2) include the compounds represented by the general formula (A3) or (A4) below. The amine compounds (a3) include the compounds represented by the general formula (A5) below.

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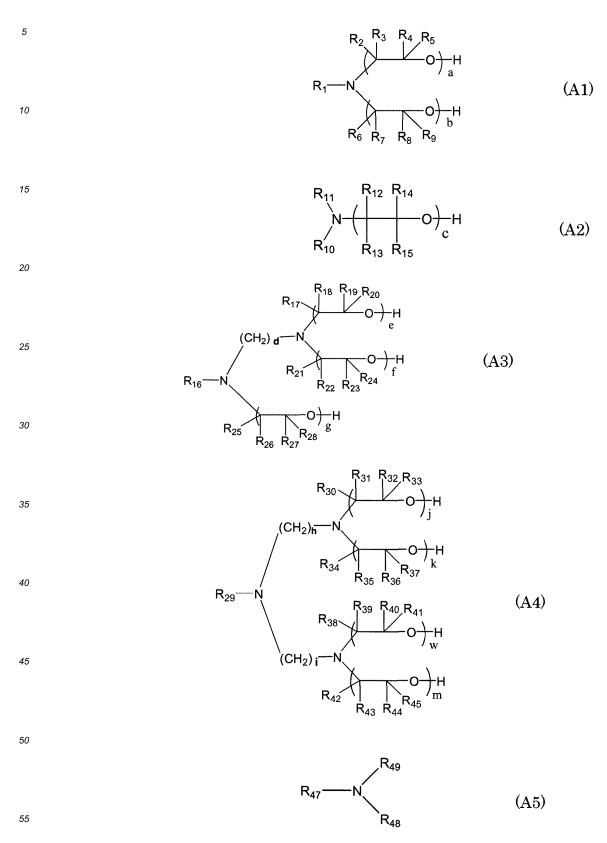
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[Chem 1]



 $\begin{tabular}{ll} \textbf{[0030]} & In formulae (A1) to (A5), R_1, R_{10}, R_{11}, R_{16}, R_{29} \ and R_{47} \ each \ represent a C_1 to C_{32} \ hydrocarbon group, and R_{10} \ and R_{11} \ may be the same or different from each other. The hydrocarbon groups may be saturated or unsaturated, and R_{11} \ may be the same or different from each other. The hydrocarbon groups may be saturated or unsaturated, and R_{12} \ may be the same or different from each other. The hydrocarbon groups may be saturated or unsaturated, and R_{12} \ may be the same or different from each other. The hydrocarbon groups may be saturated or unsaturated, and R_{12} \ may be the same or different from each other. The hydrocarbon groups may be saturated or unsaturated, and R_{13} \ may be the same or different from each other. The hydrocarbon groups may be saturated or unsaturated, and R_{13} \ may be the same or different from each other. The hydrocarbon groups may be saturated or unsaturated, and R_{13} \ may be the same of the following the foll$

aliphatic or aromatic, and linear, branched or cyclic. The examples of the hydrocarbon group is an aliphatic hydrocarbon group, such as an alkyl group or alkenyl group, or an aromatic hydrocarbon group.

[0031] Specific examples of the hydrocarbon groups include aliphatic hydrocarbon groups such as methyl group, ethyl group, propyl group, butyl group, butyl group, hexpl group, hexenyl group, octyl group, octenyl group, 2-ethylhexyl group, nonyl group, decyl group, undecyl group, decenyl group, dodecyl group, dodecenyl group, tridecyl group, tetradecyl group, tetradecyl group, pentadecyl group, hexadecyl group, hexadecenyl group, heptadecyl group, octadecyl group, octadecyl group, isostearyl group, oleyl group, linoleic group, nonadecyl group, icosyl group, eicosyl group, heneicosyl group, docosyl group, tricosyl group, tetracosyl group, pentacosyl group, hexacosyl group, heptacosyl group, octacosyl group, nonacosyl group, triacontyl group, hentriacontyl group, dotriacontyl group, decenetrimer group, polybutene group, cyclopentyl group, cyclohexyl group, methylcyclohexyl group, ethylcyclohexyl group, propylcyclohexyl group, dimethylcyclohexyl group, and trimethylcyclohexyl group, propylphenyl group, trimethylphenyl group, butylphenyl group, group; and so on.

[0032] The hydrocarbon groups are preferably C_4 to C_{22} hydrocarbon groups, more preferably C_6 to C_{18} hydrocarbon groups.

[0033] Symbols R_2 to R_9 , R_{12} to R_{15} , R_{17} to R_{28} , R_{30} to R_{45} and R_{48} to R_{49} each represent a hydrogen atom, a C_1 to C_{18} hydrocarbon group, or an oxygen-containing hydrocarbon group containing an ether bond or ester bond. These may be the same or different from each other, and each of them is preferably a hydrogen atom or a hydrocarbon group.

[0034] The hydrocarbon groups may be saturated or unsaturated, aliphatic or aromatic, and linear, branched or cyclic, and the examples of hydrocarbon group include an aliphatic hydrocarbon group such as an alkyl group or alkenyl group, or an aromatic hydrocarbon group. More specific examples thereof include aliphatic hydrocarbon groups such as methyl group, ethyl group, propyl group, butyl group, butenyl group, hexyl group, hexenyl group, octyl group, octenyl group, 2-ethylhexyl group, nonyl group, decyl group, undecyl group, decenyl group, dodecyl group, dodecenyl group, tridecyl group, tetradecyl group, tetradecenyl group, pentadecyl group, hexadecyl group, hexadecenyl group, heptadecyl group, octadecyl group, octadecyl group, stearyl group, isostearyl group, oleyl group, linoleic group, cyclopentyl group, cyclohexyl group, methylcyclohexyl group, ethylcyclohexyl group, propylcyclohexyl group, dimethylcyclohexyl group, ethylphenyl group, dimethylphenyl group, propylphenyl group, trimethylphenyl group, butylphenyl group and naphthyl group; and so on.

[0035] The hydrocarbon groups are preferably C_1 to C_{18} hydrocarbon groups, more preferably C_1 to C_{16} hydrocarbon groups, especially preferably C_1 to C_{12} hydrocarbon groups.

[0036] The oxygen-containing hydrocarbon group containing an ether bond or ester bond has 1 to 18 carbon atoms, and examples thereof include methoxymethyl group, ethoxymethyl group, propoxymethyl group, isopropoxymethyl group, n-butoxymethyl group, t-butoxymethyl group, hexyloxymethyl group, octyloxymethyl group, 2-ethyl-hexyloxymethyl group, decyloxymethyl group, dodecyloxymethyl group, 2-butyloctyloxymethyl group, tetradecyloxymethyl group, hexadecyloxymethyl group, 2-hexyldodecyloxymethyl group, allyloxymethyl group, phenoxy group, benzyloxy group, methoxyethyl group, methoxypropyl group, (1-methyl-2-methoxy)propyl group, acetyloxymethyl group, propanoyloxymethyl group, butanoyloxymethyl group, hexanoyloxymethyl group, octanoyloxymethyl group, 2-ethylhexanoyloxymethyl group, decanoyloxymethyl group, dodecanoyloxymethyl group, 2-butyloctanoyloxymethyl group, tetradecanoyloxymethyl group, hexadecanoyloxymethyl group, 2-hexyldodecanoyloxymethyl group, benzoyloxymethyl group, and the like.

[0037] In addition, a, b, c, e, f, g, j, k, w and m each represent an integer of 0 to 20, d, h and i each represent an integer of 1 to 6, provided that a+b is 1 to 20, e+f+g is 0 to 20 and j+k+w+m is 0 to 20.

[0038] Preferably (a+b) is 1 to 12, more preferably 1 to 10. Each of c, (e+f+g) and (j+k+w+m) is preferably 0 to 12, more preferably 0 to 10. Each of d, h, and i is preferably 2 to 4.

[0039] Preferably, in the general formula (A1), R_2 to R_5 and R_6 to R_9 are all hydrogen atoms, or R_2 to R_4 and R_6 to R_8 are all hydrogen atoms and either or both of R_5 and R_9 are hydrocarbon groups.

[0040] Preferably, in the general formula (A2), R_{12} to R_{14} are all hydrogen atoms and R_{15} is a hydrogen atom or hydrocarbon group.

[0041] Preferably, in the general formula (A3), each of e, f and g is 1 or greater and R₁₇ to R₂₈ are all hydrogen atoms. More preferably, e, f and g are all 1. As a matter of course, e, f and g may be all 0 in the general formula (A3), in which case no hydroxyl group is contained.

[0042] Preferably, in the general formula (A4), j, k, w and m are all 0.

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[0043] In addition, in the general formula (A5), R_{47} is preferably an alkyl group, and at least one of R_{48} and R_{49} may be a hydrocarbon group, in which case the hydrocarbon group is preferably an alkyl group.

[0044] Specific examples of the compounds of the general formula (A1) include amine compounds having one 2-hydroxyalkyl group, e.g., hydroxyethyl group, such as octyl ethanolamine, decyl ethanolamine, dodecyl ethanolamine, tetradecyl ethanolamine, hexadecyl ethanolamine, stearyl ethanolamine, oleyl ethanolamine, coconut oil ethanolamine, palm oil ethanolamine, rapeseed oil ethanolamine and beef tallow ethanolamine; amine compounds having two 2-

hydroxyalkyl groups such as octyl diethanolamine, decyl diethanolamine, dodecyl diethanolamine, tetradecyl diethanolamine, hexadecyl diethanolamine, stearyl diethanolamine, oleyl diethanolamine, coconut oil diethanolamine, palm oil diethanolamine, rapeseed oil diethanolamine, beef tallow diethanolamine, 1-[cyclohexyl(2-hydroxyethyl)amino]-dodecane-2-ol, N,N-bis(2-hydroxydodecyl)-N-cyclohexylamine, 1-[N-(2-hydroxyethyl)anilino]dodecane-2-ol, N,N-bis(2-hydroxydodecyl)-N-oleylamine and N,N-bis(2-hydroxydodecyl)-aniline; and amine compounds having a polyalkylene oxide structure such as polyoxyethylene octylamine, polyoxyethylene decylamine, polyoxyethylene tetradecylamine, polyoxyethylene hexadecylamine, polyoxyethylene stearylamine, polyoxyethylene oleylamine, polyoxyethylene beef tallow amine, polyoxyethylene coconut oil amine, polyoxyethylene palm oil amine, polyoxyethylene laurylamine, polyoxyethylene stearylamine, polyoxyethylene oleylamine and ethylene oxide-propylene oxide stearylamine. Among these, the amine compounds having two 2-hydroxyalkyl groups are preferred.

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[0045] In the case of the amine compounds having two 2-hydroxyalkyl groups, the ratio of an integrated value of peaks in the 5 to 25 ppm chemical shift to an integrated value of peaks in the -10 to 25 ppm chemical shift can be increased relatively easily. For example, the ratio can be 0.70 or higher and even 0.80 or higher. The amine compounds having two 2-hydroxyalkyl groups are compounds of formula (A1) in which each of a and b is 1.

[0046] As commercial products of the compounds represented by the general formula (A1), Amiet 105, Amiet 308 and Amiet 320 (these are manufactured by Kao Corporation), Nymeen L-202, Nymeen L-207, Nymeen S-202, Nymeen S-204, Nymeen S-210, Nymeen S-215, Nymeen S-220, Nymeen T2-206, Nymeen T2-210, Nymeen T2-230, Nymeen T2-260, Nymeen DT-203 and Nymeen DT-208 (these are manufactured by NOF CORPORATION), Kawasoft EP59S (manufactured by Kawaken Fine Chemicals Co., Ltd.), Ethomeen T/15, Ethomeen S/15 and Ethomeen O/15 (these are manufactured by Lion Corporation) and so on are available.

[0047] Specific examples of the compounds of the general formula (A2) include alkylamine compounds having one 2-hydroxyalkyl group, e.g., hydroxyethyl group, such as N-methyl-octyl ethanolamine, N-methyl-decyl ethanolamine, N-methyl-decyl ethanolamine, N-methyl-stearyl ethanolamine, N-methyl-oleyl ethanolamine, N-methyl-coconut oil ethanolamine, N-methyl-palm oil ethanolamine, N-methyl-rapeseed oil ethanolamine and N-methyl-beef tallow ethanolamine; and alkylamine compounds having a polyalkylene oxide structure such as polyoxyethylene N-methyl-decylamine, polyoxyethylene N-methyl-decylamine, polyoxyethylene N-methyl-tetradecylamine, polyoxyethylene N-methyl-hexadecylamine, polyoxyethylene N-methyl-stearylamine and polyoxyethylene N-methyl-oleylamine.

[0048] Specific examples of the compounds of the general formula (A3) include alkyl or alkenyl diamines such as N-lauryl-1,3-diaminopropane, N-stearyl-1,3-diaminopropane, N-oleyl-1,3-diaminopropane, N-coconut oil alkyl-1,3-diaminopropane, N-beef tallow alkyl-1,3-diaminopropane and N-hardened beef tallow-1,3-diaminopropane, as the amine compounds having two or more amino groups (a2).

[0049] As commercial products of the alkyl or alkenyl diamines, Duomeen CD, Duomeen T and Duomeen HT Flake (these are manufactured by Lion Corporation), and Nissan Amine DT, Nissan Amine DT-H and Nissan Amine DOB-R (these are manufactured by NOF CORPORATION), and so on are commercially available.

[0050] Specific examples of the compounds of the general formula (A3) also include diamine compounds having three 2-hydroxyalkyl groups, e.g., 2-hydroxyethyl groups, such as tris(2-hydroxyethyl)lauryl propylenediamine, tris(2-hydroxyethyl)stearyl propylenediamine, tris(2-hydroxyethyl)oleyl propylenediamine, tris(2-hydroxyethyl)coconut oil propylenediamine, tris(2-hydroxyethyl)palm oil propylenediamine and tris(2-hydroxyethyl)beef tallow propylenediamine; and diamine compounds having a polyalkylene oxide structure such as polyoxyethylene lauryl propylenediamine, polyoxyethylene stearyl propylenediamine, polyoxyethylene oleyl propylenediamine, polyoxyethylene coconut oil propylenediamine, polyoxyethylene palm oil propylenediamine and polyoxyethylene beef tallow propylenediamine, as the compounds having one or more hydroxyl groups and one or more amino groups (a1). Ethoduomeen T/13 and Ethoduomeen T/25 (these are manufactured by Lion Corporation), and Nymeen DT-203 and Nymeen DT-208 (these are manufactured by NOF CORPORATION) and so on are available as commercial products thereof.

[0051] Specific examples of the compounds represented by the general formula (A4) include alkyl or alkenyl triamines such as lauryl dipropylene triamine, stearyl dipropylene triamine, oleyl dipropylene triamine, coconut oil alkyl dipropylene triamine, palm oil alkyl dipropylene triamine, beef tallow alkyl dipropylene triamine and hardened beef tallow alkyl dipropylene triamine, as the compounds having two or more amino groups (a2). Triameen Y12D and Triameen T (these are manufactured by Lion Corporation) and so on are available as commercial products thereof.

[0052] Examples of the compounds represented by the general formula (A5) include amines having an alkyl or alkenyl group having 6 to 18 carbon atoms including primary amines such as hexylamine, octylamine, decylamine, dodecylamine, tetradecylamine, hexadecylamine, octadecylamine or oleylamine; secondary amines such as dihexylamine, dioctylamine, didodecylamine, didodecylamine, dihexadecylamine, dioctadecylamine and dioleylamine; and tertiary amine such as trihexylamine, trioctylamine, tridecylamine, tridecylamine, tridecylamine, tritetradecylamine, trihexadecylamine, trioctadecylamine and trioleylamine, as the amine compounds having one amino group only (a3). Preferred are the secondary or tertiary amines, and more preferred are the tertiary amines.

[0053] Among the amine compounds (A) described above, the amine compounds having hydroxyl group(s) and amino group(s) (e.g. amino alcohol compounds) (a1) preferably has 12 or more carbon atoms in total in the hydrocarbon group(s) in view of the solubility in the lubricant base oil. In addition, among the above amine compounds (A), the compounds represented by the general formula (A1) are preferred from the standpoint of the solubility in the lubricant base oil and the ability to further reduce the friction coefficient.

[0054] The amine compounds having hydroxyl group(s) and amino group(s) (e.g. amino alcohol compounds) (a1) are preferably compounds obtained by reacting a compound having at least one primary amino group and/or at least one secondary amino group (Aa) with a compound having an epoxy group (Ae).

[0055] Specific examples of the compound having at least one primary amino group and/or at least one secondary amino group (Aa) include primary amines and secondary amines. The primary amines preferably have a hydrocarbon group having 1 or more and 32 or less carbon atoms in total and may additionally contain an oxygen atom. The secondary amines preferably have 2 or more and 40 or less carbon atoms in total in the hydrocarbon groups, and may additionally contain an oxygen atom.

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[0056] When the total number of carbon atoms in the hydrocarbon group(s) in the primary amines or secondary amines is equal to or greater than the above lower limit, the amino alcohol compounds that are obtained from the primary amines or secondary amines are sufficiently soluble in the lubricant base oil or the like. When the total number of carbon atoms is equal to or smaller than the upper limit, amino alcohol compounds having a high base number are obtained.

[0057] The hydrocarbon group(s) that the primary and secondary amines have may be saturated or unsaturated, aliphatic or aromatic, and linear, branched or cyclic. Specific examples include aliphatic hydrocarbon groups such as methyl group, ethyl group, propyl group, butyl group, butenyl group, hexyl group, hexenyl group, octyl group, octenyl group, 2-ethylhexyl group, nonyl group, decyl group, undecyl group, decenyl group, dodecyl group, dodecenyl group, tridecyl group, tetradecyl group, tetradecenyl group, pentadecyl group, hexadecyl group, hexadecenyl group, octadecenyl group, stearyl group, isostearyl group, oleyl group, linoleic group, nonadecyl group, icosyl group, eicosyl group, heneicosyl group, docosyl group, tricosyl group, tetracosyl group, pentacosyl group, hexacosyl group, heptacosyl group, octacosyl group, nonacosyl group, triacontyl group, hentriacontyl group, dotriacontyl group, decenetrimer group, polybutene group, cyclopentyl group, cyclohexyl group, methylcyclohexyl group, ethylcyclohexyl group, propylcyclohexyl group, and dimethylcyclohexyl group, propylphenyl group, trimethylphenyl group, butylphenyl group and naphthyl group; and so on.

[0058] Specific examples of the primary amines include methylamine, ethylamine, butylamine, hexylamine, octylamine, decylamine, dodecylamine, tetradecylamine, hexadecylamine, octadecylamine, 2-ethylhexylamine, 2-decyltetradecylamine, oleylamine, ethanolamine, propanolamine, octadecyloxyethylamine, 3-(2-ethylhexyloxy)propylamine, 12-hydroxystearylamine, cyclohexylamine, aniline, and the like.

[0059] Specific examples of the secondary amines include dimethylamine, diethylamine, dibutylamine, dihexylamine, dioctylamine, didecylamine, didecylamine, didecylamine, didecylamine, didecylamine, didecylamine, diethylamine, dioctadecylamine, di2-ethylhexylamine, dioleylamine, methylstearylamine, methylstearylamine, diethanolamine, dipropanolamine, 2-butylaminoethanol, hexylaminoethanol, phenylaminoethanol and cyclohexylethanolamine. The examples also include cyclic secondary amines such as piperidine, piperazine and morpholine.

[0060] The compound having an epoxy group (Ae) is preferably a compound in which an epoxy group is directly bonded to a hydrocarbon group. The hydrocarbon group may be saturated or unsaturated, aliphatic or aromatic, and linear, branched or cyclic. The example of the hydrocarbon group include an aliphatic hydrocarbon group such as an alkyl group or alkenyl group, or an aromatic hydrocarbon group. More specifically, examples of the hydrocarbon group include aliphatic hydrocarbon groups such as methyl group, ethyl group, propyl group, butyl group, pentyl group, hexpl group, hexenyl group, octyl group, octenyl group, decyl group, decenyl group, dodecyl group, dodecenyl group, tetradecyl group, tetradecenyl group, hexadecyl group, hexadecenyl group, octadecyl group, octadecenyl group, isostearyl group, decenetrimer group, polybutene group, cyclopentyl group, cyclohexyl group, methylcyclohexyl group and dimethylcyclohexyl group; aromatic hydrocarbon groups such as phenyl group, methylphenyl group, ethylphenyl group, dimethylphenyl group, propylphenyl group, trimethylphenyl group, butylphenyl group and naphthyl group; and so on.

[0061] Specific examples of the compound having an epoxy group (Ae) include ethylene oxide, propylene oxide, 1,2-epoxybutane, 1,2-epoxypentane, 1,2-epoxyhexane, 1,2-epoxyoctane, 1,2-epoxydecane, 1,2-epoxydodecane, 1,2-epoxytetradecane, 1,2-epoxyhexadecane, 1,2-epoxyoctadecane, 1,2-epoxyetradecane, 1,2-epoxyhexadecane, 1,2-epoxyoctadecene, 1,2-epoxy-2-octyldodecane, and the like.

[0062] The amino alcohol compounds are preferably compounds obtained by reacting an amine compound (Aa) with an epoxy compound (Ae) at a ratio (molar ratio) of 1:0.7 to 1:12, more preferably at a ratio of 1:1 to 10:1.

[0063] The reaction between the amine compound (Aa) and the epoxy compound (Ae) is preferably carried out at a temperature of approximately 50°C to 250°C, more preferably at a temperature of approximately 80°C to 200°C.

[Amide compounds (B)]

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[0064] The amide compounds having a hydroxyl group (B) are amide alcohol compounds having one or more amide groups in each molecule and one or more hydroxyl groups in each molecule, and preferably are compounds represented by the general formula (B1) or the general formula (B2) below.

[Chem 2]

[0065] In formulae (B1) and (B2), R_{51} , R_{61} and R_{62} each represent a C_1 to C_{32} hydrocarbon group, and R_{61} and R_{62} may be the same or different from each other.

[0066] The hydrocarbon groups for R₅₁, R₆₁ and R₆₂ may be saturated or unsaturated, aliphatic or aromatic, and linear, branched or cyclic. The examples of the hydrocarbon groups include an aliphatic hydrocarbon group such as an alkyl group or alkenyl group, or an aromatic hydrocarbon group. Specific examples thereof include aliphatic hydrocarbon groups such as methyl group, ethyl group, propyl group, butyl group, butenyl group, hexyl group, hexenyl group, octyl group, octenyl group, 2-ethylhexyl group, nonyl group, decyl group, undecyl group, decenyl group, dodecyl group, tetradecyl group, tetradecyl group, pentadecyl group, hexadecyl group, hexadecyl group, hexadecyl group, octadecyl group, stearyl group, isostearyl group, oleyl group, linoleic group, nonadecyl group, icosyl group, eicosyl group, heneicosyl group, docosyl group, tricosyl group, tetracosyl group, pentacosyl group, hexacosyl group, heptacosyl group, octacosyl group, nonacosyl group, tricosyl group, hentriacontyl group, dotriacontyl group, decenetrimer group, polybutene group, cyclopentyl group, cyclohexyl group, methylcyclohexyl group, group, imethylcyclohexyl group, propylcyclohexyl group, methylcyclohexyl group, dimethylcyclohexyl group, dimethylcyclohexyl group, propylphenyl group, trimethylphenyl group, butylphenyl group and naphthyl group; and so on.

[0067] The hydrocarbon groups for R_{51} and R_{61} preferably have 4 to 22 carbon atoms, more preferably 6 to 18 carbon atoms. The hydrocarbon group for R_{62} preferably has 1 to 18 carbon atoms, more preferably 1 to 10 carbon atoms, especially preferably 1 to 4 carbon atoms.

[0068] Each of R_{52} to R_{59} and R_{63} to R_{66} is a hydrogen atom or C_1 to C_{18} hydrocarbon group, and these may be the same or different from each other. The hydrocarbon groups may be saturated or unsaturated, aliphatic or aromatic, and linear, branched or cyclic. The examples of the hydrocarbon group include an aliphatic hydrocarbon group such as an alkyl group or alkenyl group, or an aromatic hydrocarbon group. More specifically, examples thereof include aliphatic hydrocarbon groups such as methyl group, ethyl group, propyl group, butyl group, butenyl group, hexyl group, hexenyl group, octyl group, octenyl group, 2-ethylhexyl group, nonyl group, decyl group, undecyl group, decenyl group, dodecenyl group, tetradecyl group, tetradecenyl group, pentadecyl group, hexadecyl group, hexadecyl group, octadecyl group, octadecyl group, stearyl group, isostearyl group, oleyl group, linoleic group, cyclopentyl group, cyclohexyl group, methylcyclohexyl group, ethylcyclohexyl group, propylcyclohexyl group, methylcyclohexyl group, ethylcyclohexyl group, butylphenyl group, methylphenyl group, ethylphenyl group, butylphenyl group, butylphenyl group, butylphenyl group, butylphenyl group, butylphenyl group, propylphenyl group, trimethylphenyl group, butylphenyl group and naphthyl group; and so on.

[0069] The hydrocarbon groups for R_{52} to R_{59} , R_{63} to R_{66} preferably have 1 to 16 carbon atoms, more preferably 1

to 12 carbon atoms, especially preferably 1 to 10 carbon atoms.

[0070] Here, n represents an integer of 0 to 20, o and p each represent an integer of 1 to 20. In addition, n+o=1 to 20, and each of (n+o) and p is preferably 1 to 12, more preferably 1 to 10.

[0071] Preferably, in the general formula (B1), R_{52} to R_{55} and R_{56} to R_{59} are all hydrogen atoms, or R_{52} to R_{54} and R_{56} to R_{58} are all hydrogen atoms and either or both of R_{55} and R_{59} are hydrocarbon groups.

[0072] Preferably, in the general formula (B2), R_{63} to R_{65} are all hydrogen atoms and R_{66} is a hydrogen atom or hydrocarbon group.

[0073] Specific examples of the compounds of the general formula (B1) include fatty acid dialkanolamides, including fatty acid diethanolamides, such as octyl diethanolamide, decyl diethanolamide, dodecyl diethanolamide, stearyl diethanolamide, oleyl diethanolamide, coconut oil fatty acid diethanolamide, palm oil fatty acid diethanolamide, rapeseed oil fatty acid diethanolamide and beef tallow fatty acid diethanolamide.

[0074] As the fatty acid dialkanolamides, Stafoam F, Stafoam T, Stafoam FK, Stafoam DL, Stafoam DF-1, Stafoam DF-2, Stafoam DF-4, Stafoam DFC, Stafoam DO and Stafoam DOS (these are manufactured by NOF CORPORATION), Amizol CDE, Amizol FDE, Amizol KD-1, Amizol KD-3, Amizol CD, Amizol FD, Amizol CDC, Amizol MDE, Amizol SDE, Amizol SDHE, Amizol ODE and Amizol ODHE (these are manufactured by Kawaken Fine Chemicals Co., Ltd.), Polinon DAO, Polinon DAL-C and Polinon DAT (these are manufactured by Shoei Yakuhin Co., Ltd.), and so on are available as commercial products.

[0075] Specific examples of the compounds represented by the general formula (B1) include fatty acid amides having a polyalkylene oxide structure, including polyoxyalkylene fatty acid amides, such as polyoxyethylene octanoic acid amide, polyoxyethylene decanoic acid amide, polyoxyethylene lauric acid amide, polyoxyethylene myristic acid amide, polyoxyethylene palmitic acid amide, polyoxyethylene stearic acid amide, polyoxyethylene oleic acid amide, polyoxyethylene coconut oil fatty acid amide, polyoxyethylene palm oil fatty acid amide, polyoxyethylene rapeseed oil fatty acid amide and polyoxyethylene beef tallow fatty acid amide. For these compounds, a monoalkanolamide, in which only either n or o is 1 or greater, is usually used but a dialkanolamide, in which n and o are both 1 or greater, may be used. A mixture of these compounds may be used.

[0076] Specific examples of the compounds of the general formula (B2) include fatty acid N-alkyl monoalkanolamides, including fatty acid N-alkyl ethanolamides, such as N-methyl-octyl ethanolamide, N-methyl-decyl ethanolamide, N-methyl-dodecyl ethanolamide, N-methyl-oleyl ethanolamide, coconut oil fatty acid N-methyl ethanolamide, palm oil fatty acid N-methyl ethanolamide, rapeseed oil fatty acid N-methyl ethanolamide and beef tallow fatty acid N-methyl ethanolamide. As fatty acid N-alkyl monoalkanolamides, Ethomid HT/15, Ethomid HT/60 and Ethomid O/15 (these are manufactured by Lion Corporation), Amizol CME, Amizol SME, Amizol PCME, Amizol PLME-A, Amizett 2C, Amizett 10C, Amizett 2L-Y and Aminex HO (these are manufactured by Kawaken Fine Chemicals Co., Ltd.), Stafoam MF Pellet and Stafoam LIPA (these are manufactured by NOF CORPORATION), and Aminon C-11S (manufactured by Kao Corporation) are available as commercial products.

[0077] Among the above amide compounds (B), the compounds represented by the general formula (B2) are preferred from the standpoint of the ability to further reduce the friction coefficient.

[Imide compounds (C)]

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[0078] The imide compounds (C) have one or more imide groups in each molecule, and are preferably succinimides. Examples of the succinimides include the compounds represented by the general formula (C1) below.

R₆₉

$$N - \left(CH_2CH_2NH\right)_s CH_2CH_2NH_2$$
(C1)

[0079] In formula (C1), R_{69} represents a C_{20} to C_{150} hydrocarbon group. The hydrocarbon group may be saturated or unsaturated, aliphatic or aromatic, and linear or branched. The examples of the hydrocarbon group is an alkenyl group or alkyl group. In addition, s represents an integer of 0 to 5.

[0080] As the C_{20} to C_{150} alkenyl group or alkyl group, a polymer or copolymer of a C_2 to C_{16} monoolefin or diolefin, or a hydrogenation product thereof is usually used. Specific examples of the monoolefin include ethylene, propylene, butene, butadiene, decene, dodecene and hexadecene. Among these monoolefins, butene is especially preferred in

the present invention because of its ability to improve cleanness at high temperatures and easy availability. Its polymer, i.e., a polybutenyl group and a hydrogenated alkyl group thereof, i.e., a hydrogenated polybutenyl group, are preferred. When the alkenyl group or alkyl group has 20 or more carbon atoms, the succinimides are sufficiently soluble in the lubricant base oil and can exhibit their intended function, i.e., dispersion performance. When the number of carbon atoms is 150 or less, an increase in viscosity can be prevented.

[0081] In the case of the imide compounds (C), the above-mentioned ratio of an integrated value of peaks in the 5 to 25 ppm chemical shift to an integrated value of peaks in the -10 to 25 ppm chemical shift tends to be relatively low, and for example, the ratio is more preferably adjusted to 0.50 to 0.80, especially preferably to 0.55 to 0.70.

[0082] A succinimide represented by the general formula (C1) is obtained by reacting a C_{20} to C_{150} alkene with maleic acid anhydride at a reaction temperature of 50 to 280°C and then heating and stirring the reaction product together with a polyamine represented by the general formula (C2) below at a reaction temperature of 50 to 250°C.

[0083] At this time, the molar ratio of the C_{20} to C_{150} alkene to maleic acid anhydride is adjusted to 1:5 to 5:1, and the ratio of the alkenyl succinic anhydride to the polyamine (C2) is adjusted to 1:5 to 5:1.

[Chem 4]

$$N - \left(-CH_2CH_2NH \right) - CH_2CH_2NH_2$$
 (C2)

(wherein t represents an integer of 0 to 5).

[0084] Examples of the polyamine (C2) include alkylene diamines such as ethylenediamine, propanediamine, butane-diamine, N-methyl -1,3-propanediamine and N,N-dimethyl-1,3-propanediamine; polyalkylene polyamines such as diethylenetriamine, triethylenetetramine and tetraethylenepentamine; and polyalkylene polyamines having a cyclic alkylene amine such as aminoethylpiperazine.

[Ester compounds (D)]

[0085] The ester compounds (D) are compounds having one or more hydroxyl groups in each molecule. Examples of the ester compounds (D) include the compounds represented by the general formula (D1) or (D2) below.

[Chem 5]

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$$R_{101}$$
 R_{102}
 R_{104}
 R_{106}
 R_{1

[0086] In formulae (D1) and (D2), R_{81} and R_{101} each represent a C_1 to C_{32} hydrocarbon group. The hydrocarbon groups may be saturated or unsaturated, aliphatic or aromatic, and linear, branched or cyclic. The examples of the hydrocarbon group include an aliphatic hydrocarbon group such as an alkyl group or an alkenyl group, or an aromatic hydrocarbon group.

[0087] Examples of the hydrocarbon groups for R₈₁ and R₁₀₁ include aliphatic hydrocarbon groups such as methyl group, ethyl group, propyl group, butyl group, butenyl group, hexyl group, hexenyl group, octyl group, octenyl group, 2-ethylhexyl group, nonyl group, decyl group, undecyl group, decenyl group, dodecyl group, dodecenyl group, tridecyl group, tetradecyl group, tetradecyl group, pentadecyl group, hexadecyl group, hexadecenyl group, heptadecyl group, octadecyl group, octadecenyl group, stearyl group, isostearyl group, linoleic group, nonadecyl group, icosyl group, eicosyl group, heneicosyl group, docosyl group, tricosyl group, tetracosyl group, pentacosyl group, hexacosyl

group, heptacosyl group, octacosyl group, nonacosyl group, triacontyl group, hentriacontyl group, dotriacontyl group, decenetrimer group, polybutene group, cyclopentyl group, cyclohexyl group, methylcyclohexyl group, ethylcyclohexyl group, propylcyclohexyl group, dimethylcyclohexyl group and trimethylcyclohexyl group; aromatic hydrocarbon groups such as phenyl group, methylphenyl group, ethylphenyl group, dimethylphenyl group, propylphenyl group, trimethylphenyl group, butylphenyl group and naphthyl group; and so on.

[0088] The hydrocarbon groups for R_{81} and R_{101} preferably have 8 to 32 carbon atoms, more preferably 12 to 24 carbon atoms.

[0089] Each of R_{82} to R_{85} and R_{102} to R_{106} is a hydrogen atom or a C_1 to C_{18} hydrocarbon group, and these may be the same or different from each other. The hydrocarbon groups may be saturated or unsaturated, aliphatic or aromatic, and linear, branched or cyclic. The examples of the hydrocarbon group include an aliphatic hydrocarbon group such as alkyl group or alkenyl group, or an aromatic hydrocarbon group. More specifically, examples include aliphatic hydrocarbon groups such as methyl group, ethyl group, propyl group, butyl group, butenyl group, hexyl group, hexenyl group, octyl group, octenyl group, 2-ethylhexyl group, nonyl group, decyl group, undecyl group, decenyl group, dodecyl group, tetradecyl group, tetradecenyl group, pentadecyl group, hexadecyl group, hexadecyl group, hexadecyl group, octadecyl group, octadecyl group, stearyl group, isostearyl group, oleyl group, linoleic group, cyclopentyl group, cyclohexyl group, methylcyclohexyl group, ethylcyclohexyl group, propylcyclohexyl group, methylcyclohexyl group, ethylcyclohexyl group, butylphenyl group, methylphenyl group, ethylphenyl group, trimethylphenyl group, butylphenyl group and naphthyl group; and so on.

[0090] In addition, r represents an integer of 1 to 20, preferably 1 to 12, more preferably 1 to 10.

[0091] Preferably, in the general formula (D1), R_{82} to R_{85} are all hydrogen atoms, or R_{82} to R_{84} are all hydrogen atoms and R_{85} is a hydrocarbon group. Preferably, in the general formula (D2), R_{102} to R_{106} are all hydrogen atoms.

[0092] A compound represented by the general formula (D1) is obtained by a reaction of a fatty acid with an alkylene oxide, for example.

[0093] Examples of the fatty acid that is used to obtain a compound represented by the general formula (D1) include lauric acid, myristic acid, palmitic acid, oleic acid, beef tallow fatty acid, coconut oil fatty acid, and the like.

[0094] As the alkylene oxide, there are C_2 to C_{12} alkylene oxides, and specific examples thereof include ethylene oxide, propylene oxide, butylene oxide, hexylene oxide, octylene oxide, decylene oxide, dodecylene oxide, and the like. **[0095]** Examples of the compounds of the general formula (D1) include polyoxyethylene monolaurate, polyoxyethylene monostearate and polyoxyethylene monooleate. As the compounds of the general formula (D1), Nonion (L-2, L-4, S-2, S-4, S-6, S-10, S-15, S-15K, S-15.4, O-2, O-3, O-4, O-5 and O-6) (manufactured by Lion Corporation), and lonet series (MS-400, MS-1000, DL-200 and DS-4000, MO-400, MO-600, DO-400 and DO-600) (manufactured by Sanyo Chemical Industries, Ltd.) are available as commercial products.

[0096] Examples of the compounds represented by the general formula (D2) include glycerin fatty acid monoesters such as glycerol monolaurate, glycerol monostearate, glycerin monomyristate and glycerin monooleate. As commercial products of the compounds represented by the general formula (D2), Unigly GO-102R, Unigly GO-106, Unigly GL-106 and Unigly GS-106 (these are manufactured by NOF CORPORATION), Ryoto Polyglyester L-10D, Ryoto Polyglyester L-7D, Ryoto Polyglyester M-10D, Ryoto Polyglyester M-7D, Ryoto Polyglyester P-8D, Ryoto Polyglyester SWA-20D, Ryoto Polyglyester SWA-10D, Ryoto Polyglyester O-50D, Ryoto Polyglyester O-15D and Ryoto Polyglyester ER-60D (these are manufactured by Mitsubishi-Kagaku Foods Corporation), NIKKOL DGMO-CV, NIKKOL DGMO-90V, NIKKOL Tetraglyn1-OV, NIKKOL Decaglyn1-OV, NIKKOL Decaglyn 1-LN, NIKKOL Decaglyn 5-HS, NIKKOL Decaglyn 5-OV, NIKKOL Decaglyn 7-OV, NIKKOL Decaglyn 10-OV, NIKKOL Decaglyn 10-MAC, NIKKOL Decaglyn PR-20 (these are manufactured by Nikko Chemicals Co., Ltd.), and so on are available.

45 [Alcohol compounds (E)]

[0097] The alcohol compounds (E) are compounds having one or more hydroxyl groups in each molecule, examples of which include the compounds represented by the general formulae (E1), (E2) and (E3) below.

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[Chem 6]

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$$R_{71} = O = \begin{pmatrix} R_{72} & R_{74} \\ R_{71} & O = H \end{pmatrix}$$

$$R_{73} & R_{75}$$
(E1)

$$R_{91}$$
 R_{94}
 R_{94}
 R_{95}
 R_{96}
 R_{96}
 R_{98}
 R_{98}
 R_{98}
 R_{98}
 R_{98}
 R_{98}
 R_{98}
 R_{98}
 R_{98}

$$R_{111}$$
 R_{112}
 R_{113}
 R_{114}
 OH
 OH
 OH
 OH
 R_{115}
 R_{116}
 R_{116}
 R_{117}
 R_{118}
 R_{118}

[0098] In the general formulae (E1) to (E3), R_{71} , R_{91} and R_{111} each represent a C_1 to C_{32} hydrocarbon group. The hydrocarbon groups may be saturated or unsaturated, aliphatic or aromatic, and linear, branched or cyclic. The examples of the hydrocarbon group include an aliphatic hydrocarbon group such as alkyl group or alkenyl group, an aromatic hydrocarbon group, or the like. Examples of the hydrocarbon groups for R_{71} , R_{91} and R_{111} include aliphatic hydrocarbon groups such as methyl group, ethyl group, propyl group, butyl group, butenyl group, hexyl group, hexenyl group, octyl group, octenyl group, 2-ethylhexyl group, nonyl group, decyl group, undecyl group, decenyl group, dodecyl group, tetradecyl group, tetradecyl group, pentadecyl group, hexadecyl group, hexadecyl group, hexadecyl group, nonadecyl group, octadecyl group, octadecenyl group, stearyl group, isostearyl group, oleyl group, linoleic group, nonadecyl group, icosyl group, heptacosyl group, heneicosyl group, nonacosyl group, tricosyl group, tetracosyl group, dotriacontyl group, heptacosyl group, octacosyl group, nonacosyl group, triacontyl group, hentriacontyl group, dotriacontyl group, decenetrimer group, polybutene group, cyclopentyl group, cyclohexyl group, methylcyclohexyl group, ethylcyclohexyl group, propylcyclohexyl group, dimethylcyclohexyl group, group, propylcyclohexyl group, methylcyclohexyl group, dimethylcyclohexyl group, propylphenyl group, trimethylphenyl group, butylphenyl group and naphthyl group; and so on.

[0099] The hydrocarbon groups for R_{71} , R_{91} and R_{111} are preferably C_8 to C_{24} hydrocarbon groups, more preferably C_8 to C_{18} hydrocarbon groups.

[0100] Each of R_{72} to R_{75} , R_{92} to R_{98} and R_{112} to R_{118} is a hydrogen atom or C_1 to C_{18} hydrocarbon group, and these may be the same or different from each other. The hydrocarbon groups may be saturated or unsaturated, aliphatic or aromatic, and linear, branched or cyclic. The example of the hydrocarbon group include an aliphatic hydrocarbon group such as alkyl group or alkenyl group, or an aromatic hydrocarbon group. More specific examples include aliphatic hydrocarbon groups such as methyl group, ethyl group, propyl group, butyl group, butenyl group, hexyl group, hexenyl group, octyl group, octenyl group, 2-ethylhexyl group, nonyl group, decyl group, undecyl group, decenyl group, dodecyl group, tetradecyl group, tetradecyl group, pentadecyl group, hexadecyl group, hexadecyl group, octadecyl group, octadecenyl group, stearyl group, isostearyl group, linoleic group, cyclopentyl group, cyclohexyl group, methylcyclohexyl group, ethylcyclohexyl group, propylcyclohexyl group, methylcyclohexyl group, ethylphenyl group, dimethylcyclohexyl group, propylphenyl group, trimethylphenyl group, butylphenyl group, and naphthyl group; and so on.

[0101] The hydrocarbon groups are preferably C_1 to C_{16} hydrocarbon groups, more preferably C_1 to C_{12} hydrocarbon groups, especially preferably C_1 to C_{10} hydrocarbon groups.

[0102] In addition, q represents an integer of 0 to 20, preferably 0 to 12, more preferably 0 to 10.

[0103] Preferably, in the general formula (E1), R_{72} to R_{75} are all hydrogen atoms, or any one of them is a hydrocarbon

group and the others are hydrogen atoms. Preferably, in the general formula (E2), A is an oxygen atom and R_{92} to R_{98} are all hydrogen atoms. Preferably, in the general formula (E3), R_{112} to R_{117} are all hydrogen atoms and R_{117} is a hydrocarbon group.

[0104] Specific examples of the compounds of the general formula (E1), as the compounds in which q is 0 and which has one hydroxyl group in each molecule, include alcohol compounds, including linear alkanols, such as hexanol, heptanol, octanol, nonaol, decanol, undecanol, dodecanol, tridecanol, tetradecanol, pentadecanol, hexadecanol, nonadecanol, eicosanol, heneicosanol, docosanol, tricosanol, tetracosanol, pentacosanol, hexacosanol, heptacosanol, octacosanol, nonacosanol and triacontanol; linear alkenols such as hexenol, heptenol, octenol, nonenol, decenol, undecenol, dodecenol, tridecenol, tetradecenol, pentadecenol, pentacosenol, pentacosenol, hexacosenol, heptacosenol, octadecenol, nonacosenol, docosenol, tetracosenol, pentacosenol, hexacosenol, heptacosenol, octacosenol, nonacosenol and triacontenol; branched alkanols such as 2-ethylhexanol, 1-methylheptadecanol, 1-hexylheptanol, isodecanol and isotridecanol; and branched alkenols such as isohexenol, 2-ethylhexenol, isotridecenol, 1-methylheptadecenol, 1-hexylheptanol, isotridecenol and isooctadecenol, and so on.

[0105] Specific examples of the compounds represented by the general formula (E1), as the compounds in which q is 1 to 20, include alkylene oxide adducts and styrene oxide adducts of the above-mentioned various alcohol compounds. Here, examples of the alkylene oxide include C_2 to C_8 alkylene oxides, specific examples of which include ethylene oxide, propylene oxide, butylene oxide, hexylene oxide, octylene oxide, decylene oxide and dodecylene oxide.

[0106] The number of added moles of the alkylene oxide is preferably 1 to 20, more preferably 1 to 10, especially preferably 1 to 7, per mole of alcohol. When the number of added moles is in this range, the solubility in mineral oils is much better.

[0107] One kind of alkylene oxide may be added, or a mixture of two or more kinds of alkylene oxides may be added. When a mixture of alkylene oxides is added, the alkylene oxides may be in any form such as block, random or a combination thereof. Specific examples thereof include polyoxyethylene glycol, polyoxypropylene glycol, polyoxypropylene-ethylene glycol (binding mode of oxypropylene and oxyethylene: random), polyoxybutylene-ethylene glycol (binding mode of oxypropylene and oxyethylene: random), and polyoxyethylene-polyoxypropylene-polyoxyethylene glycol (binding mode of oxypropylene and oxyethylene: block).

[0108] The alcohol compounds (E) preferably has 12 to 30 carbon atoms, more preferably 12 to 24 carbon atoms, especially preferably 12 to 18 carbon atoms. When the number of carbon atoms in this range, the solubility in mineral oils is high.

[0109] Examples of commercial products of the compounds represented by the general formula (E1) in which q is 1 to 20 include Nonion (K-204, K-220 and K-230), Persoft (NK-60, NK-60C, NK-100 and NK-100C), and Nonion (P-208, P-210, P-213, E-202, E-202S, E-205S, E-205S, E-212, E-215, E-230, S-202, S-207, S-215, S-220, EH-204, EH-208, ID-203, ID-206, ID-209, EAD-13, TA-405, TA-407, TA-409, TA-411, TA-412, TA-413, TA-415 and TA-418) (these are manufactured by Lion Corporation), BLAUNON series (EL-1303, EN-1502, EN-905, EH-2, EH-4, DAL-2, DAH-3, NDB-2040, DAP-1008, ELP-0809B, ELP-1608B, EHP-4 and BPO-3), FINESURF series (NDB-800, IDEP-608, IDEP-604, IDEP-802, IDEP-560, IDEP-580, IDEP-9161, IDEP-5010, IDEP-9010, IDEP-7045, IDEP-7085, 5010, 7045, 7085, 9010, TDP-0108, TDP-0109, TDP-0255, TDP-0633K, TDP-1033 and TDP-1055), and WONDERSURF series (100,140, NDR-800, NDR-1000, NDR-1400, RL-80, RL-100, RL-140, ID-50, ID-70, ID-90, S-800, S-1000 and S-1400) (these are manufactured by AOKI OIL INDUSTRIAL CO., LTD.), Lionol series (TDL-20, TDL-30 and TDL-50) (these are manufactured by Lion Corporation), and New Pole series (LB-285, LB-625, LB-1715, 50HB-100, 50HB-260, 50HB-400, 50B-2000, 50HB-5100 and PP-400) (these are manufactured by Sanyo Chemical Industries, Ltd.).

[0110] Examples of the compounds represented by the general formula (E2) include 3-(dodecyloxy)propane-1,2-diol, 3-(tetradecyloxy)propane-1,2-diol, 3-(octadecyloxy)propane-1,2-diol, 3-(oley-loxy)propane-1,2-diol, 1, 2-dodecanediol, 1, 2-tetradecanediol, 1, 2-hexadecanediol and 1,2-octadecanediol.

[0111] Examples of the compounds represented by the general formula (E3) include lauryl glycol hydroxypropyl ether, myristyl glycol hydroxypropyl ether, stearyl glycol hydroxypropyl ether, oleyl glycol hydroxypropyl ether, and the like. Commercial products of the compounds represented by the general formula (E3) include Viscosafe LPE and Viscosafe LMPE (these are manufactured by Kawaken Fine Chemicals Co., Ltd.) and so on.

[0112] Among the above-mentioned alcohol compounds (E), the compounds represented by the general formula (E2) are preferred from the standpoint of the ability to further reduce the friction coefficient.

[0113] Among the above-mentioned compounds (A) to (E), the compounds (A) to (C) are preferred from the standpoint of friction reducing ability and improvement of the antiwear properties.

[Method for producing boron-containing compound]

[0114] The boron-containing compound according to the present invention is a boron-containing compound obtained by heating and stirring an organic compound having a hydroxyl group or an amino group and a boric acid or a boric acid derivative at a temperature of 100°C or higher and 150°C or lower, and usually contains a mixture of a three-coordinate

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boron-containing compound and a four-coordinate boron-containing compound, but it may be composed only of a three-coordinate boron-containing compound. Furthermore, the resulting boron-containing compound includes a boric acid or a boric acid derivative that is bonded to the compound having a hydroxyl group or an amino group or that is not bonded to but dispersed in the compound having a hydroxyl group or an amino group.

[0115] When the above-mentioned organic compound and boric acid are heated and stirred at a temperature higher than 150°C, the production of a three-coordinate boron compound decreases. In this case, a sufficient friction coefficient reducing effect cannot be obtained and the viscosity may increase. In particular, in case of the amide compounds (B), when the process is carried out at a high temperature, the friction coefficient is more likely to increase.

[0116] When heating and stirring is not carried out at a temperature of 100°C or higher, water generated by a dehydration reaction remains and causes hydrolysis of other additives in the fuel oil, lubricant oil, or the like. This accelerates deterioration of the fuel oil, lubricant oil, or the like and causes unstable performance.

[0117] In the present invention, the organic compound having a hydroxyl group or an amino group and a boric acid are preferably heated and stirred in a temperature range of 100 to 150°C for 30 minutes or longer and 30 hours or shorter, more preferably for 30 minutes or longer and 10 hours or shorter.

[0118] In the present invention, it is preferable that the organic compound having a hydroxyl group or an amino group and boric acid or a boric acid derivative be heated and stirred at a temperature of 100°C or higher and 130°C or lower to obtain the boron-containing compound of the present invention with acting the boric acid or the boric acid derivative on the organic compound.

[0119] Specifically, in the present invention, the organic compound having a hydroxyl group or an amino group and a boric acid or a boric acid derivative are first mixed, and the mixture is then dehydrated while being mixed and stirred at a temperature lower than 100°C (approximately 80°C, for example) to obtain a boric acid salt or boric acid dispersion. After that, the temperature is increased gradually or stepwise and the mixture is further dehydrated while being mixed and stirred in a temperature range of 100 to 150°C, preferably 100 to 130°C. As a result, a boron-containing compound that is composed of any one of a boric acid salt, a boric acid ester, a boric acid amide and a dispersion of boric acid and a dehydration condensation product of boric acid, or a boron-containing compound that is composed of a mixture of these is obtained.

[0120] In this process, the temperature is preferably increased stepwise or gradually also in the temperature range of 100 to 150°C.

[0121] The heating and stirring of the organic compound having a hydroxyl group or an amino group and a boric acid or a boric acid derivative may be carried out in the presence of a solvent; for example, an organic solvent such as a hydrocarbon oil.

[0122] The timing of addition of the solvent is not particularly limited. The solvent may be added to the organic compound before a boric acid or a boric acid derivative is mixed with the organic compound or may be added after a boric acid or a boric acid derivative is mixed with the organic compound. Alternatively, after heating is carried out in a temperature range of 100 to 150°C for a predetermined period of time, the solvent may be added and then heating may be subsequently continued in a temperature range of 100 to 150°C for a predetermined period of time.

[0123] When the heating and stirring is carried out, the pressure may be reduced so that water can be easily removed. **[0124]** However, in the present invention, in the series of steps of mixing the organic compound having a hydroxyl group or an amino group and a boric acid or a boric acid derivative and heating and stirring the mixture to obtain a boroncontaining compound, the mixture is not heated to a temperature higher than 150°C, and is not heated to a temperature higher than 130°C preferably. In the present invention, because the mixture is not subjected to heating at a high temperature, the production of a three-coordinate boron compound can be increased.

[0125] The boron-containing compound is preferably a compound obtained by heating and stirring the organic compound having a hydroxyl group or an amino group and a boric acid or a boric acid derivative with the ratio of the number of moles of the organic compound having a hydroxyl group or an amino group to the number of moles of boric acid or a boric acid derivative being from 1:0.01 to 1:10, more preferably with the ratio being from 1:0.05 to 1:8. Here, when the ratio of the numbers of moles is 1:0.01 or greater, a boron-containing compound having a low friction coefficient and excellent antiwear properties can be obtained. On the other hand, when the ratio of the numbers of moles is 1:10 or smaller, the boron-containing compound has high solubility in the lubricant base oil or fuel oil.

[Lubricant base oil]

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[0126] The base oil used in the present invention is not specifically limited, and any conventionally known and used mineral oil and/or synthetic oil can be used.

[0127] Examples of the mineral oil include distillate oils obtained by subjecting a paraffin-base crude oil, intermediate base crude oil or naphthene-base crude oil to atmospheric distillation or obtained by subjecting an atmospheric distillation residue oil to distillation under a reduced pressure; and purified oils obtained by purifying the distillate oils according to an ordinary method, such as solvent-purified oils, hydrogenation-purified oils, hydrocracked oils, dewaxing treated oils

and white clay treated oils. In addition, isomerized oils of waxes (such as slack wax) can be also used.

[0128] Examples of the synthetic oil include poly α -olefins which are C_8 to C_{14} α -olefin oligomers, polybutene, polyol esters, alkylbenzenes, and so on.

[0129] In the present invention, the above-mentioned mineral oils may be used singly or in combination of two or more kinds as a base oil. Alternatively, the above-mentioned synthetic oils may be used singly or in combination of two or more kinds. Further, one or more kinds of the mineral oils and one or more kinds of the synthetic oils may be used in combination.

[0130] The lubricant oil composition of the present invention usually contains the base oil in addition to the above-described boron-containing compound, and the content of the base oil in the composition is preferably 70% by mass or higher, more preferably 80% by mass or higher.

[0131] The base oil usually has a kinetic viscosity at 100°C of 1.5 to 50 mm²/s, preferably 3 to 30 mm²/s. When the kinetic viscosity at 100°C is 1.5 mm²/s or higher, the evaporation loss will be small. When the kinetic viscosity is 50 mm²/s or lower, power loss due to viscosity resistance is reduced and a high fuel efficiency improving effect can be achieved.

[0132] In addition, the base oil has a viscosity index of 80 or higher, preferably 90 or higher, especially preferably 100 or higher. When the viscosity index is 80 or higher, the change in viscosity of the base oil with change in temperature will be so small that stable lubricating performance can be achieved.

[0133] The base oil preferably has a sulfur content, as measured according to JIS K 2541, of 50 ppm by mass or lower. When the sulfur content is 50 ppm by mass or lower, the base oil is effective in improving the wear resistance of low-friction sliding materials. The sulfur content is more preferably 30 ppm by mass or lower, much more preferably 20 ppm by mass or lower.

[0134] The lubricant oil composition of the present invention is preferably prepared by additionally blending one or more of a viscosity index improver, a molybdenum compound, a zinc dialkyldithiophosphate, an antioxidant, a metal-based detergent and an ashless-type dispersant into the base oil. The boron-containing compound of the present invention can sufficiently reduce the friction coefficient of a lubricant oil composition in which these additives are blended. **[0135]** Each additive is described in detail below.

[Viscosity index improver]

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[0136] Examples of the viscosity index improver include non-dispersion type polymethacrylates, dispersion-type polymethacrylates, olefin-based copolymers (such as ethylene-propylene copolymer), dispersion-type olefin-based copolymers, styrene-based copolymers (such as styrene-diene hydrogenated copolymer), and so on, and the non-dispersion type polymethacrylates such as polyalkyl methacrylates are preferred. The viscosity index improver has a weight-average molecular weight of 10,000 to 1,000,000, for example, preferably 50,000 to 600,000.

[0137] The viscosity index improver is usually blended in the lubricant oil composition in an amount of 0.5 to 30% by mass, preferably 2 to 20% by mass.

[Molybdenum compound]

[0138] The molybdenum compound is used as a friction modifier, and one example is a compound represented by the general formula (F) below.

[Chem 7]

$$R_{151}$$
 R_{152} R_{152} R_{154} R_{154} R_{154} R_{154} R_{154} R_{154} R_{154} R_{154}

[0139] In formula (F), R_{151} to R_{154} each represent a C_4 to C_{22} hydrocarbon group, and R_{151} to R_{154} may be the same or different from each other. When the number of carbon atoms is 4 or greater, the molybdenum compound has high oil solubility. When the number of carbon atoms is 22 or smaller, the molybdenum compound not only has a proper melting point and therefore is easy to handle but also has excellent friction reducing ability. From the above standpoint,

the number of carbon atoms is preferably 4 to 18.

[0140] Examples of the hydrocarbon group include alkyl groups, alkenyl groups, alkylaryl groups, cycloalkyl groups and cycloalkenyl groups. Branched or linear alkyl or alkenyl groups are preferred, and branched or linear alkyl groups are more preferred. Examples of the C_4 to C_{18} branched or linear alkyl groups include various kinds of butyl groups, various kinds of hexyl groups, various kinds of octyl groups, various kinds of nonyl groups, various kinds of decyl groups, various kinds of hexadecyl groups, various kinds of octadecyl groups, various kinds of hexadecyl groups, various kinds of octadecyl groups, and so on. From the standpoint of solubility in the base oil, storage stability and friction reducing ability, it is preferred that R_{151} and R_{152} be the same alkyl group, R_{153} and R_{154} be the same alkyl group, and the alkyl group for R_{151} and R_{152} and the alkyl group for R_{153} and R_{154} be different from each other.

[0141] In formula (F), X^1 to X^4 each represent a sulfur atom or oxygen atom, and X^1 to X^4 may be the same or different from each other. Preferably, the ratio of the sulfur atom content to the oxygen atom content (sulfur atom/oxygen atom) is 1/3 to 3/1, more preferably 1.5/2.5 to 3/1. When the ratio is in the above range, good performance can be obtained in terms of corrosion resistance and solubility in the base oil. Alternatively, all of X^1 to X^4 may be sulfur atoms or oxygen atoms.

[0142] In the present invention, the friction coefficient of the lubricant oil composition can be successfully reduced by blending the molybdenum compound in addition to the above-mentioned boron-containing compound.

[0143] The organic molybdenum compound is usually blended in the lubricant oil composition in an amount, expressed as molybdenum content, of 0.005 to 0.5% by mass, preferably 0.01 to 0.15% by mass.

[Zinc dialkyldithiophosphate]

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[0144] The zinc dialkyldithiophosphate is used as an antiwear agent, and one example is the compound represented by the general formula (G) below.

[Chem 8]

$$R_{155}O$$
 P S S OR_{157} $R_{156}O$ S S OR_{158} (G)

[0145] In the general formula (G), R_{155} to R_{158} each independently represent an alkyl group, preferably a C_1 to C_{24} alkyl group.

[0146] The C₁ to C₂₄ alkyl group may be linear, branched or cyclic. Specific examples thereof include methyl group, ethyl group, various kinds of propyl groups, various kinds of butyl groups, various kinds of pentyl groups, various kinds of hexyl groups, various kinds of octyl groups, various kinds of nonyl groups, various kinds of decyl groups, various kinds of undecyl groups, various kinds of dodecyl groups, various kinds of tridecyl groups, various kinds of tetradecyl groups, various kinds of pentadecyl groups, various kinds of hexadecyl groups, various kinds of heptadecyl groups, various kinds of octadecyl groups, various kinds of nonadecyl groups, various kinds of icosyl groups, various kinds of heneicosyl groups, various kinds of docosyl groups, various kinds of tricosyl groups and various kinds of tetracosyl groups; and cyclopentyl group, cyclohexyl group, cycloheptyl group, and alkyl substitution products thereof; and the like.

[0147] The zinc dialkyldithiophosphate is usually blended in the lubricant oil composition in an amount of 0.01 to 10% by mass, preferably 0.1 to 5% by mass.

[Antioxidant]

[0148] Examples of the antioxidant include phenol-based antioxidants and amine-based antioxidants.

[0149] Examples of the phenol-based antioxidants include 4,4'-methylenebis(2,6-di-t-butylphenol); 4,4'-bis(2,6-di-t-butylphenol); 2,2'-methylenebis(4-ethyl-6-t-butylphenol); 2,2'-methylenebis(4-methyl-6-t-butylphenol); 4,4'-bis(2-methylenebis (3-methyl-6-t-butylphenol); 4,4'-isopropylidenebis(2,6-di-t-butylphenol); 2,2'-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4-methylenebis(4

[0150] Examples of the amine-based antioxidants include monoalkyldiphenylamines such as monooctyldiphenylamine and monononyldiphenylamine; dialkyldiphenylamines such as 4,4'-dibutyldiphenylamine 4,4'-dipentyldiphenylamine, 4,4'-dipentyldiphenylamine, 4,4'-dibutyldiphenylamine and 4,4'-dinonyldiphenylamine; polyalkyldiphenylamines such as tetrabutyldiphenylamine, tetrahexyldiphenylamine, tetraoctyldiphenylamine and tetranonyldiphenylamine; naphthylamines such as α -naphthylamine; phenyl- α -naphthylamine; alkyl-substituted phenyl- α -naphthylamines such as butylphenyl- α -naphthylamine, pentylphenyl- α -naphthylamine, hexylphenyl- α -naphthylamine, octylphenyl- α -naphthylamine and nonylphenyl- α -naphthylamine, and the like.

[0151] One kind of the above-mentioned antioxidants may be selected and used or two or more kinds of the above-mentioned antioxidants may be used in combination. Among the above-mentioned antioxidants, dialkyldiphenylamines are preferred.

[0152] The antioxidant is usually blended in the lubricant oil composition in an amount of 0.05 to 10% by mass, preferably 0.1 to 5% by mass.

[Metal-based detergent]

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[0153] As the metal-based detergent, any alkaline-earth metal-based detergent that has been used in lubricant oils can be used. Examples thereof include alkaline-earth metal sulfonates, alkaline-earth metal phenates and alkaline-earth metal salicylates, mixtures of two or more kinds selected from these compounds, and the like.

[0154] Examples of the alkaline-earth metal sulfonates include alkaline-earth metal salts, especially, magnesium salts and/or calcium salts, of alkyl aromatic sulfonic acids obtained by sulfonation of an alkyl aromatic compound having a molecular weight of 300 to 1,500, preferably 400 to 700. Above these, calcium salts are preferably used.

[0155] Examples of the alkaline-earth metal phenates include alkaline-earth metal salts, especially magnesium salts and/or calcium salts, of alkylphenols, alkylphenol sulfides, Mannich reaction products of alkylphenols, and the like. Above these, calcium salts are especially preferably used.

[0156] Examples of the alkaline-earth metal salicylates include alkaline-earth metal salts, especially magnesium salts and/or calcium salts, of alkyl salicylic acids. Above these, calcium salts are preferably used.

[0157] Among these metal-based detergents, the use of a calcium salicylate is preferred.

[0158] In the present invention the metal-based detergent usually has a total base number of 10 to 500 mgKOH/g, preferably 20 to 450 mgKOH/g.

[0159] The term "total base number" used herein refers to the total base number measured according to the perchloric acid method of JIS K-2501.

[0160] The metal-based detergent is usually blended in the lubricant oil composition in an amount of 0.1 to 10% by mass, preferably 0.5 to 5.0% by mass.

35 [Ashless-type dispersant]

[0161] As the ashless-type dispersant, an imide-based dispersant that is free of boron is preferably used. As the imide-based dispersant, the succinimides represented by the general formula (C1) are preferred, and a polybutenyl succinimide is more preferred, in which R_{69} is a polybutenyl group.

[0162] The ashless-type dispersant is usually blended in the lubricant oil composition in an amount of 0.1 to 10% by mass, preferably 1 to 5% by mass.

[Other additives]

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[0163] The lubricant composition of the present invention may be prepared by blending, in addition to the above-mentioned additive(s), a pour-point depressant, an antirust, a corrosion inhibitor, an antifoaming agent and so on therein.
[0164] As the pour-point depressant, a polymethacrylate having a weight-average molecular weight of approximately 10,000 to 150,000 and so on can be used, for example. Examples of the antirust include alkylbenzene sulfonates, dinonylnaphthalene sulfonates, alkenyl succinic acid esters, polyhydric alcohol esters, and the like. Examples of the corrosion inhibitor include benzotriazole-based corrosion inhibitors, benzimidazole-based corrosion inhibitors, thiadiazole-based corrosion inhibitors, and the like. Examples of the antifoaming agent include dimethylpolysiloxanes, polyacrylates, and the like.

[0165] The lubricant oil composition that is prepared by blending the boron-containing compound and optionally blending various additives selected from a viscosity index improver, a molybdenum compound, a zinc dialkyldithiophosphate, an antioxidant, a metal-based detergent, an ashless-type dispersant and other additives as described above usually contains the blended compound(s) itself, but in some cases at least some portion of the blended compound(s) may be reacted to form different compounds.

[0166] A method for producing a lubricant oil composition according to the present invention comprises: blending the

above-mentioned boron-containing compound into a base oil to produce a lubricant oil composition. Alternatively, the method for producing a lubricant oil composition of the present invention may comprise: blending a boron-containing compound obtained by the method for producing a boron-containing compound as described above into a base oil to produce a lubricant oil composition. In these production methods, at least one of the above-mentioned various kinds of additives, including a viscosity index improver, a molybdenum compound, a zinc dialkyldithiophosphate, an antioxidant, a metal-based detergent, an ashless-type dispersant and other additives, may be additionally blended into the base oil, if necessary.

[0167] The lubricant oil composition of the present invention usually has a kinetic viscosity at 100°C of 1 to 30 mm²/s, preferably 2 to 15 mm²/s, and usually has a kinetic viscosity at 40°C of 5 to 100 mm²/s, preferably 20 to 80 mm²/s. The lubricant oil composition usually has a viscosity index of 90 or higher, preferably 100 or higher.

[0168] The lubricant oil composition of the present invention is improved in friction reducing ability and has excellent antiwear properties by the effect of the boron-containing compound. Thus, the lubricant oil composition of the present invention can be suitably used in internal combustion engines. The lubricant oil composition of the present invention can improve fuel efficiency by reducing friction between sliding parts in engines for example.

[Examples]

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[0169] While the present invention is described in more detail by way of examples, the present invention is by no means limited by these examples. The various physical properties of boron-containing compounds, base oils, and lubricant oil compositions were measured according to the following methods.

(1) ¹¹B-NMR

[0170] The ¹¹B-NMR measurement was conducted in deuterochloroform at a pulse width of 90 degrees using BF₃·OEt₂ as an external standard (0 ppm). (2) Measurement of boron content and nitrogen content in boron-containing compound [0171] The boron content is a value measured in compliance with ASTM D4951. The nitrogen content is a value measured in compliance with JIS K2609.

(3) Kinetic viscosity and viscosity index

[0172] The kinetic viscosity and viscosity index are values measured in compliance with JIS K2283.

(4) Friction coefficient

[0173] Using an MTM (Mini Traction Machine) tester (PCS Instruments), a preliminary operation (rubbing) was carried out for 60 minutes at an oil temperature of 80°C, a load of 30 N, a speed of 1000 mm/s and a slide roll ratio (SRR) of 50%, and then the friction coefficient was measured while the test was conducted at an oil temperature of 80°C, a load of 30N, a speed of 10 m/s and an SRR of 50%.

40 Production Example 1

[0174] 550 Grams of polybutene (Mn: 960), 1.5 g of cetyl bromide and 59 g (0.6 mol) of maleic anhydride were placed in a 1L autoclave and then after nitrogen substitution the reaction was carried out at 240°C for 5 hours. Then, the temperature was lowered to 215°C, and unreacted maleic anhydride and cetyl bromide were removed by distillation under a reduced pressure. Then, the temperature was lowered to 140°C and the residue was filtered. The yield of the obtained polybutenyl succinic anhydride were 550 g and the saponification value thereof 86 mgKOH/g.

[0175] 200 Grams of the polybutenyl succinic anhydride, 7.2 g (0.056 mol) of aminoethylpiperazine (AEP), 4.3 g (0.042 mol) of diethylenetriamine (DETA), 6.1 g (0.042 mol) of triethylene tetraamine (TETA) and 100 g of a 150 neutral mineral oil were placed in a 500 mL separable flask, and the reaction was carried out under a nitrogen stream at 150°C for 2 hours. Then, the temperature was raised to 200°C, and unreacted AEP and DETA and formed water were removed by distillation under a reduced pressure. The yield of the obtained polybutenyl succinimide was 315 g, and the base value (hydrochloric acid method) thereof was 30.8 mgKOH/g.

[0176] 110 Grams of the polybutenyl succinimide was charged in a separable flask reactor equipped with a stirrer, a heater, a thermometer, a Dean-Stark tube, a Dimroth condenser tube and a nitrogen blowing tube, and heated to 70 to 80°C under a nitrogen stream. To this mixture, 19.1 g (0.308 mol) of boric acid was gradually added, and then, the mixture was stirred and heated at 80°C for 2 hours. The mixture was further heated and stirred at 90 to 100°C for 2 hours and then at 150°C for 2 hours while being dehydrated. The obtained mixture was subjected to pressure filtration (pressure: 4.0 kg/cm²) using a filtration plate (NA-600, retention particle diameter: 0.4 µm, manufactured by Advantec

Toyo Kaisha, Ltd.), which gave 110 g of a boron-containing polybutenyl succinimide (boron-containing compound 1). The boron-containing polybutenyl succinimide had a boron content of 2.1% by mass, and a boron atom/nitrogen atom mass ratio of 1.0.

5 Production Example 2

[0177] The same procedure as in Production Example 1 was repeated except that the amount of boric acid was changed to 13.6 g (0.220 mol) to obtain a boron-containing polybutenyl succinimide (boron-containing compound 2). The boron-containing polybutenyl succinimide had a boron content of 1.5% by mass, a nitrogen content 2.1% by mass, and a boron atom/nitrogen atom mass ratio of 0.7.

Production Example 3

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[0178] 129.2 Grams (1.00 mol) of 1-octylamine and 368.6 g (2.00 mol) of 1,2-epoxydodecane were charged in a separable flask reactor equipped with a stirrer, a heater, a thermometer, a Dean-Stark tube, a Dimroth condenser tube and a nitrogen blowing tube. The mixture was heated and stirred under a nitrogen atmosphere at 80°C for 2 hours and then at 120°C for 2 hours and then further heated and stirred at 160°C for 12 hours to obtain N,N-bis(2-hydroxydodecyl)-N-octylamine.

[0179] To the reaction product, 61.8 g (1.00 mol) of boric acid was gradually added under a nitrogen stream, and then the mixture was stirred at 80°C for 2 hours. The mixture was further heated and stirred at 90 to 100° C for 2 hours and then at 110° C to 120° C for 3 hours while being dehydrated. Then, 113 Grams of mineral oil A (kinetic viscosity at 100° C = 4.12 mm^2 /s, viscosity index = 134) was added, and the mixture was heated and stirred at 120° C for 30 minutes. The obtained mixture was subjected to pressure filtration (pressure 4.0 kg/cm^2) using a filtration plate (NA-600, retention particle diameter: $0.4 \mu \text{m}$, manufactured by Advantec Toyo Kaisha, Ltd.), which gave 609 g of a boron-containing N,N-bis (2-hydroxydodecyl)-N-octylamine (boron-containing compound 3). The obtained boron-containing compound had a boron content of 1.7% by mass, a nitrogen content of 2.1% by mass and a boron atom/nitrogen atom mass ratio of 0.79.

Production Comparative Example 1

[0180] The same procedure as in Production Example 2 was repeated except that after the addition of boric acid and 2 hour stirring at 80°C, the mixture was heated and stirred at 90 to 100°C for 2 hours, at 110 to 120°C for 2 hours and at 150 to 160°C for 3 hours and then filtration was carried out in the same manner as in Production Example 2 to obtain a boron-containing polybutenyl succinimide (boron-containing compound 4).

35 Production Comparative Example 2

[0181] 123.78 Grams (0.35 mol) of 2-decyltetradecane-1-amine was charged in a separable flask reactor equipped with a stirrer, a heater, a thermometer, a Dean-Stark tube, a Dimroth condenser tube and a nitrogen blowing tube, and heated to 70 to 80°C under a nitrogen stream. To the mixture, 21.64 g (0.35 mol) of boric acid was gradually added, and then, the mixture was stirred at 80°C for 2 hours. Then, the mixture was further heated and stirred at 90 to 100°C for 2 hours, at 110°C to 120°C for 2 hours and at 150 to 160°C for 3 hours while being dehydrated. The obtained mixture was subjected to pressure filtration (pressure: 4.0 kg/cm²) using a filtration plate (NA-600, retention particle diameter: 0.4 μm, manufactured by Advantec Toyo Kaisha, Ltd.), which gave 125 g of a boron-containing 2-decyltetradecane-1-amine (boron-containing compound 5). The boron-containing 2-decyltetradecane-1-amine had a boron content of 2.8% by mass, and a boron atom/nitrogen atom mass ratio of 1.0.

[0182] Each of the boron-containing compounds 1 to 5 obtained in Production Examples 1 to 3 and Comparative Examples 1 and 2 was subjected to measurement for boron atom content and nitrogen atom content, and the ratio of a boron atom content to a nitrogen atom content (on a mass basis) was obtained. Also, each of the boron-containing compounds 1 to 5 obtained in Production Examples 1 to 3 and Comparative Example 1 and 2 was subjected to ¹¹B-NMR measurement, and the ratio [integrated value of peaks in the 5 to 25 ppm chemical shift / integrated value of peaks in the -10 to 25 ppm chemical shift] of each boron-containing compound was calculated. The integrated value ratios are shown in Table 1.

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Table 1

	Example			Comparative Example		
	1	2	3	1	2	
Boron atom content (% by mass)	2.1	1.5	1.7	1.0	2.7	
Nitrogen atom content (% by mass)	2.1	2.1	2.1	2.1	2.8	
Integrated value ratio from boron NMR	0.63	0.56	1.00	0.47	0.36	
Boron atom content / nitrogen atom content	1.0	0.7	0.8	0.5	1.0	

Examples 4 to 6, and Comparative Examples 3 and 4

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[0183] Using the boron-containing compounds 1 to 5 produced in Production Examples 1 to 3 and Comparative Examples 1 and 2, lubricant oil compositions having a formulation shown in Table 2 below were prepared. For each lubricant oil composition, the friction coefficient was calculated according to the above-mentioned method. The results are shown in Table 2.

Table 2

		Example			Comparative Example		
		4	5	6	3	4	
Base oil	Base oil 1) (% by mass)	82.68	82.78	82.29	82.00	83.05	
Additives	Viscosity index improver ²⁾ (% by mass)	6.00	5.40	6.20	5.20	6.00	
	Pour-point depressant 3) (% by mass)	0.10	0.10	0.10	0.10	0.10	
	Antioxidant ⁴⁾ (% by mass)	0.85	0.85	0.85	0.85	0.85	
	Metal-based detergent ⁵⁾ (% by mass)	3.00	3.00	3.00	3.00	3.00	
	Ashless-type dispersant ⁶⁾ (% by mass)	2.50	2.50	2.50	2.50	2.50	
	Antiwear agent 7) (% by mass)	1.00	1.00	1.00	1.00	1.00	
	Friction modifier 8) (% by mass)	0.70	0.70	0.70	0.70	0.70	
	Boron-containing compound 1 (% by mass)	1.34	0.00	0.00	0.00	0.00	
	Boron-containing compound 2 (% by mass)	0.00	1.84	0.00	0.00	0.00	
	Boron-containing compound 3 (% by mass)	0.00	0.00	1.53	0.00	0.00	
	Boron-containing compound 4 (% by mass)	0.00	0.00	0.00	2.82	0.00	
	Boron-containing compound 5 (% by mass)	0.00	0.00	0.00	0.00	0.97	
	Other additives ⁹⁾ (% by mass)	1.83	1.83	1.83	1.83	1.83	
Properties	Kinetic viscosity (40°C) (mm²/s)	36.8	36.4	35.4	37.0	35.0	
	Kinetic viscosity (100°C) (mm²/s)	8.7	8.5	8.6	8.5	8.5	
	Viscosity index	226	224	235	220	232	
	Boron content (% by mass)	0.03	0.03	0.03	0.03	0.03	
Friction coefficient		0.07	0.07	0.06	0.09	0.10	

[0184] The components in Table 2 are as follows.

- (1) Base oil: mineral oil (100 N), 40°C kinetic viscosity: 19.5mm²/s, 100°C kinetic viscosity: 4.2 mm²/s, viscosity index: 120, sulfur content: 10 ppm by mass or lower
- (2) Viscosity index improver: polyalkyl methacrylate (weight-average molecular weight: 475,000)
- (3) Pour-point depressant: polyalkyl methacrylate (weight-average molecular weight: 62,000)
- (4) Antioxidant: dialkyldiphenylamine (nitrogen content: 4.6% by mass)

- (5) Metal-based detergent: calcium salicylate (total base number: 225 mgKOH/g, calcium content: 7.8% by mass)
- (6) Ashless-type dispersant: polybutenyl succinimide (nitrogen content: 0.7% by mass)
- (7) Antiwear agent: zinc dialkyldithiophosphate (Zn content: 0.11% by mass, phosphorus content: 0.10% by mass, alkyl group: mixture of secondary butyl group and secondary hexyl group)
- (8) Friction modifier: molybdenum dithiocarbamate (Mo content: 10% by mass)
- (9) Other additives: antirust, corrosion inhibitor and antifoaming agent

[0185] As is obvious from Table 1 and Table 2, in each Example, in which the integrated value ratio from boron NMR was 0.5 to 1.0 and the ratio of the boron atom content to the nitrogen atom content was 0.6 or higher, the friction coefficient was 0.06 to 0.07, which means the friction reducing ability of the lubricant oil composition was able to be improved. On the other hand, in each Comparative Example, in which the integrated value ratio from boron NMR was lower than 0.50, the friction coefficient was 0.09 to 0.10, which means the friction reducing ability of the lubricant oil composition was unable to be sufficiently improved.

15 [Industrial applicability]

[0186] According to the present invention, a lubricant oil composition that is reduced in friction coefficient and thereby improved in the ability to reduce friction, and that has improved antiwear properties can be obtained. The lubricant oil composition can be suitably used as a lubricant oil especially for internal combustion engines.

Claims

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- 1. A lubricant oil composition that is prepared by blending a boron-containing compound therein, the boron-containing compound having a ratio of an integrated value of peaks in the 5 to 25 ppm chemical shift to an integrated value of peaks in the -10 ppm to 25 ppm chemical shift, as determined by ¹¹B-NMR measurement in deuterochloroform using BF₃·OEt₂ as an external standard (0 ppm), of 0.5 or higher and 1.0 or lower.
- 2. The lubricant oil composition according to Claim 1, wherein the boron-containing compound has a ratio of a boron atom content to a nitrogen atom content of 0.6 or higher on a mass basis.
 - 3. The lubricant oil composition according to Claim 1 or 2, wherein the boron-containing compound is obtained by acting a boric acid or a boric acid derivative on an organic compound having a hydroxyl group or an amino group.
- 35 **4.** The lubricant oil composition according to Claim 3, wherein the organic compound is at least one compound selected from amine compounds (A), amide compounds having a hydroxyl group (B), imide compounds having an amino group (C), ester compounds having a hydroxyl group (D), and alcohol compounds having a hydroxyl group (E).
 - **5.** The lubricant oil composition according to any one of Claims 1 to 4, wherein the boron-containing compound is blended therein in an amount of 0.01 to 30% by weight.
 - 6. The lubricant oil composition according to any one of Claims 1 to 5, prepared by blending, in addition to the boron-containing compound, at least one of a viscosity index improver, a molybdenum compound, a zinc dialkyldithio-phosphate, an antioxidant, a metal-based detergent, and an ashless-type dispersant into a base oil composed of a mineral oil and/or a synthetic oil.
 - 7. The lubricant oil composition according to any one of Claims 1 to 6, being a lubricant oil composition for internal combustion engines.
- 50 **8.** A lubricant oil composition that is prepared by blending a boron-containing compound therein, the boron-containing compound obtained by heating and stirring an organic compound having a hydroxyl group or an amino group and a boric acid or a boric acid derivative at a temperature of 100°C or higher and 150°C or lower.

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INTERNATIONAL SEARCH REPORT International application No. PCT/JP2013/083991 A. CLASSIFICATION OF SUBJECT MATTER 5 C10M139/00(2006.01)i, C10M141/12(2006.01)i, C10M159/12(2006.01)i, C10M163/00(2006.01)i, C10M137/10(2006.01)n, C10N30/06(2006.01)n, C10N40/25(2006.01)n According to International Patent Classification (IPC) or to both national classification and IPC FIELDS SEARCHED 10 Minimum documentation searched (classification system followed by classification symbols) C10M139/00, C10M141/12, C10M159/12, C10M163/00, C10M137/10, C10N30/06, C10N40/25 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Jitsuyo Shinan Koho 1922-1996 Jitsuyo Shinan Toroku Koho 1996-2014 15 Kokai Jitsuyo Shinan Koho 1971-2014 Toroku Jitsuyo Shinan Koho 1994-2014 Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) 20 DOCUMENTS CONSIDERED TO BE RELEVANT Category* Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. Χ JP 7-316578 A (Ethyl Corp.), 1-8 05 December 1995 (05.12.1995) claims 1 to 6; paragraphs [0008] to [0022]; 25 examples & US 5612295 A & EP 683220 A2 & AU 2008795 A & CA 2148975 A JP 5-112578 A (Nippon Oil Co., Ltd.), 1-8 Χ 07 May 1993 (07.05.1993), 30 claims 1 to 3; paragraphs [0008] to [0017]; examples & US 5543081 A 35 Further documents are listed in the continuation of Box C. See patent family annex. 40 Special categories of cited documents 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 "T "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing 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 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) "L" 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 45 "O" document referring to an oral disclosure, use, exhibition or other means document published prior to the international filing date but later than the document member of the same patent family priority date claimed Date of the actual completion of the international search Date of mailing of the international search report 50 03 March, 2014 (03.03.14) 11 March, 2014 (11.03.14) Name and mailing address of the ISA/ Authorized officer Japanese Patent Office Telephone No. 55 Form PCT/ISA/210 (second sheet) (July 2009)

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5	C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT							
	Category*	Citation of document, with indication, where appropriate, of the relevan		Relevant to claim No.				
10	Х	JP 56-49799 A (Toa Nenryo Kogyo Kabushiki Kaisha), 06 May 1981 (06.05.1981), claim 1; example 2 (Family: none)		1-8				
15	X A	JP 57-61089 A (Edwin Cooper, Inc.), 13 April 1982 (13.04.1982), claims 1 to 10; examples 6 to 8 & US 4295983 A & EP 42270 A2 & BR 8103758 A & CA 1148925 A		1-3,5-8 4				
20	A	JP 61-97294 A (Ciba-Geigy AG.), 15 May 1986 (15.05.1986), claims 1 to 9 & US 4689162 A & EP 183642 A1 & CA 1251218 A		1-8				
25	А	JP 58-129099 A (The Lubrizol Corp.), 01 August 1983 (01.08.1983), claims 1 to 37 & US 5240624 A & GB 2114116 A & DE 3301104 A & FR 2520363 A & CA 1223274 A		1-8				
30	A	JP 59-157087 A (Standard Oil Co.), 06 September 1984 (06.09.1984), claims 1 to 20; examples 1 to 6 & US 4519926 A & EP 118203 A2 & AU 2429984 A & CA 1221373 A		1-8				
35	A	JP 8-504855 A (Exxon Chemical Patents Inc 28 May 1996 (28.05.1996), claims 1 to 10; examples & US 5430105 A & EP 674692 A1 & WO 94/013762 A1 & AU 5802594 A & BR 9307683 A & SG 52776 A & CA 2111056 A & MX 9307829 A	c.),	1-8				
40		& CA 2111056 A & MX 9307829 A & CN 1090877 A						
45								
50								
55								

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REFERENCES CITED IN THE DESCRIPTION

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Patent documents cited in the description

• JP 2011012213 A **[0005]**

• JP 2006502287 A [0005]