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(54) **LUBRICATING OIL COMPOSITION FOR INDUSTRIAL MACHINERY AND EQUIPMENT**

(57) A lubricative composition for industrial machinery and equipment which comprises a base oil selected from among mineral oils, fats and oils, synthetic oils and mixture of two or more of them, and at least one additive selected from among the following components (A) to (D): component (A): (A-1) a phosphorus-containing carboxylic acid and/or (A-2) a thiophosphoric ester, component (B): a dispersant viscosity index improver, compo-

nent (C): (C-1) a specific amidocarboxylic acid compound and/or (C-2) a specific carboxylic acid compound, and component (D): an ester oiliness improver. The composition is favorable as a lubricating oil composition and useful as a gear oil composition, a lubricating oil composition for paper machines or a lubricating oil composition for slide guides, a hydraulic oil, or the like.

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**Description**Technical Field

**[0001]** The present invention relates to a novel lubricative composition for industrial machinery and equipment, more specifically, to a lubricating oil composition with excellent lubricative characteristics such as the abrasion resistance, the extreme pressure property, the frictional property and the like, as well as excellent in characteristics required according to the use of industrial machinery and equipment, and even more specifically, to a gear oil composition, a lubricating oil composition for paper machines, a lubricating oil composition for slide guides and a hydraulic oil. In particular, the gear oil composition is excellent in the sludge resistance and the extreme pressure property, the lubricating oil composition for paper machines is excellent in the sludge resistance and the extreme pressure property, the lubricating oil composition for slide guides is excellent in both aspects of the frictional property and the anti-stick-slip property, and the hydraulic oil is excellent in all the sludge suppression property, the abrasion resistance and the frictional property.

Background Art

**[0002]** With respect to the gear oil composition:

**[0003]** High efficiency and energy-saving has been improved remarkably accompanied with the development in the industry, but desires to these improvements are still strong as the endless assignment. High efficiency and low cost are required also to the gears used in various industrial machines. Therefore, the composition having the tendency for high-speed and high-output as the gear oil, tolerant in higher temperature, and tolerant in the operation under higher load is strongly desired.

**[0004]** For example, combining sulfur-based extreme pressure additives or phosphorus-based extreme pressure additives to the gear oil is described in the Patent Document 1 (JP-A No. 10-259394) and Patent Document 2 (JP-A No. 9-208976) related to the gear oil.

**[0005]** However, the conventional gear oil, when used in the gear of the recent industrial machines, was not always sufficient in the sludge resistance and the extreme pressure property. That is, for the use required to have high extreme pressure property and abrasion resistance, a lubricating oil added with the extreme pressure agent such as zinc dialkyldithiophosphate is widely used, but even if these conventional extreme pressure agents are added in small amount, thermal load is increased and a great amount of sludge is generated, and there is tendency to lower thermal and oxidation stability. Therefore, it is difficult for the lubricating oil added with the sulfur-based extreme pressure agent to reach sufficient thermal and oxidation stability in the gear of the recent industrial machines. Meanwhile, the phosphorus-based extreme pressure agent has tendency not to generate sludge easily compared with the sulfur-based extreme pressure agent, but when the phosphorus-based extreme pressure agent is used alone, it is difficult to obtain high extreme pressure property in the gear oil level.

**[0006]** With respect to the lubricating oil composition for paper machines:

**[0007]** Moreover, after making a paper in the paper machine, there is a step of drying the paper, but in the conventional paper machine, the drying time is comparatively long and the drying temperature is also comparatively low. However, in the recent paper machines, the drying time became short in the drying step to improve the production efficiency, as a result, the drying temperature also became very high, and demand for the heat resistance in the lubricating oil used in the paper machine has increased. Also, in the same manner, the demand level of abrasion resistance in the lubricating oil has increased. Furthermore, when making a paper in the paper machine, strong acidic white water is used, but since the white water has tendency to corrode metals, the lubricating oil for paper machines requires the prevention of machine corrosion by the white water. Thus, there is a big demand for performance of heat resistance, abrasion resistance and anticorrosion property in the lubricating oil for paper machines.

**[0008]** Here, in order for the conventional lubricating oil for paper machines to improve abrasion resistance, using the extreme pressure agent is general, but if the extreme pressure agent is used, the heat resistance is deteriorated, and sludge, cocking or the like is easily generated in the lubricating portion. In order to inhibit sludge, using a great amount of metal-based refining agent or ashless dispersant has been considered, but if these are used, the anticorrosion property is lowered, and problems such as the main body of the paper machine being corroded and the like are generated. As a result, it is difficult to satisfy all the required performances such as the heat resistance, abrasion resistance and anticorrosion property. For example, the lubricating oil for paper machines with excellent heat resistance, abrasion resistance and anticorrosion property is suggested in the Patent Document 3 (JP-A No. 2002-97485), but it does not always satisfy the performances.

**[0009]** With respect to the lubricating oil composition for slide guides:

**[0010]** Further, when stick-slip is generated on the slide guide such as a process table of machine tool, its frictional vibration is transferred to the product as is; as a result, problems such as the processing accuracy being decreased and the tool life being reduced from the vibration are generated. Therefore, high anti-stick-slip property between the metal

surfaces is demanded in the lubricating oil for slide guides, in addition to the low frictional (the frictional index being low) property.

**[0011]** With respect to the anti-stick-slip property, for example, using the lubricating oil composition which combined a sulfur compound or ester and fatty acid (e.g. see Patent Document 4 (JP-A No. 57-67693)), the lubricating oil composition which combined a sulfur compound and an amine salt of phosphorus compound (e.g., see Patent Document 5 (JP-A No. 51-74005)), and the like are suggested.

**[0012]** Further, with respect to the frictional property, for example, using the lubricating oil composition which combined acidic phosphoric acid ester or its alkyl ammonium salt, phosphorus acid, fatty acid and a straight-chained alkylamine (e.g., see Patent Document 6 (JP-A No. 8-134488)), the lubricating oil composition for slide guides which used phosphorus compound (e.g. see Patent Document 7 (JP-A No. 8-209175)), the lubricating oil for operation machines which combined glycerin ether compounds and phosphoric acid esters or their amine salts (e.g. see Patent Document 8 (JP-A No. 11-209775)), and the like are suggested.

**[0013]** However, having both the frictional property and anti-stick-slip property is not always easy, and the conventional lubricating oil composition for slide guides did not have sufficiently satisfying performances.

**[0014]** With respect to the hydraulic oil:

**[0015]** Recently, the hydraulic system has become highly efficient, and in order to carry out high speed and highly precise control, a case for controlling the flow amount and direction of the hydraulic system by a valve such as a spool valve, and a case installing a servo valve has been increased. The performance of such the spool valve or servo valve may be deteriorated to the great extent if sludge is generated in the hydraulic oil, thus the hydraulic oil used in the recent highly efficient system is strongly required to be a sludgeless hydraulic oil which does not generate sludge, in addition to have an excellent abrasive resistance.

**[0016]** Meanwhile, due to the revision of the energy-saving laws, to the factories appointed as the energy supervision appointed factories, energy-saving has become an essential problem. Every year a target value is set, and it is necessary to reduce energy. Therefore, as a part of energy-saving, reducing consumption power of the operation motor in a hydraulic machines used widely in the factories became vary important object. Therefore, the hydraulic oil is required to improve its characteristic in the viewpoint of saving energy.

**[0017]** Accordingly, in order to respond to the above demands, development of a novel hydraulic oil is on process. For example, as the abrasion resistant of the hydraulic oil, a zinc-based abrasion resistant such as zinc dithiophosphate (ZnDTP) is widely used, but the zinc-based abrasion resistant can cause sludge generation. Further, the abrasion resisting effect by using ZnDTP or the like may form rigid film such as iron phosphate on the surface of the metal, and by such film formation, the frictional index of the sliding part is increased, thus it is said to be not preferable in the viewpoint of energy-saving. Here, the investigation on the hydraulic oil which used non-zinc-based abrasion resistant is carried out, for example, the use of non-zinc-based hydraulic oil obtained by combining non-zinc-based abrasion resistant such as aromatic phosphoric ester, phosphorus acid ester and its amine salt, thiophosphate and  $\beta$ -dithiophosphorylated propionic acid compound is suggested instead of ZnDTP as a purpose to secure the sludge resistance and the abrasion resistance (e.g., see Patent Document 9 (JP-A No. 10-67993), Patent Document 10 (JP-A No. 11-217577) and Patent Document 11 (JP-A No. 2002-265971)).

**[0018]** Further, the hydraulic oil obtained by combining complete ester or partial ester of polyhydric alcohol or an oiliness improver such as fatty acid amide, together with an amine-based antioxidant, a phenol-based antioxidant and phosphoric ester into a certain base oil in the purpose of improving the oxidation stability under high pressure and lubricating performances is described (see Patent Document 12 (JP-A No. 09-111277)).

#### Disclosure of the Invention

**[0019]** The first aspect of the present invention provides a lubricative composition for industrial machinery and equipment which comprises a base oil selected from mineral oils, fats and oils, synthetic oils and mixtures of two or more of them, and at least one additive selected from the following components (A) to (D):

Component (A):

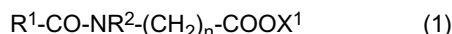
(A-1) a phosphorus-containing carboxylic acid compound and/or (A-2) a thiophosphoric ester;

Component (B): a dispersant viscosity index improver;

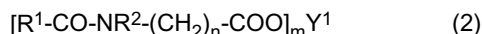
Component (C):

the following component (C-1) and/or component (C-2):

component (C-1): at least one kind of a compound represented by the following formulas (1) to (3):



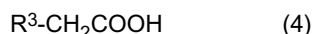
wherein R<sup>1</sup> is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms, R<sup>2</sup> is an alkyl group having 1 to 4 carbon atoms, X<sup>1</sup> is hydrogen, an alkyl group having 1 to 30 carbon atoms or an alkenyl group having 1 to 30 carbon atoms, and n is an integer of 1 to 4,



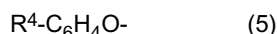
wherein R<sup>1</sup> is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms, R<sup>2</sup> is an alkyl group having 1 to 4 carbon atoms, Y<sup>1</sup> is an alkali metal or an alkali earth metal, n is an integer of 1 to 4, and m is 1 when Y<sup>1</sup> is an alkali metal and 2 when Y<sup>1</sup> is an alkali earth metal, and



wherein R<sup>1</sup> is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms, R<sup>2</sup> is an alkyl group having 1 to 4 carbon atoms, Z is a residue having a hydroxyl group removed from a polyhydric alcohol with two or more valences, m is an integer of 1 or more, m' is an integer of 0 or more, m + m' is a valence number of Z, and n is an integer of 1 to 4,  
component (C-2): a compound represented by the following formula (4):



wherein R<sup>3</sup> is an alkyl group having 7 to 29 carbon atoms, an alkenyl group having 7 to 29 carbon atoms or a group represented by the formula (5):



wherein R<sup>4</sup> is an alkyl group having 1 to 20 carbon atoms or hydrogen; and component (D): an ester oiliness improver.

**[0020]** The second aspect of the invention provides the lubricative composition in the first aspect of the invention as a gear oil composition, wherein at least one kind of the additive is selected from the component (A) to component (C).

**[0021]** The gear oil composition is excellent in the sludge resistance and the extreme pressure property.

**[0022]** The third aspect of the invention provides the lubricative composition in the first aspect of the invention as a lubricating oil composition for paper machines, wherein the additive is at least one selected from the component (A) to component (C).

**[0023]** The lubricating oil composition for paper machines is excellent in the sludge resistance and the extreme pressure property.

**[0024]** The fourth aspect of the invention provides the lubricative composition in the first aspect of the invention as a lubricating oil composition for slide guides, wherein the additive is at least one selected from the component (A) to component (C).

**[0025]** The lubricating oil composition for slide guides is excellent in both aspects of the frictional property and the anti-stick-slip property.

**[0026]** The fifth aspect of the invention provides the lubricative composition in the first aspect of the invention as a lubricating oil composition, wherein the additive comprises the phosphorus-containing carboxylic acid compound of the component (A-1) and the dispersant viscosity index improver of the component (B).

**[0027]** The sixth aspect of the invention provides the lubricative composition in the first aspect of the invention as a lubricating oil composition, wherein the additive comprises the thiophosphoric ester of the component (A-2) and the dispersant viscosity index improver of the component (B).

**[0028]** The seventh aspect of the invention provides the lubricative composition in the first aspect of the invention as a lubricating oil composition, wherein the additive comprises the ester oiliness improver of the component (D) which is an ester of a polyhydric alcohol and a fatty acid of monobasic acids.

**[0029]** The eighth aspect of the invention provides the lubricative composition in the seventh aspect of the invention, wherein the ester oiliness improver of the component (D) which is an ester of a polyhydric alcohol and a fatty acid of monobasic acids is any one selected from the following esters of (D-1) to (D-3):

(D-1): an ester of a polyhydric alcohol and an unsaturated fatty acid containing a partial ester with the degree of

esterification of 1 and a partial ester with the degree of esterification of 2 or more;

(D-2): a whole ester of a polyhydric alcohol and a mixture of fatty acids, wherein the fatty acids are short-chained fatty acids and long-chained fatty acids; and

(D-3): an ester of a polyhydric alcohol and a branched saturated fatty acid containing a partial ester with the degree of esterification of 1 and a partial ester with the degree of esterification of 2 or more.

**[0030]** The ninth aspect of the invention provides the lubricative composition in the eighth aspect of the invention, wherein the lubricating oil composition is a hydraulic oil.

**[0031]** The hydraulic oil is excellent in all the sludge suppression property, the abrasion resistance and the frictional property.

## BRIEF DESCRIPTION OF THE DRAWINGS

**[0032]**

Fig. 1 is an outline block diagram showing the friction coefficient measurement system used in Example concerning a lubricating oil composition for slide guides in one embodiment of the invention.

Fig. 2 is an outline block diagram showing the anti-stick-slip property evaluation equipment used in Example concerning a lubricating oil composition for slide guides in one embodiment of the invention.

Fig. 3 is a graph showing an example of the correlation with the friction coefficient and time which are obtained using the equipment of the Fig. 2.

Fig. 4 is an illustrative diagram showing the separation state of each layer in the evaluation test of separability over water soluble cutting fluid on an Example concerning a lubricating oil composition for slide guides in one embodiment of the invention.

Fig. 5 is a diagram illustrating the arrangement and the operation of the disk and ball in the SRV (micro-scale reciprocating friction) test on an Example concerning a hydraulic oil composition in one embodiment of the invention.

## Best Mode for Carrying Out the Invention

**[0033]** As a lubricative composition of the present invention, at least one kind selected among mineral oils, fats and oils, and synthetic oils are used as a base oil.

**[0034]** Here, examples of mineral oils may include paraffinic, naphthenic or the like mineral oil obtained by subjecting a lubricating oil distillate, which was obtained by the normal pressure distillation or the reduced pressure distillation of a crude oil, to suitably combined one kind or two or more kinds of refining means such as solvent deasphalting, solvent extraction, hydrogenolysis, solvent dewaxing, contact dewaxing, hydrotreating, sulfuric acid washing, white clay treatment and the like. Further, a wax isomeric base oil, a base oil prepared by the means of isomerization of GTL WAX (Gas-to-liquid Wax) can be also used.

**[0035]** Moreover, examples of fats and oils include a tallow, a lard, a sunflower oil, a soybean oil, a canola oil, a rice bran oil, a coconut oil, a palm oil, palm kernel oil, or hydrogen additives thereof, and the like.

**[0036]** Also, examples of synthetic oils include poly- $\alpha$ -olefin (ethylene-propylene copolymer, polybutene, 1-octene oligomer, 1-decene oligomer and hydrides thereof), alkyl benzene, alkyl naphthalene, monoester (butyl stearate, octyl laurate), diester (di-tridecyl glutarate, di-2-ethylhexyl adipate, di-isodecyladipate, di-tridecyl adipate, di-2-ethylhexyl sebacate, etc.), polyester (trimellitic acid ester, etc.), polyol ester (trimethylolpropane caprylate, trimethylolpropane pentalonate, pentaerythritol-2-ethyl hexanoate, pentaerythritol pentalonate, etc.), polyoxyalkylene glycol, polyphenyl ether, dialkyl diphenyl ether, phosphoric acid ester (tricresyl phosphate, etc.), fluorine-containing compound (perfluoropolyether, fluorinated polyolefin, etc.), a silicone oil and the like.

**[0037]** Among these base oils, mineral oils, poly- $\alpha$ -olefin, polyol ester, polyoxyalkylene glycol are preferred.

**[0038]** As the base oil with lubricative composition of the invention, the above-mentioned base oil may be used alone or in combination of two or more.

**[0039]** The kinematic viscosity of these base oils is arbitrary and not particularly limited, but the minimum value of the kinematic viscosity at 40°C in the aspect of pitch resistance is 10 mm<sup>2</sup>/s or higher, preferably 20 mm<sup>2</sup>/s or higher, more preferably 40 mm<sup>2</sup>/s or higher and most preferably 60 mm<sup>2</sup>/s or higher. The maximum value thereof in the point of small friction loss by agitation resistance is 10000 mm<sup>2</sup>/s or less, preferably 5000 mm<sup>2</sup>/s or less and more preferably 1000 mm<sup>2</sup>/s or less. Further, the viscosity index of the base oil is also arbitrary, but in the point of inhibiting oil film degradation in high temperatures, the minimum value of the viscosity index is 80 or higher, preferably 90 or higher, and in general, the value is 500 or less. Further, the flow point is also arbitrary, but in the point of low temperature property, the flow point, in general, is preferably -5°C or lower and more preferably -15°C or lower.

**[0040]** For a lubricative composition, at least one kind of additive selected from the components (A) to (D) is mixed

with the base oil.

**[0041]** Hereinbelow, the components (A) to (D) will be further described.

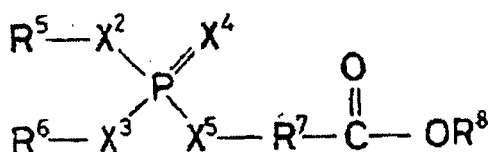
**[0042]** The component (A) is (A-1) a phosphorus-containing carboxylic acid compound, (A-2) a thiophosphoric acid ester or a mixture thereof.

Component (A-1):

(A-1) a phosphorus-containing carboxylic acid compound will be described.

**[0043]** As (A-1) a phosphorus-containing carboxylic acid compound, the compound may have both carboxyl group and phosphorus atom in the same molecule, and its structure is not particularly limited. However, in the point of extreme pressure property and thermo-oxidative stability, phosphorylated carboxylic acid is preferred.

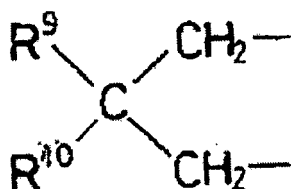
**[0044]** Examples of phosphorylated carboxylic acid include the compounds represented by the following formula (6).



(6)

wherein  $\text{R}^5$  and  $\text{R}^6$  may be identical with or different from each other, and they are each independently a hydrogen atom or a hydrocarbon group having 1 to 30 carbon atoms,  $\text{R}^7$  is an alkylene group having 1 to 20 carbon atoms,  $\text{R}^8$  is a hydrogen atom or a hydrocarbon group having 1 to 30 carbon atoms, and  $\text{X}^1$ ,  $\text{X}^2$ ,  $\text{X}^3$  and  $\text{X}^4$  may be identical with or different from each other, and they are each independently an oxygen atom or a sulfur atom.

**[0045]** In the formula (6),  $\text{R}^5$  and  $\text{R}^6$  are each independently a hydrogen atom or a hydrocarbon group having 1 to 30 carbon atoms. Examples of the hydrocarbon group having 1 to 30 carbon atoms include an alkyl group, an alkenyl group, a cycloalkyl group, a bicycloalkyl group, a tricycloalkyl group, an alkylcycloalkyl group, an alkylbicycloalkyl group, an alkyltricycloalkyl group, a cycloalkylalkyl group, a bicycloalkylalkyl group, a tricycloalkylalkyl group, an aryl group, an alkylaryl group, an arylalkyl group and the like. Moreover,  $\text{R}^5$  and  $\text{R}^6$  may be bonded to each other to form a divalent group represented by the following formula (7). Two bonds of the divalent group are bonded with  $\text{X}^1$  and  $\text{X}^2$  respectively.



(7)

wherein  $\text{R}^9$  and  $\text{R}^{10}$  may be identical with or different from each other, and they are each independently a hydrogen atom or an alkyl group having 1 to 4 carbon atoms; and, both of  $\text{R}^9$  and  $\text{R}^{10}$  being a methyl group is preferred.

**[0046]** Among these, as  $\text{R}^5$  and  $\text{R}^6$ , an alkyl group, a cycloalkyl group, a cycloalkylalkyl group, tricycloalkylalkyl group, an aryl group, an alkylaryl group, a divalent group represented by the formula (7) bonded with  $\text{R}^9$  and  $\text{R}^{10}$  are preferred, and an alkyl group is more preferred.

**[0047]** An alkyl group as  $\text{R}^5$  and  $\text{R}^6$  may be any one of straight-chained or branched, and the alkyl group having 1 to 18 carbon atoms is preferred. Examples of such an alkyl group include a methyl group, an ethyl group, a propyl group, an isopropyl group, an n-butyl group, an isobutyl group, a tert-butyl group, a pentyl group, an isopentyl group, a hexyl group, a heptyl group, a 3-heptyl group, an octyl group, a 2-ethylhexyl group, a nonyl group, a decyl group, an undecyl group, a dodecyl group, a tridecyl group, a tetradecyl group, a pentadecyl group, a hexadecyl group, a heptadecyl group, an octadecyl group, a 2-ethylbutyl group, a 1-methylphenyl group, a 1,3-dimethylbutyl group, a 1,1,3,3-tetramethylbutyl group, a 1-methylhexyl group, an isoheptyl group, a 1-methylheptyl group, a 1,1,3-trimethylhexyl group, a 1-methylundecyl group, and the like. Among these, an alkyl group having 3 to 18 carbon atoms is preferred, and an alkyl group having 3 to 8 carbon atoms is more preferred.

**[0048]** Examples of the cycloalkyl group as  $R^5$  and  $R^6$  include a cyclopentyl group, a cyclohexyl group, a cycloheptyl group, a cyclooctyl group, a cyclododecyl group and the like. Among these, a cycloalkyl group having 5 or 6 carbon atoms (a cyclopentyl group and a cyclohexyl group) is preferred, and a cyclohexyl group is particularly preferred.

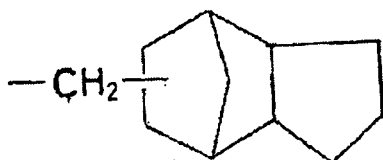
**[0049]** For the cycloalkylalkyl group as  $R^5$  and  $R^6$ , a cycloalkylmethyl group is preferred, a cycloalkylmethyl group having 6 or 7 carbon atoms is more preferred, and a cyclopentylmethyl group and a cyclohexylmethyl group are particularly preferred.

**[0050]** For the bicycloalkylalkyl group as  $R^5$  and  $R^6$ , a bicycloalkylmethyl group is preferred, a bicycloalkylmethyl group having 9 to 11 carbon atoms is more preferred, and a decalinylmethyl group is particularly preferred.

**[0051]** For the tricycloalkylalkyl group as  $R^5$  and  $R^6$ , a tricycloalkylmethyl group is preferred, a tricycloalkylmethyl group having 9 to 15 carbon atoms is more preferred, and a group represented by the following formula (8) or (9) is particularly preferred.



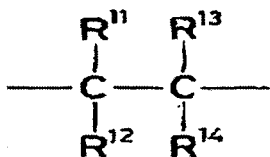
(8)



(9)

**[0052]** Examples of the aryl group and the alkylaryl group as  $R^5$  and  $R^6$  include a phenyl group, a tolyl group, a xylyl group, an ethylphenyl group, a vinylphenyl group, a methylphenyl group, a dimethylphenyl group, a trimethylphenyl group, an ethylphenyl group, an isopropylphenyl group, a tert-butylphenyl group, a di-tert-butylphenyl group, 2,6-di-tert-butyl-4-methylphenyl group and the like. Among these, an aryl group and an alkylaryl group having 6 to 15 carbon atoms are preferred.

**[0053]**  $R^7$  is an alkylene group having 1 to 20 carbon atoms. The carbon numbers of such an alkylene group is preferably 1 to 10, more preferably 2 to 6, and even more preferably 3 to 4. Further, for such an alkylene group, a group represented by the following formula (10) is preferred.



(10)

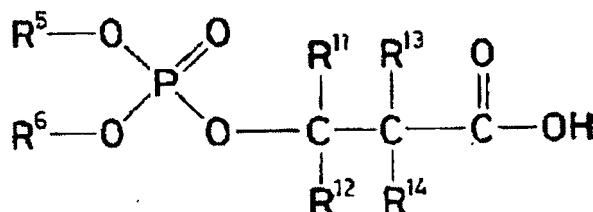
wherein  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$  and  $R^{14}$  may be identical with or different from each other and are each independently a hydrogen atom or a hydrocarbon group having 1 to 4 carbon atoms, and the total carbon number of  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$  and  $R^{14}$  is 6 or lower. Preferably,  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$  and  $R^{14}$  may be identical with or different from each other and are each independently a hydrogen atom or a hydrocarbon group having 1 to 3 carbon atoms, and the total carbon number of  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$  and  $R^{14}$  is 5 or lower. More preferably,  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$  and  $R^{14}$  may be identical with or different from each other and are each independently a hydrogen atom or a hydrocarbon group having 1 or 2 carbon atoms, and the total carbon number of  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$  and  $R^{14}$  is 4 or lower. Particularly preferably,  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$  and  $R^{14}$  may be identical with or different from each other and are each independently a hydrogen atom or a hydrocarbon group having 1 to 2 carbon atoms, and the total carbon number of  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$  and  $R^{14}$  is 3 or lower. Most preferably,  $R^{13}$  and  $R^{14}$  are each independently a methyl group, and the remaining 3 groups are hydrogen atoms.

**[0054]** In the formula (6),  $R^8$  is a hydrogen atom or a hydrocarbon group having 1 to 30 carbon atoms. Examples of

such a hydrocarbon group include the hydrocarbon group exemplified in the description of R<sup>5</sup> and R<sup>6</sup>.

**[0055]** Also, in the formula (6), X<sup>2</sup>, X<sup>3</sup>, X<sup>4</sup> and X<sup>5</sup> may be identical or different from each other and are each independently an oxygen atom or a sulfur atom. In the point of the extreme pressure property, one or more among X<sup>2</sup>, X<sup>3</sup>, X<sup>4</sup> and X<sup>5</sup> is preferably a sulfur atom, two or more is more preferably a sulfur atom, and two among X<sup>2</sup>, X<sup>3</sup>, X<sup>4</sup> and X<sup>5</sup> being sulfur atoms and the remaining two being oxygen atoms is even more preferred. In this case, any among X<sup>2</sup>, X<sup>3</sup>, X<sup>4</sup> and X<sup>5</sup> being a sulfur atom is arbitrary, but X<sup>2</sup> and X<sup>3</sup> being oxygen atoms and X<sup>4</sup> and X<sup>5</sup> being sulfur atoms is preferred.

**[0056]** Heretofore, each group in the formula (6) has been described, but for more excellent extreme pressure property,  $\beta$ -dithiophosphorylpropionic acid represented by the following formula (11) is preferably used.



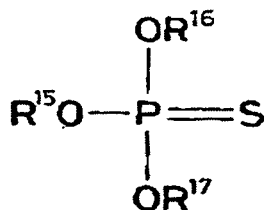
(11)

wherein  $R^5$  and  $R^6$  each independently have the same definition as  $R^5$  and  $R^6$  in the formula (6), and  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$  and  $R^{14}$  each independently have the same definition as  $R^{11}$ ,  $R^{12}$ ,  $R^{13}$  and  $R^{14}$  in the formula (10).

**[0057]** Next, (A-2) a thiophosphoric ester will be described.

Component (A-2): thiophosphoric ester

**[0058]** (A-2) Thiophosphoric ester according to the invention is a compound represented by the following formula (12):



(12)

wherein R<sup>15</sup> to R<sup>17</sup> may be identical with or different from each other and are each independently a hydrocarbon group having 1 to 24 carbon atoms.

**[0059]** Examples of the hydrocarbon group having 1 to 24 carbon atoms represented by R<sup>15</sup> to R<sup>17</sup> include an alkyl group, a cycloalkyl group, an alkenyl group, an alkylcycloalkyl group, an aryl group, an alkylaryl group, an arylalkyl group and the like.

**[0060]** Examples of the alkyl group include alkyl groups (wherein the alkyl group may be straight-chained or branched) such as a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, an undecyl group, a dodecyl group, a tridecyl group, a tetradecyl group, a pentadecyl group, a hexadecyl group, a heptadecyl group and an octadecyl group.

**[0061]** Examples of the cycloalkyl group include cycloalkyl groups having 5 to 7 carbon atoms such as a cyclopentyl group, a cyclohexyl group and a cycloheptyl group. Further, examples of the alkylcycloalkyl group include alkylcycloalkyl groups having 6 to 11 carbon atoms (wherein the substitution site of an alkyl group in the cycloalkyl group is arbitrary) such as a methylcyclopentyl group, a dimethylcyclopentyl group, a methylethylcyclopentyl group, a diethylcyclopentyl group, a methylcyclohexyl group, a dimethylcyclohexyl group, a methylethylcyclohexyl group, a diethylcyclohexyl group, a methylcycloheptyl group, a dimethylcycloheptyl group, a methylethylcycloheptyl group and a diethylcycloheptyl group.

**[0062]** Examples of the alkenyl group include alkenyl groups (wherein the alkenyl group may be straight-chained or branched, and the position of the double bond is arbitrary) such as a butenyl group, a pentenyl group, a hexenyl group, a heptenyl group, an octenyl group, a nonenyl group, a decenyl group, an undecenyl group, a dodecenyl group, a tridecenyl group, a tetradecenyl group, a pentadecenyl group, a hexadecenyl group, a heptadecenyl group and an octadecenyl group.

**[0063]** Examples of the aryl group include aryl groups such as a phenyl group, a naphthyl group. Further, examples



of the alkylaryl group include alkylaryl groups having 7 to 18 carbon atoms (wherein the alkyl group may be straight-chained or branched, and the substitution site in the aryl group is arbitrary) such as a tolyl group, a xylyl group, an ethylphenyl group, a propylphenyl group, a butylphenyl group, a pentylphenyl group, a hexylphenyl group, a heptylphenyl group, an octylphenyl group, a nonylphenyl group, a decylphenyl group, an undecylphenyl group and a dodecylphenyl group.

**[0064]** Examples of the arylalkyl group include arylalkyl groups having 7 to 12 carbon atoms (wherein the alkyl group may be straight-chained or branched) such as a benzyl group, a phenylethyl group, a phenylpropyl group, a phenylbutyl group, a phenylpentyl group and a phenylhexyl group.

**[0065]** The hydrocarbon group having 1 to 24 carbon atoms represented by  $R^{15}$  to  $R^{17}$  is preferably an alkyl group, an aryl group and an alkylaryl group, and more preferably an alkyl group having 4 to 18 carbon atoms, an alkylaryl group having 7 to 24 carbon atoms and a phenyl group.

**[0066]** Specific examples of the thiophosphoric ester represented by the formula (12) include tributyl phosphorothionate, tripentyl phosphorothionate, trihexyl phosphorothionate, triheptyl phosphorothionate, trioctyl phosphorothionate, trinonyl phosphorothionate, tridecyl phosphorothionate, triundecyl phosphorothionate, tridodecyl phosphorothionate, tridecyl phosphorothionate, tritetradecyl phosphorothionate, tripentadecyl phosphorothionate, trihexadecyl phosphorothionate, triheptadecyl phosphorothionate, trioctadecyl phosphorothionate, trioleyl phosphorothionate, triphenyl phosphorothionate, tricresyl phosphorothionate, trixylenyl phosphorothionate, cresyldiphenyl phosphorothionate, xylenyldiphenyl phosphorothionate, tris(n-propylphenyl)phosphorothionate, tris(isopropylphenyl)phosphorothionate, tris(n-butylphenyl)phosphorothionate, tris(isobutylphenyl)phosphorothionate, tris(s-butylphenyl)phosphorothionate, tris(t-butylphenyl)phosphorothionate and the like. Moreover, a mixture thereof can be also used.

**[0067]** In the lubricative composition of the invention, the (A-1) phosphorus-containing carboxylic acid compound or (A-2) thiophosphoric ester alone, or both of them in combination can be used.

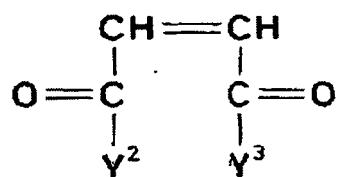
Component (B): dispersant viscosity index improver

**[0068]** The component (B) in the lubricative composition of the invention is a dispersant viscosity index improver. Here, as the dispersant viscosity index improver, an arbitrary compound used as a dispersant viscosity index improver of a lubricating oil can be used, but specific examples include a copolymer obtained by copolymerizing (B-1) a monomer of one kind or two or more kinds selected from compounds represented by the following formulas (13), (14) and (15) and (B-2) a nitrogen-containing monomer of one kind or two or more kinds selected from compounds represented by the following formulas (16) and (17), a hydride thereof, and the like.

Component (B-1)

**[0069]**

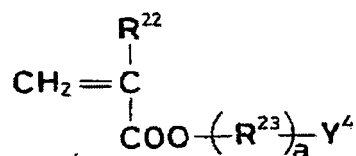




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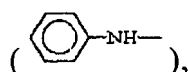
Component (B-2)

[0070]

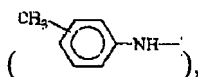


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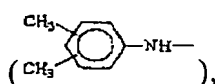
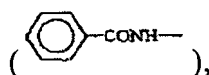
wherein as  $\text{Y}^4$ , each independently, specific examples include a dimethylamino group, a diethylamino group, a dipropylamino group, a dibutylamino group, an anilino group



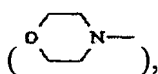
a toluidino group



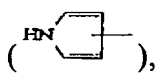
a xylydino group

an acetylamino group ( $\text{CH}_3\text{CONH}-$ ), a benzoylamino group

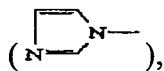
a morpholino group



a pyrrolyl group

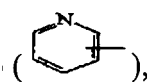


5 a pyrrolino group



10

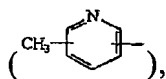
a pyridyl group



15

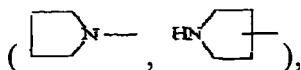
a methylpyridyl group

20



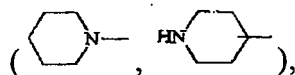
25

a pyrrolidinyl group



30

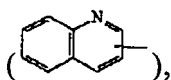
a piperidinyl group



35

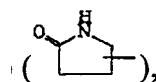
a quinonyl group

40



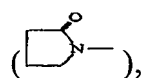
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a pyrrolidonyl group



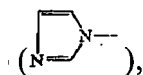
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a pyrrolidono group

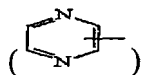


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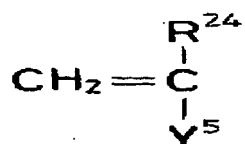
an imidazolino group



a pyrrazino group

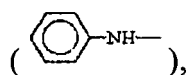


and the like.

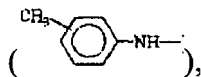


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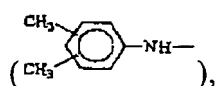
wherein as  $\text{Y}^5$ , each independently, specific examples include a dimethylamino group, a diethylamino group, a dipropylamino group, a dibutylamino group, an anilino group



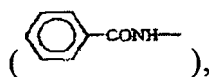
a toluidino group



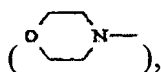
a xylydino group



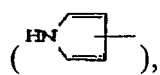
an acetylamino group ( $\text{CH}_3\text{CONH-}$ ), a benzoylamino group



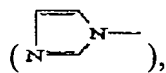
a morpholino group



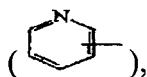
a pyrrolyl group



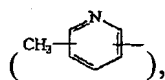
5 a pyrrolino group



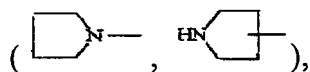
10 a pyridyl group



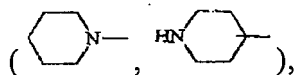
15 a methylpyridyl group



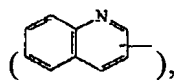
20 a pyrrolidinyl group



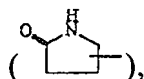
30 a piperidinyl group



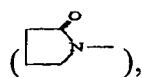
35 a quinonyl group



40 a pyrrolidonyl group



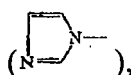
45 a pyrrolidono group



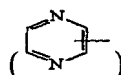
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an imidazolino group



a pyrazino group



and the like.

**[0071]** In formulas (13), (14), and (15),  $R^{18}$  and  $R^{20}$  are each independently a hydrogen atom or a methyl group,  $R^{19}$  is an alkyl group having 1 to 18 carbon atoms,  $R^{21}$  is a hydrocarbon group having 1 to 12 carbon atoms,  $Y^2$  and  $Y^3$  is each independently a hydrogen atom, a remaining group of alkylalcohol having 1 to 18 carbon atoms ( $-OR^{25}$ :  $R^{25}$  is an alkyl group having 1 to 18 carbon atoms) or a remaining group of monoalkylamine having 1 to 18 carbon atoms ( $-NHR^{26}$ :  $R^{26}$  is an alkyl group having 1 to 18 carbon atoms, respectively).

**[0072]** Specific examples of the alkyl group having 1 to 18 carbon atoms represented by  $R^{19}$ ,  $R^{25}$  and  $R^{26}$  each independently include alkyl groups (wherein the alkyl group may be straight-chained or branched) such as a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, an undecyl group, a dodecyl group, a tridecyl group, a tetradecyl group, a pentadecyl group, a hexadecyl group, a heptadecyl group and an octadecyl group.

**[0073]** Specific examples of  $R^{21}$  include alkyl groups (wherein the alkyl group may be straight-chained or branched) such as a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, an undecyl group and a dodecyl group; alkenyl groups (wherein the alkenyl group may be straight-chained or branched) such as a butenyl group, a pentenyl group, a hexenyl group, a heptenyl group, an octenyl group, a nonenyl group, a decenyl group, an undecenyl group and a dodecenyl group; cycloalkyl groups having 5 to 7 carbon atoms such as a cyclopentyl group, a cyclohexyl group, a cycloheptyl group; alkylcycloalkyl groups having 6 to 11 carbon atoms (wherein the alkyl group may be straight-chained or branched, and the bonding site to the cycloalkyl group is arbitrary) such as a methylcyclopentyl group, a dimethylcyclopentyl group, a methylethylcyclopentyl group, a diethylcyclopentyl group, a methylcyclohexyl group, a dimethylcyclohexyl group, a methylethylcyclohexyl group, a diethylcyclohexyl group, a methylcycloheptyl group, a dimethylcycloheptyl group, a methylethylcycloheptyl group and a diethylcycloheptyl group; aryl groups such as phenyl group, naphthyl group; alkylaryl groups having 7 to 12 carbon atoms (wherein the alkyl group may be straight-chained or branched, and the bonding site to the aryl group is arbitrary) such as a tolyl group, a xylyl group, an ethylphenyl group, a propylphenyl group, a butylphenyl group, a pentylphenyl group and a hexylphenyl group; arylalkyl groups having 7 to 12 carbon atoms (wherein the alkyl group may be straight-chained or branched, and the bonding site of the aryl group to the alkyl group is arbitrary) such as a benzyl group, a phenylethyl group, a phenylpropyl group, a phenylbutyl group, a phenylpentyl group and a phenylhexyl group; and the like.

**[0074]** Specific examples of the monomer of the component (B-1) preferably include alkyl acrylate having 1 to 18 carbon atoms, alkyl methacrylate having 1 to 18 carbon atoms, olefin having 2 to 20 carbon atoms, styrene, methylstyrene, anhydrous maleic acid ester, anhydrous maleic acid amide, and mixtures thereof, and the like. Among these, as the monomer of the component (B-1), a monomer represented by the formula (13), for example, alkyl (meth)acrylate having 1 to 18 carbon atoms of the alkyl group is mentioned.

**[0075]**  $R^{22}$  and  $R^{24}$ , in the formulas (16) and (17) as the component (B-2), are each independently a hydrogen atom or a methyl group,  $R^{23}$  is an alkylene group having 2 to 18 carbon atoms, and  $a$  is an integer of 0 or 1.

**[0076]**  $Y^4$  and  $Y^5$  are each independently a nitrogen-containing organic group having 1 to 30 carbon atoms.

**[0077]** As  $Y^4$  and  $Y^5$ , a group having a ring is preferred, and the group having an aliphatic ring than that having an aromatic ring as the ring is more preferred.

**[0078]** As  $Y^4$  and  $Y^5$ , a group having a 6-membered ring is preferred in the point of the sludge resistance.

**[0079]** As  $Y^4$  and  $Y^5$ , a group having an oxygen atom-containing ring is preferred in the point of the sludge resistance.

**[0080]** As  $Y^4$  and  $Y^5$ , a group having one nitrogen atom is preferred.

**[0081]** As  $Y^4$  and  $Y^5$ , a morpholino group is most preferred in the point of the sludge resistance.

**[0082]** Specific examples of  $R^{23}$  include alkylene groups (wherein the alkylene group may be straight-chained or branched) such as an ethylene group, a propylene group, a butylene group, a pentylene group, a hexylene group, a heptylene group, an octylene group, a nonylene group, a decylene group, an undecylene group, a dodecylene group,

a tridecylene group, a tetradecylene group, a pentadecylene group, a hexadecylene group, a heptadecylene group and an octadecylene.

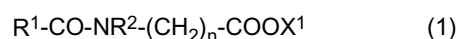
**[0083]** As the monomer of the component (B-2) among the formulas (16) and (17), a monomer represented by the formula (16) is preferred in the point of the sludge resistance. Specific examples thereof include, dimethylaminomethyl methacrylate, diethylaminomethyl methacrylate, dimethylaminoethyl methacrylate, diethylaminoethyl methacrylate, 2-methyl-5-vinyl pyridine, morpholinomethyl methacrylate, morpholinoethyl methacrylate, and mixtures thereof, and the like.

**[0084]** As from the above, the dispersant viscosity index improver as the component (B) means a copolymer which has, as a comonomer, a nitrogen-containing monomer such as the component (B-2). The dispersant viscosity index improver as the component (B) can be obtained by copolymerizing one kind or two or more kinds of monomer selected among the component (B-1) and one kind or two or more kinds of nitrogen-containing monomer selected among the component (B-2). When carrying out the copolymerization, the copolymerization molar ratio of the component (B-1) and the component (B-2) is arbitrary, but, in general, about 80:20 to 95:5. Although no particular limitation is imposed on the copolymerization method, such copolymers are generally obtained with ease by radical-solution polymerization of components (B-1) with components (B-2) in the presence of a polymerization initiator such as benzoyl peroxide. Further, the number average molecular weight of the dispersant viscosity index improver as the component (B) is arbitrary, but, in general, the number average molecular weight of 1,000 to 1,500,000, and preferably 10,000 to 200,000 can be desirably used.

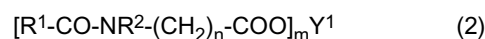
Component (C):

**[0085]** The components (C-1) and (C-2) combined with the lubricative composition of the invention will be shown again, and described in detail below.

**[0086]** That is, the component (C-1) is at least one kind of a compound represented by the following formulas (1) to (3) as mentioned above.



wherein  $R^1$  is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms,  $R^2$  is an alkyl group having 1 to 4 carbon atoms,  $X^1$  is hydrogen, an alkyl group having 1 to 30 carbon atoms or an alkenyl group having 1 to 30 carbon atoms, and  $n$  is an integer of 1 to 4,



wherein  $R^1$  is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms,  $R^2$  is an alkyl group having 1 to 4 carbon atoms,  $Y^1$  is an alkali metal or an alkali earth metal,  $n$  is an integer of 1 to 4, and  $m$  is 1 when  $Y^1$  is an alkali metal and 2 when  $Y^1$  is an alkali earth metal, and



wherein  $R^1$  is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms,  $R^2$  is an alkyl group having 1 to 4 carbon atoms,  $Z$  is a residue having a hydroxyl group removed from a polyhydric alcohol with two or more valences,  $m$  is an integer of 1 or more,  $m'$  is an integer of 0 or more,  $m + m'$  is a valence number of  $Z$ , and  $n$  is an integer of 1 to 4.

**[0087]** In the formulas (1) to (3),  $R^1$  is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms. In the point of solubility or the like to a base oil, it is necessary that the group is an alkyl group or an alkenyl group having 6 or more carbon atoms, preferably 7 or more carbon atoms, and more preferably 8 or more carbon atoms. Moreover, in the point of the storage stability or the like, it is necessary that the group is an alkyl group or an alkenyl group having 30 or less carbon atoms, preferably 24 or less carbon atoms, and more preferably 20 or less carbon atoms. Specific examples of such the alkyl group or the alkenyl group include alkyl groups (wherein the alkyl group may be straight-chained or branched) such as a hexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, an undecyl group, a dodecyl group, a tridecyl group, a tetradecyl group, a pentadecyl group, a hexadecyl group, a heptadecyl group, an octadecyl group, a nonadecyl group and an icosyl group; alkenyl groups (wherein the alkenyl group may be straight-chained or branched, and the position of the double bond is arbitrary) such as a hexenyl group, a heptenyl group, an octenyl group, a nonenyl group, a decenyl group, an undecenyl group, a dodecenyl group, a tridecenyl group, a tetradecenyl group, a pentadecenyl group, a hexadecenyl group, a heptadecenyl group, an octadecenyl group, a nonadecenyl group and an icosenyl group; and the like.

**[0088]** In the formulas (1) to (3),  $R^2$  is an alkyl group having 1 to 4 carbon atoms. In the point of the storage stability

or the like, it is necessary that the group is an alkyl group having 4 or less carbon atoms, preferably 3 or less carbon atoms, and more preferably 2 or less carbon atoms. In the formulas (1) to (3), n is an integer of 1 to 4. On the point of the storage stability, it is necessary that n is an integer of 4 or less, preferably 3 or less, and more preferably 2 or less.

**[0089]** In the formula (1), X<sup>1</sup> is hydrogen, an alkyl group having 1 to 30 carbon atoms or an alkenyl group having 1 to 30 carbon atoms. When X<sup>1</sup> represents an alkyl group or an alkenyl group, in the point of storage stability, it is necessary that the groups have 30 or less carbon atoms, preferably 20 or less carbon atoms, and more preferably 10 or less carbon atoms. Specific examples of such the alkyl group or the alkenyl group include alkyl groups (wherein the alkyl group may be straight-chained or branched) such as a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, a heptyl group, an octyl group, a nonyl group and a decyl group; alkenyl groups (wherein the alkenyl group may be straight-chained or branched, and the position of the double bond is arbitrary) such as an ethenyl group, a propenyl group, a butenyl group, a pentenyl group, a hexenyl group, a heptenyl group, an octenyl group, a nonenyl group and a decenyl group; and the like. Further, in the point of excellence in the sludge resistance or the like, an alkyl group is preferred. As X<sup>1</sup>, in the point of improving the frictional property, improving persistent effect of the frictional property or the like, hydrogen, an alkyl group having 1 to 20 carbon atoms or an alkenyl group having 1 to 20 carbon atoms is preferred; hydrogen or an alkyl group having 1 to 20 carbon atoms is more preferred; and hydrogen or an alkyl group having 1 to 10 carbon atoms is even more preferred.

**[0090]** In the formula (2), Y<sup>1</sup> is an alkali metal or an alkali earth metal, and specific examples include sodium, potassium, magnesium, calcium and the like. Among these, in the point of improving persistent effect of the frictional property, an alkali earth metal is preferred. In formula (2), m is 1 when Y<sup>1</sup> is an alkali metal and 2 when Y<sup>1</sup> is an alkali earth metal.

**[0091]** In the formula (3), Z is a residue having a hydroxyl group removed from a polyhydric alcohol with two or more valences. Specific examples of such the polyhydric alcohol include dihydric alcohols such as ethylene glycol, propylene glycol, 1,4-butanediol, 1,2-butanediol, neopentyl glycol, 1,6-hexandiol, 1,2-octandiol, 1,8-octandiol, isoprene glycol, 3-methyl-1,5-pentanediol, sorbite, catechol, resorcin, hydroxynone, bisphenol A, bisphenol F, hydrogenated bisphenol A, hydrogenated bisphenol F and dimmer diol; trihydric alcohols such as glycerin, 2-(hydroxymethyl)-1,3-propandiol, 1,2,3-butanetriol, 1,2,3-pentanetriol, 2-methyl-1,2,3-propanetriol, 2-methyl-2,3,4-butanetriol, 2-ethyl-1,2,3-butanetriol, 2,3,4-pentanetriol, 2,3,4-hexanetriol, 4-propyl-3,4,5-heptanetriol, 2,4-dimethyl-2,3,4-pentanetriol, 1,2,4-butanetriol, 1,2,4-pentanetriol, trimethylol ethane and trimethylol propane; tetrahydric alcohols such as pentaerythritol, erythritol, 1,2,3,4-pentantetrol, 2,3,4,5-hexantetrol, 1,2,4,5-pentantetrol, 1,3,4,5-hexantetrol, diglycerin and sorbitan; pentahydric alcohols such as adonitol, arabitol, xylitol and triglycerin; hexahydric alcohols such as dipentaerythritol, sorbitol, mannitol, iditol, inositol, dulcitol, talose and allose; polyglycerin or a dehydrated condensate thereof; and the like.

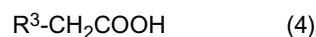
**[0092]** In the formula (3), m is an integer of 1 or more, m' is an integer of 0 or more, and m + m' is the same as a valence number of Z. That is, among hydroxyl groups of the polyhydric alcohol of Z, all of them may be substituted or a part of them may be substituted.

**[0093]** The component (C-1) is at least a compound selected from the formulas (1) to (3), but in the point of improving persistent effect of the frictional property, at least a compound selected from the formulas (1) and (2). In addition, at least a compound selected from the formulas (1) to (3) may be used alone or used in a mixture of two or more compounds.

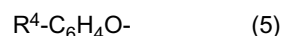
**[0094]** As a preferred example of a compound represented by the formula (1), N-oleoyl sarcosine may be mentioned, wherein R<sup>1</sup> is an alkenyl group having 17 carbon atoms, R<sup>2</sup> is a methyl group, X<sup>1</sup> is hydrogen and n is 1.

Component (C-2):

**[0095]** Here, the component (C-2) is a compound represented by the following formula (4).



wherein R<sup>3</sup> is an alkyl group having 7 to 29 carbon atoms, an alkenyl group having 7 to 29 carbon atoms or a group represented by the formula (5).



wherein R<sup>4</sup> is an alkyl group having 1 to 20 carbon atoms or hydrogen.

**[0096]** In the formula (4), R<sup>3</sup> is an alkyl group having 7 to 29 carbon atoms, an alkenyl group having 7 to 29 carbon atoms or a group represented by the formula (5). When R<sup>3</sup> represents an alkyl group, it is necessary that the alkyl group has 7 to 29 carbon atoms. In the point of the solubility to a base oil or the like, it is necessary that the alkyl group has 7 or more carbon atoms, and preferably 9 or more carbon atoms. Moreover, in the point of the storage stability or the like, it is necessary that the alkyl group has 29 or less carbon atoms, preferably 22 or less carbon atoms, and more preferably 19 or less carbon atoms. Specific examples of such the alkyl group include a heptyl group, an octyl group, a nonyl group, a decyl group, an undecyl group, a dodecyl group, a tridecyl group, a tetradecyl group, a pentadecyl group, a hexadecyl



group, a heptadecyl group, an octadecyl group, a nonadecyl group and the like (wherein the alkyl group may be straight-chained or branched).

[0097] When  $R^3$  represents an alkenyl group, it is necessary that the alkenyl group has 7 to 29 carbon atoms. In the point of the solubility to a base oil or the like, it is necessary that the alkenyl group has 7 or more carbon atoms, and preferably 9 or more carbon atoms. Moreover, in the point of the storage stability or the like, it is necessary that the alkenyl group has 29 or less carbon atoms, preferably 22 or less carbon atoms, and more preferably 19 or less carbon atoms. Specific examples of such the alkenyl group include a heptenyl group, an octenyl group, a nonenyl group, a decenyl group, an undecenyl group, a dodecenyl group, a tridecenyl group, a tetradecenyl group, a pentadecenyl group, a hexadecenyl group, a heptadecenyl group, an octadecenyl group, a nonadecenyl group and the like (wherein the alkenyl group may be straight-chained or branched).

[0098] In the formula (5),  $R^4$  is an alkyl group having 1 to 20 carbon atoms or hydrogen. When  $R^4$  is an alkyl group, in the point of the storage stability or the like, it is necessary that the alkyl group has 20 or less carbon atoms, preferably 19 or less carbon atoms, and more preferably 15 or less carbon atoms. Moreover, in the point of solubility to a base oil or the like, it is preferable that the alkyl group has 3 or more carbon atoms, and more preferably 5 or more carbon atoms. In addition, in the case where  $R^4$  is an alkyl group, its substitution site is arbitrary, but in the point of excelling in improving effect of the frictional property, the para position or the meta position is preferred, and the para position is more preferred. In the formula (4),  $R^3$  may be an alkyl group having 7 to 29 carbon atoms, an alkenyl group having 7 to 29 carbon atoms or a group represented by the formula (5) as described above, but in the point of excelling in the frictional property or the like, a group represented by the formula (5) is preferred.

Component (D): ester oiliness improver

[0099] To the lubricative composition of the invention, an ester oiliness improver as the component (D) is added.

[0100] The ester oiliness improver as the component (D) can be obtained by reacting alcohol with carboxylic acid. As an alcohol, a monohydric alcohol as well as a polyhydric alcohol may be used. Further, as a carboxylic acid, a monobasic acid as well as a polybasic acid may be used.

[0101] As a monohydric alcohol composing the ester oiliness improver, in general, the one having 1 to 24 carbon atoms, preferably 1 to 12, and more preferably 1 to 8 can be used. Such an alcohol may be straight-chained or branched, and may be saturated or unsaturated. Specific examples of the alcohol having 1 to 24 carbon atoms include methanol, ethanol, straight-chained or branched propanol, straight-chained or branched butanol, straight-chained or branched pentanol, straight-chained or branched hexanol, straight-chained or branched heptanol, straight-chained or branched octanol, straight-chained or branched nonanol, straight-chained or branched decanol, straight-chained or branched undecanol, straight-chained or branched dodecanol, straight-chained or branched tridecanol, straight-chained or branched tetradecanol, straight-chained or branched pentadecanol, straight-chained or branched hexadecanol, straight-chained or branched heptadecanol, straight-chained or branched octadecanol, straight-chained or branched nonadecanol, straight-chained or branched icosanol, straight-chained or branched heneicosanol, straight-chained or branched tricosanol, straight-chained or branched tetracosanol, and mixtures thereof, and the like.

[0102] As a polyhydric alcohol composing the ester oiliness improver, in general, 2 to 10, preferably 2 to 6 valent alcohol can be used. Specific examples of the polyhydric alcohol of 2 to 10 valences include dihydric alcohols such as ethylene glycol, diethylene glycol, polyethylene glycol (3 to 15-meric ethylene glycol), propylene glycol, dipropylene glycol, polypropylene glycol (3 to 15-meric propylene glycol), 1,3-propandiol, 1,2-propandiol, 1,3-butandiol, 1,4-butandiol, 2-methyl-1,2-propandiol, 2-methyl-1,3-propandiol, 1,2-pentandiol, 1,3-pentandiol, 1,4-pentandiol, 1,5-pentandiol and neopentyl glycol; polyhydric alcohols such as glycerin, polyglycerin (2 to 8-meric glycerin, e.g., diglycerin, triglycerin, tetraglycerin, etc.), trimethylol alkane (trimethylol ethane, trimethylol propane, trimethylol butane, etc.) and 2 to 8-mers thereof, pentaerythritol and 2 to 4-mers thereof, 1,2,4-butanetriol, 1,3,5-pentanetriol, 1,2,6-hexanetriol, 1,2,3,4-butanetetrol, sorbitol, sorbitan, sorbitol glycerin condensate, adonitol, arabitol, xylitol and mannitol; sugars such as xylose, arabinose, ribose, rhamnose, glucose, fructose, galactose, mannose, sorbose, cellobiose, maltose, isomaltose, trehalose, sucrose and mixtures thereof; and the like.

[0103] Among these polyhydric alcohols, polyhydric alcohols of 2 to 6 valences such as ethylene glycol, diethylene glycol, polyethylene glycol (3 to 10-meric ethylene glycol), propylene glycol, dipropylene glycol, polypropylene glycol (3 to 10-meric propylene glycol), 1,3-propandiol, 2-methyl-1,2-propandiol, 2-methyl-1,3-propandiol, neopentyl glycol, glycerin, diglycerin, triglycerin, trimethylol alkane (trimethylol ethane, trimethylol propane, trimethylol butane, etc.) and 2 to 4-mers thereof, pentaerythritol, dipentaerythritol, 1,2,4-butanetriol, 1,3,5-pentanetriol, 1,2,6-hexanetriol, 1,2,3,4-butanetetrol, sorbitol, sorbitan, sorbitol glycerin condensate, adonitol, arabitol, xylitol, mannitol and mixtures thereof, are preferred. More preferred are ethylene glycol, propylene glycol, neopentyl glycol, glycerin, trimethylol ethane, trimethylol propane, pentaerythritol, sorbitan and mixtures thereof, and glycerin is particularly preferred.

[0104] The alcohol composing the ester oiliness improver in the component (D) of the invention may be a monohydric alcohol or a polyhydric alcohol, as mentioned above. However, in the point of excelling in the frictional property, a

polyhydric alcohol is preferred.

**[0105]** Further, among acids composing the ester oiliness improver in the component (D) of the invention, an aliphatic acid having 1 to 24 carbon atoms in general, can be used as a monobasic acid. Such the aliphatic acid may be straight-chained or branched, and may be saturated or unsaturated. Specific examples thereof include saturated aliphatic acids such as formic acid, acetic acid, propionic acid, straight-chained or branched butanoic acid, straight-chained or branched pentanoic acid, straight-chained or branched hexanoic acid, straight-chained or branched heptanoic acid, straight-chained or branched octanoic acid, straight-chained or branched nonanoic acid, straight-chained or branched decanoic acid, straight-chained or branched undecanoic acid, straight-chained or branched dodecanoic acid, straight-chained or branched tridecanoic acid, straight-chained or branched tetradecanoic acid, straight-chained or branched pentadecanoic acid, straight-chained or branched hexadecanoic acid, straight-chained or branched heptadecanoic acid, straight-chained or branched octadecanoic acid, straight-chained or branched hydroxyoctadecanoic acid, straight-chained or branched nonadecanoic acid, straight-chained or branched icosanoic acid, straight-chained or branched heneicosanoic acid, straight-chained or branched docosanoic acid, straight-chained or branched tricosanoic acid and straight-chained or branched tetracosanoic acid; unsaturated aliphatic acids such as acrylic acid, straight-chained or branched butenoic acid, straight-chained or branched pentenoic acid, straight-chained or branched hexenoic acid, straight-chained or branched heptenoic acid, straight-chained or branched octenoic acid, straight-chained or branched nonenoic acid, straight-chained or branched decenoic acid, straight-chained or branched undecenoic acid, straight-chained or branched dodecenoic acid, straight-chained or branched tridecenoic acid, straight-chained or branched tetradecenoic acid, straight-chained or branched pentadecenoic acid, straight-chained or branched hexadecenoic acid, straight-chained or branched heptadecenoic acid, straight-chained or branched octadecenoic acid, straight-chained or branched hydroxyoctadecenoic acid, straight-chained or branched nonadecenoic acid, straight-chained or branched icosenoic acid, straight-chained or branched heneicosenoic acid, straight-chained or branched docosenoic acid, straight-chained or branched tricosenoic acid and straight-chained or branched tetracosenoic acid and mixtures thereof and the like.

**[0106]** As a polyhydric acid, a dibasic acid, trimellitic acid and the like can be mentioned, but a dibasic acid is preferred. The dibasic acid may be a chain dibasic acid or a cyclic dibasic acid. In the case of the chain dibasic acid, it may be straight-chained or branched, and may be saturated or unsaturated. As the chain dibasic acid, a chain dibasic acid having 2 to 16 carbon atoms is preferred, and specific examples include ethane diacid, propane diacid, straight-chained or branched butane diacid, straight-chained or branched pentane diacid, straight-chained or branched hexane diacid, straight-chained or branched heptane diacid, straight-chained or branched octane diacid, straight-chained or branched nonane diacid, straight-chained or branched decane diacid, straight-chained or branched undecane diacid, straight-chained or branched dodecane diacid, straight-chained or branched tridecane diacid, straight-chained or branched tetradecane diacid, straight-chained or branched heptadecane diacid, straight-chained or branched hexadecane diacid, straight-chained or branched hexene diacid, straight-chained or branched heptene diacid, straight-chained or branched octene diacid, straight-chained or branched nonene diacid, straight-chained or branched decene diacid, straight-chained or branched undecene diacid, straight-chained or branched dodecene diacid, straight-chained or branched tridecene diacid, straight-chained or branched tetradecene diacid, straight-chained or branched heptadecene diacid, straight-chained or branched hexadecene diacid, and mixtures thereof, and the like. Further, examples of the cyclic dibasic acid include 1,2-cyclohexane dicarboxylic acid, 4-cyclohexene-1,2-dicarboxylic acid, aromatic dicarboxylic acid and the like. Among these, in the point of stability, chain dibasic acids are preferred.

**[0107]** The acid composing the ester oiliness improver may be a monobasic acid or a polybasic acid, as mentioned above. However, in the point of the frictional property, a monobasic acid is preferred.

**[0108]** The combination of alcohol and acid in the ester oiliness improver is arbitrary and not particularly limited, but, for example, the esters according to the following combinations (i) to (vii) can be mentioned.

- (i) an ester of monohydric alcohol and monobasic acid,
- (ii) an ester of polyhydric alcohol and monobasic acid,
- (iii) an ester of monohydric alcohol and polybasic acid,
- (iv) an ester of polyhydric alcohol and polybasic acid,
- (v) a mixed ester of a mixture of monohydric alcohol with polyhydric alcohol and polybasic acid,
- (vi) a mixed ester of polyhydric alcohol and a mixture of monobasic acid with polybasic acid, and
- (vii) a mixed ester of a mixture of monohydric alcohol with polyhydric alcohol and a mixture of monobasic acid with polybasic acid.

**[0109]** Each ester of (ii) to (vii) above may be a complete ester which esterified all hydroxyl groups of polyhydric alcohol or carboxyl groups of polybasic acid, or a partial ester in which a part thereof remains as hydroxyl groups or carboxyl groups.

**[0110]** Among the esters of (i) to (vii) above, (ii) an ester of polyhydric alcohol and monobasic acid is preferred as the ester oiliness improver of the component (D). This ester is very high in the improving effect of the frictional property.

**[0111]** The esters of (ii) may be a complete ester in which all hydroxyl groups of polyhydric alcohol is esterified or a partial ester in which a part remains as hydroxyl groups. The complete ester is preferred in the point of precipitation resistance and the partial ester is preferred in the point of the frictional property.

**[0112]** When the esters of (ii) contains a partial ester, the partial ester is preferably a mixture of two or more kind of partial esters having different esterification degree, and more preferably a mixture of partial esters having the esterification degree of 1 and partial esters having the esterification degree of 2 or more in the point of the precipitation resistance, the abrasion resistance and the frictional property. Moreover, the esterification degree herein means the number of ester bonds in one molecule. For example, when the polyhydric alcohol constituting the esters of (ii) is sorbitan, a partial ester having the esterification degree of 1 is sorbitan monoester, one the other hand, a partial ester having the esterification degree of 2 or more includes sorbitan diester and sorbitan triester.

**[0113]** Furthermore, when the esters of (ii) contains both partial esters having the esterification degree of 1 and partial esters having the esterification degree of 2 or more, the proportion of the partial ester having the esterification degree of 1 is preferably 10 to 70% by mole, and more preferably 20 to 50% by mole based on the total amount of both. Also, the proportion of the partial ester having the esterification degree of 2 or higher is preferably 30 to 70% by mole, and more preferably 50 to 80% by mole based on the total amount of both.

**[0114]** Monobasic acid constituting the esters of (ii) may be any one of a straight-chained fatty acid or branched fatty acid, but in the point of the frictional property, a straight-chained fatty acid is preferred, and in the point of the precipitation resistance, a branched fatty acid is preferred.

**[0115]** Moreover, monobasic acid constituting the esters of (ii) may be any one of a saturated fatty acid or an unsaturated fatty acid, but in the point of the frictional property and abrasion resistance, a saturated fatty acid is preferred, and in the point of the precipitation resistance, an unsaturated fatty acid is preferred. Further, when monobasic acid contains both saturated and unsaturated fatty acids the proportion of saturated fatty acids in the total of both fatty acids is preferably 60% by mole or higher, more preferably 75% by mole or higher, even more preferably 90% by mole or higher, and still more preferably 95% by mole or higher in the point of the friction property (i.e. reduction of the frictional index).

**[0116]** The number of carbon atoms in monobasic acid constituting ester of the (ii) is not particularly limited, but in the point of the precipitation resistance, a mixture of short-chained fatty acid having 1 to 4 carbon atoms, preferably 2 carbon atoms and long-chained fatty acid having 10 to 24 carbon atoms, preferably 12 to 18 carbon atoms is preferably used. Further, when combining the short-chained fatty acid and the long-chained fatty acid, the proportion of the short-chained fatty acid in the total of both is preferably 50 to 80% by mole, more preferably 60 to 75% by mole, and even more preferably 65 to 70% by mole.

**[0117]** Among the esters of (ii), esters represented in the following (ii-1) to (ii-4) are preferably used in the point of achieving various characteristic such as the frictional property and the precipitation resistance in good balance.

(ii-1) monoester of polyhydric alcohol and unsaturated fatty acid ester,

(ii-2) ester of polyhydric alcohol and unsaturated fatty acid, which is a mixture of partial ester having the esterification degree of 1 and partial ester having the esterification degree of 2 or more,

(ii-3) complete ester of polyhydric alcohol and saturated fatty acid, wherein the constituting fatty acid is a mixture of short-chained fatty acid having 1 to 4 carbon atoms and long-chained fatty acid having 10 to 24 carbon atoms, and

(ii-4) ester of polyhydric alcohol and brached saturated fatty acid, which is a mixture of partial ester having the esterification degree of 1 and partial ester having the esterification degree of 2 or more.

**[0118]** The esters of (ii-1) are preferred in the point that both the frictional property and the precipitation resistance can be achieved in high degree. The number of carbon atoms in the unsaturated fatty acid constituting the esters of (ii-1) is preferably 10 or higher, more preferably 12 or higher, and even more preferably 14 or higher in the point of further improving the frictional property. Further, the number of carbon atoms in the unsaturated fatty acid is preferably 28 or lower, more preferably 26 or lower and even more preferably 24 or lower in the point of the precipitation resistance. Examples of such esters include glycerin monoleate, sorbitan monoleate and the like.

**[0119]** The esters of (ii-2) are preferred in the point of achieving all the frictional property, the precipitation resistance, the abrasion resistance and the rust resistance in good balance with high degree. Further polyhydric alcohol constituting ester of the (ii-2) may be one kind or a mixture of two or more kinds, but when the polyhydric alcohol contains dihydric alcohol, it is preferable to further contain polyhydric alcohols of 3 or more valence.

**[0120]** In the esters of (ii-2), the proportion of partial ester having the esterification degree of 1 is preferably 10 to 70% by mole, and more preferably 20 to 50 % by mole based on the total amount of partial ester having the esterification degree of 1 and partial ester having the esterification degree of 2 or more. Further, the proportion of partial ester having the esterification degree of 2 or more is preferably 30 to 70% by mole, and more preferably 50 to 80 % by mole based on the total amount of partial ester having the esterification degree of 1 and partial ester having the esterification degree of 2 or more. In addition, the esters of (ii-2) may further contain complete esters, but in the point of achieving all the frictional property, the precipitation resistance, the abrasion resistance and the rust resistance in good balance with high

degree, the content of the complete ester is 10% by mole or lower, preferably 5% by mole or lower, and particularly preferably contains substantially no complete esters based on the esters of (ii-2).

**[0121]** Esters of (ii-3) are preferred in the point of achieving all the frictional property (i.e. reduction of the frictional index and energy-saving property), the precipitation resistance, the abrasion resistance and the rust resistance in good balance with high degree. In the esters of (ii-3), it is preferable to combine short-chained fatty acid having 2 carbon atoms with long-chained fatty acid having 12 to 18 carbon atoms in the point of further improving the precipitation resistance. Moreover, the proportion of the short-chained fatty acid in the total amount of the constituting fatty acids is preferably 60 to 80% by mole, more preferably 60 to 75% by mole, and even more preferably 65 to 70% by mole. When the proportion of the short-chained fatty acid is less than 50% by mole, there is tendency to lower the solubility to the base oil. Moreover, when the proportion of the short-chained fatty acid exceeds 80% by mole, there is tendency to lower the reduction effect of the friction. Further, in the point of further improving the frictional property (i.e. reduction of the frictional index), the proportion of the saturated fatty acid in the total amount of the constituting fatty acids is preferably 60% by mole or higher, more preferably 75% by mole or higher, even more preferably 90% by mole or higher, and still more preferably 95% by mole or higher. When the proportion of the saturated fatty acid is less than 60% by mole, there is tendency that the reduction effect of the friction becomes insufficient.

**[0122]** Esters of (ii-4) are preferred in the point of achieving all the frictional property, the precipitation resistance, the abrasion resistance and the rust resistance in good balance with high degree. Further, polyhydric alcohols constituting the esters of (ii-4) may be one kind or a mixture of two or more kinds, but when the polyhydric alcohol contains dihydric alcohol, it is preferable to further contain polyhydric alcohols of 3 or more valence.

**[0123]** In the esters of (ii-4), the proportion of partial ester having the esterification degree of 1 is preferably 10 to 70% by mole, and more preferably 20 to 50 % by mole based on the total amount of partial ester having the esterification degree of 1 and partial ester having the esterification degree of 2 or more. Further, the proportion of partial ester having the esterification degree of 2 or more is preferably 30 to 70% by mole, and more preferably 50 to 80 % by mole based on the total amount of partial ester having the esterification degree of 1 and partial ester having the esterification degree of 2 or more. In addition, the esters of (ii-4) may further contain complete esters, but in the point of achieving all the frictional property, the precipitation resistance, the abrasion resistance and the rust resistance in good balance with high degree, the content of the complete ester is 10% by mole or lower, preferably 5% by mole or lower, and particularly preferably contains substantially no complete esters based on the esters of (ii-4).

**[0124]** Among the esters of (ii-1) to (ii-4), in the point of having good balance of the frictional property, the precipitation resistance and the abrasion resistance, and excellent rust resistance, the esters of (ii-2), (ii-3) and (ii-4) are preferred as (D-1), (D-2) and (D-3) of the component (D), respectively. More preferably, esters of (D-1) are preferred.

(D-1) ester of polyhydric alcohol and unsaturated fatty acid, which is a mixture of partial ester having the esterification degree of 1 and partial ester having the esterification degree of 2 or more,

(D-2) complete ester of polyhydric alcohol and saturated fatty acid, wherein the constituting fatty acid is a mixture of short-chained fatty acid having 1 to 4 carbon atoms and long-chained fatty acid having 10 to 24 carbon atoms, and

(D-3) ester of polyhydric alcohol and brached saturated fatty acid, which is a mixture of partial ester having the esterification degree of 1 and partial ester having the esterification degree of 2 or more.

#### <Specific Embodiment>

**[0125]** The lubricative composition of the invention is specifically suitable as a lubricating oil composition. As more specific embodiment, it is suitable for a gear oil composition, a lubricating oil composition for paper machines, a lubricating oil composition for slide guides, a hydraulic oil, and the like.

#### (Embodiment of gear oil composition)

**[0126]** Gear oil composition is more specifically a composition containing at least one kind of the component (A), the component (B) and the component (C) in the base oil.

**[0127]** The mixing ratio of the component (A), the component (B) and the component (C) concerning the gear oil composition is as in the following.

**[0128]** The content of a phosphorus-containing carboxylic acid compound as the component (A-1) in the gear oil composition of the invention is not particularly limited, but it is preferably 0.001 to 5% by weight, more preferably 0.002 to 3% by weight, and even more preferably 0.003 to 1% by weight based on the total amount of the composition. The content of the phosphorus-containing carboxylic acid compound lower than the minimum value has tendency not to obtain sufficient lubricity. On the other hand, the content exceeding the maximum value has tendency not to obtain appropriate improving effect of lubricity that meet with the content, and may have concerns of lowering thermal and oxidation stability or hydrolysis stability, thus not preferred. Further, among phosphorylated carboxylic acid represented

by the formula (1), the content of the compound (including  $\beta$ -dithiophosphorylated propionic acid represented by the formula (6)) whose  $R^4$  is a hydrogen atom, is preferably 0.001 to 0.1 % by weight, more preferably 0.002 to 0.08% by weight, even more preferably 0.003 to 0.07% by weight, much more preferably 0.004 to 0.06% by weight, and particularly preferably 0.005 to 0.05% by weight. When the content is less than 0.001, there may be a concern of having insufficient improving effect of the extreme pressure property, and on the other hand, when the content exceeds 0.1% by weight, there may be a concern of lowering the thermal and oxidation stability.

**[0129]** The content of thiophosphoric ester (phosphorothionate) as the component (A-2) in the gear oil composition of the invention is not particularly limited, but usually the content is 0.001 to 10% by weight, preferably 0.005 to 5% by weight, and more preferably 0.01 to 3% by weight based on the total amount of the gear oil composition (based on the total amount of a base oil and all the combined additives).

**[0130]** The gear oil composition of the invention can be obtained by (A-1) the phosphorus-containing carboxylic acid compound or (A-2) the thiophosphoric ester alone, or in combination of both.

**[0131]** The maximum value of the content of the dispersant viscosity index improver as an arbitrary component (B) in the gear oil composition of the invention is 10% by weight, preferably 5% by weight, and more preferably 2% by weight based on the total amount of the composition. When the content exceeds 10% by weight, further improvement in the sludge resisting effect that meet with the content cannot be seen, and lowering of viscosity by shearing is generated, thus not preferred. On the other hand, the minimum value of the content of the dispersant viscosity index improver is 0.01 % by weight, preferably 0.05% by weight, and more preferably 0.1 % by weight based on the total amount of the composition. When the content of the dispersant viscosity index improver is less than 0.01% by weight, its additive effect cannot be seen, and there may be a concern of worsening the sludge resisting effect of the gear oil composition, thus not preferred.

**[0132]** The maximum value of the content of the component (C-1) as an arbitrary component in the gear oil composition of the invention is 5% by weight, preferably 2% by weight, and more preferably 1% by weight based on the total amount of the composition. When the content exceeds 5% by weight, further improvement in the frictional property that meet with the content cannot be seen, and lowering of the storage stability is generated, thus not preferred. On the other hand, the minimum value of the content of the component (C-1) is 0.001% by weight, preferably 0.003% by weight, and more preferably 0.005% by weight based on the total amount of the composition. When the content of the component (C-1) is less than 0.001% by weight, the improving effect of the frictional property cannot be seen, thus not preferred.

**[0133]** The content of the component (C-2), when combining in the gear oil composition of the invention, is arbitrary, but since there may be a concern of lowering the sludge resistance when a great amount is combined, the content is 5% by weight or lower, preferably 1% by weight or lower, and more preferably 0.5% by weight or lower based on the total amount of the composition. On the other hand, in the point of exhibiting sufficient improving effect of the frictional property, the content is 0.001 % by weight or higher, preferably 0.003% by weight or higher, and more preferably 0.005% by weight or higher based on the total amount of the composition.

#### (Embodiment of lubricating oil for paper machines)

**[0134]** The lubricative composition of the invention is specifically suitable as a lubricating oil composition for paper machines.

**[0135]** Lubricating oil composition for paper machines is more specifically a composition containing at least one kind of the component (A), the component (B) and the component (C) in the base oil.

**[0136]** The mixing ratio of the component (A), the component (B) and the component (C) concerning the lubricating oil composition for paper machines is as in the following.

**[0137]** The content of (A-1) a phosphorus-containing carboxylic acid compound in the lubricating oil composition for paper machines of the invention is not particularly limited, but it is preferably 0.001 to 5% by weight, more preferably 0.002 to 3% by weight, and even more preferably 0.003 to 1% by weight based on the total amount of the composition. The content of a phosphorus-containing carboxylic acid compound lower than the minimum value has tendency not to obtain sufficient lubricity. On the other hand, the content exceeding the maximum value has tendency not to obtain appropriate improving effect of lubricity that meet with the content, and may have concerns of lowering thermal and oxidation stability or hydrolysis stability, thus not preferred. Further, among phosphorylated carboxylic acid represented by the formula (1), the content of the compound (including  $\beta$ -dithiophosphorylated propionic acid represented by the formula (6)) whose  $R^4$  is a hydrogen atom, is preferably 0.001 to 0.1% by weight, more preferably 0.002 to 0.08% by weight, even more preferably 0.003 to 0.07% by weight, much more preferably 0.004 to 0.06% by weight, and particularly preferably 0.005 to 0.05% by weight. When the content is less than 0.001, there may be a concern of having insufficient improving effect of the extreme pressure property, and on the other hand, when the content exceeds 0.1 % by weight, there may be a concern of lowering the thermal and oxidation stability.

**[0138]** The content of thiophosphoric ester (phosphorothionate) as the component (A-2) is not particularly limited, but usually the content is 0.001 to 10% by weight, preferably 0.005 to 5% by weight, and more preferably 0.01 to 3% by

weight based on the total amount of the lubricating oil composition for paper machines (based on the total amount of a base oil and all the combined additives).

**[0139]** The lubricating oil composition for paper machines of the invention can be obtained by (A-1) the phosphorus-containing carboxylic acid compound or (A-2) the phosphorothionate alone, or in combination of both.

**[0140]** The maximum value of the content of the component (B), the dispersant viscosity index improver, as an arbitrary component in the lubricating oil composition for paper machines of the invention is 10% by weight, preferably 5% by weight, and more preferably 2% by weight based on the total amount of the composition. When the content exceeds 10% by weight, further improvement in the sludge resisting effect that meet with the content cannot be seen, and lowering of viscosity by shearing is generated, thus not preferred. On the other hand, the minimum value of the content of the component (B) is 0.01% by weight, preferably 0.05% by weight, and more preferably 0.1 % by weight based on the total amount of the composition. When the content of the component (B) is less than 0.01 % by weight, its additive effect cannot be seen, and there may be a concern of worsening the sludge resisting effect of the lubricating oil composition for paper machines, thus not preferred.

**[0141]** The maximum value of the content of the component (C-1) as an arbitrary component in the lubricating oil composition for paper machines of the invention is 5% by weight, preferably 2% by weight, and more preferably 1% by weight based on the total amount of the composition. When the content exceeds 5% by weight, further improvement in the frictional property that meet with the content cannot be seen, and lowering of the storage stability is generated, thus not preferred. On the other hand, the minimum value of the content of the component (C-1) is 0.001% by weight, preferably 0.003% by weight, and more preferably 0.005% by weight based on the total amount of the composition. When the content of the component (C-1) is less than 0.001% by weight, the improving effect of the frictional property cannot be seen, thus not preferred.

**[0142]** The content of the component (C-2), when combining in the lubricating oil composition for paper machines of the invention, is arbitrary, but since there may be a concern of lowering the sludge resistance when a great amount is combined, the content is 5% by weight or lower, preferably 1% by weight or lower, and more preferably 0.5% by weight or lower based on the total amount of the composition. On the other hand, in the point of exhibiting sufficient improving effect of the frictional property, the content is 0.001% by weight or higher, preferably 0.003% by weight or higher, and more preferably 0.005% by weight or higher based on the total amount of the composition.

#### (Embodiment of lubricating oil for slide guides)

**[0143]** The lubricative composition of the invention is specifically suitable as a lubricating oil composition for slide guides.

**[0144]** Lubricating oil composition for slide guides is more specifically a composition containing at least one kind of the component (A), the component (B) and the component (C) in the base oil.

**[0145]** The mixing ratio of the component (A), the component (B) and the component (C) concerning the lubricating oil composition for slide guides is as in the following.

**[0146]** The content of (A-1) a phosphorus-containing carboxylic acid compound in the lubricating oil composition for slide guides of the invention is not particularly limited, but it is preferably 0.001 to 5% by weight, more preferably 0.002 to 3% by weight, and even more preferably 0.003 to 1% by weight based on the total amount of the lubricating oil composition for slide guides. The content of a phosphorus-containing carboxylic acid compound lower than the minimum value has tendency not to obtain sufficient low frictional property and anti-stick-slip property. On the other hand, the content exceeding the maximum value has tendency not to obtain low frictional property and anti-stick-slip property that meet with the content, and may have concerns of lowering thermal and oxidation stability or hydrolysis stability, thus not preferred. Further, among phosphorylated carboxylic acid represented by the formula (1), the content of the compound (including  $\beta$ -dithiophosphorylated propionic acid represented by the formula (6)) whose  $R^4$  is a hydrogen atom, is preferably 0.001 to 0.1% by weight, more preferably 0.002 to 0.08% by weight, even more preferably 0.003 to 0.07% by weight, much more preferably 0.004 to 0.06% by weight, and particularly preferably 0.005 to 0.05% by weight. When the content is less than 0.001, there may be a concern of having insufficient improving effect of low frictional property and anti-stick-slip property, and on the other hand, when the content exceeds 0.1 % by weight, there may be a concern of lowering the thermal and oxidation stability.

**[0147]** The content of thiophosphoric ester (phosphorothionate) as the component (A-2) is not particularly limited, but usually the content is 0.001 to 10% by weight, preferably 0.005 to 5% by weight, and more preferably 0.01 to 3% by weight based on the total amount of the lubricating oil composition for slide guides (based on the total amount of a base oil and all the combined additives).

**[0148]** The lubricating oil composition for slide guides of the invention can be obtained by (A-1) the phosphorus-containing carboxylic acid compound or (A-2) the thiophosphoric ester alone, or in combination of both.

**[0149]** The maximum value of the content of the component (B), the dispersant viscosity index improver, as an arbitrary component in the lubricating oil composition for slide guides of the invention is 10% by weight, preferably 5% by weight, and more preferably 2% by weight based on the total amount of the composition. When the content exceeds 10% by

weight, further improvement in the frictional property and separability from a soluble cutting oil that meet with the content cannot be seen, and lowering of viscosity by shearing is generated, thus not preferred. On the other hand, the minimum value of the content of the component (B) is 0.01% by weight, preferably 0.05% by weight, and more preferably 0.1% by weight based on the total amount of the composition. When the content of the component (B) is less than 0.01 % by weight, its additive effect cannot be seen, and there may be a concern of worsening the frictional property of the lubricating oil composition for slide guides and separability from soluble cutting oil, thus not preferred.

**[0150]** The maximum value of the content of the component (C-1) as an arbitrary component in the lubricating oil composition for slide guides of the invention is 5% by weight, preferably 2% by weight, and more preferably 1% by weight based on the total amount of the composition. When the content exceeds 5% by weight, further improvement in the frictional property that meet with the content cannot be seen, and lowering of the storage stability is generated, thus not preferred.

**[0151]** On the other hand, the minimum value of the content of the component (C-1) is 0.001% by weight, preferably 0.003% by weight, and more preferably 0.005% by weight based on the total amount of the composition. When the content of the component (C-1) is less than 0.001% by weight, the improving effect of the frictional property cannot be seen, thus not preferred.

**[0152]** The content of the component (C-2), when combining in the lubricating oil composition for slide guides of the invention, is arbitrary, but since there may be a concern of lowering the sludge resistance when a great amount is combined, the content is 5% by weight or lower, preferably 1% by weight or lower, and more preferably 0.5% by weight or lower based on the total amount of the composition. On the other hand, in the point of exhibiting sufficient improving effect of the frictional property, the content is 0.001 % by weight or higher, preferably 0.003% by weight or higher, and more preferably 0.005% by weight or higher based on the total amount of the composition.

#### (Embodiment of hydraulic oil)

**[0153]** The lubricative composition of the invention is specifically suitable as a hydraulic oil.

**[0154]** Hydraulic oil composition is more specifically a composition containing at least one kind of the component (A) and the component (B) in the base oil. Further, hydraulic oil composition is even more specifically a composition containing the component (D) in the base oil.

**[0155]** The mixing ratio of the component (A), the component (B) and the component (D) concerning the hydraulic oil composition is as in the following.

**[0156]** The content of (A-1) a phosphorus-containing carboxylic acid compound in the hydraulic oil of the invention is not particularly limited, but it is preferably 0.001 to 1% by weight, and more preferably 0.002 to 0.5% by weight based on the total amount of the composition. The content of a phosphorus-containing carboxylic acid compound lower than the minimum value has tendency to be insufficient in improving effect of abrasion resistance and frictional property. On the other hand, the content exceeding the maximum value has tendency to lower sludge resistance. Further, among phosphorylated carboxylic acid represented by the formula (1), the content of the compound (including  $\beta$ -dithiophosphorylated propionic acid represented by the formula (6)) whose  $R^4$  is a hydrogen atom, is preferably 0.001 to 0.1% by weight, more preferably 0.002 to 0.08% by weight, even more preferably 0.003 to 0.07% by weight, much more preferably 0.004 to 0.06% by weight, and particularly preferably 0.005 to 0.05% by weight. When the content is less than 0.001, there is tendency to be insufficient in improving effect of abrasion resistance and frictional property, and on the other hand, when the content exceeds 0.1 % by weight, there is tendency to lower sludge resistance.

**[0157]** The content of the component (A-2) in the hydraulic oil of the invention is preferably 5% by weight or lower, more preferably 2% by weight or lower, and even more preferably 1.5% by weight or lower based on the total amount of the composition. When the content exceeds 5% by weight, the thermal stability of the hydraulic oil becomes insufficient, and there is tendency to lower the sludge resistance. Moreover, the content of the component (A-2) is preferably 0.005% by weight or higher, more preferably 0.01 % by weight or higher, and even more preferably 0.05% by weight or higher based on the total amount of the composition. When the content is lower than 0.005% by weight, there is tendency to be insufficient in the abrasion resistance.

**[0158]** The hydraulic oil of the invention can be obtained by (A-1) the phosphorus-containing carboxylic acid compound or (A-2) the thiophosphoric ester alone, or in combination of both.

**[0159]** The content of (B) the dispersant viscosity index improver in the hydraulic oil of the invention is preferably 10% by weight or lower, more preferably 5% by weight or lower, and even more preferably 2% by weight or lower based on the total amount of the composition. When the content exceeds 10% by weight, further improvement in the sludge resistance, the abrasion resistance and the frictional property that meet with the content cannot be seen, and there is tendency to easily generate lowering of viscosity by shearing. Moreover, the content of (B) the dispersant viscosity index improver in the hydraulic oil is preferably 0.01% by weight or higher, more preferably 0.05% by weight or higher, and even more preferably 0.1 % by weight or higher based on the total amount of the composition. When the content is less than 0.01 % by weight, there is tendency to lower the sludge resistance, the abrasion resistance and the frictional property.

**[0160]** The content of (D) an ester oiliness improver in the hydraulic oil of the invention is arbitrary, but in the point of excelling in the friction reducing effect, the content is preferably 0.01% by weight or higher, more preferably 0.05% by weight or higher, and even more preferably 0.1 % by weight or higher based on the total amount of the composition. Further, the content, in the point of precipitation resistance, is preferably 10% by weight or lower, more preferably 7.5%

**[0161]** In the case where the hydraulic oil composition is a composition containing the component (A) and component (B) in the base oil, when a sulfur-based abrasion resistant or a phosphorus-based abrasion resistant is used, there may be a concern of increasing the friction index or worsening the sludge resistance; thus, the content of the sulfur-based abrasion resistant or the phosphorus-based abrasion resistant is 5% by weight or lower, preferably 1% by weight or lower, more preferably 0.5% by weight or lower based on the total amount of the composition, and it is the most preferable that no such abrasion resistant is contained therein.

**[0162]** As described above, the hydraulic oil composition is, more specifically, a composition containing the component (D) in the base oil, but in this case, when a sulfur-based abrasion resistant is used, there may be a concern of increasing the friction index or worsening the sludge resistance; thus, the content of the sulfur-based abrasion resistant is 5% by weight or lower, preferably 1% by weight or lower, more preferably 0.5% by weight or lower based on the total amount of the composition, and it is the most preferable that no such abrasion resistant is contained therein.

(Description of arbitrary components in lubricative composition)

**[0163]** Next, in addition to the components (A) to (D), the components (E) to (K) that are arbitrarily contained in the lubricative composition of the invention will be described below.

Component (E):

**[0164]** In the lubricative composition of the invention, the sulfur-based extreme pressure agent is preferably used as an arbitrary component (E) in the point of improving the extreme pressure property.

**[0165]** Specific examples of such the sulfur-based extreme pressure agent include sulfurized fats and oils, sulfurized fatty acids, sulfurized esters, sulfurized olefins, dihydrocarbyl (poly)sulfides, thiadiazole compounds, alkylthiocarbamoyl compounds, thiocarbamate compounds, thioterpenes compounds, dialkyl thiodipropionate compounds, sulfurized mineral oils, zinc dithiophosphate compounds, zinc dithiocarbamate compounds, molybdenum dithiophosphate compounds, molybdenum dithiocarbamate and the like. These sulfur-based extreme pressure agents may be used alone or in a mixture of two or more.

**[0166]** Sulfurized fats and oils are obtained by reacting sulfur or a sulfur-containing compound with fat and oil (lard oil, whale oil, plant oil, fish oil, etc.). The content of the sulfur is not particularly limited, but generally 5 to 30% by weight is preferred. Specific examples thereof include, sulfurized lard, sulfurized seed oil, sulfurized castor oil, sulfurized soybean oil, sulfurized rice bran oil, and mixtures thereof, and the like.

**[0167]** Examples of sulfurized fatty acids include sulfurized oleic acid and the like, and examples of sulfurized esters include the ones obtained by sulfurizing unsaturated fatty acid esters and mixtures thereof obtained by reacting unsaturated fatty acids (including oleic acid, linoleic acid or fatty acids extracted from the animal and plant fats and oils) and various alcohols, with an arbitrary method. Specific examples of sulfurized esters include sulfurized oleic acid methyl or sulfurized rice bran fatty acid octyl, and mixtures thereof, and the like.

**[0168]** Examples of sulfurized olefins include compounds represented by the following formula (18).

**[0169]** These compounds are obtained by reacting olefins having 2 to 15 carbon atoms or 2 to 4-mers thereof with sulfurizing agent such as sulfur and sulfur chloride, and for such olefins, propylene, isobutene, diisobutene and the like are preferred.



wherein  $R^{27}$  is an alkenyl group having 2 to 15 carbon atoms,  $R^{28}$  is an alkyl group or an alkenyl group having 2 to 15 carbon atoms, and a is an integer of 1 to 8.

**[0170]** Further, dihydrocarbyl (poly)sulfide is a compound represented by the following formula (19). Here, when  $R^{29}$  and  $R^{30}$  are an alkyl group, they may be referred to as alkyl sulfide.



wherein  $R^{29}$  and  $R^{30}$  may be identical with or different from each other and are each independently an alkyl group having 1 to 20 carbon atoms, a branched or cyclic alkyl group, an aryl group having 6 to 20 carbon atoms, an alkylaryl group having 7 to 20 carbon atoms or an arylalkyl group having 7 to 20 carbon atoms, and b is an integer of 1 to 8.

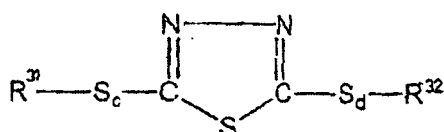


**[0171]** In the formula (19), specific examples of R<sup>29</sup> and R<sup>30</sup> include straight-chained or branched alkyl groups such as an n-propyl group, an isopropyl group, an n-butyl group, an isobutyl group, a sec-butyl group, a tert-butyl group, a straight-chained or branched pentyl group, a straight-chained or branched hexyl group, a straight-chained or branched heptyl group, a straight-chained or branched octyl group, a straight-chained or branched nonyl group, a straight-chained or branched decyl group, a straight-chained or branched undecyl group, a straight-chained or branched dodecyl group, a straight-chained or branched tridecyl group, a straight-chained or branched tetradecyl group, a straight-chained or branched pentadecyl group, a straight-chained or branched hexadecyl group, a straight-chained or branched heptadecyl group, a straight-chained or branched octadecyl group, a straight-chained or branched nonadecyl group and a straight-chained or branched icosyl group; aryl groups such as a phenyl group and a naphthyl group; alkylaryl groups such as a tolyl group (including all structural isomers), an ethylphenyl group (including all structural isomers), a straight-chained or branched propylphenyl group (including all structural isomers), a straight-chained or branched butylphenyl group (including all structural isomers), a straight-chained or branched pentylphenyl group (including all structural isomers), a straight-chained or branched hexylphenyl group (including all structural isomers), a straight-chained or branched heptylphenyl group (including all structural isomers), a straight-chained or branched octylphenyl group (including all structural isomers), a straight-chained or branched nonylphenyl group (including all structural isomers), a straight-chained or branched decylphenyl group (including all structural isomers), a straight-chained or branched undecylphenyl group (including all structural isomers), a straight-chained or branched dodecylphenyl group (including all structural isomers), a xylyl group (including all structural isomers), an ethylmethylphenyl group (including all structural isomers), a diethylphenyl group (including all structural isomers), a di(straight-chained or branched)propylphenyl group (including all structural isomers), a di(straight-chained or branched)butylphenyl group (including all structural isomers), a methylnaphthyl group (including all structural isomers), an ethylnaphthyl group (including all structural isomers), a straight-chained or branched propylnaphthyl group (including all structural isomers), a straight-chained or branched butylnaphthyl group (including all structural isomers), a dimethylnaphthyl group (including all structural isomers), an ethylmethylnaphthyl group (including all structural isomers), a diethylnaphthyl group (including all structural isomers), a di(straight-chained or branched)propylnaphthyl group (including all structural isomers) and a di(straight-chained or branched)butylnaphthyl group (including all structural isomers); arylalkyl groups such as a benzyl group, a phenylethyl group (including all isomers) and a phenylpropyl group (including all isomers); and the like. Among these, R<sup>29</sup> and R<sup>30</sup> in the formula (19) are preferably an alkyl group having 3 to 18 carbon atoms derived from propylene, 1-butene or isobutylene, an aryl group, an alkylaryl group or an arylalkyl group having 6 to 8 carbon atoms. Examples of these groups include alkyl groups such as a branched hexyl group (including all branched isomers) derived from an isopropyl group or a propylene dimer, a branched nonyl group (including all branched isomers) derived from a propylene trimer, a branched dodecyl group (including all branched isomers) derived from a propylene tetramer, a branched pentadecyl group (including all branched isomers) derived from a propylene pentamer, a branched octadecyl group (including all branched isomers) derived from a propylene hexamer, a branched octyl group (including all branched isomers) derived from a sec-butyl group, a tert-butyl group or a 1-butene dimer, a branched octyl group (including all branched isomers) derived from an isobutylene dimer, a branched dodecyl group (including all branched isomers) derived from a 1-butene trimer, a branched dodecyl group (including all branched isomers) derived from an isobutylene trimer, a branched hexadecyl group (including all branched isomers) derived from a 1-butene tetramer and a branched hexadecyl group (including all branched isomers) derived from an isobutylene tetramer; alkylaryl groups such as a phenyl group, a tolyl group (including all structural isomers), an ethylphenyl group (including all structural isomers) and a xylyl group (including all structural isomers); arylalkyl groups such as a benzyl group and a phenylethyl group (including all isomers).

**[0172]** In addition, in the point of improving the processing efficiency and the tool life, R<sup>29</sup> and R<sup>30</sup> in the formula (19) are each independently preferably a branched alkyl group having 3 to 18 carbon atoms derived from ethylene or propylene, and particularly preferably a branched alkyl group having 6 to 15 carbon atoms derived from ethylene or propylene.

**[0173]** Preferred examples of dihydrocarbyl (poly)sulfide include dibenzyl polysulfide, various dinonyl polysulfides, various didodecyl polysulfides, various dibutyl polysulfides, various dioctyl polysulfides, diphenyl polysulfide, dicyclohexyl polysulfide, and mixtures thereof, and the like.

**[0174]** Examples of thiadiazole compounds include 1,3,4-thiadiazole represented by the following formula (20), 1,2,4-thiadiazole compounds represented by the following formula (21) and 1,4,5-thiadiazole compounds represented by the following formula (22).



(20)



**[0175]** Preferred specific examples of such the thiadiazole compound include 2,5-bis(n-hexyldithio)-1,3,4-thiadiazole, 2,5-bis(n-octyldithio)-1,3,4-thiadiazole, 2,5-bis(n-nonyldithio)-1,3,4-thiadiazole, 2,5-bis(1,1,3,3-tetramethylbutyldithio)-1,3,4-thiadiazole, 3,5-bis(n-hexyldithio)-1,2,4-thiadiazole, 3,5-bis(n-octyldithio)-1,2,4-thiadiazole, 3,5-bis(n-nonyldithio)-1,2,4-thiadiazole, 3,5-bis(1,1,3,3-tetramethylbutyldithio)-1,2,4-thiadiazole, 4,5-bis(n-hexyldithio)-1,2,3-thiadiazole, 4,5-bis(n-octyldithio)-1,2,3-thiadiazole, 4,5-bis(n-nonyldithio)-1,2,3-thiadiazole, 4,5-bis(1,1,3,3-tetramethylbutyldithio)-1,2,3-thiadiazole, and mixtures thereof, and the like.

**[0176]** Examples of alkylthiocarbamoyl compounds include compounds represented by the following formula (23).



**[01777]** Preferred specific examples of such alkylthiocarbamoyl compounds include bis(dimethyl thiocarbamoyl)monosulfide, bis(dibutyl thiocarbamoyl)monosulfide, bis(dimethyl thiocarbamoyl)disulfide, bis(dibutyl thiocarbamoyl)disulfide, bis(diamyl thiocarbamoyl)disulfide, bis(dioctyl thiocarbamoyl)disulfide, and mixtures thereof, and the like.

**[0178]** Examples of alkylcarbamate compounds include compounds represented by the following formula (24).



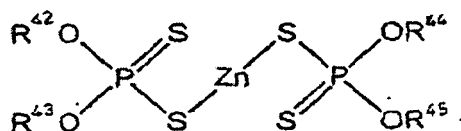
wherein R<sup>37</sup> to R<sup>40</sup> may be identical with or different from each other and are each independently an alkyl group having 1 to 20 carbon atoms, and R<sup>41</sup> is an alkyl group having 1 to 10 carbon atoms.

[0179] Preferred specific examples of such alkylcarbamate compounds include methylene bis(dibutyl dithiocarbamate), methylene bis[di(2-ethylhexyl)dithiocarbamate] and the like.

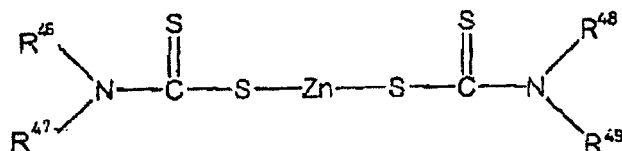
[0180] Further, examples of thioterpene compounds include reactants of phosphorus pentasulfide and pinene, and examples of dialkyl thiodipropionate compounds include dilauryl thiodipropionate, distearyl thiodipropionate, and mixtures thereof, and the like.

[0181] Sulfurized mineral oil means what dissolved a simple sulfur in mineral oil. Here, mineral oil used for the sulfurized mineral oil concerning the invention is not particularly limited, but specifically include paraffinic, naphthenic or the like mineral oil obtained by subjecting a lubricating oil distillate, which was obtained by the normal pressure distillation or the reduced pressure distillation of a crude oil, to suitably combined refining treatment such as solvent deasphalting, solvent extraction, hydrogenolysis, solvent dewaxing, contact dewaxing, hydrotreating, sulfuric acid washing, white clay treatment and the like. Moreover, as a simple sulfur, any form of clusters, powders, melt liquid and the like may be used, but if a simple sulfur of powder or melt liquid form is used dissolution to a base oil can be effectively carried out, thus preferred. Further, since a melt liquid simple sulfur is mixed with the same liquid, it has an advantage that the dissolution operation can be carried out in very short time. However, the mixture must be handled in the temperature higher than or equal to the melting point of the simple sulfur, thus special equipments such as a heating equipment or the like are necessary, and since handling is done in high temperature atmosphere, danger is accompanied, and the like, thereby the handling is not necessarily easy. On the contrary, the powdered simple sulfur is easily handled due to the cheap price, and since the time needed for the dissolution is sufficiently short, thus particularly preferred. In addition, the content of sulfur in sulfurized mineral oil concerning the invention is not particularly limited, but it is preferably 0.05 to 1.0% by weight, and more preferably 0.1 to 0.5% by weight based on the total amount of sulfurized mineral oil.

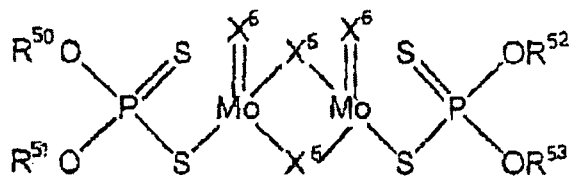
[0182] Zinc dithiophosphate compounds, zinc dithiocarbamate compounds, molybdenum dithiophosphate compounds and molybdenum dithiocarbamate compounds are referred to as the compounds represented by the following formulas (25) to (28), respectively.



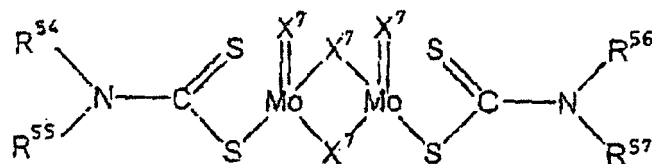
(25)



(26)



(27)



(28)

in the formulas (25) to (28),  $R^{42}$ ,  $R^{43}$ ,  $R^{44}$ ,  $R^{45}$ ,  $R^{46}$ ,  $R^{47}$ ,  $R^{48}$ ,  $R^{49}$ ,  $R^{50}$ ,  $R^{51}$ ,  $R^{52}$ ,  $R^{53}$ ,  $R^{54}$ ,  $R^{55}$ ,  $R^{56}$  and  $R^{57}$  may be identical with or different from each other and are each independently a hydrocarbon group having 1 or more carbon atoms, and  $X^6$  and  $X^7$  are each independently an oxygen atom or a sulfur atom.

**[0183]** Here, specific examples of the hydrocarbon group represented by  $R^{42}$ ,  $R^{43}$ ,  $R^{44}$ ,  $R^{45}$ ,  $R^{46}$ ,  $R^{47}$ ,  $R^{48}$ ,  $R^{49}$ ,  $R^{50}$ ,  $R^{51}$ ,  $R^{52}$ ,  $R^{53}$ ,  $R^{54}$ ,  $R^{55}$ ,  $R^{56}$  and  $R^{57}$  include alkyl groups such as a methyl group, an ethyl group, a propyl group (including all branched isomers), a butyl group (including all branched isomers), a pentyl group (including all branched isomers), a hexyl group (including all branched isomers), a heptyl group (including all branched isomers), an octyl group (including all branched isomers), a nonyl group (including all branched isomers), a decyl group (including all branched isomers), an undecyl group (including all branched isomers), a dodecyl group (including all branched isomers), a tridecyl group (including all branched isomers), a tetradecyl group (including all branched isomers), a pentadecyl group (including all branched isomers), a hexadecyl group (including all branched isomers), a heptadecyl group (including all branched isomers), an octadecyl group (including all branched isomers), a nonadecyl group (including all branched isomers), an icosyl group (including all branched isomers), a hencicosyl group (including all branched isomers), a docosyl group (including all branched isomers), a tricosyl group (including all branched isomers) and a tetracosyl group (including all branched isomers); cycloalkyl groups such as a cyclopentyl group, a cyclohexyl group and a cycloheptyl group; alkylcycloalkyl groups such as a methylcyclopentyl group (including all substituted isomers), an ethylcyclopentyl group (including all substituted isomers), a dimethylcyclopentyl group (including all substituted isomers), a propylcyclopentyl group (including all branched isomers and substituted isomers), a methylethylcyclopentyl group (including all substituted isomers), a trimethylcyclopentyl group (including all substituted isomers), a butylcyclopentyl group (including all branched isomers and substituted isomers), a methylpropylcyclopentyl group (including all branched isomers and substituted isomers), a diethylcyclopentyl group (including all substituted isomers), a dimethylethylcyclopentyl group (including all substituted isomers), a methylcyclohexyl group (including all substituted isomers), an ethylcyclohexyl group (including all substituted isomers), a dimethylcyclohexyl group (including all substituted isomers), a propylcyclohexyl group (including all branched isomers and substituted isomers), a methylethylcyclohexyl group (including all substituted isomers), a trimethylcyclohexyl group (including all substituted isomers), a butylcyclohexyl group (including all branched isomers and substituted isomers), a methylpropylcyclohexyl group (including all branched isomers and substituted isomers), a diethylcyclohexyl group (including all substituted isomers), a dimethylethylcyclohexyl group (including all substituted isomers), a methylcycloheptyl group (including all substituted isomers), an ethylcycloheptyl group (including all substituted isomers), a dimethylcycloheptyl group (including all substituted isomers), a propylcycloheptyl group (including all branched isomers and substituted isomers), a methylethylcycloheptyl group (including all substituted isomers), a trimethylcycloheptyl group (including all substituted isomers), a butylcycloheptyl group (including all branched isomers and substituted isomers), a methylpropylcycloheptyl group (including all branched isomers and substituted isomers), a diethylcycloheptyl group (including all substituted isomers) and a dimethylethylcycloheptyl group (including all substituted isomers); aryl groups such as a phenyl group and a naphthyl group; alkylaryl groups such as a tolyl group (including all substituted isomers), a xylyl group (including all substituted isomers), an ethylphenyl group (including all substituted isomers), a propylphenyl group (including all branched isomers and substituted isomers), a methylethylphenyl group (including all substituted isomers), a trimethylphenyl group (including all substituted isomers), a butylphenyl group (including all branched isomers and substituted isomers), a methylpropylphenyl group (including all branched isomers and substituted isomers), a diethylphenyl group (including all substituted isomers), a dimethylethylphenyl group (including all substituted isomers), a pentylphenyl group (including all branched isomers and substituted isomers), a hexylphenyl group (including all branched isomers and substituted isomers), a heptylphenyl group (including all branched isomers and substituted isomers), an octylphenyl group (including all branched isomers and substituted isomers), a nonylphenyl group (including all branched isomers and substituted isomers), a decylphenyl group (including all branched isomers and substituted isomers), an undecylphenyl group (including all branched isomers and substituted isomers), a dodecylphenyl group (including all branched isomers and substituted isomers), a tridecylphenyl group (including all branched isomers and substituted isomers), a tetradecylphenyl group (including all branched isomers and substituted isomers), a pentadecylphenyl group (including all branched isomers and substituted isomers), a hexadecylphenyl group (including all branched isomers and substituted isomers), a heptadecylphenyl group (including all branched isomers and substituted isomers), an octadecylphenyl group (including all branched isomers and substituted isomers), a nonadecylphenyl group (including all branched isomers and substituted isomers), an icosylphenyl group (including all branched isomers and substituted isomers), a hencicosylphenyl group (including all branched isomers and substituted isomers), a docosylphenyl group (including all branched isomers and substituted isomers), and a tetracosylphenyl group (including all branched isomers and substituted isomers).

isomers) and an octadecylphenyl group (including all branched isomers and substituted isomers); arylalkyl groups such as a benzyl group, a phenethyl group, a phenylpropyl group (including all branched isomers) and phenylbutyl group (including all branched isomers); and the like.

**[0184]** The content of a sulfur-based extreme pressure agent as the component (E) in the lubricative composition of the invention is arbitrary, but in the point of improving the extreme pressure property of the obtained lubricative composition, the minimum value of the content is preferably 0.01% by weight or higher, more preferably 0.05% by weight or higher, and even more preferably 0.1 % by weight or higher based on the total amount of the composition. Further, the maximum value of the content of the sulfur-based extreme pressure agent, in the point that an effect to the extent that meet with the added amount is not acquired even if more than such the amount is combined, is preferably 10% by weight or lower, more preferably 5% by weight or lower, even more preferably 3% by weight or lower, and most preferably 1% by weight or lower based on the total amount of the composition.

Component (F): Epoxy compound

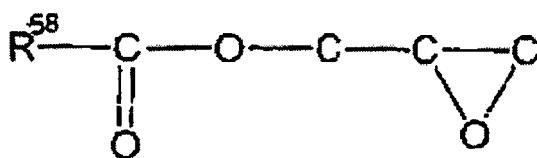
**[0185]** The component (F) in the lubricative composition of the invention is an arbitrary component in the point of the sludge resistance, but combining an epoxy compound is possible. Examples of epoxy compounds include the following compounds:

- (1) phenylglycidyl ether-type epoxy compounds,
- (2) alkylglycidyl ether-type epoxy compounds,
- (3) glycidyl ester-type epoxy compounds,
- (4) aryloxirane compounds,
- (5) alkyloxirane compounds,
- (6) alicyclic epoxy compounds,
- (7) epoxidized fatty acid monoester, and
- (8) epoxidized plant oil.

**[0186]** Specific examples of (1) phenylglycidyl ether-type epoxy compounds include phenylglycidyl ether or alkylphenylglycidyl ether. Here, as alkylphenylglycidyl ether, the one having 1 to 3 alkyl groups having 1 to 13 carbon atoms can be mentioned, and among these, the one having one alkyl group having 4 to 10 carbon atoms is preferred. Examples thereof include n-butylphenylglycidyl ether, i-butylphenylglycidyl ether, sec-butylphenylglycidyl ether, tert-butylphenylglycidyl ether, pentylphenylglycidyl ether, hexylphenylglycidyl ether, heptylphenylglycidyl ether, octylphenylglycidyl ether, nonylphenylglycidyl ether, decylphenylglycidyl ether and the like.

**[0187]** Specific examples of (2) alkylglycidyl ether-type epoxy compounds include decylglycidyl ether, undecylglycidyl ether, dodecyl glycidyl ether, tridecylglycidyl ether, tetradecylglycidyl ether, 2-ethylhexylglycidyl ether, neopentylglycol diglycidyl ether, trimethylolpropane triglycidyl ether, pentaerythritol tetraglycidyl ether, 1,6-hexanediol diglycidyl ether, sorbitol polyglycidyl ether, polyalkyleneglycol monoglycidyl ether, polyalkyleneglycol diglycidyl ether and the like.

**[0188]** Specific examples of (3) glycidyl ester-type epoxy compounds include the compounds represented by the following formula (29):



(29)

wherein R<sup>58</sup> is a hydrocarbon group having 1 to 18 carbon atoms.

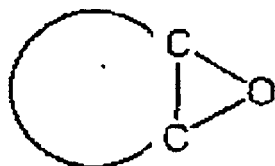
**[0189]** In the formula (29), R<sup>58</sup> is a hydrocarbon group having 1 to 18 carbon atoms, and examples of such the hydrocarbon group include an alkyl group having 1 to 18 carbon atoms, an alkenyl group having 2 to 18 carbon atoms, a cycloalkyl group having 5 to 7 carbon atoms, an alkylcycloalkyl group having 6 to 18 carbon atoms, an aryl group having 6 to 10 carbon atoms, an alkylaryl group having 7 to 18 carbon atoms, an arylalkyl group having 7 to 18 carbon atoms and the like. Among these, an alkyl group having 5 to 15 carbon atoms, an alkenyl group having 2 to 15 carbon atoms and an alkylphenyl group having a phenyl group and an alkyl group having 1 to 4 carbon atoms are preferred.

**[0190]** Preferred specific examples among glycidyl ester-type epoxy compounds include glycidyl-2,2-dimethyl octanoate, glycidyl benzoate, glycidyl-tert-butyl benzoate, glycidyl arylate, glycidyl methacrylate and the like.

[0191] Specific examples of (4) aryloxirane compounds include 1,2-epoxystyrene, alkyl-1,2-epoxystyrene and the like.

[0192] Specific examples of (5) alkyloxirane compounds include 1,2-epoxybutane, 1,2-epoxypentane, 1,2-epoxyhexane, 1,2-epoxyheptane, 1,2-epoxyoctane, 1,2-epoxynonane, 1,2-epoxydecane, 1,2-epoxyundecane, 1,2-epoxydodecane, 1,2-epoxytridecane, 1,2-epoxytetradecane, 1,2-epoxypentadecane, 1,2-epoxyhexadecane, 1,2-epoxyheptadecane, 1,1,2-epoxyoctadecane, 2-epoxynonadecane, 1,2-epoxyicosaen and the like.

[0193] Examples of (6) alicyclic epoxy compounds include the compound in which carbon atoms constituting the epoxy group directly constitutes the alicyclic ring, such as the compounds represented by the following formula (30):



(30)

[0194] Specific examples of alicyclic epoxy compounds include 1,2-epoxycyclohexane, 1,2-epoxycyclopentane, 3,4-epoxycyclohexylmethyl-3,4-epoxycyclohexane carboxylate, bis(3,4-epoxycyclohexylmethyl)adipate, exoxo-2,3-epoxynorbornene, bis(3,4-epoxy-6-methylcyclohexylmethyl)adipate, 2-(7-oxabicyclo[4.1.0]hepto-3-yl)-spiro(1,3-dioxane-5,3'-[7]oxabicyclo[4.1.0]heptane, 4-(1'-methylepoxyethyl)-1,2-epoxy-2-methylcyclohexane, 4-epoxyethyl-1,2-epoxycyclohexane and the like.

[0195] Specific examples of (7) epoxidized fatty acid monoester include esters of epoxidized fatty acid having 12 to 20 carbon atoms and alcohol having 1 to 8 carbon atoms, phenol or alkylphenol, and the like. Particularly, butyl epoxystearate, hexyl epoxystearate, benzyl epoxystearate, cyclohexyl epoxystearate, methoxyethyl epoxystearate, octyl epoxystearate, phenyl epoxystearate and butylphenyl ester epoxystearate are preferably used.

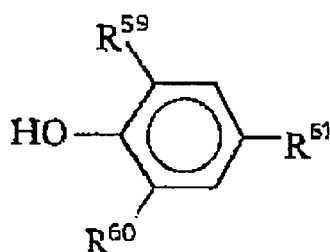
[0196] Specific examples of (8) epoxidized plant oil include an epoxy compound of plant oils such as soybean oil, linseed oil and cottonseed oil.

[0197] When combining these epoxy compounds as the component (F) to the lubricative composition of the invention, the combined amount is not particularly limited, but usually, the content of the epoxy compound combined therein is preferably 0.1 to 5.0% by weight, and more preferably 0.2 to 2.0% by weight based on the total amount of the lubricative composition (based on the total amount of base oil and all combined additives).

Component (G): Antioxidant

[0198] The component (G), in the point of oxidation stability in the lubricative composition of the invention, is an arbitrary component, but it may be (G-A) a phenol-based antioxidant, (G-B) an amine-based antioxidant or an antioxidant containing both of them.

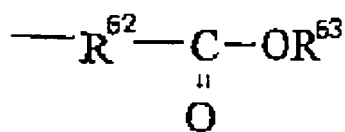
[0199] As (G-A) the phenol-based antioxidant, an arbitrary phenol-based compound used as an antioxidant of a lubricating oil can be used without any particular limitation, but for example, an alkylphenol compounds of one kind or two or more kinds selected from the compound represented by the following formula (31) or the formula (32) are preferably mentioned.



(31)

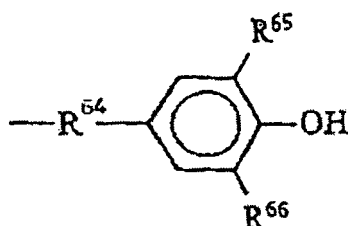
wherein  $R^{59}$  is an alkyl group having 1 to 4 carbon atoms,  $R^{60}$  is a hydrogen atom or an alkyl group having 1 to 4 carbon atoms, and  $R^{61}$  is a hydrogen atom or an alkyl group having 1 to 4 carbon atoms, a group represented by the following

formula (31-i) or a group represented by the following formula (31-ii).



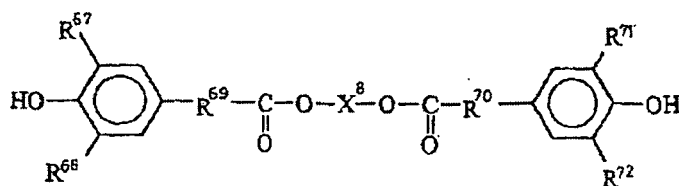
(31-i)

wherein R<sup>62</sup> is an alkylene group having 1 to 6 carbon atoms, and R<sup>63</sup> is an alkyl group or akenyl group having 1 to 24 carbon atoms.



(31-ii)

wherein R<sup>64</sup> is an alkylene group having 1 to 6 carbon atoms, R<sup>65</sup> is an alkyl group having 1 to 4 carbon atoms, and R<sup>66</sup> is a hydrogen atom or an alkyl group having 1 to 4 carbon atoms.



(32)

wherein R<sup>67</sup> and R<sup>71</sup> are each independently an alkyl group having 1 to 4 carbon atoms, R<sup>68</sup> and R<sup>72</sup> are each independently a hydrogen atom or an alkyl group having 1 to 4 carbon atoms, R<sup>69</sup> and R<sup>70</sup> are each independently an alkylene group having 1 to 6 carbon atoms, and X<sup>8</sup> is an alkylene group having 1 to 18 carbon atoms or a group represented by the following formula (32-i).



wherein R<sup>73</sup> and R<sup>74</sup> are each independently an alkylene group having 1 to 6 carbon atoms.

**[0200]** In the formula (31), specific examples of R<sup>59</sup> include a methyl group, an ethyl group, an n-propyl group, an isopropyl group, an n-butyl group, an isobutyl group, a sec-butyl group, a tert-butyl group and the like, but in the point of excelling in the oxidation stability, a tert-butyl group is preferred. Further, examples of R<sup>60</sup> include a hydrogen atom or an alkyl group having 1 to 4 carbon atoms as described above, but in the point of excelling in the oxidation stability, a methyl group or a tert-butyl group is preferred.

**[0201]** In the formula (31), when R<sup>61</sup> is an alkyl group having 1 to 4 carbon atoms, specific examples of R<sup>61</sup> include a methyl group, an ethyl group, an n-propyl group, an isopropyl group, an n-butyl group, an isobutyl group, a sec-butyl group, a tert-butyl group and the like, but in the point of excelling in the oxidation stability, a methyl group or an ethyl group is preferred.

**[0202]** Among the alkylphenol compound represented by the formula (31), particularly preferred example of the compound when R<sup>61</sup> is an alkyl group having 1 to 4 carbon atoms is 2,6-di-tert-butyl-p-cresol, 2,6-di-tert-butyl-4-ethylphenol, and mixtures thereof, and the like.

**[0203]** When R<sup>61</sup> in the formula (31) is a group represented by the formula (31-i), an alkylene group having 1 to 6

carbon atoms represented by  $R^{62}$  in the formula (31-i) may be straight-chained or branched, and specific examples thereof include a methylene group, a methylmethylene group, an ethylene group (a dimethylene group), an ethylmethylene group, a propylene group (a methylethylene group), a trimethylene group, a straight-chained or branched butylenes group, a straight-chained or branched pentylene group, a straight-chained or branched hexylene group and the like.

**[0204]** In the point that the compound represented by the formula (31) is prepared with less reaction steps,  $R^{62}$  is more preferably an alkylene group having 1 to 2 carbon atoms, and specific examples thereof include a methylene group, a methylmethylene group, an ethylene group (dimethylene group) and the like.

**[0205]** On the other hand, an alkyl group or alkenyl group having 1 to 24 carbon atoms represented by  $R^{63}$  in the formula (31-i) may be straight-chained or branched, and specific examples thereof include alkyl groups (wherein the alkyl group may be straight-chained or branched) such as a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, an undecyl group, a dodecyl group, a tridecyl group, a tetradecyl group, a pentadecyl group, a hexadecyl group, a heptadecyl group, an octadecyl group, a nonadecyl group, an icosyl group, a hencosyl group, a docosyl group, a tricosyl group and a tetracosyl group; an alkenyl groups (wherein the alkenyl group may be straight-chained or branched, and the position of the double bond is arbitrary) such as vinyl group, a propenyl group, an isopropenyl group, a butenyl group, a pentenyl group, a hexenyl group, a heptenyl group, an octenyl group, a nonenyl group, a decenyl group, an undecenyl group, a dodecenyl group, a tridecenyl group, a tetradecenyl group, a pentadecenyl group, a hexadecenyl group, a heptadecenyl group, an octadecenyl group, an octadecadienyl group, a nonadecenyl group, an icosenyl group, a henicosenyl group, a docosenyl group, a tricosenyl group and a tetracosenyl group; and the like.

**[0206]** As  $R^{63}$ , in the point of excelling in the solubility to a base oil, an alkyl group having 4 to 18 carbon atoms is preferred, and specific examples include alkyl groups (wherein the alkyl group may be straight-chained or branched) such as a butyl group, a pentyl group, a hexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, an undecyl group, a dodecyl group, a tridecyl group, a tetradecyl group, a pentadecyl group, a hexadecyl group, a heptadecyl group and an octadecyl group, a straight-chained or branched alkyl group having 6 to 12 carbon atoms is more preferred, and a branched alkyl group having 6 to 12 carbon atoms is particularly preferred.

**[0207]** Among phenol compounds represented by the formula (31), as the compound when  $R^{61}$  is a group represented by the formula (31-i), it is more preferable in that  $R^{62}$  in the formula (31-i) is an alkylene group having 1 to 2 carbon atoms and  $R^{63}$  is a straight-chained or branched alkyl group having 6 to 12 carbon atoms, and it is particularly preferable in that  $R^{62}$  in the formula (31-i) is an alkylene group having 1 to 2 carbon atoms and  $R^{63}$  is a branched alkyl group having 6 to 12 carbon atoms.

**[0208]** More specifically, more preferred examples include n-hexyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, isohexyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, n-heptyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, isoheptyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, n-octyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, isooctyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, 2-ethylhexyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, n-nonyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, isononyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, n-decyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, isodecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, n-undecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, isoundecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, n-dodecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, isododecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)acetate, n-hexyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, isohexyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, n-heptyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, isoheptyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, n-octyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, isooctyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, 2-ethylhexyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, n-nonyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, isononyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, n-decyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, isodecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, n-undecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, isoundecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, n-dodecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, isododecyl (3-methyl-5-tert-butyl-4-hydroxyphenyl)propionate, n-hexyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, isohexyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, n-heptyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, isoheptyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, n-octyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, isooctyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, 2-ethylhexyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, n-nonyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, isononyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, n-decyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, isodecyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, n-undecyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, isoundecyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, n-dodecyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, isododecyl (3,5-di-tert-butyl-4-hydroxyphenyl)acetate, n-hexyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, isohexyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, n-heptyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, isoheptyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, n-octyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, isooctyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, 2-ethylhexyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, n-nonyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, isononyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, n-decyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, isodecyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, n-undecyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate,



isoundecyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, n-dodecyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, isododecyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate, and mixtures thereof, and the like.

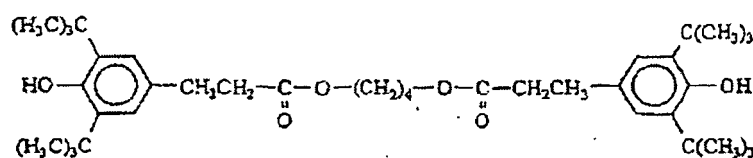
**[0209]** When  $R^{61}$  in the formula (31) is a group represented by the formula (31-ii),  $R^{64}$  in the formula (31-ii) is an alkylene group having 1 to 6 carbon atoms. Such the alkylene group may be straight-chained or branched, and specific examples include various alkylene groups as exemplified for  $R^{63}$  in the above. In the point that the compound of the formula (31) is prepared with less reaction steps and its raw material is easily obtained,  $R^{64}$  is more preferably an alkylene group having 1 to 3 carbon atoms, and specific examples include a methylene group, a methylenemethylene group, an ethylene group (a dimethylene group), an ethylmethylene group, a propylene group (a methylethylene group), a trimethylenemethylene group and the like. In addition, specific examples of  $R^{65}$  in the formula (31-ii) include a methyl group, an ethyl group, an n-propyl group, an isopropyl group, an n-butyl group, an isobutyl group, a sec-butyl group, a tert-butyl group and the like, but in the point of excelling in the oxidation stability, a tert-butyl group is preferred. Also, examples of  $R^{66}$  include a hydrogen atom or an alkyl group having 1 to 4 carbon atoms as described above, but in the point of excelling in the oxidation stability, a methyl group or a tert-butyl group is preferred.

**[0210]** Among alkylphenol compounds represented by the formula (31), as the compound when  $R^{61}$  is a group represented by the formula (31-ii), preferred specific examples include bis(3,5-di-tert-butyl-4-hydroxyphenyl)methane, 1,1-bis(3,5-di-tert-butyl-4-hydroxyphenyl)ethane, 1,2-bis(3,5-di-tert-butyl-4-hydroxyphenyl)ethane, 1,1-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane, 1,2-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane, 1,3-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane, 2,2-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane, and mixtures thereof, and the like.

**[0211]** Meanwhile, in the formula (32) above,  $R^{67}$  and  $R^{71}$  are each independently an alkyl group having 1 to 4 carbon atoms, and specifically they are a methyl group, an ethyl group, an n-propyl group, an isopropyl group, an n-butyl group, an isobutyl group, a sec-butyl group, a tert-butyl group and the like, but in the point of excelling in the oxidation stability, a tert-butyl group is preferred. Further, examples of  $R^{68}$  and  $R^{72}$  include, each independently, a hydrogen atom or an alkyl group having 1 to 4 carbon atoms as described above, but in the point of excelling in the oxidation stability, a methyl group or a tert-butyl group is preferred.

**[0212]** In the formula (32), an alkylene group having 1 to 6 carbon atoms representing  $R^{69}$  and  $R^{70}$  may be straight-chained or branched, and specific examples include, each independently, various alkylene groups described above for  $R^{62}$ . In the point that the compound represented by the formula (32) is prepared with less reaction steps and its raw material is easily obtained,  $R^{69}$  and  $R^{70}$  are each independently and more preferably, an alkylene group having 1 to 2 carbon atoms, and specific examples include a methylene group, a methylenemethylene group, an ethylene group (a dimethylene group) and the like.

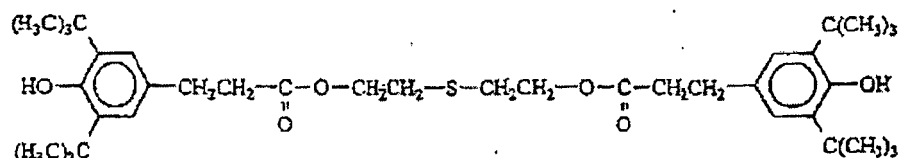
**[0213]** In the formula (32), specific examples of an alkylene group having 1 to 18 carbon atoms representing  $X^8$  include a methylene group, a methylenemethylene group, an ethylene group (a dimethylene group), an ethylmethylene group, a propylene group (a methylethylene group), a trimethylenemethylene group, a butylenemethylene group, a pentylene group, a hexylene group, a heptylene group, an octylene group, a nonylene group, a decylene group, an undecylene group, a dodecylene group, a tridecylene group, a tetradecylene group, a pentadecylene group, a hexadecylene group, a heptadecylene group, an octadecylene group and the like (wherein the alkylene group may be straight-chained or branched). However, in the point of easily obtaining the raw material or the like, an alkylene group having 1 to 6 carbon atoms, specifically for example, a methylene group, a methylenemethylene group, an ethylene group (a dimethylene group), an ethylmethylene group, a propylene group (a methylethylene group), a trimethylenemethylene group, a butylenemethylene group, a pentylene group, a hexylene group and the like (wherein the alkylene group may be straight-chained or branched), are more preferred; and a straight-chained alkylene group having 2 to 6 carbon atoms such as an ethylene group (a dimethylene group), a trimethylenemethylene group, a straight-chained butylene group (a tetramethylenemethylene group), a straight-chained pentylene group (a pentamethylenemethylene group), and a straight-chained hexylene group (a hexamethylenemethylene group), are particularly preferred. Among alkylphenol compounds represented by the formula (32), particularly preferred compound when  $X^8$  is an alkylene group having 1 to 18 carbon atoms, is the compound represented by the following formula (33).



(33)

**[0214]** Further, when  $X^8$  in the formula (32) is a group represented by the formula (32-i), an alkylene group having 1 to 6 carbon atoms represented by  $R^{73}$  and  $R^{74}$  in the formula (32-i) may be straight-chained or branched, and specific

examples include, each independently, various alkylene groups described above for R<sup>62</sup>. In the point that the raw material is easily obtained when preparing the compound of the formula (32), R<sup>73</sup> and R<sup>74</sup> are each independently and more preferably, an alkylene group having 1 to 3 carbon atoms, and specific examples include a methylene group, a methylmethylene group, an ethylene group (a dimethylene group), an ethylmethylene group, a propylene group (a methylethylene group), a trimethylene group and the like. Among alkylphenols represented by the formula (32), particularly preferred compound when X<sup>8</sup> is a group represented by the formula (32-i), is the compound represented by the following formula (34).



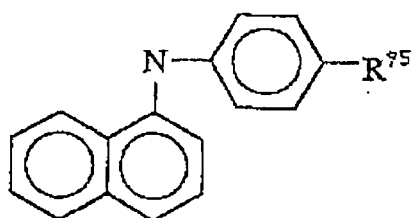
(34)

**[0215]** Moreover, although it is natural, as the component (G-A) as an arbitrary component of the invention, a compound selected from alkylphenol compounds represented by the formula (31) and the formula (32) may be used alone, or a mixture of two or more compounds selected from the alkylphenol compounds may be used in an arbitrary mixing ratio.

**[0216]** The maximum value of the component (G-A) in the lubricative composition of the invention is 3% by weight, preferably 2% by weight, and more preferably 1% by weight based on the total amount of the composition. When the content exceeds 3% by weight further improvement in the oxidation stability and sludge resisting effect that meet with the content cannot be seen, and the solubility to the base oil may be lowered, thus not preferred.

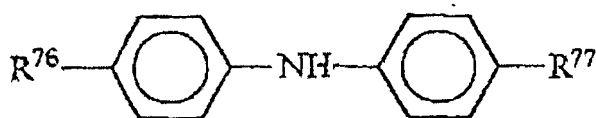
**[0217]** On the other hand, the minimum value of the content of the component (G-A) is 0.01 % by weight, preferably 0.1% by weight, and more preferably 0.2% by weight based on the total amount of the composition. When the content of the component (G-A) is less than 0.01 % by weight, its addition effect cannot be seen, and there may be a concern of worsening the oxidation stability of the gear oil composition or sludge resisting effect, thus not preferred.

**[0218]** As (G-B) an amine-based antioxidant which can be added as an arbitrary component in the component (G), an arbitrary amine-based compound used as an antioxidant of the lubricating oil can be used without any particular limitation, but for example, it is preferably one kind or two or more kinds of aromatic amine selected from (N-p-alkyl) phenyl- $\alpha$ -naphthyl amine represented by the following formula (35) or p,p'-dialkyldiphenyl amine represented by the formula (36).



(35)

wherein R<sup>75</sup> is a hydrogen atom or an alkyl group having 1 to 16 carbon atoms.



(36)

wherein R<sup>76</sup> and R<sup>77</sup> are each independently an alkyl group having 1 to 16 carbon atoms.

**[0219]** In the formula (35) representing (N-p-alkyl)phenyl- $\alpha$ -naphthyl amine, R<sup>75</sup> is a hydrogen atom or a straight-chained or branched alkyl group having 1 to 16 carbon atoms. When the number of carbon atoms in R<sup>75</sup> exceeds 16,

the proportion of the functional group occupied in a molecule becomes small, and there may be a concern of weakening the oxidation resisting ability. Specific examples of an alkyl group in  $R^{75}$  include a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, an undecyl group, a dodecyl group, a tridecyl group, a tetradecyl group, a pentadecyl group, a hexadecyl group and the like (wherein the alkyl group may be straight-chained or branched).

**[0220]** Among the compound represented by the formula (35), when  $R^{75}$  is an alkyl group, a branched alkyl group having 8 to 16 carbon atoms is preferred, and a branched alkyl group having 8 to 16 carbon atoms derived from an olefin oligomer having 3 or 4 carbon atoms is more preferred, in the point of excelling in solubility of its oxidized product to the base oil. Here, specific examples of olefin having 3 or 4 carbon atoms include propylene, 1-butene, 2-butene and isobutylene, but in the point of excelling in solubility of its oxidized product to the base oil, propylene or isobutylene is preferred.

**[0221]** When using (N-p-alkyl)phenyl- $\alpha$ -naphthyl amine represented by the formula (35) as the component (G-B) amine-based antioxidant,  $R^{75}$  is particularly preferably a hydrogen molecule or a branched octyl group derived from an isobutylene dimer, a branched nonyl group derived from a propylene trimer, a branched dodecyl group derived from an isobutylene trimer, a branched dodecyl group derived from a propylene tetramer or a branched pentadecyl group derived from a propylene pentamer, and particularly preferably a hydrogen molecule or a branched octyl group derived from an isobutylene dimer, a branched dodecyl group derived from an isobutylene trimer, a branched dodecyl group derived from a propylene tetramer.

**[0222]** As an aromatic amine represented by the formula (35), when using N-p-alkylphenyl- $\alpha$ -naphthyl amine whose  $R^{75}$  is an alkyl group, the commercially available N-p-alkylphenyl- $\alpha$ -naphthyl amine may be used. Further, it can be easily synthesized by reacting phenyl- $\alpha$ -naphthyl amine with a halogenated alkyl compound having 1 to 16 carbon atoms and an olefin having 2 to 16 carbon atoms, or an olefin oligomer having 2 to 16 carbon atoms with phenyl- $\alpha$ -naphthyl amine using a Fiedel-Craft catalyst. At this time, specific examples of the Fiedel-Craft catalyst including metal halides such as aluminum chloride, zinc chloride and iron chloride; acidic catalysts such as sulfuric acid, phosphoric acid, phosphorus pentoxide, boron fluoride, acid earth and activated earth; and the like can be used.

**[0223]** Meanwhile, in the formula (36) representing p,p'-dialkyldiphenyl amine,  $R^{76}$  and  $R^{77}$  are each independently an alkyl group having 1 to 16 carbon atoms. When either one or both of  $R^{76}$  and  $R^{77}$  are a hydrogen atom, there may be a concern of sedimenting as sludge by its oxidized products, on the contrary, when the number of carbon atoms exceeds 16, the proportion of the functional group occupied in a molecule becomes small, and there may be a concern of weakening the oxidation resisting ability.

**[0224]** Specific examples of  $R^{76}$  and  $R^{77}$  include a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, an undecyl group, a dodecyl group, a tridecyl group, a tetradecyl group, a pentadecyl group, a hexadecyl group and the like (wherein the alkyl group may be straight-chained or branched). Among these,  $R^{76}$  and  $R^{77}$  are preferably a branched alkyl group having 3 to 16 carbon atoms, and more preferably a branched alkyl group having 3 to 16 carbon atoms derived from olefin having 3 or 4 carbon atoms or its oligomer, in the point of excelling in solubility of its oxidized product to the base oil. Here, specific examples of olefin having 3 or 4 carbon atoms include propylene, 1-butene, 2-butene and isobutylene, but in the point of excelling in solubility of its oxidized product to the base oil of the lubricating oil, propylene or isobutylene is preferred.

**[0225]** When using p,p'-dialkyldiphenyl amine represented by the formula (36) as the component (G-B) amine-based antioxidant,  $R^{76}$  and  $R^{77}$  are particularly preferably an isopropyl group derived from propylene, a tert-butyl group derived from isobutylene, a branched hexyl group derived from a propylene dimer, a branched octyl group derived from an isobutylene dimer, a branched nonyl group derived from a propylene trimer, a branched dodecyl group derived from an isobutylene trimer, a branched dodecyl group derived from a propylene tetramer or a branched pentadecyl group derived from a propylene pentamer, and particularly preferably a tert-butyl group derived from isobutylene, a branched hexyl group derived from a propylene dimer, a branched octyl group derived from an isobutylene dimer, a branched nonyl group derived from a propylene trimer, a branched dodecyl group derived from an isobutylene trimer or a branched dodecyl group derived from a propylene tetramer.

**[0226]** As p,p'-dialkyldiphenyl amine represented by the formula (36), the commercially available one may be used. Further, in the same manner as for N-p-alkylphenyl- $\alpha$ -naphthyl amine represented by the formula (35), it can be easily synthesized by reacting diphenyl amine with a halogenated alkyl compound having 1 to 16 carbon atoms and an olefin having 2 to 16 carbon atoms, or olefin having 2 to 16 carbon atoms or its oligomer with diphenyl amine using a Fiedel-Craft catalyst. At this time, specific examples of the Fiedel-Craft catalyst including metal halides and acidic catalysts exemplified in the synthesis of N-p-alkylphenyl- $\alpha$ -naphthyl amine can be used. Moreover, although it is natural, as the component (G-B) as an arbitrary component of the invention, a compound selected from aromatic amines represented by the formula (35) and the formula (36) may be used alone, or a mixture of two or more compounds selected from the aromatic amines may be used in an arbitrary mixing ratio.

**[0227]** The maximum value of (G-B) amine-based antioxidant in the lubricative composition of the invention is 3% by weight, preferably 2% by weight, and more preferably 1% by weight based on the total amount of the composition. When

the content exceeds 3% by weight further improvement in the oxidation stability and sludge resisting effect that meet with the content cannot be seen, and the solubility to the base oil may be lowered, thus not preferred. On the other hand, the minimum value of the content of (G-B) amine-based antioxidant is 0.01 % by weight, preferably 0.1 % by weight, and more preferably 0.2% by weight based on the total amount of the composition. When the content of (G-B) amine-based antioxidant is less than 0.01% by weight, its addition effect cannot be seen, and there may be a concern of worsening the oxidation stability of the lubricative composition or sludge resisting effect, thus not preferred.

Component (H): Phosphorus-based compound

**[0228]** Although it is arbitrary, (H) a phosphorus-based compound as the component (H) may be contained for the improvement of the extreme pressure property of the lubricative composition of the invention.

**[0229]** Specific examples of the phosphorus-based compound contained herein include phosphoric acid esters such as phosphoric acid monoester, phosphoric acid diester and phosphoric acid triester; phosphorus acid esters such as phosphorus acid monoester, phosphorus acid diester and phosphorus acid triester; salts of these phosphoric acid esters or phosphorus acid esters; and mixtures thereof; and the like. The phosphoric acid esters and phosphorous acid esters described above are generally a compound containing a hydrocarbon having 2 to 30 carbon atoms, and preferably 3 to 20 carbon atoms.

**[0230]** Specific examples of the hydrocarbon atoms having 2 to 30 carbon atoms include alkyl groups (wherein the alkyl group may be straight-chained or branched) such as an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, a heptyl group, an octyl group, a nonyl group, a decyl group, an undecyl group, a dodecyl group, a tridecyl group, a tetradecyl group, a pentadecyl group, a hexadecyl group, a heptadecyl group and an octadecyl group; alkenyl groups (wherein the alkenyl group may be straight-chained or branched, and the position of the double bond is arbitrary) such as a butenyl group, a pentenyl group, a hexenyl group, a heptenyl group, an octenyl group, a nonenyl group, a decenyl group, an undecenyl group, a dodecenyl group, a tridecenyl group, a tetradecenyl group, a pentadecenyl group, a hexadecenyl group, a heptadecenyl group and an octadecenyl group; cycloalkyl groups having 5 to 7 carbon atoms such as a cyclopentyl group, a cyclohexyl group and a cycloheptyl group; alkylcycloalkyl groups having 6 to 11 carbon atoms (the substitution site of the alkyl group to the cycloalkyl group is arbitrary) such as a methylcyclopentyl group, a dimethylcyclopentyl group, a methylethylcyclopentyl group, a diethylcyclopentyl group, a methylcyclohexyl group, a dimethylcyclohexyl group, a methylethylcyclohexyl group, a diethylcyclohexyl group, a methylcycloheptyl group, a dimethylcycloheptyl group, a methylethylcycloheptyl group and a diethylcycloheptyl group; aryl groups such as a phenyl group and a naphthyl group; each alkylaryl group having 7 to 18 carbon atoms (wherein the alkyl group may be straight-chained or branched, and the substitution site to the aryl group is arbitrary) such as a tolyl group, a xylyl group, an ethylphenyl group, a propylphenyl group, a butylphenyl group, a pentylphenyl group, a hexylphenyl group, a heptylphenyl group, an octylphenyl group, a nonylphenyl group, a decylphenyl group, an undecylphenyl group and a dodecylphenyl group; each arylalkyl group having 7 to 12 carbon atoms (wherein the alkyl group may be straight-chained or branched) such as a benzyl group, a phenylethyl group, a phenylpropyl group, a phenylbutyl group, a phenylpentyl group and a phenylhexyl group; and the like.

**[0231]** Specific examples of preferred compounds as the component (H) phosphorus compound as an arbitrary component, which may be contained, include phosphoric acid monoalkyl esters (the alkyl group may be straight-chained or branched) such as monopropyl phosphate, monobutyl phosphate, monopentyl phosphate, monoheptyl phosphate, monooctyl phosphate; phosphoric acid mono(alkyl)aryl esters such as monophenyl phosphate and monocresyl phosphate; phosphoric acid dialkyl esters (the alkyl group may be straight-chained or branched) such as dipropyl phosphate, dibutyl phosphate, dipentylphosphate, dihexyl phosphate, diheptyl phosphate and dioctyl phosphate; phosphoric acid di(alkyl)aryl esters such as diphenyl phosphate and dicresyl phosphate; phosphoric acid trialkyl esters (the alkyl group may be straight-chained or branched) such as tripropyl phosphate, tributyl phosphate, tripentyl phosphate, trihexyl phosphate, triheptyl phosphate and trioctyl phosphate; phosphoric acid tri(alkyl)aryl esters such as triphenyl phosphate and tricresyl phosphate; phosphorus acid monoalkyl esters (the alkyl group may be straight-chained or branched) such as monopropyl phosphite, monobutyl phosphite, monopentyl phosphite, monoheptyl phosphate and monooctyl phosphite; phosphorus acid mono(alkyl)aryl esters such as monophenyl phosphate and monocresyl phosphite; phosphorus acid dialkyl esters (the alkyl group may be straight-chained or branched) such as dipropyl phosphite, dibutyl phosphate, dipentyl phosphite, dihexyl phosphate, diheptyl phosphate and dioctylphosphite; phosphorus acid di(alkyl)aryl esters such as diphenyl phosphate and dicresyl phosphite; phosphorus acid trialkyl esters (the alkyl group may be straight-chained or branched) such as tripropyl phosphite, tributyl phosphite, tripentyl phosphite, trihexyl phosphite, triheptyl phosphate and trioctyl phosphite; phosphorus acid tri(alkyl)aryl esters such as triphenyl phosphate and tricresyl phosphite; and mixtures thereof; and the like.

**[0232]** Specific examples of salts of phosphoric acid esters or phosphorus acid esters include salts obtained by reacting nitrogen-containing compound such as amine compounds having only ammonium, a hydrocarbon group having 1 to 8 carbon atoms or a hydrocarbon group having a hydroxyl group in the molecule to phosphoric acid monoester, phosphoric

acid diester, phosphorus acid monoester, phosphorus acid diester and the like, and then neutralizing a part or all the remaining acidic hydrogen.

[0233] Specific examples of such the nitrogen-containing compound include ammonia; alkyl amines (the alkyl group may be straight-chained or branched) such as monomethyl amine, monoethyl amine, monopropyl amine, monobutyl amine, monopentyl amine, monohexyl amine, monoheptyl amine, monooctyl amine, dimethyl amine, methylethyl amine, diethyl amine, methylpropyl amine, ethylpropyl amine, dipropyl amine, methylbutyl amine, ethylbutyl amine, propylbutyl amine, dibutyl amine, dipentyl amine, dihexyl amine, diheptyl amine and dioctyl amine; alkanol amines (the alkanol group may be straight-chained or branched) such as monomethanol amine, monoethanol amine, monopropanol amine, monobutanolamine, monopentanolamine, monohexanolamine, monoheptanolamine, monooctanolamine, monononanolamine, dimethanol amine, methanolethanol amine, diethanol amine, methanolpropanol amine, ethanolpropanol amine, dipropanol amine, methanolbutanolamine, ethanolbutanolamine, propanolbutanolamine, dibutanolamine, dipentanolamine, dihexanolamine, diheptanol amine and dioctanol amine; and mixtures thereof; and the like.

[0234] Moreover, although it is natural, the phosphorus-based compound which may be contained as the component (H) may be used a compound selected from the above-described phosphoric acid esters, phosphorus acid esters or salts thereof alone, or a mixture of two or more compounds selected therefrom may be used in an arbitrary mixing ratio.

[0235] The maximum value of the content of the phosphorus-based compound (the component (H)) in the lubricative composition of the invention is 10% by weight, preferably 5% by weight, and more preferably 3% by weight based on the total amount of the composition. When the content exceeds 5% by weight further improvement in the lubricity that meet with the content cannot be seen, and the oxidation stability may be lowered, thus not preferred. On the other hand, the minimum value of the content of the phosphorus-based compound is 0.01% by weight, preferably 0.05% by weight, and more preferably 0.1% by weight based on the total amount of the composition. When the content of the phosphorus-based compound is less than 0.01 % by weight, its addition effect cannot be seen.

Component (I): Oiliness improver

[0236] Although it is arbitrary, an oiliness improver as the component (I) can be added to the lubricative composition of the invention in the point of improving the frictional property.

[0237] Examples of the oiliness improver include an ester oiliness improver, an alcohol oiliness improver, a carboxylic acid oiliness improver, an ether oiliness improver, an amine oiliness improver, an amide oiliness improver and the like.

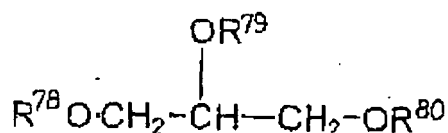
[0238] As an ester oiliness improver, the compounds given as the component (D) can be exemplified.

[0239] As an alcohol oiliness improver, alcohols exemplified in the description of ester oiliness improver can be mentioned. The number of carbon atoms in the alcohol oiliness improver is preferably 6 or higher, more preferably 8 or higher, and most preferably 10 or higher in the point of improving the frictional property. Moreover, in case of too many number of carbon atoms, there may be a concern of easy precipitation, thus the number of carbon atoms is preferably 24 or less, more preferably 20 or less, and most preferably 18 or less.

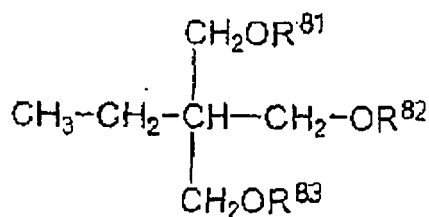
[0240] The carboxylic acid oiliness improver may be mono basic acid or polybasic acid. As such the carboxylic acid, for example, monobasic acids and polybasic acids exemplified in the description of ester oiliness improver can be mentioned. Among those, monobasic acids are preferred in the point of improving the frictional property. Further, the number of carbon atoms in the carboxylic acid oiliness improver is preferably 6 or higher, more preferably 8 or higher, and most preferably 10 or higher in the point of improving the frictional property. Moreover, in case of too many number of carbon atoms, there may be a concern of easy precipitation, thus the number of carbon atoms is preferably 24 or less, more preferably 20 or less, and most preferably 18 or less.

[0241] As an ether oiliness improver, an ether compound of an aliphatic polyhydric alcohol having 3 to 6 valences, an ether compound of bimolecular condensate or termolecular condensate of an aliphatic polyhydric alcohol having 3 to 6 valences and the like are mentioned.

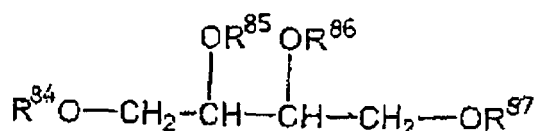
[0242] Examples of the ether compound of an aliphatic polyhydric alcohol having 3 to 6 valences are represented by the following formulas (37) to (42).



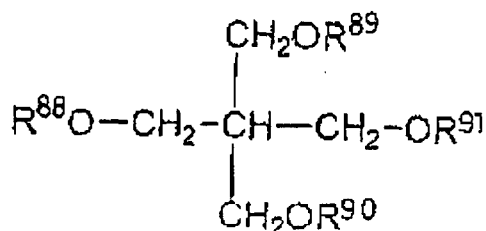
(37)



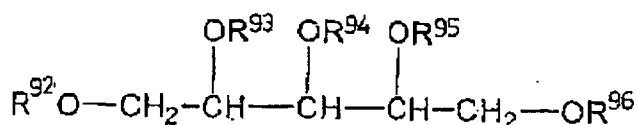
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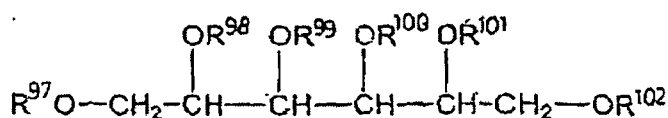
(39)



(40)



(41)



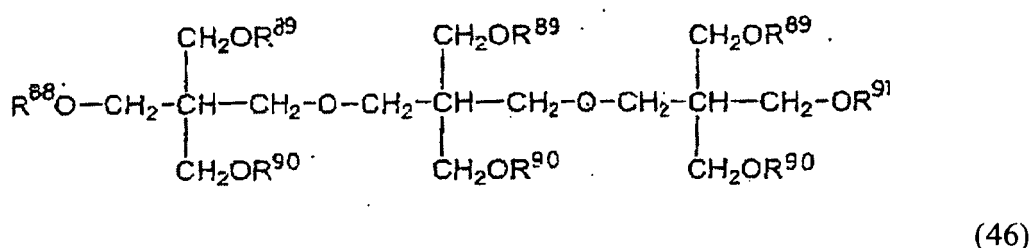
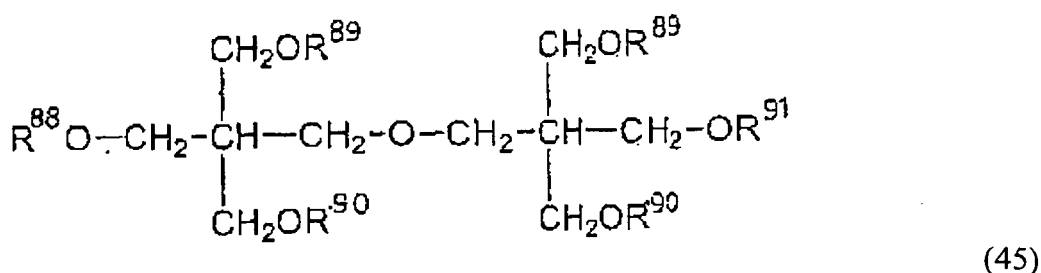
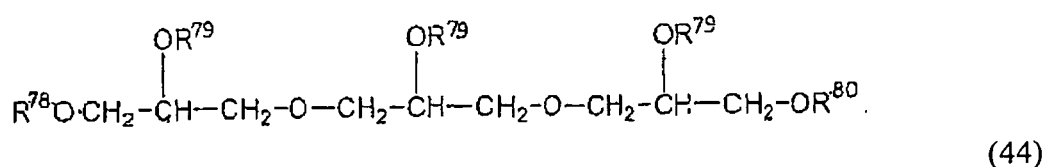
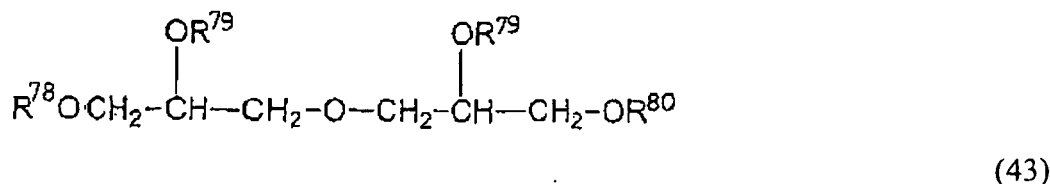
(42)

wherein  $\text{R}^{78}$  to  $\text{R}^{102}$  may be identical with or different from each other and are each independently a hydrogen atom, a straight-chained or branched alkyl group, aryl group or aralkyl group having 1 to 18 carbon atoms, or a glycol ether remaining group represented by  $-(\text{R}^a\text{O})_n-\text{R}^b$  (wherein  $\text{R}^a$  is an alkylene group having 2 to 6 carbon atoms,  $\text{R}^b$  is an alkyl group, aryl group or aralkyl group having 1 to 20 carbon atoms, and  $n$  is an integer of 1 to 10).

**[0243]** Specific examples of the ether compound of an aliphatic polyhydric alcohol having 3 to 6 valences include glycerin, trimethylol propane, erythritol, pentaerythritol, arabitol, sorbitol, mannitol and the like. Examples of  $\text{R}^{77}$  to  $\text{R}^{101}$  in the formulas (37) to (42) include a methyl group, an ethyl group, an n-propyl group, an isopropyl group, various butyl groups, various pentyl groups, various hexyl groups, various heptyl groups, various octyl groups, various nonyl groups, various decyl groups, various undecyl groups, various dodecyl groups, various tridecyl groups, various tetradecyl groups, various pentadecyl groups, various hexadecyl groups, various heptadecyl groups, various octadecyl groups, a phenyl group, a benzyl group and the like. The ether compound also include partial ether compound whose part of  $\text{R}^{77}$  to  $\text{R}^{101}$

is a hydrogen atom.

**[0244]** As the ether compound of bimolecular condensate or termolecular condensate of an aliphatic polyhydric alcohol having 3 to 6 valences, the same kind or different kind condensates from the compound represented by the formulas (37) to (42) are mentioned. For example, the ether compounds of bimolecular condensate or termolecular condensate of alcohol represented by the formula (37) are represented by the formulas (43) and (44), respectively. Further, the ether compounds of bimolecular condensate or termolecular condensate of alcohol represented by the formula (39) are represented by the formulas (45) and (46), respectively.



wherein  $\text{R}^{78}$  to  $\text{R}^{80}$  and  $\text{R}^{88}$  to  $\text{R}^{91}$  have the same definition as  $\text{R}^{77}$  to  $\text{R}^{79}$  of the formula (37) and  $\text{R}^{88}$  to  $\text{R}^{91}$  of the formula (40), respectively.

**[0245]** Specific examples of bimolecular condensate and termolecular condensate of an aliphatic polyhydric alcohol having 3 to 6 valences include diglycerin, ditrimethylolpropane, dipentaerythritol, disorbitol, triglycerin, tritrimethylolpropane, tripentaerythritol, trisorbitol and the like.

**[0246]** Specific examples of the ether oiliness improver represented by the formulas (37) to (46) include glycerin trihexyl ether, glycerin dimethyloctyl triether, glycerin di(methyloxyisopropylene)dodecyl triether, glycerin diphenyloctyl triether, glycerin di(phenyloxyisopropylene)dodecyl triether, trimethylolpropane trihexyl ether, trimethylolpropane dimethyloctyl triether, trimethylolpropane di(methyloxyisopropylene)dodecyl triether, pentaerythritol tetrahexyl ether, pentaerythritol trimethyloctyl tetraether, pentaerythritol tri(methyloxyisopropylene)dodecyl tetraether, sorbitol hexapropyl ether, sorbitol tetramethyloctyl pentaether, sorbitol hexa(methyloxyisopropylene) ether, diglycerin tetrabutyl ether, diglycerin dimethyldioctyl tetraether, diglycerin tri(methyloxyisopropylene)dodecyl tetraether, triglycerin pentaethyl ether, triglycerin trimethyldioctyl pentaether, triglycerin tetra(methyloxyisopropylene)decyl pentaether, ditrimethylolpropane tetrabutyl

ether, ditrimethylolpropane dimethyldioctyl tetraether, ditrimethylolpropane tri(methyloxyisopropylene)dodecyl tetraether, tritrimethylolpropane pentaethyl ether, tritrimethylolpropane trimethyldioctyl pentaether, tritrimethylolpropane tetra(methyloxyisopropylene)decyl pentaether, dipentaerythritol hexapropyl ether, dipentaerythritol pentamethyloctyl hexaether, dipentaerythritol hexa(methyloxyisopropylene)ether, tripentaerythritol octapropyl ether, tripentaerythritol pentamethyloctyl hexaether, tripentaerythritol hexa(methyloxyisopropylene) ether, disorbitol octamethyldioctyl decaether, disorbitol deca(methyloxyisopropylene) ether and the like.

**[0247]** Among these, glycerin diphenyloctyl triether, trimethylolpropane di(methyloxyisopropylene)dodecyl triether, pentaerythritol tetrahexyl ether, sorbitol hexapropyl ether, diglycerin dimethyldioctyl tetraether, triglycerin tetra(methyloxyisopropylene)decyl pentaether, dipentaerythritol hexapropyl ether, tripentaerythritol pentamethyloctyl hexaether are preferred.

**[0248]** For the oiliness improver as the component (I) which can be used in the lubricative composition of the invention, additionally, (I-1) an amine oiliness improver, (I-2) an amide oiliness improver and the like are mentioned.

**[0249]** Examples of (I-1) the amine oiliness improver include monoamine, polyamine, alkanol amine, but among these, monoamine is preferred in the point of improving the frictional property.

**[0250]** Specific examples of monoamine include alkyl amines such as monomethyl amine, dimethyl amine, trimethyl amine, monoethyl amine, diethyl amine, triethyl amine, monopropyl amine (including all isomers), dipropyl amine (including all isomers), tripropyl amine (including all isomers), monobutyl amine (including all isomers), dibutyl amine (including all isomers), tributyl amine (including all isomers), monopentyl amine (including all isomers), dipentyl amine (including all isomers), tripentyl amine (including all isomers), monohexyl amine (including all isomers), dihexyl amine (including all isomers), monoheptyl amine (including all isomers), diheptyl amine (including all isomers), monooctyl amine (including all isomers), dioctyl amine (including all isomers), monononyl amine (including all isomers), monodecyl amine (including all isomers), monoundecyl (including all isomers), monododecyl amine (including all isomers), monotridecyl amine (including all isomers), monotetradecyl amine (including all isomers), monopentadecyl amine (including all isomers), monohexadecyl amine (including all isomers), monoheptadecyl amine (including all isomers), monooctadecyl amine (including all isomers), monononadecyl amine (including all isomers), monoicosyl amine (including all isomers), monohenicosyl amine (including all isomers), monodocosyl amine (including all isomers), monotricosyl amine (including all isomers), dimethyl(ethyl) amine, dimethyl(propyl) amine (including all isomers), dimethyl(butyl) amine (including all isomers), dimethyl(pentyl) amine (including all isomers), dimethyl(hexyl) amine (including all isomers), dimethyl(heptyl) amine (including all isomers), dimethyl(octyl) amine (including all isomers), dimethyl(nonyl) amine (including all isomers), dimethyl(decyl) amine (including all isomers), dimethyl(undecyl) amine (including all isomers), dimethyl(dodecyl) amine (including all isomers), dimethyl(tridecyl) amine (including all isomers), dimethyl(tetradecyl) amine (including all isomers), dimethyl(pentadecyl) amine (including all isomers), dimethyl(hexadecyl) amine (including all isomers), dimethyl(heptadecyl) amine (including all isomers), dimethyl(octadecyl) amine (including all isomers), dimethyl(nonadecyl) amine (including all isomers), dimethyl(icosyl) amine (including all isomers), dimethyl(henicosyl) amine (including all isomers) and dimethyl(tricosyl) amine (including all isomers);

alkenyl amines such as monovinyl amine, divinyl amine, trivinyl amine, monopropenyl amine (including all isomers), dipropenyl amine (including all isomers), tripropenyl amine (including all isomers), monobutenyl amine (including all isomers), dibutenyl amine (including all isomers), tributenyl amine (including all isomers), monopentenyl amine (including all isomers), dipentenyl amine (including all isomers), tripentenyl amine (including all isomers), monohexenyl amine (including all isomers), dihexenyl amine (including all isomers), monoheptenyl amine (including all isomers), diheptenyl amine (including all isomers), monooctenyl amine (including all isomers), dioctenyl amine (including all isomers), monononenyl amine (including all isomers), monodecenyl amine (including all isomers), monoundecenyl (including all isomers), monododecenyl amine (including all isomers), monotridecenyl amine (including all isomers), monotetradecenyl amine (including all isomers), monopentadecenyl amine (including all isomers), monohexadecenyl amine (including all isomers), monoheptadecenyl amine (including all isomers), monooctadecenyl amine (including all isomers), monononadecenyl amine (including all isomers), monoicosenyl amine (including all isomers), monohenicosenyl amine (including all isomers), monodocosenyl amine (including all isomers) and monotricosenyl amine (including all isomers);

monoamines having an alkyl group and an alkenyl group such as dimethyl(vinyl) amine, dimethyl(propenyl) amine (including all isomers), dimethyl(butenyl) amine (including all isomers), dimethyl(pentenyl) amine (including all isomers), dimethyl(hexenyl) amine (including all isomers), dimethyl(heptenyl) amine (including all isomers), dimethyl(octenyl) amine (including all isomers), dimethyl(nonenyl) amine (including all isomers), dimethyl(decenyl) amine (including all isomers), dimethyl(undecenyl) amine (including all isomers), dimethyl(dodecenyl) amine (including all isomers), dimethyl(tridecenyl) amine (including all isomers), dimethyl(tetradecenyl) amine (including all isomers), dimethyl(pentadecenyl) amine (including all isomers), dimethyl(hexadecenyl) amine (including all isomers), dimethyl(heptadecenyl) amine (including all isomers), dimethyl(octadecenyl) amine (including all isomers), dimethyl(nonadecenyl) amine (including all isomers), dimethyl(icosenyl) amine (including all isomers), dimethyl(henicosenyl) amine (including all isomers) and dimethyl(tricosenyl) amine (including all isomers);

aromatic substituted alkyl amines such as monobenzyl amine, (1-phenylethyl) amine, (2-phenylethyl) amine (also known



as monophenethyl amine), dibenzyl amine, bis(1-phenylethyl) amine and bis(2-phenylethylene) amine (also known as diphenethyl amine);

cycloalkyl amines having 5 to 16 carbon atoms such as monocyclopentyl amine, dicyclopentyl amine, tricyclopentyl amine, monocyclohexyl amine, dicyclohexyl amine, monocycloheptyl amine and dicycloheptyl amine;

monoamines having an alkyl group and a cycloalkyl group such as dimethyl(cyclopentyl) amine, dimethyl(cyclohexyl) amine and dimethyl(cycloheptyl) amine; and

alkylcycloalkyl amines such as (methylcyclopentyl) amine (including all substituted isomers), bis(methylcyclopentyl) amine (including all substituted isomers), (dimethylcyclopentyl) amine (including all substituted isomers), bis(dimethylcyclopentyl) amine (including all substituted isomers), (ethylcyclopentyl) amine (including all substituted isomers), bis(ethylcyclopentyl) amine (including all substituted isomers), (methylethylcyclopentyl) amine (including all substituted isomers), bis(methylethylcyclopentyl) amine (including all substituted isomers), (diethylcyclopentyl) amine (including all substituted isomers), (methylcyclohexyl) amine (including all substituted isomers), bis(methylcyclohexyl) amine (including all substituted isomers), (dimethylcyclohexyl) amine (including all substituted isomers), bis(dimethylcyclohexyl) amine (including all substituted isomers), (ethylcyclohexyl) amine (including all substituted isomers), bis(ethylcyclohexyl) amine (including all substituted isomers), (methylethylcyclohexyl) amine (including all substituted isomers), (diethylcyclohexyl) amine (including all substituted isomers), (methylcycloheptyl) amine (including all substituted isomers), bis(methylcycloheptyl) amine (including all substituted isomers), (dimethylcycloheptyl) amine (including all substituted isomers), (ethylcycloheptyl) amine (including all substituted isomers), (methylethylcycloheptyl) amine (including all substituted isomers) and (diethylcycloheptyl) amine (including all substituted isomers); and the like. Further, monoamines derived from fats and oils such as tallow amines are also included in the above-mentioned monoamines.

**[0251]** Among the above-mentioned monoamines, in the point of improving the frictional property, alkyl amines, monoamines having an alkyl group and an alkenyl group, monoamines having an alkyl group and a cycloalkyl group, cycloalkyl amines and alkylcycloalkyl amines are particularly preferred, and alkyl amines and monoamines having an alkyl group and an alkenyl group are more preferred.

**[0252]** The number of carbon atoms in monoamine is not particularly limited, but it is preferably 8 or higher, and more preferably 12 or higher in the point of the rust resistance. Moreover, in the point of improving the frictional property, it is preferably 24 or lower, and more preferably 18 or lower.

**[0253]** In addition, the number of hydrocarbon groups bonded to a nitrogen atom in monoamine is not particularly limited, but it is preferably 1 to 2 hydrocarbon groups, and more preferably 1 hydrocarbon group in the point of improving the frictional property.

**[0254]** As (I-2) an amide oiliness improver, amides obtained by reacting a fatty acid having 6 to 30 carbon atoms or its acidic salt with ammonia, or a nitrogen-containing compound such as an amine compound containing only a hydrocarbon group having 1 to 8 carbon atoms or a hydrocarbon group having a hydroxyl group in the molecule are mentioned.

**[0255]** Here, the fatty acid may be straight-chained or branched, and may be saturated fatty acid or unsaturated fatty acid. Further, the number of carbon atoms is 6 to 30, and preferably 9 to 24.

**[0256]** Specific examples of such the fatty acid include saturated fatty acids (wherein the saturated fatty acid may be straight-chained or branched) such as heptanoic acid, octanoic acid, nonanoic acid, decanoic acid, undecanoic acid, dodecanoic acid, tridecanoic acid, tetradecanoic acid, pentadecanoic acid, hexadecanoic acid, heptadecanoic acid, octadecanoic acid, nonadecanoic acid, icosanoic acid, heneicosanoic acid, docosanoic acid, tricosanoic acid, tetracosanoic acid, pentacosanoic acid, hexacosanoic acid, heptacosanoic acid, octacosanoic acid, nonacosanoic acid and triacontanoic group; unsaturated fatty acids (wherein the unsaturated fatty acid may be straight-chained or branched, and the position of the double bond is arbitrary) such as heptenoic acid, octenoic acid, nonenoic acid, decenoic acid, undecenoic acid, dodecenoic acid, tridecenoic acid, tetradecenoic acid, pentadecenoic acid, hexadecenoic acid, heptadecenoic acid, octadecenoic acid (including oleic acid), nonadecenoic acid, icosenoic acid, heneicosenoic acid, docosenoic acid, tricosenoic acid, tetracosenoic acid, pentacosenoic acid, hexacosenoic acid, heptacosenoic acid, octacosenoic acid, nonacosenoic acid and triacontenoic acid; and the like. However, straight-chained fatty acids of straight-chained fatty acids derived from lauric acid, myristic acid, palmitic acid, stearic acid, oleic acid and various fats and oils (such as coconut oil fatty acid, etc.) or a mixture of straight-chained fatty acids and brached fatty acids synthesized by an oxo method or the like are preferably used.

**[0257]** Specific examples of the nitrogen-containing compound reacted with the fatty acids include ammonia; alkyl amines (the alkyl group may be straight-chained or branched) such as monomethyl amine, monoethyl amine, monopropyl amine, monobutyl amine, monopentyl amine, monohexyl amine, monoheptyl amine, monooctyl amine, dimethyl amine, methylethyl amine, diethyl amine, methylpropyl amine, ethylpropyl amine, dipropyl amine, methylbutyl amine, ethylbutyl amine, propylbutyl amine, dibutyl amine, dipentyl amine, dihexyl amine, diheptyl amine and dioctyl amine; alkanol amines (the alkanol group may be straight-chained or branched) such as monomethanol amine, monoethanol amine, monopropanol amine, monobutanol amine, monopentanol amine, monohexanol amine, monoheptanol amine, monooctanol amine, monononanol amine, dimethanol amine, methanolethanol amine, diethanol amine, methanolpropanol amine, ethanolpropanol amine, dipropanol amine, methanolbutanol amine, ethanolbutanol amine, propanolbutanol amine, dib-

utanol amine, dipentanol amine, dihexanol amine, diheptanol amine and dioctanol amine; and mixtures thereof; and the like.

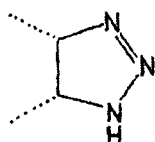
[0258] Specific examples of the fatty acid amide including lauric acid amide, lauric acid diethanol amide, lauric acid monopropanol amide, myristic acid amide, myristic acid diethanol amide, myristic acid monopropanol amide, palmitic acid amide, palmitic acid diethanol amide, palmitic acid monopropanol amide, stearic acid amide, stearic acid diethanol amide, stearic acid monopropanol amide, oleic acid amide, oleic acid diethanol amide, oleic acid monopropanol amide, coconut oil fatty acid amide, coconut oil fatty acid diethanol amide, coconut oil fatty acid monopropanol amide, synthetic mixture fatty acid amide having 12 to 13 carbon atoms, synthetic mixture fatty acid diethanol amide having 12 to 13 carbon atoms, synthetic mixture fatty acid monopropanol amide having 12 to 13 carbon atoms, and mixtures thereof and the like, are preferably used.

[0259] Among the oiliness improvers, polyhydric alcohol partial esters and aliphatic amides are preferred in the point of improving effect on the frictional property.

[0260] The content of the oiliness improver as the component (I) in the lubricative composition is arbitrary, but in the point of excelling in the improving effect of the frictional property, it is preferably 0.01 % by weight or higher, more preferably 0.05% by weight or higher, and even more preferably 0.1% by weight or higher based on the total amount of the lubricative composition. Further, the content, in the point of the precipitation resistance, is preferably 10% by weight or lower, more preferably 7.5% by weight or lower, and even more preferably 5% by weight or lower based on the total amount of the composition.

Component(J): Triazole and/or its derivatives

[0261] Although it is arbitrary, triazole and/or its derivatives represented by the formula (47) as the composition (J) can be added to the lubricative composition of the invention in the point of improving thermal and oxidation stability.

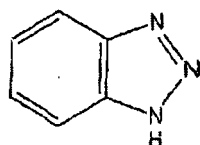


(47)

wherein the two dotted lines are the same or different substituents substituted to the triazole ring and are preferably each independently a hydrocarbon group, and they can be bonded to each other to form a ring group, for example, to form a condensed benzene ring.

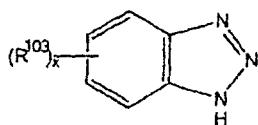
[0262] Preferred compound as triazole and/or its derivatives is benzotriazole and/or its derivatives.

[0263] Examples of the benzotriazole include a compound represented by the following formula (48).

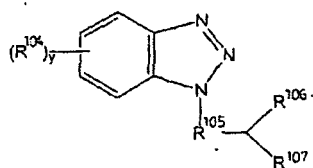


(48)

[0264] Further, examples of the benzotriazole derivatives include alkylbenzotriazole represented by the following formula (49) and (alkyl)aminoalkylbenzotriazole represented by the formula (50).



(49)



(50)

**[0265]** In the formula (49),  $R^{103}$  is a straight-chained or branched alkyl group having 1 to 4 carbon atoms and preferably a methyl group or an ethyl group, and  $x$  is a number of 1 to 3, and preferably 1 or 2. Examples of  $R^{103}$  include a methyl group, an ethyl group, an *n*-propyl group, an isopropyl group, an *n*-butyl group, an isobutyl group, a *sec*-butyl group, a *tert*-butyl group and the like. As the alkylbenzotriazole represented by the formula (49), a compound, wherein  $R^{103}$  is a methyl group or an ethyl group, and  $x$  is 1 or 2, is preferred, and examples include methylbenzotriazole (tolyltriazole), dimethylbenzotriazole, ethylbenzotriazole, ethylmethylbenzotriazole, diethylbenzotriazole or mixtures thereof, and the like.

**[0266]** In the formula (50),  $R^{104}$  is a straight-chained or branched alkyl group having 1 to 4 carbon atoms and preferably a methyl group or an ethyl group;  $R^{105}$  is a methylene group or an ethylene group;  $R^{106}$  and  $R^{107}$  may be identical with or different from each other and are each independently a hydrogen atom or a straight-chained or branched alkyl group having 1 to 18 carbon atoms and preferably a straight-chained or branched alkyl group having 1 to 12 carbon atoms; and  $y$  is a number of 0 to 3 and preferably 0 or 1. Examples of  $R^{103}$  include a methyl group, an ethyl group, an *n*-propyl group, an isopropyl group, an *n*-butyl group, an isobutyl group, a *sec*-butyl group, a *tert*-butyl group and the like. Examples of  $R^{106}$  and  $R^{107}$  include, each independently, a hydrogen atom and alkyl groups such as a methyl group, an ethyl group, a propyl group, an isopropyl group, an *n*-butyl group, an isobutyl group, a *sec*-butyl group, a *tert*-butyl group, a straight-chained or branched pentyl group, a straight-chained or branched hexyl group, a straight-chained or branched heptyl group, a straight-chained or branched octyl group, a straight-chained or branched nonyl group, a straight-chained or branched decyl group, a straight-chained or branched undecyl group, a straight-chained or branched dodecyl group, a straight-chained or branched tridecyl group, a straight-chained or branched tetradecyl group, a straight-chained or branched pentadecyl group, a straight-chained or branched hexadecyl group, a straight-chained or branched heptadecyl group and a straight-chained or branched octadecyl group.

**[0267]** As (alkyl)aminobenzotriazole represented by the formula (50), dialkylaminoalkylbenzotriazole, dialkylaminoalkyltriazole or mixtures thereof are preferably used in the point of excelling particularly the oxidation resistance, wherein  $R^{104}$  is a methyl group,  $y$  is 0 or 1,  $R^{105}$  is a methylene group or an ethylene group, and  $R^{106}$  and  $R^{107}$  are a straight-chained or branched alkyl group having 1 to 12 carbon atoms. Examples of such dialkylaminoalkylbenzotriazole include dimethylaminomethylbenzotriazole, diethylaminomethylbenzotriazole, di(straight-chained or branched)propylaminomethylbenzotriazole, di(straight-chained or branched)butylaminomethylbenzotriazole, di(straight-chained or branched)pentylaminomethylbenzotriazole, di(straight-chained or branched)hexylaminomethylbenzotriazole, di(straight-chained or branched)heptylaminomethylbenzotriazole, di(straight-chained or branched)octylaminomethylbenzotriazole, di(straight-chained or branched)nonylaminomethylbenzotriazole, di(straight-chained or branched)decylaminomethylbenzotriazole, di(straight-chained or branched)undecylaminomethylbenzotriazole, di(straight-chained or branched)dodecylaminomethylbenzotriazole; dimethylaminoethylbenzotriazole, diethylaminoethylbenzotriazole, di(straight-chained or branched)propylaminoethylbenzotriazole, di(straight-chained or branched)butylaminoethylbenzotriazole, di(straight-chained or branched)pentylaminoethylbenzotriazole, di(straight-chained or branched)hexylaminoethylbenzotriazole, di(straight-chained or branched)heptylaminoethylbenzotriazole, di(straight-chained or branched)octylaminoethylbenzotriazole, di(straight-chained or branched)nonylaminoethylbenzotriazole, di(straight-chained or branched)decylaminoethylbenzotriazole, di(straight-chained or branched)undecylaminoethylbenzotriazole, di(straight-chained or branched)dodecylaminoethylbenzotriazole; dimethylaminomethyltolyltriazole, diethylaminomethyltolyltriazole, di(straight-chained or branched)propylaminomethyltolyltriazole, di(straight-chained or branched)butylaminomethyltolyltriazole, di(straight-chained or branched)pentylaminomethyltolyltriazole, di(straight-chained or branched)hexylaminomethyltolyltriazole, di(straight-chained or branched)heptylaminomethyltolyltriazole, di(straight-chained or branched)octylaminomethyltolyltriazole, di(straight-chained or branched)nonylaminomethyltolyltriazole, di(straight-chained or branched)decylaminomethyltolyltriazole, di(straight-chained or branched)undecylaminomethyltolyltriazole, di(straight-chained or branched)dodecylaminomethyltolyltriazole; dimethylaminoethyltolyltriazole, diethylaminoethyltolyltriazole, di(straight-chained or branched)propylaminoethyltolyltriazole, di(straight-chained or branched)butylaminoethyltolyltriazole, di(straight-chained or branched)pentylaminoethyltolyltriazole, di(straight-chained or branched)hexylaminoethyltolyltriazole, di(straight-chained or branched)heptylaminoethyltolyltriazole, di(straight-chained or branched)octylaminoethyltolyltriazole, di(straight-chained or branched)nonylaminoethyltolyltriazole, di(straight-chained or branched)de-

cylaminoethyltolyltriazole, di(straight-chained or branched)undecylaminoethyltolyltriazole, di(straight-chained or branched)dodecylaminoethyltolyltriazole; or mixtures thereof; and the like.

**[0268]** The content of triazole and its derivatives as the composition (J), which is an arbitrary component in the lubricative composition, is arbitrary, but it is preferably 0.001% by weight or higher, and more preferably 0.005% by weight or higher based on the total amount of the composition. When the content is less than 0.001 % by weight, there may be a concern of becoming insufficient in the improving effect of the thermal and oxidation stability by containing triazole and/or its derivatives. Further, the content of triazole and/or its derivatives is preferably 1.0% by weight or lower, and more preferably 0.5% by weight or lower based on the total amount of the composition. When the content exceeds 1.0% by weight, the thermal and oxidation stability that meet with the content cannot be obtained, and there may be a concern of becoming disadvantageous economically.

**[0269]** Other arbitrary additive component (K):

**[0270]** In the purpose of further improving the performance, if necessary, various additives such as a rust inhibitor, a metal deactivator, a viscosity index improver or detergent additive in addition to the dispersant viscosity index improver of the component (B), a pour-point depressant and an defoaming agent can be contained alone or in combination of a plurality of kinds in the lubricative composition of the invention.

**[0271]** Specific examples of the rust inhibitor include metal soaps such as fatty acid metal salt, lanoline fatty acid metal salt and oxidized wax metal salt; polyhydric alcohol partial esters such as sorbitan fatty acid ester; esters such as lanoine fatty acid ester; sulfonates such as calcium sulfonate and barium sulfonate; oxidized wax; amines; phosphoric acids; phosphoric acid salts; and the like. In the invention, one kind or two or more kinds of compound arbitrarily selected from these rust inhibitors can be contained in an arbitrary amount, but in general, the content is preferably 0.01 to 1% by weight based on the total amount of the lubricative composition.

**[0272]** Specific examples of the metal deactivator include imidazole compounds other than the benzotriazole compound mentioned as the component (J). In the invention, one kind or two or more kinds of compound arbitrarily selected from these metal deactivators can be contained in an arbitrary amount, but in general, the content is preferably 0.001 to 1% by weight based on the total amount of the lubricative composition.

**[0273]** Specific examples of viscosity index improver in addition to the dispersant viscosity index improver of the component (B) include nondispersant viscosity index improvers such as a copolymer of one kind or two or more kinds of monomers selected from various methacrylic acid esters, or its hydrides, an ethylene- $\alpha$ -olefin copolymer (examples of  $\alpha$ -olefin include propylene, 1-butene, 1-pentene and the like) or its hydrides, polyisobutylene or its hydrides, a styrene-diene hydrogenated copolymer or polyalkylstyrene, and the like. Further, examples of the detergent additive in addition to the dispersant viscosity index improver of the component (B) include alkenyl succinic acid imide, sulfonate, salicylate, phenate and the like. One kind or two or more kinds of compound arbitrarily selected from these viscosity index improvers or detergent additives can be contained in an arbitrary amount, but in general, the content is preferably 0.01 to 10% by weight based on the total amount of the lubricative composition.

**[0274]** Specific examples of the pour-point depressant include a copolymer of one kind or two or more kinds of monomers selected from various acrylic acid esters or methacrylic acid esters or its hydrides. One kind or two or more kinds of compound arbitrarily selected from these pour-point depressants can be contained in an arbitrary amount, but in general, the content is preferably 0.01 to 5% by weight based on the total amount of the lubricative composition.

**[0275]** Specific examples of the defoaming agent include silicones such as dimehtylsilicone and fluorosilicone. In the invention, one kind or two or more kinds of compound arbitrarily selected from these defoaming agents can be contained in an arbitrary amount, but in general, the content is preferably 0.0001 to 0.05% by weight based on the total amount of the lubricative composition.

**[0276]** Use:

The lubricative composition of the invention is suitable in the gear oil composition, the lubricating oil composition for paper machines, the lubricating oil composition for slide guides (sliding surfaces) and the like, and used for various industrial machines, automobiles, rail vehicles and the like. In addition to these, the composition can be used as the hydraulic oil, the turbine oil, the compression machine oil, the bearing oil and the like, and particularly as the hydraulic oil.

The hydraulic oil exhibits excellent effect when used as the hydraulic oil for hydraulic equipments such as an injection machine, a machine tool, a construction machine, an iron manufacturing equipment, an industrial robot and hydraulic elevator.

#### Examples

**[0277]** Next, the present invention will be described in more detail by way of Examples. Further, the gear oil, the lubricating oil for paper machines, the lubricating oil for slide guides and the hydraulic oil which are specific embodiments of the lubricative composition of the invention will be described individually in their order by way of Examples, respectively.

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### [Gear oil composition - 1]

**[0278]** This Example is an embodiment related to the gear oil formed by combining the component (C-1), the component (C-2) or a mixture thereof into the base oil.

**[0279]** Tested base oils, the kind of additives added thereto and their amounts are listed below. Further, the test methods carried out on each combined lubricating oil composition is also listed below. The obtained test results are presented in Tables 1 to 3.

(IAE oil temperature)

**[0280]** Based on the IAE gear test specified in IP 166/77 (92), the oil temperature of the gear box was measured after 120 minutes by operating under the rotation number of 6000 rpm, the oil amount of 1250 ml and the load of 80 1b.

(Timken test)

**[0281]** Based on JIS K2519, OK load of each lubricating oil composition was measured.

(Base oil)

**[0282]**

Base oil 1: solvent refined paraffinic mineral oil (kinematic viscosity: 150 mm<sup>2</sup>/s (40°C), viscosity index: 95)

Base oil 2: 1-decene oligomer (kinematic viscosity: 150 mm<sup>2</sup>/s (40°C), viscosity index: 140)

Base oil 3: polyol ester obtained by using a mixture of pentaerythritol and dipentaerythritol and a straight-chained saturated fatty acid having 5 to 12 carbon atoms (kinematic viscosity: 150 mm<sup>2</sup>/s (40°C), viscosity index: 120)

(Additives)

**[0283]**

Component (C-1)

C-1-1: N-oleoyl sarcosine

Component (C-2)

C-2-1: nonyl phenoxyacetate

Component (E)

E1: 2,5-bis(n-nonyldithio)-1,3,4-thiadiazole

E2: Zinc dioctyldithiophosphate (ZnDTP)

Other additives

Amine: dioctyldiphenyl amine

DBPC: di-tert-butyl-p-cresol

TCP: tricresylphosphate

[Effect concerning the embodiment of gear oil composition - 1]

**[0284]** The gear oil composition of this embodiment is excellent in energy-saving. Therefore, the oil composition copes with the gear equipment used for various industrial machines with high efficiency and low cost and also keeps up with the tendency for high-speed and high-output as the gear oil, thus more energy-saving operation is possible.

Table 1

Example	1	2	3	4	5
Base oil 1	99.19	98.17	98.99	98.15	98.85
2	-	-	-	-	-
3	-	-	-	-	-
Additives					

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(continued)

Example	1	2	3	4	5
C-1 component	C-1-1 0.01	-	C-1-1 0.01	C-1-1 0.02	C-1-1 0.02
C-2 component	-	C-2-1 0.03	-	C-2-1 0.03	C-2-1 0.03
E component	-	-	E1 0.2	-	E1 0.3
DBPC	0.5	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3	0.3
Other additives					
IAE oil Temperature (°C)	130	134	123	127	120
Timken OK load (lb)	15	24	55	15	45

Table 2

Example	6	7
Base oil 1	-	-
2	98.85	-
3	-	98.85
Additives		
C-1 component	C-1-1 0.02	C-1-1 0.02
C-2 component	C-2-1 0.03	C-2-1 0.03
E component	E1 0.3	E1 0.3
DBPC	0.5	0.5
Amine	0.3	0.3
Other additives	-	-
IAE oil temperature (°C)	130	119
Timken OK load (lb)	35	50

Table 3

Test Example	1	2	3	4
Base oil 1	99.2	98.2	98.7	99.0
2	-	-	-	-
3	-	-	-	-
Additives				
C-1 component	-	-	-	-
C-2 component	-	-	-	-
E component		-	E2 0.5	E1 0.2

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(continued)

Test Example	1	2	3	4
DBPC	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3
Other additives	-	TCP 1.0	-	-
IAE oil temperature (°C)	Test aborted due to burning	159	155	151
Timken OK load (lb)	Not tested	Not tested	Not tested	Not tested

[Gear oil composition - 2]

**[0285]** This Example is an embodiment related to the gear oil formed by combining the dispersant viscosity index improver of the component (B) into the base oil.

**[0286]** Tested base oils, the kind of additives added thereto and their amounts are listed below.

**[0287]** Further, the test methods carried out on each combined lubricating oil composition is also listed below.

**[0288]** The obtained test results are presented in Tables 1 to 3.

(FZG)

**[0289]** In accordance with ASTM D 5182-91, FZG test was carried out, and the extreme pressure property of each lubricating oil composition was evaluated. During the evaluation, the unqualified stage was considered as the index.

(Thermal stability test)

**[0290]** In accordance with the "Testing method for thermal stability of lubricating oils" of JIS K2540-1989, sludge resisting effect of the lubricating oil was evaluated. That is, to a 50 ml-beaker, 45 g of the lubricating oil in Table was fed, and a copper and iron catalyst was added thereto. The mixture was left to stand in an air constant-temperature bath at 140°C for 240 hours, and then the amount of sludge in the sample oil was measured. The amount of sludge generated was determined by diluting the tested lubricating oil with n-hexane, passing through a membrane filter of 0.8 μm, and measuring the weight of the collected substance. Further, for the copper and iron catalyst, the catalyst used in the test for oxidation stability of the turbine oil (JIS K2514) that has been cut 8 times (length: about 3.5 cm) was used.

(IAE oil temperature)

**[0291]** In accordance with the IAE gear test specified in IP 166/77 (92), the oil temperature of the gear box was measured after 120 minutes by operating under the rotation number of 6000 rpm, the oil amount of 1250 ml and the load of 80 lb.

(Antiemulsifying property test)

**[0292]** In accordance with JIS K 2520, the antiemulsifying property was measured at 82°C.

(Base oil)

**[0293]**

Base oil 1: solvent refined paraffinic mineral oil (kinematic viscosity: 150 mm<sup>2</sup>/s (40°C), viscosity index: 95)

Base oil 2: 1-decene oligomer (kinematic viscosity: 150 mm<sup>2</sup>/s (40°C), viscosity index: 140)

Base oil 3: polyol ester obtained by using a mixture of pentaerythritol and dipentaerythritol and a straight-chained saturated fatty acid having 5 to 12 carbon atoms (kinematic viscosity: 150 mm<sup>2</sup>/s (40°C), viscosity index: 120)

(Additives)

Component (B)

**[0294]**

B1: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and morpholinoethyl methacrylate (10 mol%) (the average molecular weight: 80,000)

B2: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and benzoylamino methacrylate (10 mol%) (the average molecular weight: 70,000)

Component (C-1)

**[0295]** C-1-1: N-oleoyl sarcosine

Component (C-2)

**[0296]** C-2-1: nonyl phenoxyacetate (NPA)

Component (E)

**[0297]**

E1: 2,5-bis(n-nonyldithio)-1,3,4-thiadiazole

E2: Zinc dioctyldithiophosphate (ZnDTP)

Other additives

**[0298]**

Amine: dioctyldiphenyl amine

DBPC: di-tert-butyl-p-cresol

TCP: tricresylphosphate

OAP: dioleoyl acid phosphate

Imide: mono-type polybutenyl succinic acid imide obtained by reacting polybutenyl succinic anhydride with tetraethylene pentamine

[Effect concerning the embodiment of gear oil composition - 2]

**[0299]** The gear oil composition of this embodiment is excellent in the sludge resistance and water dissolubility. Therefore, the oil composition corresponds to the gear equipment used for various industrial machines with high efficiency and low cost and also to the tendency for high-speed and high-output as the gear oil, thus tolerant in higher temperature and tolerant in operation under higher load is possible.

Table 1

Example	1	2	3	4	5
Base oil	96.1	98.5	99.09	98.97	98.89
1	-	-	-	-	-
2	-	-	-	-	-
3	-	-	-	-	-
B component	B1 0.1	B2 0.2	B1 0.1	B2 0.2	B1 0.1
C-1 component	-	-	C-1-1 0.01	-	C-1-1 0.01
C-2 component	-	-	-	C-2-1 0.03	-



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(continued)

Example	1	2	3	4	5
E component	-	-	-	-	E1 0.2
DBPC	0.5	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3	0.3
Other additives	TCP 0.3	OAP 0.5	-	-	-
Unqualified stage in FZG test	10	10	10	10	>12
Amount of sludge (mg/45g)	1.8	1.5	1.2	0.7	1.5
IAE oil temperature (°C)	141	139	128	131	125
Antiemulsifying property	40-40-0 (15)	40-40-0 (15)	40-40-0 (20)	40-40-0 (20)	40-40-0 (20)

Table 2

Example	6	7	8	9
Base oil	1	2	3	
	98.95	98.65	-	-
	-	-	98.65	-
	-	-	-	98.65
Additive				
B component	B2 0.2	B2 0.2	B2 0.2	B2 0.2
C-1 component	C-1-1 0.02	C-1-1 0.02	C-1-1 0.02	C-1-1 0.02
C-2 component	C-2-1 0.03	C-2-1 0.03	C-2-1 0.03	C-2-1 0.03
E component	-	E1 0.3	E1 0.3	E1 0.3
DBPC Amine	0.5	0.5	0.5	0.5
	0.3	0.3	0.3	0.3
Other additives	-	-	-	-
Unqualified stage in FZG test	10	>12	>12	>12
Amount of sludge (mg/45 g) in the thermal stability test	1.2	1.8	0.7	2.1
IAE oil temperature (°C)	127	125	129	119
Antiemulsifying property	40-40-0 (20)	40-40-0 (20)	40-40-0 (15)	40-40-0 (30)

Table 3

Test Example	1	2	3	4	5	6
Base oil	1	2	3	4	5	6
	96.2	95.9	98.7	98.4	99.0	98.7
	-	-	-	-	-	-
	-	-	-	-	-	-

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(continued)

Test Example	1	2	3	4	5	6
Additive						
B component	-	-	-	-	-	-
C-1 component	-	-	-	-	-	-
C-2 component	-	-	-	-	-	-
E component	-	-	E2 0.5	E2 0.5	E1 0.2	E1 0.2
DBPC	0.5	0.5	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3	0.3	0.3
Other additives	TCP 3.0	TCP 3.0 Imide 0.3	-	Imide 0.3	-	Imide 0.3
Unqualified stage in FZG test	10	10	9	9	10	10
Amount of sludge (mg/45 g) in the thermal stability test	10.8	2.5	15.2	3.4	6.8	1.9
IAE oil temperature (°C)	Not tested	Not tested	Not tested	Not tested	Not tested	Not tested
Antiemulsifying property	Not tested	18-21-41 (60)	Not tested	21-23-36 (60)	Not tested	31-25-24 (60)

## [Gear oil composition - 3]

**[0300]** This Example is an embodiment related to the gear oil formed by combining the phosphorus-containing carboxylic acid compound of the component (A-1), thiophosphoric acid ester (A-2), or mixtures thereof into the base oil.

**[0301]** Tested base oils, the kind of additives added thereto and their amounts are listed below. Further, the test methods carried out on each combined lubricating oil composition is also listed below. The obtained test results are presented in Tables 1 to 5.

(FZG)

**[0302]** In accordance with ASTM D 5182-91, FZG test was carried out, and the extreme pressure property of each lubricating oil composition was evaluated. During the evaluation, the unqualified stage was considered as the index.

(Thermal stability test)

**[0303]** In accordance with the "Testing method for thermal stability of lubricating oils" of JIS K2540-1989, the sludge resisting effect of the lubricating oil was evaluated. That is, to a 50 ml-beaker, 45 g of the lubricating oil in Table was fed, and a copper and iron catalyst was added thereto. The mixture was left to stand in an air constant-temperature bath at 140°C for 240 hours, and then the amount of sludge in the sample oil was measured. The amount of sludge generated was determined by diluting the tested lubricating oil with n-hexane, passing through a membrane filter of 0.8 μm, and measuring the weight of the collected substance. Further, for the copper and iron catalyst, the catalyst used in the test for oxidation stability of the turbine oil (JIS K2514) that has been cut 8 times (length: about 3.5 cm) was used.

(IAE oil temperature)

**[0304]** In accordance with the IAE gear test specified in IP 166/77 (92), the oil temperature of the gear box was measured after 120 minutes by operating under the rotation number of 6000 rpm, the oil amount of 1250 ml and the load of 80 lb.

(Timken test)

**[0305]** In accordance with JIS K2519, OK load of each lubricating oil composition was measured.

5 (Base oil)

**[0306]**

10 Base oil 1: solvent refined paraffinic mineral oil (kinematic viscosity: 150 mm<sup>2</sup>/s (40°C), viscosity index: 95)  
Base oil 2: 1-decene oligomer (kinematic viscosity: 150 mm<sup>2</sup>/s (40°C), viscosity index: 140)  
Base oil 3: polyol ester obtained by using a mixture of pentaerythritol and dipentaerythritol and a straight-chained saturated fatty acid having 5 to 12 carbon atoms (kinematic viscosity: 150 mm<sup>2</sup>/s (40°C), viscosity index: 120)

(Additives)

15

Component (A-1)

**[0307]**

20 A-1-1:  $\beta$ -dithiophosphorylated propionic acid  
A-1-2:  $\beta$ -dithiophosphorylated propionic acid ethyl ester

Component (A-2)

25

**[0308]**

A-2-1: triphenyl phosphorothionate

Component (B)

30

**[0309]**

35 B1: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and morpholinoethyl methacrylate (10 mol%) (the average molecular weight: 80,000)  
B2: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and benzoylamino methacrylate (10 mol%) (the average molecular weight: 70,000)

Component (C-1)

40

**[0310]**

C-1-1: N-oleoyl sarcosine

Component (C-2)

45

**[0311]**

C-2-1: nonyl phenoxyacetate

50

Component (E)

**[0312]**

55 E1: 2,5-bis(n-nonyldithio)-1,3,4-thiadiazole  
E2: Zinc dioctyldithiophosphate (ZnDTP)

Other additives

**[0313]**

5 Amine: dioctyldiphenyl amine  
DBPC: di-tert-butyl-p-cresol  
TCP: tricresylphosphate

[Effect concerning the embodiment of gear oil composition - 3]

10 **[0314]** The gear oil composition of this embodiment is excellent in the sludge resistance and extreme pressure property. Therefore, the oil composition corresponds to the gear equipment used for various industrial machines with high efficiency and low cost and also to the tendency for high-speed and high-output as the gear oil, thus tolerant in higher temperature and tolerant in operation under higher load is possible.

Table 1

Example	1	2	3	4	5
Base oil	99.18	99.0	99.0	99.0	98.9
1	-	-	-	-	-
2	-	-	-	-	-
3	-	-	-	-	-
Additives					
A-1 component	A-1-1 0.02	A-1-2 0.2	-	A-1-2 0.1	A-1-2 0.2
A-2 component	-	-	A-2-1 0.2	A-2-1 0.1	-
B component	-	-	-	-	B1 0.1
C-1 component C-1-1	-	-	-	-	-
C-2 component C-2-1	-	-	-	-	-
E component E1	-	-	-	-	-
DBPC	0.5	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3	0.3
Other additives	-	-	-	-	-
Unqualified stage in FZG test	>12	>12	>12	>12	>12
Amount of sludge (mg/45 g) in the thermal stability test	5.4	4.3	3.2	5.8	0.8
IAE oil temperature (°C)	145	140	138	135	141
Timken OK load	21(1b)	24(1b)	24(1b)	30(1b)	24(1b)

Table 2

Example	6	7	8	9	10
Base oil	98.98	98.75	99.07	98.77	98.87
1	-	-	-	-	-
2	-	-	-	-	-
3	-	-	-	-	-
Additives					
A-1 component	A-1-1 0.02	A-1-2 0.2	A-1-1 0.02	A-1-2 0.1	A-1-1 0.02
A-2 component	-	A-2-1 0.05	-	A-2-1 0.1	-

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(continued)

Example	6	7	8	9	10
B component	B2 0.2	B1 0.2	B1 0.1	B2 0.2	B 1 0.1
C-1 component	-	-	C-1-1 0.01	-	C-1-1 0.01
C-2 component	-	-	-	C-2-1 0.03	-
E component	-	-	-	-	E1 0.2
DBPC	0.5	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3	0.3
Other additives Unqualified stage in FZG test Amount of sludge (mg/45 g) in the thermal stability test	-	-	-	-	-
	>12	>12	>12	>12	>12
	1.0	0.7	1.2	0.7	1.5
IAE oil temperature (°C)	143	140	128	131	125
Timken OK load	21(1b)	40(1b)	24(1b)	35(1b)	55(1b)

Table 3

Example	11	12	13	14	15
Base oil	1	2	3		
	98.75	98.45	-	-	-
	-	-	99.18	98.63	-
	-	-	-	-	99.0
Additives					
A-1 component	A-1-2 0.1	A-1-2 0.1	A-1-1 0.02	A-1-1 0.02	A-1-2 0.1
A-2 component	A-2-1 0.1	A-2-1 0.1	-	-	A-2-1 0.1
B component	B2 0.2	B2 0.2	-	B2 0.2	-
C-1 component	C-1-1 0.02	C-1-1 0.02	-	C-1-1 0.02	-
C-2 component	C-2-1 0.03	C-2-1 0.03	-	C-2-1 0.03	-
E component	-	E1 0.3	-	E1 0.3	-
DBPC	0.5	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3	0.3
Other additives	-	-	-	-	-
Unqualified stage in FZG test	>12	>12	>12	>12	>12
Amount of sludge (mg/45 g) in the thermal stability test	1.1	1.6	1.2	0.7	0.8
IAE oil temperature (°C)	129	123	128	131	132
Timken OK load	21(1b)	60(1b)	24(1b)	35(1b)	30(1b)

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Table 4

Example	16
Base oil	1
	2
	3
	-
	-
	98.45
Additives	
A-1 component	A-1-2
	0.1
A-2 component	A-2-1
	0.1
B component	B2
	0.2
C-1 component	C-1-1
C-1-1	0.02
C-2 component	C-2-1
C-2-1	0.03
E component	E1
E1	0.3
DBPC	0.5
Amine	0.3
Other additives	-
Unqualified stage in FZG test	>12
Amount of sludge (mg/45 g) in the thermal stability test	1.6
IAE oil temperature (°C)	121
Timken OK load	60(lb)

Table 5

Test Example	1	2	3	4
Base oil	1	99.2	98.2	98.7
	2	-	-	-
	3	-	-	-
Additives				
A-1 component	-	-	-	-
A-2 component	-	-	-	-
B component	-	-	-	-
C-1 component	-	-	-	-
C-2 component	-	-	-	-
E component	-	-	E2	E1
			0.5	0.2
DBPC	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3

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(continued)

Test Example	1	2	3	4
Other additives	-	TCP 1.0	-	-
Unqualified stage in FZG test	4	8	9	10
Amount of sludge (mg/45 g) in the thermal stability test	Not tested	5.2	15.2	6.8
IAE oil temperature (°C)	Not tested	Not tested	Not tested	Not tested
Timken OK load	Not tested	Not tested	Not tested	Not tested

## [Lubricating oil composition for paper machines - 1]

**[0315]** This Example is an embodiment related to the lubricating oil composition for paper machines formed by combining the component (C-1), the component (C-2), or mixtures thereof into the base oil.

**[0316]** Hereinbelow, the embodiment will be further described by way of Examples.

**[0317]** Tested base oils, the kind of additives added thereto and their amounts are listed below. Further, the test methods carried out on each combined lubricating oil composition is also listed below. The obtained test results are presented in Tables 1 to 3.

(Test method)

(Abrasion resistance: FALEX abrasion test)

**[0318]** In the FALEX abrasion test, the test machine specified in "Standard Test Methods for Measurement of Extreme Pressure Properties of Fluid Lubricants (Falex Pin Vee Block Methods)" of ASTM D 3233-93 was used to measure the abrasion amount. Further, a pin (journal) made of steel having the outer diameter of 6.35 mm (1/4 inch), the length of 31.75 mm (1 and 1/4 inch), a V-type block made of steel having the angle of 96° were used. The test was carried out under the test conditions of the rotation number of 1000 rpm, the direct load of 100 lbs and the test time of 10 hours.

(Thermal stability test)

**[0319]** In accordance with the "Testing method for thermal stability of lubricating oils" of JIS K2540-1989, the sludge resisting effect of the lubricating oil was evaluated. That is, to a 50 ml-beaker, 45 g of the lubricating oil in Table was fed, and a copper and iron catalyst was added thereto. The mixture was left to stand in an air constant-temperature bath at 150°C for 120 hours, and then the amount of sludge in the sample oil was measured. The amount of sludge generated was determined by diluting the tested lubricating oil with n-hexane, passing through a membrane filter of 0.8 μm, and measuring the weight of the collected substance. Further, for the copper and iron catalyst, the catalyst used in the test for oxidation stability of the turbine oil (JIS K2514) that has been cut 8 times (length: about 3.5 cm) was used.

(Anticorrosion property: Rust resistance test)

**[0320]** For the rust resistance test, the test specified in "Testing Method of Anticorrosion Properties of Lubricating Oil" of JIS K 2510, that was modified to evaluate anticorrosion property of the lubricating oil for paper machine, was carried out. The modified point is that usually when evaluating the lubricating oil, distilled water or artificial salt water is used, but in the case of evaluating the lubricating oil for paper machines, artificial white water is used instead.

(Base oil)

**[0321]**

Base oil 1: solvent refined paraffinic mineral oil (kinematic viscosity: 150 mm<sup>2</sup>/s (40°C), viscosity index: 95)

Base oil 2: 1-decene oligomer (kinematic viscosity: 150 mm<sup>2</sup>/s (40°C), viscosity index: 140)

Base oil 3: polyol ester obtained by using a mixture of pentaerythritol and dipentaerythritol and a straight-chained saturated fatty acid having 5 to 12 carbon atoms (kinematic viscosity: 220 mm<sup>2</sup>/s (40°C), viscosity index: 120)

(Additives)

Component (C-1)

**[0322]**

C-1-1: N-oleoyl sarcosine

Component (C-2)

**[0323]**

C-2-1: nonyl phenoxyacetate (NPA)

Component (E)

**[0324]**

E1: 2,5-bis(n-nonyldithio)-1,3,4-thiadiazole

E2: Zinc dioctyldithiophosphate (ZnDTP)

Other additives

**[0325]**

DBPC: di-tert-butyl-p-cresol

Amine: dioctyldiphenyl amine

TCP: tricresylphosphate

[Effect concerning the embodiment of lubricating oil composition for paper machines - 1]

**[0326]** The lubricating oil composition for paper machines of this embodiment is excellent in the heat resistance, the abrasion resistance and the anticorrosion property at the same time. Therefore, the lubricating oil composition for paper machines is capable of corresponding to the recently developed paper machines.

Table 1

Example	1	2	3	4	5
Base oil	99.19	98.17	98.99	99.15	98.85
1	-	-	-	-	-
2	-	-	-	-	-
3	-	-	-	-	-
Additives					
C-1 component	C-1-1 0.01	-	C-1-1 0.01	C-1-1 0.02	C-1-1 0.02
C-2 component	-	C-2-1 0.03	-	C-2-1 0.03	C-2-1 0.03
E component	-	-	E1 0.2	-	E1 0.3
DBPC	0.5	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3	0.3
Other additives	-	-	-	-	-
FALEX abrasion amount (mg)	5.7	4.9	1.5	2.3	0.8
Amount of sludge (mg) in the thermal stability test	6.5	5.9	7.9	4.8	7.1



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(continued)

Example	1	2	3	4	5
Rust resistance	Rustless	Rustless	Rustless	Rustless	Rustless

Table 2

Example	6	7
Base oil	-	-
1	-	-
2	98.85	-
3	-	98.85
Additives		
C-1 component	C-1-1 0.02	C-1-1 0.02
C-2 component	C-2-1 0.03	C-2-1 0.03
E component	E1 0.3	E1 0.3
DBPC	0.5	0.5
Amine	0.3	0.3
Other additives	-	-
FALEX abrasion amount (mg)	1.0	0.7
Amount of sludge (mg) in the thermal stability test	7.4	5.8
Rust resistance	Rustless	Rustless

Table 3

Test Example	1	2	3	4
Base oil	99.2	98.2	98.7	99.0
1	-	-	-	-
2	-	-	-	-
3	-	-	-	-
Additives				
C-1 component	-	-	-	-
C-2 component	-	-	-	-
E component	-	-	E2 0.5	E1 0.2
DBPC	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3
Other additives	-	TCP 1.0	-	
FALEX abrasion amount (mg)	22.9	8.1	9.3	7.5
Amount of sludge (mg) in the thermal stability test	54.5	5.2	22.3	9.8
Rust resistance	Rusted (high degree)	Rusted (high degree)	Rusted (high degree)	Rusted (high degree)

[Lubricating oil composition for paper machines - 2]

**[0327]** This Example is an embodiment related to the lubricating oil composition for paper machines formed by combining the dispersant viscosity index improver of the component (B) into the base oil.

**[0328]** Hereinbelow, the embodiment will be further described by way of Examples.

**[0329]** Tested base oils, the kind of additives added thereto and their amounts are listed below. Further, the test methods carried out on each combined lubricating oil composition is also listed below. The obtained test results are presented in Tables 1 to 3.

(Test method)

(Abrasion resistance: FALEX abrasion test)

**[0330]** In the FALEX abrasion test, the test machine specified in "Standard Test Methods for Measurement of Extreme Pressure Properties of Fluid Lubricants (Falex Pin Vee Block Methods)" of ASTM D 3233-93 was used to measure the abrasion amount. Further, a pin (journal) made of steel having the outer diameter of 6.35 mm (1/4 inch), the length of 31.75 mm (1 and 1/4 inch), a V-type block made of steel having the angle of 96° were used. The test was carried out under the test conditions of the rotation number of 1000 rpm, the direct load of 100 lbs and the test time of 16 hours.

(Thermal stability test)

**[0331]** In accordance with the "Testing method for thermal stability of lubricating oils" of JIS K2540-1989, the sludge resisting effect of the lubricating oil was evaluated. That is, to a 50 ml-beaker, 45 g of the lubricating oil in Table was fed, and a copper and iron catalyst was added thereto. The mixture was left to stand in an air constant-temperature bath at 150°C for 120 hours, and then the amount of sludge in the sample oil was measured. The amount of sludge generated was determined by diluting the tested lubricating oil with n-hexane, passing through a membrane filter of 0.8 μm, and measuring the weight of the collected substance. Further, for the copper and iron catalyst, the catalyst used in the test for oxidation stability of the turbine oil (JIS K2514) that has been cut 8 times (length: about 3.5 cm) was used.

(Anticorrosion property: Rust resistance test)

**[0332]** For the rust resistance test, the test specified in "Testing Method of Anticorrosion Properties of Lubricating Oil" of JIS K 2510, that was modified to evaluate anticorrosion property of the lubricating oil for paper machine, was carried out. The modified point is that usually when evaluating the lubricating oil, distilled water or artificial salt water is used, but in the case of evaluating the lubricating oil for paper machines, artificial white water is used instead.

(Base oil)

**[0333]**

Base oil 1: solvent refined paraffinic mineral oil (kinematic viscosity: 220 mm<sup>2</sup>/s (40°C), viscosity index: 95)

Base oil 2: 1-decene oligomer (kinematic viscosity: 220 mm<sup>2</sup>/s (40°C), viscosity index: 140)

Base oil 3: polyol ester obtained by using a mixture of pentaerythritol and dipentaerythritol and a straight-chained saturated fatty acid having 5 to 12 carbon atoms (kinematic viscosity: 220 mm<sup>2</sup>/s (40°C), viscosity index: 120)

(Additives)

Component (B)

**[0334]**

B1: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and morpholinoethyl methacrylate (10 mol%) (the average molecular weight: 80,000)

B2: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and benzoylamino methacrylate (10 mol%) (the average molecular weight: 70,000)

Component (C-1)

**[0335]**

5 C-1-1: N-oleoyl sarcosine

Component (C-2)

**[0336]**

10

C-2-1: nonyl phenoxyacetate (NPA)

Component (E)

15

**[0337]**

E1: 2,5-bis(n-nonyldithio)-1,3,4-thiadiazole

E2: Zinc dioctyldithiophosphate (ZnDTP)

20

Other additives

**[0338]**

25

DBPC: di-tert-butyl-p-cresol

Amine: dioctyldiphenyl amine

TCP: tricresylphosphate

OAP: dioleoyl acid phosphate

Imide: mono-type polybutenyl succinic acid imide obtained by reacting polybutenyl succinic anhydride with tetraethylene pentamine

30

[Effect concerning the embodiment of lubricating oil composition for paper machines - 2]

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**[0339]** The lubricating oil composition for paper machines of this embodiment is excellent in the heat resistance, the abrasion resistance and the anticorrosion property at the same time. Therefore, the lubricating oil composition for paper machines is capable of corresponding to the recently developed paper machines.

Table 1

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45

50

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Example	1	2	3	4	5
Base oil	96.1	98.5	99.09	98.97	98.89
1	-	-	-	-	-
2	-	-	-	-	-
3	-	-	-	-	-
B: dispersant polymer	B1 0.1	B2 0.2	B1 0.1	B2 0.2	B1 0.1
C-1: sarcosine	-	-	C-1-1 0.01	-	C-1-1 0.01
C-2: NPA	-	-	-	C-2-1 0.03	-
E: sulfur-based extreme pressure additives	-	-	-	-	E1 0.2
DBPC	0.5	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3	0.3
Other additives	TCP 3.0	OAP 0.5	-	-	-

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(continued)

Example	1	2	3	4	5
FALEX abrasion amount (mg)	8.7	6.5	4.8	3.9	2.1
Amount of sludge (mg/45 g) in the thermal stability test	2.2	3.1	1.5	1.0	1.8
Rust resistance	Rustless	Rustless	Rustless	Rustless	Rustless
Antiemulsifying property	40-40-0 (20)	40-40-0 (20)	40-40-0 (25)	40-40-0 (25)	40-40-0 (25)

Table 2

Example	6	7	8	9
Base oil	98.95	98.65	-	-
1	-	-	98.65	-
2	-	-	-	98.65
3	-	-	-	98.65
B: dispersant polymer	B2 0.2	B2 0.2	B2 0.2	B2 0.2
C-1: sarcosine	C-1-1 0.02	C-1-1 0.02	C-1-1 0.02	C-1-1 0.02
C-2: NPA	C-2-1 0.03	C-2-1 0.03	C-2-1 0.03	C-2-1 0.03
E: sulfur-based extreme pressure additives	-	E1 0.3	E1 0.3	E1 0.3
DBPC	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3
Other additives	-	-	-	-
FALEX abrasion amount (mg)	2.5	2.8	1.9	2.0
Amount of sludge (mg/45 g) in the thermal stability test	1.0	1.5	0.9	1.9
Rust resistance	Rustless	Rustless	Rustless	Rustless
Antiemulsifying	40-40-0 (25)	40-40-0 (25)	40-40-0 (15)	41-38-1 (35)

Table 3

Example	1	2	3	4	5	6
Base oil	96.2	95.9	98.7	98.4	99.0	98.7
1	-	-	-	-	-	-
2	-	-	-	-	-	-
3	-	-	-	-	-	-
Additives						
B: dispersant polymer	-	-	-	-	-	-
C-1: sarcosine	-	-	-	-	-	-
C-2: NPA	-	-	-	-	-	-
E: sulfur-based extreme pressure additives	-	-	ZnDTP 0.5	ZnDTP 0.5	F1 0.2	F1 0.2
DBPC	0.5	0.5	0.5	0.5	0.5	0.5

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(continued)

Example	1	2	3	4	5	6
Amine	0.3	0.3	0.3	0.3	0.3	0.3
Other additives	TCP 3.0	TCP 3.0 Imide 0.3	-	Imide 0.3	-	Imide 0.3
FALEX abrasion amount (mg)	8.3	9.1	17.1	19.5	14.4	15.7
Amount of sludge (mg/45g) in the thermal stability test	12.5	3.1	21.8	4.5	12.1	3.9
Rust resistance Rust existence	Rusted (high degree)	Rusted (high degree)	Rusted (high degree)	Rusted (high degree)	Rusted (high degree)	Rusted (high degree)
Antiemulsifying property	Not tested	16-23-41 (60)	Not tested	18-22-40 (60)	Not tested	25-28-27 (60)

[Lubricating oil composition for paper machines - 3]

**[0340]** This Example is an embodiment related to the lubricating oil composition for paper machines formed by combining the phosphorus-containing carboxylic acid compound of the component (A-1), thiophosphoric acid ester (A-2), or mixtures thereof into the base oil.

**[0341]** Hereinbelow, the embodiment will be further described by way of Examples.

**[0342]** Tested base oils, the kind of additives added thereto and their amounts are listed below. Further, the test methods carried out on each combined lubricating oil composition is also listed below. The obtained test results are presented in Tables 1 to 5.

(Test method)

(Abrasion resistance: FALEX abrasion test)

**[0343]** In the FALEX abrasion test, the test machine specified in "Standard Test Methods for Measurement of Extreme Pressure Properties of Fluid Lubricants (Falex Pin Vee Block Methods)" of ASTM D 3233-93 was used to measure the abrasion amount. Further, a pin (journal) made of steel having the outer diameter of 6.35 mm (1/4 inch), the length of 31.75 mm (1 and 1/4 inch), a V-type block made of steel having the angle of 96° were used. The test was carried out under the test conditions of the rotation number of 1000 rpm, the direct load of 100 lbs and the test time of 16 hours.

(Thermal stability test)

**[0344]** In accordance with the "Testing method for thermal stability of lubricating oils" of JIS K2540-1989, the sludge resisting effect of the lubricating oil was evaluated. That is, to a 50 ml-beaker, 45 g of the lubricating oil in Table was fed, and a copper and iron catalyst was added thereto. The mixture was left to stand in an air constant-temperature bath at 150°C for 120 hours, and then the amount of sludge in the sample oil was measured. The amount of sludge generated was determined by diluting the tested lubricating oil with n-hexane, passing through a membrane filter of 0.8 μm, and measuring the weight of the collected substance. Further, for the copper and iron catalyst, the catalyst used in the test for oxidation stability of the turbine oil (JIS K2514) that has been cut 8 times (length: about 3.5 cm) was used.

(Anticorrosion property: Rust resistance test)

**[0345]** For the rust resistance test, the test specified in "Testing Method of Anticorrosion Properties of Lubricating Oil" of JIS K 2510, that was modified to evaluate anticorrosion property of the lubricating oil for paper machine, was carried out. The modified point is that usually when evaluating the lubricating oil, distilled water or artificial salt water is used, but in the case of evaluating the lubricating oil for paper machines, artificial white water is used instead, and the test

time was 2 hours.

(Base oil)

5 **[0346]**

Base oil 1: solvent refined paraffinic mineral oil (kinematic viscosity: 220 mm<sup>2</sup>/s (40°C), viscosity index: 95)

Base oil 2: 1-decene oligomer (kinematic viscosity: 220 mm<sup>2</sup>/s (40°C), viscosity index: 140)

10 Base oil 3: polyol ester obtained by using a mixture of pentaerythritol and dipentaerythritol and a straight-chained saturated fatty acid having 5 to 12 carbon atoms (kinematic viscosity: 220 mm<sup>2</sup>/s (40°C), viscosity index: 120)

(Additives)

Component (A-1)

15 **[0347]**

A-1-1:  $\beta$ -dithiophosphorylated propionic acid

20 A-1-2:  $\beta$ -dithiophosphorylated propionic acid ethyl ester

Component (A-2)

**[0348]**

25 A-2-1: triphenyl phosphorothionate

Component (B)

**[0349]**

30 B1: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and morpholinoethyl methacrylate (10 mol%) (the average molecular weight: 80,000)

B2: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and benzoylamino methacrylate (10 mol%) (the average molecular weight: 70,000)

35 Component (C-1)

**[0350]**

40 C-1-1: N-oleoyl sarcosine

Component (C-2)

**[0351]**

45 C-2-1: nonyl phenoxyacetate (NPA)

Component (E)

50 **[0352]**

E1: 2,5-bis(n-nonyldithio)-1,3,4-thiadiazole

E2: Zinc dioctyldithiophosphate (ZnDTP)

55 Other additives

**[0353]**

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Amine: dioctyldiphenyl amine

DBPC: di-tert-butyl-p-cresol

TCP: tricresylphosphate

[Effect concerning the embodiment of lubricating oil composition for paper machines - 3]

**[0354]** The lubricating oil composition for paper machines of this embodiment is excellent in the heat resistance, the abrasion resistance and the anticorrosion property at the same time. Therefore, the lubricating oil composition for paper machines is capable of corresponding to the recently developed paper machines.

Table 1

Example	1	2	3	4	5
Base oil	99.18	99.0	99.0	99.0	98.9
1	-	-	-	-	-
2	-	-	-	-	-
3	-	-	-	-	-
Additives					
A-1 component	A-1-1 0.02	A-1-2 0.2	-	A-1-2 0.1	A-1-2 0.2
A-2 component	-	-	A-2-1 0.2	A-2-1 0.2	-
B component	-	-	-	-	B 1 0.1
C-1 component	-	-	-	-	-
C-2 component	-	-	-	-	-
E component	-	-	-	-	-
DBPC	0.5	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3	0.3
Other additives	-	-	-	-	-
FALEX abrasion amount (mg)	3.2	4.1	5.4	2.8	3.9
Amount of sludge (mg/45 g) in the thermal stability test	4.9	5.1	3.8	5.7	0.5
Rust resistance	Rustless	Rustless	Rustless	Rustless	Rustless

Table 2

Example	6	7	8	9	10
Base oil	98.98	98.75	99.07	98.77	98.87
1	-	-	-	-	-
2	-	-	-	-	-
3	-	-	-	-	-
Additives					
A-1 component	A-1-1 0.02	A-1-2 0.2	A-1-1 0.02	A-1-2 0.1	A-1-1 0.02
A-2 component	-	A-2-1 0.05	-	A-2-1 0.1	-
B component	B2 0.2	B1 0.2	B1 0.1	B2 0.2	B1 0.1

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(continued)

Example	6	7	8	9	10
C-1 component	-	-	C-1-1 0.01	-	C-1-1 0.01
C-2 component	-	-	-	C-2-1 0.03	-
E component	-	-	-	-	E1 0.2
DBPC	0.5	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3	0.3
Other additives	-	-	-	-	-
FALEX abrasion amount (mg)	3.5	3.1	2.1	2.0	0.9
Amount of sludge (mg/45 g) in the thermal stability test	1.2	0.9	1.1	0.5	1.8
Rust resistance	Rustless	Rustless	Rustless	Rustless	Rustless

Table 3

Example	11	12	13	14	15
Base oil	1 98.75	2 98.45	-	-	-
	2 -	-	99.18	98.63	-
	3 -	-	-	-	99.0
Additives					
A-1 component	A-1-2 0.1	A-1-2 0.1	A-1-1 0.02	A-1-1 0.02	A-1-2 0.1
A-2 component	A-2-1 0.1	A-2-1 0.1	-	-	A-2-1 0.1
B component	B2 0.2	B2 0.2	-	B2 0.2	-
C-1 component	C-1-1 0.02	C-1-1 0.02	-	C-1-1 0.02	-
C-2 component	C-2-1 0.03	C-2-1 0.03	-	C-2-1 0.03	-
E component	-	E1 0.3	-	E1 0.3	-
DBPC	0.5	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3	0.3
Other additives	-	-	-	-	-
FALEX abrasion amount (mg)	1.0	0.5	4.6	1.5	3.4
Amount of sludge (mg/45 g) in the thermal stability test	0.9	1.1	2.2	0.5	1.8
Rust resistance	Rustless	Rustless	Rustless	Rustless	Rustless



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Table 4

Example	16
Base oil	1
	2
	3
	98.45
Additives	
A-1 component	A-1-2 0.1
A-2 component	A-2-1 0.1
B component	B2 0.2
C-1 component	C-1-1 0.02
C-2 component	C-2-1 0.03
E component	E1 0.3
DBPC	0.5
Amine	0.3
Other additives	-
FALEX abrasion amount (mg)	0.9
Amount of sludge (mg/45 g) in the thermal stability test	1.2
Rust resistance	Rustless

Table 5

Test Example	1	2	3	4
Base oil	1	99.2	98.2	98.7
	2	-	-	-
	3	-	-	-
Additives				
A-1 component	-	-	-	-
A-2 component	-	-	-	-
B component	-	-	-	-
C-1 component	-	-	-	-
C-2 component	-	-	-	-
E component	-	-	E2 0.5	E1 0.2
DBPC	0.5	0.5	0.5	0.5
Amine	0.3	0.3	0.3	0.3
Other additives	-	TCP 1.0	-	-

(continued)

Test Example	1	2	3	4
FALEX abrasion amount (mg)	33.5	12.7	17.1	14.4
Amount of sludge (mg/45 g) in the thermal stability test	54.5	5.2	22.3	9.8
Rust resistance	Rusted (high degree)	Rusted (high degree)	Rusted (high degree)	Rusted (high degree)

#### [Lubricating oil composition for slide guides - 1]

**[0355]** This Example is an embodiment related to the lubricating oil composition for slide guides formed by combining the component (C-1), the component (C-2), or mixtures thereof into the base oil. Hereinbelow, the embodiment will be further described by way of Examples.

**[0356]** Tested base oils, the kind of additives added thereto and their amounts are listed below. Further, the test methods carried out on each combined lubricating oil composition is also listed below. The obtained test results are presented in Tables 1 to 3.

(Frictional property evaluation test)

**[0357]** Fig. 1 is a simplified block diagram illustrating the frictional index measuring system used in the frictional property evaluation test. In Fig. 1, a table 1 and a rolling jig 4 connected by introducing a load cell 5 is arranged on a bed 6, and a heavyweight 9 as a substituted of a working tool is further arranged on the table 1. The table 1 and the bed 6 are all made of cast iron. Moreover, the rolling jig 4 has a bearing part, where to an A/C servomotor 2 is connected by introducing a lead screw 3. By driving the lead screw 3 by the A/C servomotor 2, the rolling jig 4 can be reciprocated in the axial direction (the arrow direction in Fig. 1) of the lead screw 3. Further, the load cell 5 is connected with a computer 7 and the computer 7 and the A/C servomotor 2 are individually connected with a control board 8 electrically, thereby controlling the reciprocating movement of the rolling jig 4 and measuring the load between the table 1 and the rolling jig 4 can be carried out.

**[0358]** In such the frictional index measuring system, the lubricating oil composition is dropped on the surface of the bed 6, the surface pressure between the table 1 and the bed 6 is adjusted to 200 kPa by the selection of the table heavyweight 9, and then the rolling jig 4 is reciprocated with the screw speed of 60 mm/min and the screw length of 750 mm. At this time, the load between the table 1 and the rolling jig 4 is measured by the load cell 5 (the load meter), and based on the resulting measurement, the frictional index of the guide face (table 1/bed 6 = cast iron/cast iron) is determined. Furthermore, the test was carried out after carrying out the pre-conditioned interim operation for 3 times. The frictional index of each lubricating oil composition is presented in Tables 1 to 3.

(Anti-stick-slip property evaluation test)

**[0359]** Fig. 2 is a simplified block diagram illustrating the anti-stick-slip property evaluation apparatus (TE-77 tester manufactured by PLINT & PARTNERS LTD.). The apparatus illustrated in Fig. 2 is an apparatus in which a lower test piece 12, an upper test piece 11 and an elastic body 10 are laminated on the supporting die 110 in this order, and the test pieces 11 and 12 are slid by reciprocally moving (sliding movement) the elastic body 10 along the surface of the supporting die 110 while imposing the test pieces 11 and 12 together with a specific load. During such sliding, the frictional index between the test pieces 11 and 12 can be determined by measuring the load added to the test pieces 11 and 12 using a load detector 13. Fig. 3 is a graph showing an example of the correlation between the frictional index obtained by the operation and time.  $\Delta\mu$  in Fig. 3 represents the amplitude of the frictional index.

**[0360]** Using such the apparatus,  $\Delta\mu$ , when each lubricating oil composition was introduced between the test pieces 11 and 12, was measured based on the method illustrated in the publication (Proceedings of Japanese Society of Tribologist Tribology conference, Tokyo 1999-5 D17), except that the test pieces and the conditions were modified for the lubricating oil for slide guides evaluation. Specifically, the test was carried out using JIS G 4051 S45C as the test pieces 11, 12 and chloroprene rubber as the elastic body 10, and with the average sliding speed of 0.3 mm/s and under the load of 250 N. The obtained results are presented in Tables 1 to 3.

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(Base oil)

### [0361]

- 5 Base oil 1: solvent refined paraffinic mineral oil (kinematic viscosity: 68 mm<sup>2</sup>/s (40°C), viscosity index: 95)  
 Base oil 2: 1-decene oligomer (kinematic viscosity: 68 mm<sup>2</sup>/s (40°C), viscosity index: 140)  
 Base oil 3: polyol ester obtained by using pentaerythritol and a straight-chained saturated fatty acid having 5 to 12 carbon atoms (kinematic viscosity: 68 mm<sup>2</sup>/s (40°C), viscosity index: 120)

10 (Additives)

Component (C-1)

### [0362]

15 C-1-1: N-oleoyl sarcosine

Component (C-2)

### [0363]

C-2-1: nonyl phenoxyacetate (NPA)

Component (E)

### [0364]

E1: 2,5-bis(n-nonyldithio)-1,3,4-thiadiazole  
 E2: sulfurized ester (the content of sulfur: 11.4%)

30 Other additives

### [0365]

35 Amine: dioctyldiphenyl amine  
 DBPC: di-tert-butyl-p-cresol  
 TCP: tricresylphosphate  
 OAP: dioleoyl acid phosphate  
 OAc: oleic acid

40 [Effect concerning the embodiment of lubricating oil composition for slide guides - 1]

[0366] The lubricating oil composition for slide guides of this embodiment is excellent in both the frictional property and the anti-stick-slip property.

45

Table 1

Example	1	2	3	4	5
Base oil	1	2	3	4	5
	99.69	99.67	99.49	99.65	99.35
	2	-	-	-	-
	3	-	-	-	-
Additives					
C-1 component	C-1-1	-	C-1-1	C-1-1	C-1-1
	0.01		0.01	0.02	0.02
C-2 component	-	C-2-1	-	C-2-1	C-2-1
		0.03		0.03	0.03

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(continued)

Example	1	2	3	4	5
E component	-	-	E1 0.2	-	E1 0.3
DBPC	0.2	0.2	0.2	0.2	0.2
Amine	0.1	0.1	0.1	0.1	0.1
Other additives	-	-	-	-	-
Frictional index	0.109	0.110	0.112	0.106	0.109
Anti-stick-slip property ( $\Delta\mu$ )	0.009	0.010	0.008	0.008	0.007

Table 2

Example	6	7
Base oil	-	-
1	-	-
2	99.35	-
3	-	99.35
Additives		
C-1 component	C-1-1 0.02	C-1-1 0.02
C-2 component	C-2-1 0.03	C-2-1 0.03
E component	E1 0.3	E1 0.3
DBPC	0.2	0.2
Amine	0.1	0.1
Other additives	-	-
Frictional index	0.110	0.108
Anti-stick-slip property ( $\Delta\mu$ )	0.006	0.006

Table 3

Test Example	1	2	3	4	5
Base oil	99.2	99.2	98.7	98.7	98.7
1	-	-	-	-	-
2	-	-	-	-	-
3	-	-	-	-	-
Additives	-	-	-	-	-
C-1 component	-	-	-	-	-
C-2 component	-	-	-	-	-
E component	-	-	-	E1 1.0	E2 1.0
DBPC	0.2	0.2	0.2	0.2	0.2
Amine	0.1	0.1	0.1	0.1	0.1
Other additives	TCP 0.5	OAP 0.5	OAc 1.0	-	-

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(continued)

Test Example	1	2	3	4	5
Frictional index	0.145	0.139	0.150	0.161	0.159
Anti-stick-slip property ( $\Delta\mu$ )	>0.05	0.013	0.015	0.018	0.017

[Lubricating oil composition for slide guides - 2]

**[0367]** This Example is an embodiment related to the lubricating oil composition for slide guides formed by combining the dispersant viscosity index improver of the component (B) into the base oil. Hereinbelow, the embodiment will be further described by way of Examples.

**[0368]** Tested base oils, the kind of additives added thereto and their amounts are listed below.

**[0369]** Further, the test methods carried out on each combined lubricative composition is also listed below.

**[0370]** The obtained test results are presented in Tables 1 to 4.

(Frictional property evaluation test)

**[0371]** Fig. 1 is a simplified block diagram illustrating the frictional index measuring system used in the frictional property evaluation test. In Fig. 1, a table 1 and a rolling jig 4 connected by introducing a load cell 5 is arranged on a bed 6, and a heavyweight 9 as a substituted of a working tool is further arranged on the table 1. The table 1 and the bed 6 are all made of cast iron. Moreover, the rolling jig 4 has a bearing part, where to an A/C servmotor 2 is connected by introducing a lead screw 3. By driving the lead screw 3 by the A/C servmotor 2, the rolling jig 4 can be reciprocated in the axial direction (the arrow direction in Fig. 1) of the lead screw 3. Further, the load cell 5 is connected with a computer 7 and the computer 7 and the A/C servmotor 2 are individually connected with a control board 8 electrically, thereby controlling the reciprocating movement of the rolling jig 4 and measuring the load between the table 1 and the rolling jig 4 can be carried out.

**[0372]** In such the frictional index measuring system, the lubricating oil composition is dropped on the surface of the bed 6, the surface pressure between the table 1 and the bed 6 is adjusted to 200 kPa by the selection of the table heavyweight 9, and then the rolling jig 4 is reciprocated with the screw speed of 60 mm/min and the screw length of 750 mm. At this time, the load between the table 1 and the rolling jig 4 is measured by the load cell 5 (the load meter), and based on the resulting measurement, the frictional index of the guide face (table 1/bed 6 = cast iron/cast iron) is determined. Furthermore, the test was carried out after carrying out the pre-conditioned interim operation for 3 times. The frictional index of each lubricating oil composition is presented in Tables 1 to 4.

(Anti-stick-slip property evaluation test)

**[0373]** Fig. 2 is a simplified block diagram illustrating the anti-stick-slip property evaluation apparatus (TE-77 tester manufactured by PLINT & PARTNERS LTD.). The apparatus illustrated in Fig. 2 is an apparatus in which a lower test piece 12, an upper test piece 11 and an elastic body 10 are laminated on the supporting die 110 in this order, and the test pieces 11 and 12 are slid by reciprocally moving (sliding movement) the elastic body 10 along the surface of the supporting die 110 while imposing the test pieces 11 and 12 together with a specific load. During such sliding, the frictional index between the test pieces 11 and 12 can be determined by measuring the load added to the test pieces 11 and 12 using a load detector 13. Fig. 3 is a graph showing an example of the correlation between the frictional index obtained by the operation and time.  $\Delta\mu$  in Fig. 3 represents the amplitude of the frictional index.

**[0374]** Using such the apparatus,  $\Delta\mu$ , when each lubricating oil composition was introduced between the test pieces 11 and 12, was measured based on the method illustrated in the publication (Proceedings of Japanese Society of Tribologist Tribology conference, Tokyo 1999-5 D 17), except that the test pieces and the conditions were modified for the lubricating oil for slide guides evaluation. Specifically, the test was carried out using JIS G 4051 S45C as the test pieces 11, 12 and chloroprene rubber as the elastic body 10, and with the average sliding speed of 0.3 mm/s and under the load of 250 N. The obtained results are presented in Tables 1 to 4.

(Dissolubility with water-soluble cutting fluid evaluation test)

**[0375]** 20 mL of the lubricating oil composition and 80 mL of the water-soluble cutting fluid (emulsion type cutting fluid, manufactured by Nippon Oil Corp., equivalent product of W1 Type 1 "cutting fluid" in JISK 2241, dilution ratio: 10-folds) were extracted in a 100 mL-mess cylinder. A stopper was put on the cylinder and the cylinder was vibrated for 1 minute at room temperature. Then, the dissolubility was evaluated by observing the separated appearance when 24 hours have

been passed. The dissolubility evaluation is carried out as in the following.

**[0376]** Fig. 4 is an illustration showing an example of the separation appearance in the mess cylinder 15 when a specific time was passed after vibration. In this test, separated each layer was designated as the foam layer (F layer) 16, the oil layer (O layer) 17, the cream layer (C layer) 18, the emulsion layer (E layer) 19 in the order closest from the stopper 14, and the volume of each layer was measured. The obtained results are presented in Tables 1 and 2. The shown values in Tables represent the volume of F layer-O layer-C layer-E layer, for example, "0-20-0-80" means that F layer is 0 ml, O layer is 20 ml, C layer is 0 ml and E layer is 80 ml. Furthermore, shorter time for the volume of C layer to become small or 0 ml means that the dissolubility is good.

(Base oil)

**[0377]**

Base oil 1: solvent refined paraffinic mineral oil (kinematic viscosity: 68 mm<sup>2</sup>/s (40°C), viscosity index: 95)

Base oil 2: 1-decene oligomer (kinematic viscosity: 68 mm<sup>2</sup>/s (40°C), viscosity index: 140)

Base oil 3: polyol ester obtained by using pentaerythritol and straight-chained and branched saturated fatty acids having 5 to 12 carbon atoms (kinematic viscosity: 68 mm<sup>2</sup>/s (40°C), viscosity index: 120)

(Additives)

Component (B)

**[0378]**

B1: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and morpholinoethyl methacrylate (10 mol%) (the average molecular weight: 80,000)

B2: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and benzoylamino methacrylate (10 mol%) (the average molecular weight: 70,000)

Component (C-1)

**[0379]**

C-1-1: N-oleoyl sarcosine

Component (C-2)

**[0380]**

C-2-1: nonyl phenoxyacetate (NPA)

Component (E)

**[0381]**

E1: 2,5-bis(n-nonyldithio)-1,3,4-thiadiazole

E2: sulfurized ester (the content of sulfur: 11.4%)

Other additives

**[0382]**

Amine: dioctyldiphenyl amine

DBPC: di-tert-butyl-p-cresol

TCP: tricresylphosphate

OAP: dioleoyl acid phosphate

OAc: oleic acid

PMA: alkylmethacrylate having 1 to 18 carbon atoms (the average molecular weight: 80,000)

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Imide: mono-type polybutenyl succinic acid imide obtained by reacting polybutenyl succinic anhydride with tetraethylene pentamine

[Effect concerning the embodiment of lubricating oil composition for slide guides - 2]

**[0383]** The lubricating oil composition for slide guides of this embodiment is satisfied in all the frictional property, the anti-stick-slip property and the dissolubility with water-soluble cutting fluid.

Table 1

Example	1	2	3	4	5
Base oil	99.4	99.4	99.39	99.37	99.19
1	-	-	-	-	-
2	-	-	-	-	-
3	-	-	-	-	-
B component	B1 0.3	B2 0.3	B1 0.3	B2 0.3	B1 0.3
C1 component	-	-	C-1-1 0.01	-	C-1-1 0.01
C-2 component	-	-	-	C-2-1 0.03	-
E component	-	-	-	-	E1 0.2
DBPC	0.2	0.2	0.2	0.2	0.2
Amine	0.1	0.1	0.1	0.1	0.1
Other additives	-	-	-	-	-
Frictional index	0.117	0.116	0.101	0.105	0.110
Anti-stick-slip property ( $\Delta\mu$ )	0.013	0.014	0.011	0.010	0.010
Dissolubility with cutting fluid	0-19-1-80	0-19-1-80	0-19-1-80	0-19-1-80	0-19-1-80

Table 2

Example	6	7	8	9
Base oil	99.35	99.05	-	-
1	-	-	99.05	-
2	-	-	-	99.05
3	-	-	-	-
Additives				
B component	B2 0.3	B2 0.3	B2 0.3	B2 0.3
C-1 component	C-1-1 0.02	C-1-1 0.02	C-1-1 0.02	C-1-1 0.02
C-2 component	C-2-1 0.03	C-2-1 0.03	C-2-1 0.03	C-2-1 0.03
E component	-	E1 0.3	E1 0.3	E1 0.3
DBPC	0.2	0.2	0.2	0.2
Amine	0.1	0.1	0.1	0.1
Other additives	-	-	-	-

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(continued)

Example	6	7	8	9
Frictional index	0.099	0.105	0.102	0.100
Anti-stick-slip property ( $\Delta\mu$ )	0.010	0.007	0.008	0.006
Dissolubility with cutting fluid	0-19-1-80	0-19-1-80	0-19-1-80	0-19-1-80

Table 3

Test Example	1	2	3	4	5
Base oil	99.2	99.2	98.7	98.7	98.7
1	-	-	-	-	-
2	-	-	-	-	-
3	-	-	-	-	-
Additives	-	-	-	-	-
B component	-	-	-	-	-
C-1 component	-	-	-	-	-
C-2 component	-	-	-	-	-
E component	-	-	-	E1 1.0	E2 1.0
DBPC	0.2	0.2	0.2	0.2	0.2
Amine	0.1	0.1	0.1	0.1	0.1
Other additives	TCP 0.5	OAP 0.5	OAc 1.0	-	-
Frictional index	0.145	0.139	0.150	0.161	0.159
Anti-stick-slip property ( $\Delta\mu$ )	>0.05	0.013	0.015	0.018	0.017
Dissolubility with cutting fluid	0-19-1-80	0-17-3-80	0-18-3-79	0-19-1-80	0-19-1-80

Table 4

Test Example	6	7
Base oil	99.4	99.4
1	-	-
2	-	-
3	-	-
Additives	-	-
B component	-	-
C-1 component	-	-
C-2 component	-	-
E component	-	-
DBPC	0.2	0.2
Amine	0.1	0.1
Other additives	PMA 0.3	Imide 0.3
Frictional index	0.163	0.170
Anti-stick-slip property ( $\Delta\mu$ )	>0.05	>0.05



(continued)

Test Example	6	7
Dissolubility with cutting fluid	0-19-1-80	0-10-12-78

### [Lubricating oil composition for slide guides - 3]

**[0384]** This Example is an embodiment related to the lubricating oil composition for slide guides formed by combining the phosphorus-containing carboxylic acid compound of the component (A-1), thiophosphoric acid ester (A-2), or mixtures thereof into the base oil. Hereinbelow, the embodiment will be further described by way of Examples.

**[0385]** Tested base oils, the kind of additives added thereto and their amounts are listed below. Further, the test methods carried out on each combined lubricating oil composition is also listed below. The obtained test results are presented in Tables 1 to 5.

### (Frictional property evaluation test)

**[0386]** Fig. 1 is a simplified block diagram illustrating the frictional index measuring system used in the frictional property evaluation test. In Fig. 1, a table 1 and a rolling jig 4 connected by introducing a load cell 5 is arranged on a bed 6, and a heavyweight 9 as a substituted of a working tool is further arranged on the table 1. The table 1 and the bed 6 are all made of cast iron. Moreover, the rolling jig 4 has a bearing part, where to an A/C servmotor 2 is connected by introducing a lead screw 3. By driving the lead screw 3 by the A/C servmotor 2, the rolling jig 4 can be reciprocated in the axial direction (the arrow direction in Fig. 1) of the lead screw 3. Further, the load cell 5 is connected with a computer 7 and the computer 7 and the A/C servmotor 2 are individually connected with a control board 8 electrically, thereby controlling the reciprocating movement of the rolling jig 4 and measuring the load between the table 1 and the rolling jig 4 can be carried out.

**[0387]** In such the frictional index measuring system, the lubricating oil composition is dropped on the surface of the bed 6, the surface pressure between the table 1 and the bed 6 is adjusted to 200 kPa by the selection of the table heavyweight 9, and then the rolling jig 4 is reciprocated with the screw speed of 60 mm/min and the screw length of 750 mm. At this time, the load between the table 1 and the rolling jig 4 is measured by the load cell 5 (the load meter), and based on the resulting measurement, the frictional index of the guide face (table 1/bed 6 = cast iron/cast iron) is determined. Furthermore, the test was carried out after carrying out the pre-conditioned interim operation for 3 times. The frictional index of each lubricating oil composition is presented in Tables 1 to 5.

### (Anti-stick-slip property evaluation test)

**[0388]** Fig. 2 is a simplified block diagram illustrating the anti-stick-slip property evaluation apparatus (TE-77 tester manufactured by PLINT & PARTNERS LTD.). The apparatus illustrated in Fig. 2 is an apparatus in which a lower test piece 12, an upper test piece 11 and an elastic body 10 are laminated on the supporting die 110 in this order, and the test pieces 11 and 12 are slid by reciprocally moving (sliding movement) the elastic body 10 along the surface of the supporting die 110 while imposing the test pieces 11 and 12 together with a specific load. During such sliding, the frictional index between the test pieces 11 and 12 can be determined by measuring the load added to the test pieces 11 and 12 using a load detector 13. Fig. 3 is a graph showing an example of the correlation between the frictional index obtained by the operation and time.  $\Delta\mu$  in Fig. 3 represents the amplitude of the frictional index.

**[0389]** Using such the apparatus,  $\Delta\mu$ , when each lubricating oil composition was introduced between the test pieces 11 and 12, was measured based on the method illustrated in the publication (Proceedings of Japanese Society of Tribologist Tribology conference, Tokyo 1999-5 D 17), except that the test pieces and the conditions were modified for the lubricating oil for slide guides evaluation. Specifically, the test was carried out using JIS G 4051 S45C as the test pieces 11, 12 and chloroprene rubber as the elastic body 10, and with the average sliding speed of 0.3 mm/s and under the load of 250 N. The obtained results are presented in Tables 1 to 5.

### (Dissolubility with water-soluble cutting fluid evaluation test)

**[0390]** 20 mL of the lubricative composition and 80 mL of the water-soluble cutting fluid (emulsion type cutting fluid, manufactured by Nippon Oil Corp., equivalent product of W1 Type 1 "cutting fluid" in JISK 2241, dilution ratio: 10-folds) were extracted in a 100 mL-mess cylinder. A stopper was put on the cylinder and the cylinder was vibrated for 1 minute at room temperature. Then, the dissolubility was evaluated by observing the separated appearance when 24 hours have been passed. The dissolubility evaluation is carried out as in the following.

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**[0391]** Fig. 4 is an illustration showing an example of the separation appearance in the mess cylinder 15 when a specific time was passed after vibration. In this test, separated each layer was designated as the foam layer (F layer) 16, the oil layer (O layer) 17, the cream layer (C layer) 18, the emulsion layer (E layer) 19 in the order closest from the stopper 14, and the volume of each layer was measured. The obtained results are presented in Tables 1 and 2. The shown values in Tables represent the volume of F layer-O layer-C layer-E layer, for example, "0-20-0-80" means that F layer is 0 ml, O layer is 20 ml, C layer is 0 ml and E layer is 80 ml. Furthermore, shorter time for the volume of C layer to become small or 0 ml means that the dissolubility is good.

(Base oil)

### **[0392]**

Base oil 1: solvent refined paraffinic mineral oil (kinematic viscosity: 68 mm<sup>2</sup>/s (40°C), viscosity index: 95)

Base oil 2: 1-decene oligomer (kinematic viscosity: 68 mm<sup>2</sup>/s (40°C), viscosity index: 140)

Base oil 3: polyol ester obtained by using pentaerythritol and straight-chained and branched saturated fatty acids having 5 to 12 carbon atoms (kinematic viscosity: 68 mm<sup>2</sup>/s (40°C), viscosity index: 120)

(Additives)

Component (A-1)

### **[0393]**

A-1-1:  $\beta$ -dithiophosphorylated propionic acid

A-1-2:  $\beta$ -dithiophosphorylated propionic acid ethyl ester

Component (A-2)

### **[0394]**

A-2-1: triphenyl phosphorothionate

Component (B)

### **[0395]**

B1: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and morpholinoethyl methacrylate (10 mol%) (the average molecular weight: 80,000)

B2: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and benzoylamino methacrylate (10 mol%) (the average molecular weight: 70,000)

Component (C-1)

### **[0396]**

C-1-1: N-oleoyl sarcosine

Component (C-2)

### **[0397]**

C-2-1: nonyl phenoxyacetate

Component (E)

### **[0398]**

E1: 2,5-bis(n-nonyldithio)-1,3,4-thiadiazole

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E2: sulfurized ester (the content of sulfur: 11.4%)

Other additives

5 **[0399]**

Amine: dioctyldiphenyl amine

DBPC: di-tert-butyl-p-cresol

TCP: tricresylphosphate

10 OAP: dioleoyl acid phosphate

OAc: oleic acid

[Effect concerning the embodiment of lubricating oil composition for slide guides - 3]

15 **[0400]** The lubricating oil composition for slide guides of this embodiment is excellent in both the frictional property and the anti-stick-slip property

Table 1

20	Example	1	2	3	4	5
	Base oil	1	99.68	99.5	99.5	99.4
		2	-	-	-	-
		3	-	-	-	-
25	Additives					
	A-1 Component	A-1-1 0.02	A-1-2 0.2	-	A-1-2 0.1	A-1-2 0.2
	A-2 Component	-	-	A-2-1 0.2	A-2-1 0.1	-
30	B component	-	-	-	-	B1 0.1
	C-1 component	-	-	-	-	-
35	C-2 component	-	-	-	-	-
	E component	-	-	-	-	-
	DBPC	0.2	0.2	0.2	0.2	0.2
	Amine	0.1	0.1	0.1	0.1	0.1
40	Other additives	-	-	-	-	-
	Frictional index	0.116	0.118	0.115	0.111	0.108
	Anti-stick-slip property ( $\Delta\mu$ )	0.018	0.017	0.017	0.015	0.015
45	Dissolubility with cutting fluid	0-17-4-79	0-17-4-79	0-18-3-79	0-18-3-79	0-19-1-80

Table 2

50	Example	6	7	8	9	10
	Base oil	1	99.48	99.25	99.57	99.27
		2	-	-	-	-
		3	-	-	-	-
55	Additives					
	A-1 Component	A-1-1 0.02	A-1-2 0.2	A-1-1 0.02	A-2-1 0.1	A-1-1 0.02

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(continued)

Example	6	7	8	9	10
A-2 Component	-	A-2-1 0.05	-	A-2-1 0.1	-
B component	B2 0.2	B1 0.2	B1 0.1	B2 0.2	B1 0.1
C-1 component	-	-	C-1-1 0.01	-	C-1-1 0.01
C-2 component	-	-	-	C-2-1 0.03	-
E component	-	-	-	-	E1 0.2
DBPC	0.2	0.2	0.2	0.2	0.2
Amine	0.1	0.1	0.1	0.1	0.1
Other additives	-	-	-	-	-
Frictional index	0.107	0.106	0.099	0.099	0.101
Anti-stick-slip property ( $\Delta\mu$ )	0.013	0.012	0.009	0.010	0.011
Dissolubility with cutting fluid	0-19-1-80	0-19-1-80	0-19-1-80	0-19-1-80	0-19-1-80

Table 3

Example	11	12	13	14	15
Base oil	1	2	3		
	99.25	98.95	-	-	-
	-	-	99.68	99.23	-
	-	-	-	-	99.5
Additives					
A-1 Component	A-1-2 0.1	A-1-2 0.1	A-1-1 0.02	A-1-1 0.02	A-1-2 0.1
A-2 Component	A-2-1 0.1	A-2-1 0.1	-	-	A-2-1 0.1
B component	B2 0.2	B2 0.2	-	B2 0.2	-
C-1 component	C-1-1 0.02	C-1-1 0.02	-	C-1-1 0.02	-
C-2 component	C-2-1 0.03	C-2-1 0.03	-	C-2-1 0.03	-
E component	-	E1 0.3	-	E1 0.3	-
DBPC	0.2	0.2	0.2	0.2	0.2
Amine	0.1	0.1	0.1	0.1	0.1
Other additives	-	-	-	-	-
Frictional index	0.092	0.095	0.114	0.093	0.113
Anti-stick-slip property ( $\Delta\mu$ )	<0.005	<0.05	0.017	<0.005	0.013
Dissolubility with cutting fluid	0-19-1-80	0-19-1-80	0-17-4-79	0-19-1-80	0-16-4-80

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Table 4

Example	16
Base oil	1
	2
	3
Additives	
A-1 Component	A-1-2 0.1
A-2 Component	A-2-1 0.1
B component	B2 0.2
C-1 component	0.02
C-2 component	C-2-1 0.03
E component	E1 0.3
DBPC	0.2
Amine	0.1
Other additives	
Frictional index	0.091
Anti-stick-slip property ( $\Delta\mu$ )	<0.005
Dissolubility with cutting fluid	0-19-1-80

Table 5

Test Example	1	2	3	4	5
Base oil	1	99.2	99.2	98.7	98.7
	2	-	-	-	-
	3	-	-	-	-
Additives					
A-1 Component	-	-	-	-	-
A-2 Component	-	-	-	-	-
B component	-	-	-	-	-
C-1 component	-	-	-	-	-
C-2 component	-	-	-	-	-
E component	-	-	-	E1 1.0	E2 1.0
DBPC	0.2	0.2	0.2	0.2	0.2
Amine	0.1	0.1	0.1	0.1	0.1
Other additives	TCP 0.5	OAP 0.5	OAc 1.0	-	-

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(continued)

Test Example	1	2	3	4	5
Frictional index	0.145	0.139	0.150	0.161	0.159
Anti-stick-slip property ( $\Delta\mu$ )	>0.05	0.013	0.015	0.018	0.017
Dissolubility with cutting fluid	0-19-1-80	0-17-3-80	0-18-3-79	0-19-1-80	0-19-1-80

## [Hydraulic oil - 1]

**[0401]** This Example is an embodiment related to the hydraulic oil formed by combining thiophosphoric ester of the component (A-2) and the dispersant viscosity index improver of the component (B) into the base oil. Hereinbelow, the embodiment will be further described by way of Examples.

**[0402]** In Examples 1 to 5 and Test Examples 1 to 8 below, the lubricating oil compositions having the formulation presented in Tables 1 to 3 were prepared using the base oils and additives listed below, respectively.

Base oil:

### [0403]

Base oil 1: high degree solvent refined paraffinic base oil (kinematic viscosity: 46.1 mm<sup>2</sup>/s at 40°C, viscosity index: 100)

Base oil 2: hydrocracked paraffinic base oil (kinematic viscosity: 46.1 mm<sup>2</sup>/s at 40°C, viscosity index: 125)

(A-2) thiophosphoric ester

### [0404]

A-2-1: triphenyl phosphorothionate

A-2-2: tri(butylphenyl) phosphorothionate

(B) dispersant viscosity index improver:

### [0405]

B1: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and morpholinoethyl methacrylate (10 mol%) (the average molecular weight: 80,000)

B2: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and benzoylamino methacrylate (10 mol%) (the average molecular weight: 70,000)

(C-1) compounds represented by the formulas (11) to (13)

### [0406]

C-1-1: N-oleoyl sarcosine

(C-2) compounds represented by the formula (14)

### [0407]

C-2-1: nonyl phenoxyacetate

(G-A) phenol-based antioxidant

### [0408]

G-A1: di-tert-butyl-p-cresol

(G-B): amine-based antioxidant

G-B 1: dioctyldiphenyl amine

Other additives

**[0409]**

E1: Zinc dioctyldithiophosphate

H1: tricresylphosphate

K1: monopolymer of alkylmethacrylate having 1 to 18 carbon atoms (the average molecular weight: 150,000)

K2: mono-type polybutenyl succinic acid imide obtained by reacting polybutenyl succinic anhydride with tetraethylene pentamine

K5: calcium dinonylnaphthalene sulfonate (50 wt% solvent, carrier oil: solvent-refined paraffinic mineral oil, the base value of the solvent: 170 mgKOH/g)

[Thermal stability test]

**[0410]** With respect to each lubricating oil composition of Examples 1 to 5 and Test Examples 1 to 8, based on "Testing Method of Lubricating Oil Thermal Stability" specified in JIS K2540, 50 ml of the test oil was extracted in a 50 ml-beaker, and a coil-shaped iron and copper catalysts were added thereto. The thermal stability test was carried out in an air constant-temperature bath at 140°C for a certain period (10 days, 20 days). The test oil after the test was filtered through a filter, and the amount of sludge in the test oil was measured. The obtained results are presented in Tables 1 to 3.

[SRV (microreciprocating friction) test]

**[0411]** With respect to each lubricating oil composition of Examples 1 to 5 and Test Examples 1 to 8, SRV test was carried out and the frictional property was evaluated. More specifically, as shown in Fig. 5, to the point contact area of a disc 1 and a ball 2 arranged on the upper surface of the disc, the test oil was coated. While adding load in the vertically downward direction (the arrow A in Fig. 5) to the ball 2, the ball 2 was reciprocated relative to the linear direction (the arrow B in Fig. 5) on the upper surface of the disc 1. The frictional index at this time was measured by a load cell (not shown) mounted on a disc 1 supportor (not shown). The disc 1 of SPCC product with the diameter of 25 mm and the thickness of 8 mm was used. Further, the ball 2 of SPCC product with the diameter of 10 mm was used. Furthermore, the load added to the ball 2 was 1,200 N, the amplitude of the ball 2 was 1 mm, the frequency was 50Hz and the temperature was 80°C. The obtained results are presented in Tables 1 to 3.

[Antiemulsifying property test]

**[0412]** With respect to each lubricating oil composition of Examples 1 to 5 and Test Examples 1 to 8, based on "Testing Method of Petroleum Product-Lubricating oil-Antiemulsifying Property" specified in JIS K2520, the antiemulsifying property was evaluated at 54°C. The obtained results are presented in Tables 1 to 3. In addition, the values in Tables means oil layer (ml)-aqueous layer (ml)-emulsifying layer (ml) (time elapsed (min)).

[Abrasion resistance test]

**[0413]** With respect to each lubricating oil composition of Examples 1 to 5 and Test Examples 1 to 8, the vane pump test specified in ASTM D 2882 was performed. The weight of the vane and the ring before and after the test was calculated, and the abrasion amount was measured. The testing time was 100 hours. The obtained results are presented in Tables 1 to 3.

[Effect concerning the embodiment of hydraulic oil - 1]

**[0414]** The lubricating oil composition of this embodiment may achieve all the sludge resistance, the abrasion resistance and the frictional property in good balance with high degree, and further excellent in the water dissolubility. Therefore, the lubricating oil composition useful for improving credibility of hydraulic systems and achieving energy-saving from using the lubricating oil of this embodiment as a hydraulic oil was provided.

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Table 1

Example	1	2	3	4	5
Base oil 1	98.90	-	-	-	-
2	-	98.70	98.50	98.55	98.45
A-2-1	0.30	0.20	0.30	-	-
A-2-2	-	-	-	0.30	0.30
B1	0.30	0.30	-	-	-
B2	-	-	0.30	0.30	0.30
C-1-1	-	-	-	0.05	0.05
C-2-1	-	-	0.10	-	0.10
C-A-1	0.50	0.50	0.50	0.50	0.50
C-B-1	-	0.30	0.30	0.30	0.30
Amount of sludge (mg/45g) in the thermal stability test	10 days	1.8	1.3	2.8	1.3
	20 days	7.2	8.5	7.8	5.3
SRV (frictional index)	0.118	0.119	0.118	0.113	0.110
Antiemusifying property	40-40-0 (5)	40-40-0 (10)	40-40-0 (10)	40-40-0 (10)	40-40-0 (10)
Abrasion resistance (abrasion amount [mg])	10.5	11.3	12.4	10.8	9.8

Table 2

	Test Example 1	Test Example 2	Test Example 3	Test Example 4
Base oil 1	98.90	-	-	-
2	-	98.10	98.60	98.60
A-2-1	0.30	-	-	-
A-2-2	-	0.30	0.30	0.30
B1	-	0.30	-	-
C-A-1	0.50	0.50	0.50	0.50
C-B-1	0.30	0.30	0.30	0.30
E1	-	0.50	-	-
H1	-	-	-	0.30
K1	-	-	0.30	-
K2	-	-	-	-
Amount of sludge (mg/45g) in the thermal stability test	10 days	10.3	11.8	7.7
	20 days	28.7	35.2	12.4
SRV (frictional index)	0.120	0.132	0.122	0.120
Antiemusifying property	40-40-0 (5)	40-40-0 (20)	15-20-45 (80)	20-26-34 (80)
Abrasion resistance (abrasion amount [mg])	9.5	14.3	15.8	10.2



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Table 3

		Test Example 5	Test Example 6	Test Example 7	Test Example 8
5	Base oil 1	-	-	-	-
	2	98.60	98.30	98.60	98.30
10	A-2-1	-	-	-	
	A-2-2	0.30	0.30	0.30	0.30
	B1	-	-	-	-
15	C-A-1	0.50	0.50	0.50	0.50
	C-B-1	0.30	0.30	0.30	0.30
	E1	-	-	-	-
20	H1	-	0.30	-	0.30
	K1	0.30	0.30	-	-
	K2	-	-	0.30	0.30
25	Amount of sludge (mg/45g) in the thermal stability test	10 days	5.8	1.3	13.5
		20 days	11.2	12.7	32.4
25	SRV (frictional index)	0.123	0.122	0.124	0.124
	Antiemusifying property	20-28-34 (60)	19-28-33 (60)	24-23-53 (60)	22-18-38 (80)
	Abrasion resistance (abrasion amount [mg])	13.5	12.7	15.7	14.3

[Examples 6 to 8]

**[0415]** In Examples 6 to 8, the lubricating oil compositions having the formulation presented in Table 4 were prepared using the base oils and additives above, and the ester oiliness improver listed below, respectively.

(D) Ester oiliness improver:

**[0416]**

D1: ester of sorbitan and oleic acid (sorbitan monooleate 25 mol%, sorbitan dioleate 40 mol%, sorbitan trioleate 30 mol% and sorbitan tetraoleate 5 mol%)

D2: triester of glycerin and a mixture fatty acids of acetic acid and fatty acid having 16 to 20 carbon atoms (fatty acid composition: acetic acid 66 mol%, straight-chained saturated fatty acid having 16 to 20 carbon atoms 30 mol% and straight-chained unsaturated fatty acid having 18 carbon atoms 4 mol%)

D3: ester of glycerin and branched saturated fatty acid having 16 to 20 carbon atoms (glycerin monoester 25 mol%, glycerin diester 75 mol% and glycerin triester 0 mol%)

**[0417]** Next, with respect to each lubricating oil composition of Examples 6 to 8, the thermal stability test, the SRV (microreciprocating friction) test and the abrasion resistance test was carried out in the same manner as above. The obtained results are presented in Table 4.

Table 4

Example	6	7	8
Base oil 1	98.50	-	-
2	-	98.50	98.50
A-2-1	0.30	0.30	0.30

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(continued)

Example		6	7	8
A-2-2		-	-	-
B1		-	-	-
B2		0.30	0.30	0.30
C-1-1		-	-	-
C-2-1		0.10	0.10	0.10
D1		0.30	-	
D2		-	0.30	
D3		-	-	0.30
G1		0.50	0.50	0.50
H1		-	-	-
Amount of sludge (mg/45g) in the thermal stability test	10 days	1.8	1.3	2.4
	20 days	7.2	8.5	7.8
SRV (frictional index)		0.112	0.108	0.109
Abrasion resistance (abrasion amount [mg])		9.5	8.2	10.2

## [Hydraulic oil - 2]

**[0418]** This Example is an embodiment related to the hydraulic oil formed by combining phosphorus-containing carboxylic acid of the component (A-1) and the dispersant viscosity index improver of the component (B) into the base oil. Hereinbelow, the embodiment will be further described by way of Examples.

**[0419]** In Examples 1 to 5 and Test Examples 1 to 9 below, the lubricating oil compositions having the formulation presented in Tables 1 to 3 were prepared using the base oils and additives listed below, respectively.

Base oil:

### [0420]

Base oil 1: high degree solvent refined paraffinic base oil (kinematic viscosity: 46.1 mm<sup>2</sup>/s at 40°C, viscosity index: 100)

Base oil 2: hydrocracked paraffinic base oil (kinematic viscosity: 46.1 mm<sup>2</sup>/s at 40°C, viscosity index: 125)

Component (A-1)

### [0421]

A-1-1: β-dithiophosphorylated propionic acid

A-1-2: β-dithiophosphorylated propionic acid ethyl ester

(B) dispersant viscosity index improver:

### [0422]

B1: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and morpholinoethyl methacrylate (10 mol%) (the average molecular weight: 80,000)

B2: copolymer of alkyl methacrylate having 1 to 18 carbon atoms (90 mol%) and benzoylamino methacrylate (10 mol%) (the average molecular weight: 70,000)

(A-2) thiophosphoric ester

**[0423]**

5 A-2-1: triphenylphosphorothionate

(C-1) compounds represented by the formulas (17) to (19)

**[0424]**

10

C-1-1: N-oleoyl sarcosine

(C-2) compounds represented by the formula (20)

15

**[0425]**

C-2-1: nonyl phenoxyacetate

(G-A) phenol-based antioxidant

20

**[0426]**

G-A-1: di-tert-butyl-p-cresol

25

(G-B): amine-based antioxidant

**[0427]**

30

G-B-1: dioctyldiphenyl amine  
E1: Zinc dioctyldithiophosphate  
H1: tricresylphosphate

Other additives:

35

**[0428]**

K1 : monopolymer of alkylmethacrylate having 1 to 18 carbon atoms (the average molecular weight: 150,000)  
K2: mono-type polybutenyl succinic acid imide obtained by reacting polybutenyl succinic anhydride with tetraethylene pentamine  
40 K3: calcium dinonylnaphthalene sulfonate (50 wt% solvent, carrier oil: solvent-refined paraffinic mineral oil, the base value of the solvent: 170 mgKOH/g)

[Thermal stability test]

45

**[0429]** With respect to each lubricating oil composition of Examples 1 to 5 and Test Examples 1 to 9, based on "Testing Method of Lubricating Oil Thermal Stability" specified in JIS K2540, 50 ml of the test oil was extracted in a 50 ml-beaker, and coil-shaped iron and copper catalysts were added thereto. The thermal stability test was carried out in an air constant-temperature bath at 140°C for a certain period (10 days, 20 days). The test oil after the test was filtered through a filter, and the amount of sludge in the test oil was measured. The obtained results are presented in Tables 1 to 3.

50

[SRV (microreciprocating friction) test]

55

**[0430]** With respect to each lubricating oil composition of Examples 1 to 5 and Test Examples 1 to 9, SRV test was carried out and the frictional property was evaluated. More specifically, as shown in Fig. 5, to the point contact area of a disc 1 and a ball 2 arranged on the upper surface of the disc, the test oil was coated. While adding load in the vertically downward direction (the arrow A in Fig. 5) to the ball 2, the ball 2 was reciprocated relative to the linear direction (the arrow B in Fig. 5) on the upper surface of the disc 1. The frictional index at this time was measured by a load cell (not shown) mounted on a disc 1 supportor (not shown). The disc 1 of SPCC product with the diameter of 25 mm and the

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thickness of 8 mm was used. Further, the ball 2 of SPCC product with the diameter of 10 mm was used. Furthermore, the load added to the ball 2 was 1,200 N, the amplitude of the ball 2 was 1 mm, the frequency was 50Hz and the temperature was 80°C. The obtained results are presented in Tables 1 to 3.

[Antiemulsifying property test]

**[0431]** With respect to each lubricating oil composition of Examples 1 to 5 and Test Examples 1 to 8, based on "Testing Method of Petroleum Product-Lubricating oil-Antiemulsifying Property" specified in JIS K2520, the antiemulsifying property was evaluated at 54°C. The obtained results are presented in Tables 1 to 3. In addition, the values in Tables means oil layer (ml)-aqueous layer (ml)-emulsifying layer (ml) (time elapsed (min)).

[Abrasion resistance test]

**[0432]** With respect to each lubricating oil composition of Examples 1 to 5 and Test Examples 1 to 9, the vane pump test specified in ASTM D 2882 was performed. The weight of the vane and the ring before and after the test was calculated, and the abrasion amount was measured. The testing time was 100 hours. The obtained results are presented in Tables 1 to 3.

[Effect concerning the embodiment of hydraulic oil - 2]

**[0433]** The lubricating oil composition of this embodiment may achieve all the sludge resistance, the abrasion resistance and the frictional property in good balance with high degree, and further excellent in the water dissolubility. Therefore, the lubricating oil composition useful for improving credibility of hydraulic systems and achieving energy-saving from using the lubricating oil of this embodiment as a hydraulic oil was provided.

Table 1

Example		1	2	3	4	5
Base oil	1	98.85	-	-	-	-
	2	-	98.75	98.50	98.55	98.35
A-1-1		0.05	0.05	-		
A-1-2		-	-	0.30	0.30	0.30
B1		0.30	0.30	-	-	-
B2		-	-	0.30	0.30	0.30
A-2-1		-	0.10	-	-	0.10
C-1-1		-	-	-	0.05	0.05
C-2-1		-	-	0.10	-	0.10
G-A-1		0.50	0.50	0.50	0.50	0.50
G-B-1		0.30	0.30	0.30	0.30	0.30
Amount of sludge (mg/45g) in the thermal stability test	10 days	1.8	1.3	2.3	2.7	1.3
	20 days	7.2	5.3	6.8	6.2	6.5
SRV (frictional index)		0.116	0.119	0.115	0.113	0.110
Antiemulsifying property		40-40-0 (5)	40-40-0 (10)	40-40-0 (10)	40-40-0 (10)	40-40-0 (10)
Abrasion resistance (abrasion amount [mg])		13.5	8.3	15.8	13.4	12.2

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Table 2

Test Example	1	2	3	4	5
Base oil	98.85	-	-	-	-
1	-	98.40	98.60	98.60	98.9
2					
A-1-1	0.05	-	-	-	
A-1-2	-	-	-	0.30	0.30
B 1	-	0.30	-	-	-
B2	-	-	0.30	-	-
A-2-1	-	-	-	-	-
G-A-1	0.50	0.50	0.50	0.50	0.50
G-B-1	0.30	0.30	0.30	0.30	0.30
E1	-	0.50	-	-	-
H2	-	-	0.30	-	-
K1	0.30	-	-	-	-
K2	-	-	-	0.30	-
K3	-	-	-	-	-
Amount of sludge (mg/45g) in the thermal stability test	10 days	22.3	15.2	3.3	12.3
	20 days	48.7	48.3	12.5	8.6
SRV (frictional index)	0.127	0.132	0.124	0.122	0.120
Antiemusifying property	40-40-0 (10)	40-40-0 (20)	40-40-0 (20)	14-20-46 (80)	40-40-0 (15)
Abrasion resistance (abrasion amount [mg])	17.5	12.3	22.4	18.5	13.5

Table 3

Test Example	6	7	8	9
Base oil	-	98.90	-	-
1	98.30	-	98.60	98.30
2				
A-1-1	-	-	-	-
A-1-2	0.30	-	0.30	0.30
B1	-	-	-	-
B2	-	0.30	-	-
A-2-1	-	-	-	-
G-A-1	0.50	0.50	0.50	0.50
G-B-1	0.30	0.30	0.30	0.30
E1	-	-	-	-
H1	-	-	-	-
K1	0.30	-	-	0.30
K2	0.30	-	-	-
K3	-	-	0.30	0.30

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(continued)

Test Example		6	7	8	9
Amount of sludge (mg/45g) in the thermal stability test	10 days	4.4	2.2	13.6	14.5
	20 days	13.2	5.9	33.8	35.4
SRV (frictional index)		0.122	0.120	0.128	0.127
Antiemusifying property		18-25-37 (60)	40-40-0 (10)	32-27-18 (60)	31-26-23 (60)
Abrasion resistance (abrasion amount [mg])		18.7	135.8	19.3	18.4

[Examples 6 to 8]

**[0434]** In Examples 6 to 8, the lubricating oil compositions having the formulation presented in Table 4 were prepared using the base oils and additives above, and the ester oiliness improver listed below, respectively.

(D) Ester oiliness improver:

**[0435]**

D1: ester of sorbitan and oleic acid (sorbitan monooleate 25 mol%, sorbitan dioleate 40 mol%, sorbitan trioleate 30 mol% and sorbitan tetraoleate 5 mol%)

D2: triester of glycerin and a mixture fatty acids of acetic acid and fatty acid having 16 to 20 carbon atoms (fatty acid composition: acetic acid 66 mol%, straight-chained saturated fatty acid having 16 to 20 carbon atoms 30 mol% and straight-chained unsaturated fatty acid having 18 carbon atoms 4 mol%)

D3: ester of glycerin and branched saturated fatty acid having 16 to 20 carbon atoms (glycerin monoester 25 mol%, glycerin diester 75 mol% and glycerin triester 0 mol%)

**[0436]** Next, in the same manner as the above examples, the tests were carried out. The obtained results are presented in Table 4.

Table 4

Example	6	7	8
Base oil	1	-	-
	2	98.45	98.45
A-1-1	0.05	0.05	0.05
A-1-2	-	-	-
B1	0.30	0.30	0.30
B2	-	-	-
A-2-1	-	-	-
C-1-1	-	-	-
C-2-1	0.10	0.10	0.10
D1	0.30	-	-
D2	-	0.30	-
D3	-	-	0.30
G-A-1	0.50	0.50	0.50
G-B-1	0.30	0.30	0.30
Amount of sludge (mg/45g) in the thermal stability test	10 days	1.4	1.2
	20 days	5.2	4.3

(continued)

Example	6	7	8
SRV (frictional index)	0.113	0.108	0.110
Abrasion resistance (abrasion amount [mg])	12.5	8.1	13.7

[Hydraulic oil - 3]

**[0437]** This Example is an embodiment related to the hydraulic oil formed by combining ester oiliness improver of the component (D) into the base oil. Hereinbelow, the embodiment will be further described by way of Examples.

**[0438]** In Examples 1 to 4 and Test Examples 1 to 4 below, the lubricating oil compositions having the formulation presented in Tables 1 and 2 were prepared using the base oils and additives listed below, respectively.

Base oil:

**[0439]**

Base oil 1: high degree solvent refined paraffinic base oil (kinematic viscosity: 46.1 mm<sup>2</sup>/s at 40°C, viscosity index: 100)

Base oil 2: hydrocracked paraffinic base oil (kinematic viscosity: 46.1 mm<sup>2</sup>/s at 40°C, viscosity index: 125)

(D) polyhydric alcohol ester

**[0440]**

D1: triester of glycerin and a mixture fatty acid of acetic acid, n-octadecanoic acid and oleic acid (fatty acid composition: acetic acid 66.6 mol%, n-octadecanoic acid 30.0 mol% and oleic acid 3.4 mol%)

D2: triester of glycerin and a mixture fatty acid of acetic acid, n-octadecanoic acid and oleic acid (fatty acid composition: acetic acid 67.0 mol%, n-octadecanoic acid 28.0 mol% and oleic acid 5.0 mol%)

D3: triester of glycerin and a mixture fatty acid of acetic acid, n-hexadecanoic acid and oleic acid (fatty acid composition: acetic acid 66.7 mol%, n-hexadecanoic acid 30.1 mol% and oleic acid 3.2 mol%)

(I) other ester oiliness improver

**[0441]**

I1: monoester of glycerin and n-octadecanoic acid

I2: monoester of glycerin and oleic acid

(A-1) phosphorous-containing carboxylic acid compound

**[0442]**

A-1-1:  $\beta$ -dithiophosphorylated propionic acid

(B) dispersant viscosity index improver:

**[0443]**

(H) phosphoric ester

H1: tricresylphosphate

(A-2) thiophosphoric ester

**[0444]**

A-2-1: triphenylphosphorothionate

(G-A) phenol-based antioxidant

**[0445]**

G-A-1: 2,6-di-tert-butyl-p-cresol

(G-B): amine-based antioxidant

G-B-1: dioctyldiphenyl amine

(K) dispersant pour-point depressant

K1: mono-type polybutenyl succinic acid imide obtained by reacting polybutenyl succinic anhydride with tetraethylene pentamine

[Thermal stability test]

**[0446]** With respect to each lubricating oil composition of Examples 1 to 4 and Test Examples 1 to 4, based on "Testing Method of Lubricating Oil Thermal Stability" specified in JIS K2540, 50 ml of the test oil was extracted in a 50 ml-beaker, and coil-shaped iron and copper catalysts were added thereto. The thermal stability test was carried out in an air constant-temperature bath at 140°C for a certain period (10 days, 20 days). The test oil after the test was filtered through a filter, and the amount of sludge in the test oil was measured. The obtained results are presented in Tables 1 and 2.

[SRV (microreciprocating friction) test]

**[0447]** With respect to each lubricating oil composition of Examples 1 to 4 and Test Examples 1 to 4, SRV test was carried out and the frictional property was evaluated. More specifically, as shown in Fig. 5, to the point contact area of a disc 1 and a ball 2 arranged on the upper surface of the disc, the test oil was coated. While adding load in the vertically downward direction (the arrow A in Fig. 5) to the ball 2, the ball 2 was reciprocated relative to the linear direction (the arrow B in Fig. 5) on the upper surface of the disc 1. The frictional index at this time was measured by a load cell (not shown) mounted on a disc 1 supportor (not shown). The disc 1 of SUJ2 product with the diameter of 25 mm and the thickness of 8 mm was used. Further, the ball 2 of SUJ2 product with the diameter of 10 mm was used. Furthermore, the load added to the ball 2 was 1,200 N, the amplitude of the ball 2 was 1 mm, the frequency was 50Hz and the temperature was 80°C. The obtained results are presented in Tables 1 and 2.

[Abrasion resistance test]

**[0448]** With respect to each lubricating oil composition of Examples 1 to 4 and Test Examples 1 to 4, the vane pump test specified in ASTM D 2882 was performed. The weight of the vane and the ring before and after the test was calculated, and the abrasion amount was measured. The testing time was 100 hours. The obtained results are presented in Tables 1 and 2.

[Low temperature storage stability test]

**[0449]** 100 ml of each lubricating oil composition of Examples 1 to 4 and Test Examples 1 to 4 were fed to respective 100 ml-all glass containers. The glass containers were sealed and stored in an ice chamber at 0°C. The appearance of the lubricating oil compositions after 60 days was observed, and the precipitation was observed. The obtained results are presented in Tables 1 and 2.

[Effect concerning the embodiment of hydraulic oil - 3]

**[0450]** The lubricating oil composition of this embodiment may achieve all the sludge resistance, the abrasion resistance and the precipitation resistance in good balance with high degree. Therefore, the lubricating oil composition useful for improving credibility of hydraulic systems and achieving energy-saving from using the lubricating oil of this embodiment as a hydraulic oil was provided.

Table 1

Example	1	2	3	4
Base oil	97.80	-	97.90	97.67
	-	98.30	-	-



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(continued)

Example	1	2	3	4
D1	0.20		-	-
D2	-	0.20	-	-
D3	-	-	0.30	0.30
A-1-1	-	-	-	0.03
H1	1.00	-	0.60	1.00
A-2-1	-	0.50	0.20	-
G-A-1	0.50	0.50	0.50	0.50
G-B-1	0.20	0.20	0.20	0.20
K1	0.30	0.30	0.30	0.30
Amount of sludge (mg/45g) in the thermal stability test	10 days	1.8	1.3	0.110
	20 days	5.2	6.5	8.4
SRV (frictional index)	0.108	0.108	0.110	0.106
Abrasion resistance (abrasion amount[mg])	10.2	9.3	8.4	4.8
Low temperature storage stability (precipitation)	None	None	None	None

Table 2

Example	1	2	3	4
Base oil	1	97.70	-	-
	2	-	98.20	97.70
1-1	0.30	0.30	-	-
I-2	-	-	0.30	0.30
A-1-1	-	-	-	-
H1	1.00	-	0.80	1.00
A-2-1	-	0.50	0.20	-
G-A-1	0.50	0.50	0.50	0.50
G-B-1	0.20	0.20	0.20	0.20
K1	0.30	0.30	0.30	0.30
Amount of sludge (mg/45g) in the thermal stability test	10 days	5.8	5.3	4.8
	20 days	13.2	16.5	19.7
SRV (frictional index)	0.102	0.103	0.106	0.109
Abrasion resistance (abrasion amount [mg])	17.8	19.3	12.4	13.7
Low temperature storage stability (precipitation)	Precipitated	Precipitated	None	None

## Industrial Applicability

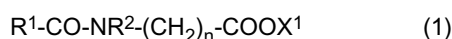
**[0451]** The composition of the present invention is used for lubricative composition for industrial machinery and equipment, which comprises a base oil selected from mineral oils, fats and oils, synthetic oils and mixtures of two or more of them, and at least one additive selected from the component (A), the component (B) or the component (C). Such lubricative composition for industrial machinery and equipment is used as a gear oil composition, a lubricating oil composition for paper machines or a lubricating oil composition for slide guides.

**[0452]** Further, the lubricative composition for industrial machinery and equipment comprises a base oil selected from mineral oils, fats and oils, synthetic oils and mixtures of two or more of them, and is combined therewith (A-1) a phosphorus-containing carboxylic acid compound and/or (A-2) a thiophosphoric ester as the component (A), and the dispersant viscosity index improver of the component (B), or instead combined therewith an ester oiliness improver as the component (D). Such lubricative composition for industrial machinery and equipment is used as a lubricating oil, and particularly a hydraulic oil. component (A):

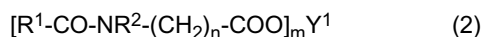
(A-1) a phosphorus-containing carboxylic acid and/or (A-2) a thiophosphoric ester;  
component (B): a dispersant viscosity index improver;  
component (C):

the following component (C-1) and/or component (C-2):

component (C-1): at least one kind of a compound represented by the following formulas (1) to (3):



wherein  $R^1$  is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms,  $R^2$  is an alkyl group having 1 to 4 carbon atoms,  $X^1$  is hydrogen, an alkyl group having 1 to 30 carbon atoms or an alkenyl group having 1 to 30 carbon atoms, and  $n$  is an integer of 1 to 4,

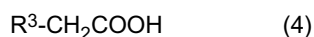


wherein  $R^1$  is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms,  $R^2$  is an alkyl group having 1 to 4 carbon atoms,  $Y^1$  is an alkali metal or an alkali earth metal,  $n$  is an integer of 1 to 4, and  $m$  is 1 when  $Y^1$  is an alkali metal and 2 when  $Y^1$  is an alkali earth metal, and

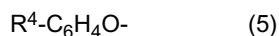


wherein  $R^1$  is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms,  $R^2$  is an alkyl group having 1 to 4 carbon atoms,  $Z$  is a residue having a hydroxyl group removed from a polyhydric alcohol with two or more valences,  $m$  is an integer of 1 or more,  $m'$  is an integer of 0 or more,  $m + m'$  is a valence number of  $Z$ , and  $n$  is an integer of 1 to 4,

component (C-2): a compound represented by the following formula (4):



wherein  $R^3$  is an alkyl group having 7 to 29 carbon atoms, an alkenyl group having 7 to 29 carbon atoms or a group represented by the formula (5):



wherein  $R^4$  is an alkyl group having 1 to 20 carbon atoms or hydrogen; and component (D): an ester oiliness improver.

## Claims

1. A lubricative composition for industrial machinery and equipment, said composition comprising a base oil selected from mineral oils, fats and oils, synthetic oils and mixtures of two or more of them, and at least one additive selected from the following components (A) to (D):

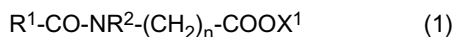
component (A):

(A-1) a phosphorus-containing carboxylic acid and/or (A-2) a thiophosphoric ester;

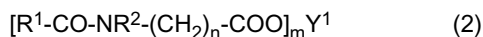
component (B): a dispersant viscosity index improver;  
component (C):

the following component (C-1) and/or component (C-2):

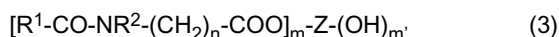
component (C-1): at least one kind of a compound represented by the following formulas (1) to (3):



wherein R<sup>1</sup> is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms, R<sup>2</sup> is an alkyl group having 1 to 4 carbon atoms, X<sup>1</sup> is hydrogen, an alkyl group having 1 to 30 carbon atoms or an alkenyl group having 1 to 30 carbon atoms, and n is an integer of 1 to 4,

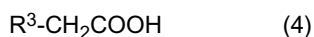


wherein R<sup>1</sup> is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms, R<sup>2</sup> is an alkyl group having 1 to 4 carbon atoms, Y<sup>1</sup> is an alkali metal or an alkali earth metal, n is an integer of 1 to 4, and m is 1 when Y<sup>1</sup> is an alkali metal and 2 when Y<sup>1</sup> is an alkali earth metal, and

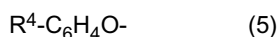


wherein R<sup>1</sup> is an alkyl group having 6 to 30 carbon atoms or an alkenyl group having 6 to 30 carbon atoms, R<sup>2</sup> is an alkyl group having 1 to 4 carbon atoms, Z is a residue having a hydroxyl group removed from a polyhydric alcohol with two or more valences, m is an integer of 1 or more, m' is an integer of 0 or more, m + m' is a valence number of Z, and n is an integer of 1 to 4,

component (C-2): a compound represented by the following formula (4):



wherein R<sup>3</sup> is an alkyl group having 7 to 29 carbon atoms, an alkenyl group having 7 to 29 carbon atoms or a group represented by the formula (5):



wherein R<sup>4</sup> is an alkyl group having 1 to 20 carbon atoms or hydrogen; and component (D): an ester oiliness improver.

2. The lubricative composition according to claim 1 as a gear oil composition, wherein the additive is at least one selected from the component (A) to component (C).
3. The lubricative composition according to claim 1 as a lubricating oil composition for paper machines, wherein the additive is at least one selected from the component (A) to component (C).
4. The lubricative composition according to claim 1 as a lubricating oil composition for slide guides, wherein the additive is at least one selected from the component (A) to component (C).
5. The lubricative composition according to claim 1 as a lubricating oil composition, wherein the additive comprises the phosphorus-containing carboxylic acid compound of the component (A-1) and the dispersant viscosity index improver of the component (B).
6. The lubricative composition according to claim 1 as a lubricating oil composition, wherein the additive comprises the thiophosphoric ester of the component (A-2) and the dispersant viscosity index improver of the component (B).
7. The lubricative composition according to claim 1 as a lubricating oil composition, wherein the additive comprises the ester oiliness improver of the component (D) which is an ester of a polyhydric alcohol and a fatty acid of monobasic acids.
8. The lubricative composition according to claim 7, wherein the ester oiliness improver of the component (D) which is an ester of a polyhydric alcohol and a fatty acid of monobasic acids is any one selected from the following esters of (D-1) to (D-3):

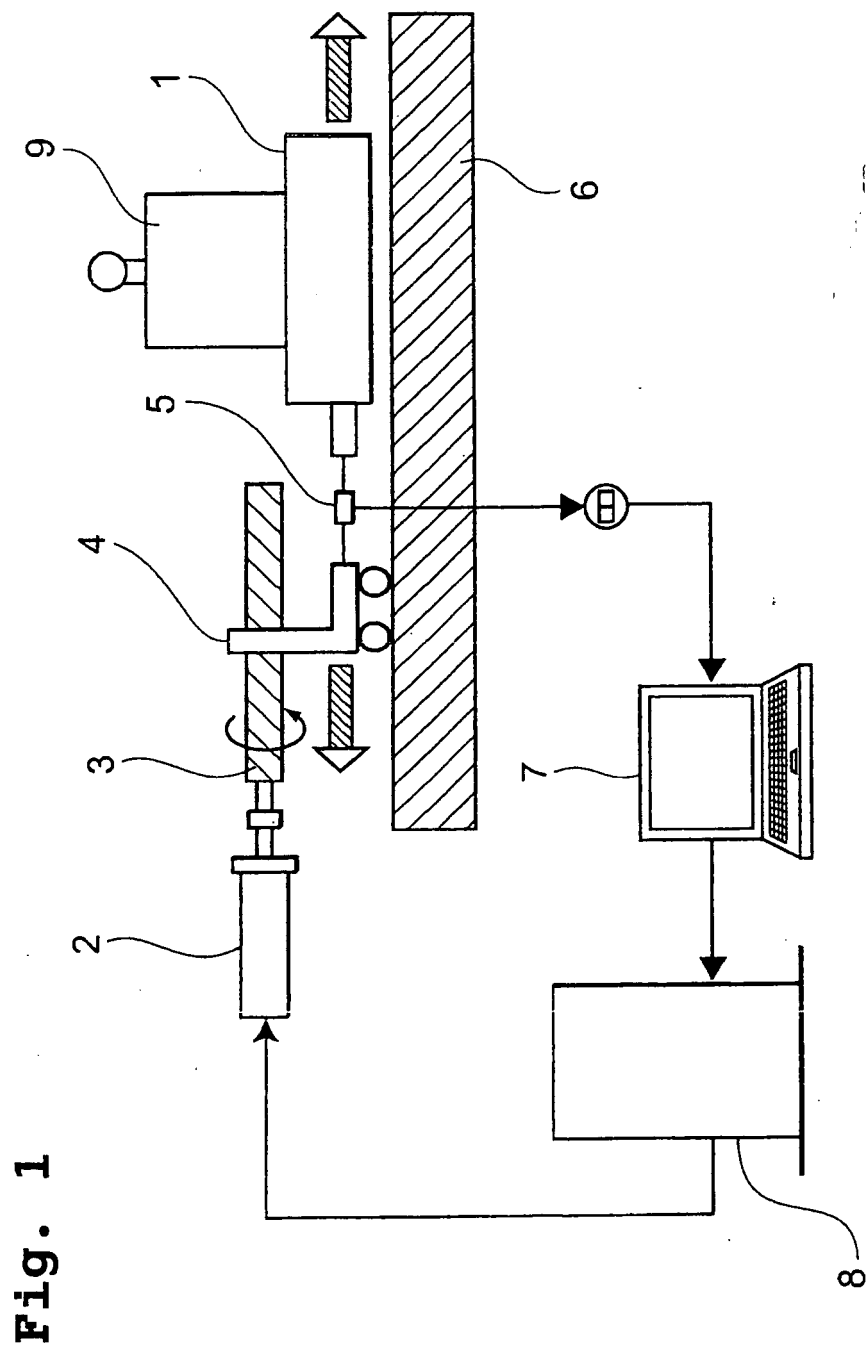
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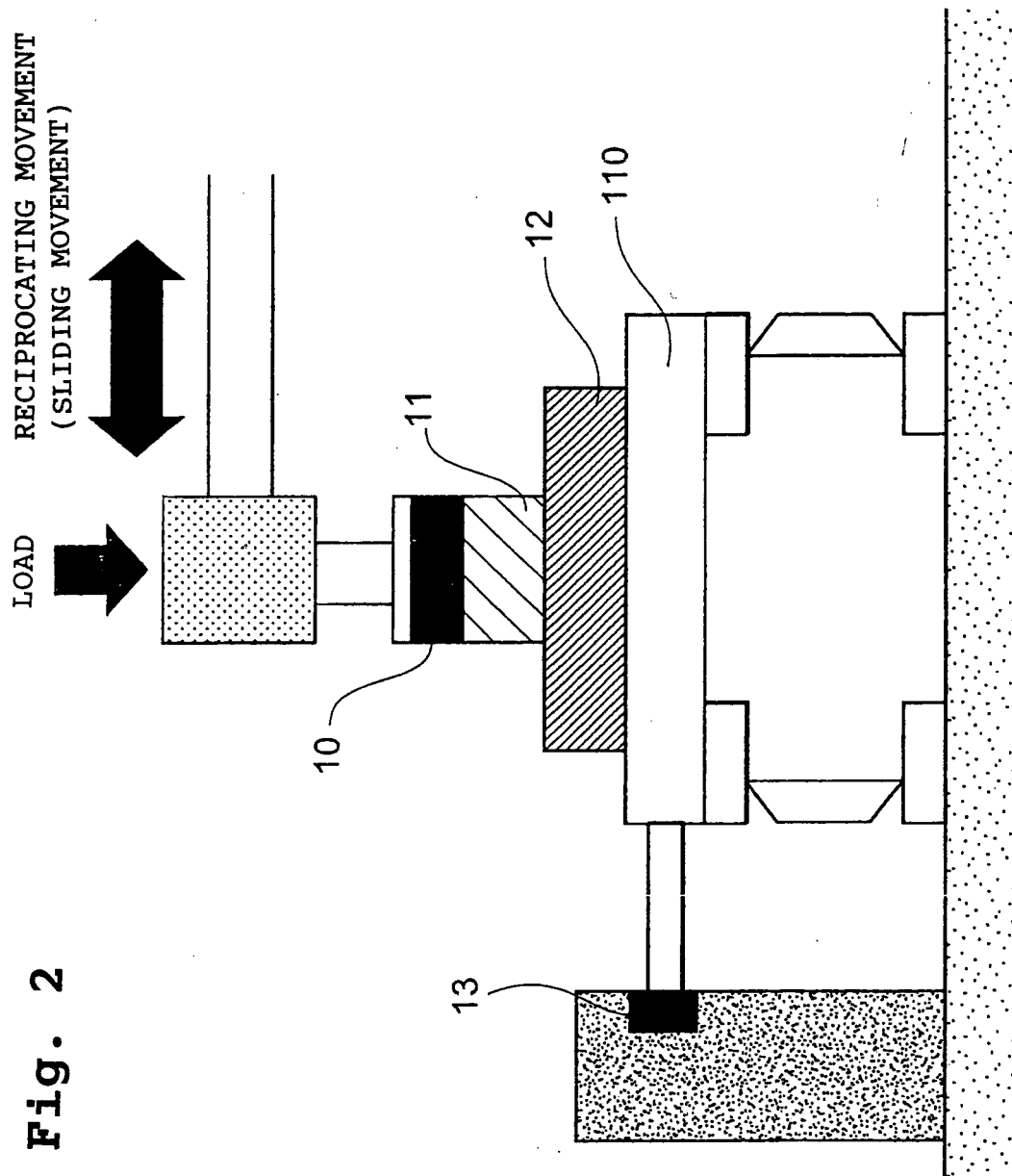
(D-1): an ester of a polyhydric alcohol and an unsaturated fatty acid containing a partial ester with the degree of esterification of 1 and a partial ester with the degree of esterification of 2 or more;

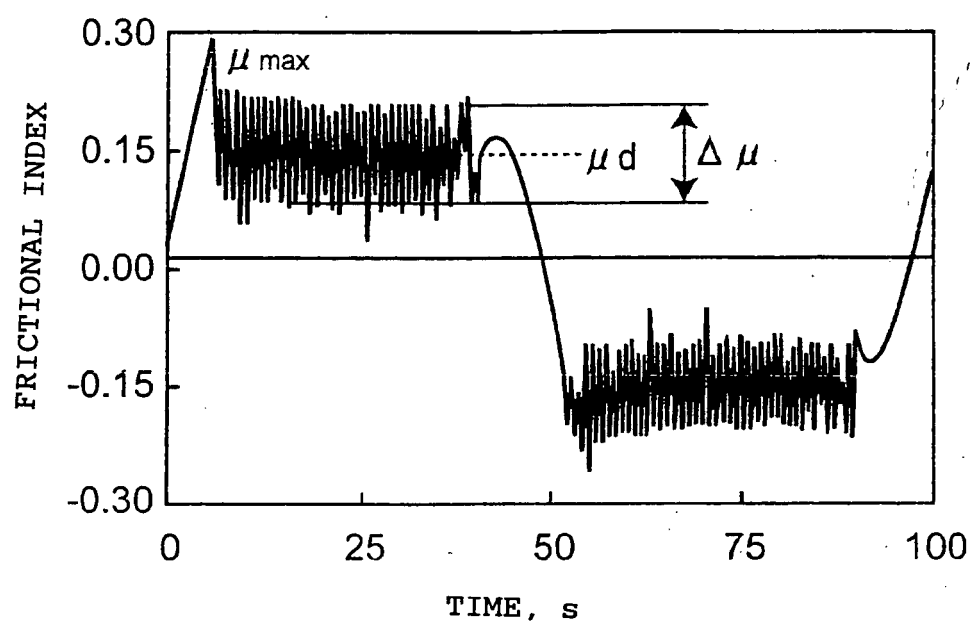
(D-2): a whole ester of a polyhydric alcohol and a mixture of fatty acids, wherein the fatty acids are short-chained fatty acids and long-chained fatty acids; and

(D-3): an ester of a polyhydric alcohol and a branched saturated fatty acid containing a partial ester with the degree of esterification of 1 and a partial ester with the degree of esterification of 2 or more.

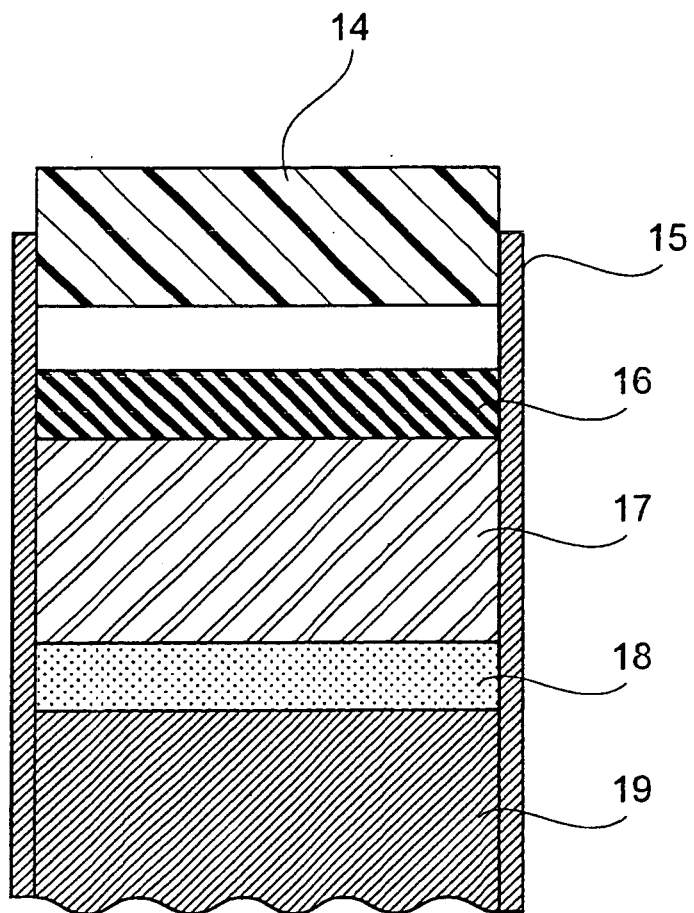
9. The lubricative composition according to claim 8, wherein the lubricating oil composition is a hydraulic oil.





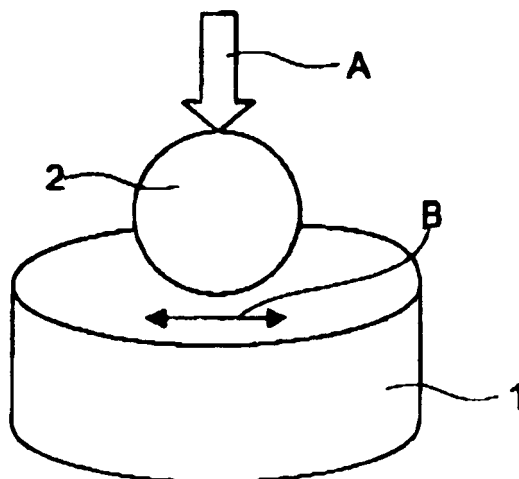
**Fig. 3**

**Fig. 4**





**Fig. 5**



## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2005/006410

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> Int.Cl. <sup>7</sup> C10M129/40, 129/68, 129/74, 129/76, 133/16, 137/04, 137/10, 149/02, 161/00// (C10M161/00, 137:04, 137:10, 149:02), C10N10:02, 10:04, 30:00, 30:04, 30:06, 30:08, 30:10, 30:12, 40:02, 40:04, According to International Patent Classification (IPC) or to both national classification and IPC																	
<b>B. FIELDS SEARCHED</b> Minimum documentation searched (classification system followed by classification symbols) Int.Cl. <sup>7</sup> C10M129/40, 129/68-129/84, 133/16, 137/04-137/10, 149/02-149/10, 161/00, C10N10:02-10:04, 30:00, 30:04-30:12, 40:02-40:04, 40:08 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 1996-2005 Kokai Jitsuyo Shinan Koho 1971-2005 Toroku Jitsuyo Shinan Koho 1994-2005 Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)																	
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b> <table border="1"> <thead> <tr> <th>Category*</th> <th>Citation of document, with indication, where appropriate, of the relevant passages</th> <th>Relevant to claim No.</th> </tr> </thead> <tbody> <tr> <td>X Y</td> <td>JP 2000-63866 A (Showa Shell Sekiyu Kabushiki Kaisha), 29 February, 2000 (29.02.00)</td> <td>1-2, 4-9 3</td> </tr> <tr> <td>X Y</td> <td>JP 2002-265971 A (Showa Shell Sekiyu Kabushiki Kaisha), 18 September, 2002 (18.09.02)</td> <td>1-2, 4-9 3</td> </tr> <tr> <td>X Y</td> <td>JP 2002-294268 A (Showa Shell Sekiyu Kabushiki Kaisha), 09 October, 2002 (09.10.02)</td> <td>1-2, 4-9 3</td> </tr> <tr> <td>X Y</td> <td>JP 2002-338983 A (Showa Shell Sekiyu Kabushiki Kaisha), 27 November, 2002 (27.11.02)</td> <td>1-2, 4-9 3</td> </tr> </tbody> </table>			Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.	X Y	JP 2000-63866 A (Showa Shell Sekiyu Kabushiki Kaisha), 29 February, 2000 (29.02.00)	1-2, 4-9 3	X Y	JP 2002-265971 A (Showa Shell Sekiyu Kabushiki Kaisha), 18 September, 2002 (18.09.02)	1-2, 4-9 3	X Y	JP 2002-294268 A (Showa Shell Sekiyu Kabushiki Kaisha), 09 October, 2002 (09.10.02)	1-2, 4-9 3	X Y	JP 2002-338983 A (Showa Shell Sekiyu Kabushiki Kaisha), 27 November, 2002 (27.11.02)	1-2, 4-9 3
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X Y	JP 2000-63866 A (Showa Shell Sekiyu Kabushiki Kaisha), 29 February, 2000 (29.02.00)	1-2, 4-9 3															
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X Y	JP 2002-338983 A (Showa Shell Sekiyu Kabushiki Kaisha), 27 November, 2002 (27.11.02)	1-2, 4-9 3															
<input checked="" type="checkbox"/> Further documents are listed in the continuation of Box C. <input checked="" type="checkbox"/> See patent family annex.																	
* Special categories of cited documents: "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 date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family																	
Date of the actual completion of the international search 12 July, 2005 (12.07.05)		Date of mailing of the international search report 02 August, 2005 (02.08.05)															
Name and mailing address of the ISA/ Japanese Patent Office		Authorized officer															
Facsimile No.		Telephone No.															

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2005/006410

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	JP 59-24794 A (CIBA-Geigy AG.), 08 February, 1984 (08.02.84)	1-2,4-9 3
X Y	JP 10-67993 A (Ciba Specialty Chemicals Holding Inc.), 10 March, 1998 (10.03.98)	1-2,4-9 3
X Y	JP 11-217577 A (Ciba Specialty Chemicals Holding Inc.), 10 August, 1999 (10.08.99)	1-2,4-9 3
X Y	JP 10-338892 A (R.T. Vandarbilt Co., Inc.), 22 December, 1998 (22.12.98)	1-2,4-9 3
X Y	JP 2000-336384 A (NOK Kuryuba Kabushiki Kaisha), 05 December, 2000 (05.12.00)	1-2,4-9 3
X Y	JP 51-74005 A (Mitsubishi Oil Co., Ltd.), 26 June, 1976 (26.06.76)	1-2,4-9 3
X Y	JP 2001-279286 A (Nippon Mitsubishi Oil Corp.), 10 October, 2001 (10.10.01)	1-2,4-9 3
X Y	JP 8-134488 A (Shell Internationale Research Maatschappij B.V.), 28 May, 1996 (28.05.96)	1-2,4-9 3
X Y	JP 9-111277 A (Idemitsu Kosan Co., Ltd.), 28 April, 1997 (28.04.97)	1-2,4-9 3
Y	JP 2002-97485 A (Nippon Mitsubishi Oil Corp.), 02 April, 2002 (02.04.02)	3

Form PCT/ISA/210 (continuation of second sheet) (January 2004)

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2005/006410

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

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

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
  
2. ☐ Claims Nos.:  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
  
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

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

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

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☒ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
  
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

**Remark on Protest**

- ☐ The additional search fees were accompanied by the applicant's protest.  
☐ No protest accompanied the payment of additional search fees.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2005/006410

Continuation of A. CLASSIFICATION OF SUBJECT MATTER  
(International Patent Classification (IPC))Int.Cl<sup>7</sup> 40:08

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

Continuation of Box No.III of continuation of first sheet (2)

Claims 1-9 contain the following groups (A) to (D) of inventions:

(A) a lubricative composition for industrial machinery and equipment which comprises a base oil selected from among mineral oils, fats and oils, synthetic oils, and mixtures of two or more of them, and an additive consisting of a phosphorus-containing carboxylic acid and/or a phosphoric ester,

(B) a lubricative composition for industrial machinery and equipment which comprises a base oil selected from among mineral oils, fats and oils, synthetic oils, and mixtures of two or more of them, and an additive consisting of a dispersant viscosity index improver,

(C) a lubricative composition for industrial machinery and equipment which comprises a base oil selected from among mineral oils, fats and oils, synthetic oils, and mixtures of two or more of them, and an additive consisting of a compound represented by any of the general formulae (1) to (4) in claim 1, and

(D) a lubricative composition for industrial machinery and equipment which comprises a base oil selected from among mineral oils, fats and oils, synthetic oils, and mixtures of two or more of them, and an additive consisting of an ester oiliness improver.

A lubricative composition for industrial machinery and equipment which comprises a base oil selected from among mineral oils, fats and oils, synthetic oils, and mixtures of two or more of them, and an additive, which is the feature common to groups (A) to (D) of inventions, is publicly known as disclosed in documents cited in Box C of second sheet and remains within the bounds of prior art, thus not corresponding to a special technical feature as provided for in PCT Rule 13.2.

Therefore, there is no technical relationship among the four groups of inventions involving one or more of the same or corresponding special technical features, and this international application includes four groups of inventions which are not so linked as to form a single general inventive concept.

# EP 1 734 103 A1

## INTERNATIONAL SEARCH REPORT Information on patent family members

International application No.

PCT/JP2005/006410

JP 2000-63866 A	2000.02.29	AU 9957377 A	2000.03.14
		BR 9913469 A	2001.06.05
		CN 1319126 A	2001.10.24
		DE 69902181 T2	2002.08.22
		EP 1109882 A1	2001.06.27
		ES 2181471 T3	2003.02.16
		KR 2001099626 A	2001.11.09
		NZ 509838 A	2003.09.26
		US 6756346 B1	2004.06.29
		WO 00/11122 A1	2000.03.02
JP 2002-265971 A	2002.09.18	AU 2002224973 A	2002.07.16
		BR 200206298 A	2004.01.13
		CN 1484688 A	2004.03.24
		EP 1354023 A2	2003.10.22
		KR 2003065589 A	2003.08.06
		US 2004/0053794 A1	2004.03.18
		WO 02/053687 A2	2002.07.11
JP 2002-294268 A	2002.10.09	AU 2002257729 A	2002.10.15
		WO 02/079358 A2	2002.10.10
JP 2002-338983 A	2002.11.27	AU 2002316899 A	2002.11.25
		CN 1522295 A	2004.08.18
		EP 1385926 A1	2004.02.04
		KR 2003096370 A	2003.12.24
		US 2004/0214733 A1	2004.10.28
		WO 02/092735 A1	2002.11.21
JP 59-24794 A	1984.02.08	CA 1219273 A	1987.03.17
		DE 3374011 T2	1987.11.12
		EP 98809 A2	1984.01.18
JP 10-67993 A	1998.03.10	BR 9703974 A	1998.11.17
		CA 2210216 A1	1998.01.15
		DE 59703420 T2	2001.05.31
		EP 819754 A1	1998.01.21
		ES 2157549 T3	2001.08.16
		KR 98009432 A	1998.04.30
		MX 9705300 A	1998.08.01
		US 5922657 A	1999.07.13
JP 11-217577 A	1999.08.10	EP 903399 A1	1999.03.24
		KR 99029924 A	1999.04.26
		US 2002/0016226 A1	2002.02.07
JP 10-338892 A	1998.12.22	AU 698079 B	1998.10.22
		CN 1190669 A	1998.08.19
		CN 1285394 A	2001.02.28
		EP 821053 A2	1998.01.28
		HU 9702214 A	1999.03.29
		US 6046144 A	2000.04.04
JP 2000-336384 A	2000.12.05	(Family: none)	
JP 51-74005 A	1976.06.26	(Family: none)	

Form PCT/ISA/210 (patent family annex) (January 2004)

# EP 1 734 103 A1

## INTERNATIONAL SEARCH REPORT Information on patent family members

International application No.  
PCT/JP2005/006410

JP 2001-279286 A	2001.10.10	CN 1315493 A	2001.10.03
		GB 2362389 A	2001.11.21
		US 2001/0034305 A1	2001.10.25
JP 8-134488 A	1996.05.28	EP 711822 A2	1996.05.15
JP 9-111277 A	1997.04.28	AU 9673335 A	1997.05.07
		CN 1166180 A	1997.11.26
		DE 69615654 T2	2001.11.08
		EP 799883 A1	1997.10.08
		KR 98700401 A	1998.03.30
		US 5902777 A	1999.05.11
		WO 97/14776 A1	1997.04.24
JP 2002-97485 A	2002.04.02	CN 1346872 A	2002.05.01
		KR 2002023127 A	2002.03.28

**REFERENCES CITED IN THE DESCRIPTION**

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**Patent documents cited in the description**

- JP 10259394 A [0004]
- JP 9208976 A [0004]
- JP 2002097485 A [0008]
- JP 57067693 A [0011]
- JP 51074005 A [0011]
- JP 8209175 A [0012]
- JP 11209775 A [0012]
- JP 10067993 A [0017]
- JP 11217577 A [0017]
- JP 2002265971 A [0017]
- JP 9111277 A [0018]