LUBRICANT ADDITIVE AND LUBRICATING OIL COMPOSITION

Provided are (i) a lubricant additive including a borazine compound represented by the following general formula (1), (ii) a lubricating oil composition including a lubricating base oil and the lubricant additive, and (iii) a lubricating oil composition further including a friction modifier represented by the following general formula (2).

![Chemical Structures](image-url)
The present invention relates to a lubricant additive and a lubricating oil composition.

For various machines represented by internal combustion engines, transmissions and the like, lubrication is an essential element for improving energy efficiency and protecting machine parts from wear. A lubricant such as lubricating oil and a grease reduces friction and wear by forming a lubricating film on a friction surface. For the purpose of reducing friction and wear even under a condition in which the friction surface is subjected to a large load and the thickness of the lubricating film is easily reduced, various load-resistant additives such as: oiliness agents which are adsorbed on the friction surface to form an oil film (e.g. higher fatty acids such as oleic acid, and higher alcohol esters); organic molybdenum based friction modifiers which react with the friction surface to form a film (e.g. molybdenum dithiocarbamate (MoDTC), and molybdenum dithiophosphate (MoDTP)); and friction-reducing agents (e.g. dithiophosphate (ZnDTP), phosphoric acid ester, and disulfide) are incorporated in the lubricant.

Regarding friction-reducing agents, phosphorus compounds such as the above-mentioned ZnDTP and phosphoric esters are known to show a good performance as friction-reducing agents. However, for lubrication of internal combustion engines, a new friction-reducing agent is required which does not contain phosphorus, preferably does not contain phosphorus or sulfur, in view of inhibiting poisoning of an exhaust gas treatment catalyst.

Regarding organic molybdenum based friction modifiers, sulfur-containing oil-soluble organic molybdenum compounds such as the above-mentioned MoDTC and MoDTP are known to show a good friction reducing effect under a boundary lubricating condition, and are widely used in combination with oiliness agent based friction modifiers so as to further improve the friction reducing performance of the lubricating oil. However, MoDTC contains a metal (ash) component and sulfur, and MoDTP contains ash, sulfur, and phosphorus. Therefore, it is pointed out that these organic molybdenum based friction modifiers can be an interfering element in recycling the lubricating oil for example, or that these organic molybdenum based friction modifiers can badly affect an exhaust gas purifier in a case where they are used for lubrication of an internal combustion engine for example. From such viewpoints, it is required to reduce the additive amount of the organic molybdenum based friction modifier, and at the same time, it is required to improve the performance of the oiliness agent based friction modifier, especially to improve the friction reducing performance under boundary lubricating conditions.

As lubricant components not including phosphorus or sulfur, inorganic solid lubricant components such as a graphite powder and a hexagonal boron nitride powder (h-BN) are known (Patent Literature 1), and a grease composition containing such an inorganic solid lubricant component is also suggested (Patent Literatures 2 to 4). Each of these inorganic solid lubricant components microscopically has a layered structure, and interaction between adjacent layers is weak. Therefore, it is considered that the adjacent layers easily change their relative positions when being subjected to a shearing force at the friction surface, thereby showing a lubricating effect.

However, since these inorganic solid lubricant components do not have solubility or dispersibility, these inorganic solid lubricant components precipitate when incorporated in a base oil as additive agents, which causes a serious problem of unevenness. Therefore, it is extremely difficult to incorporate the inorganic solid lubricant components in ordinary liquid lubricants represented by lubricating oil for internal combustion engines. Greases (semisolid lubricant) disclosed

Citation List

Patent Literatures

[0005]


SUMMARY OF INVENTION

Technical Problem

As lubricant components not including phosphorus or sulfur, inorganic solid lubricant components such as a graphite powder and a hexagonal boron nitride powder (h-BN) are known (Patent Literature 1), and a grease composition containing such an inorganic solid lubricant component is also suggested (Patent Literatures 2 to 4). Each of these inorganic solid lubricant components microscopically has a layered structure, and interaction between adjacent layers is weak. Therefore, it is considered that the adjacent layers easily change their relative positions when being subjected to a shearing force at the friction surface, thereby showing a lubricating effect.

However, since these inorganic solid lubricant components do not have solubility or dispersibility, these inorganic solid lubricant components precipitate when incorporated in a base oil as additive agents, which causes a serious problem of unevenness. Therefore, it is extremely difficult to incorporate the inorganic solid lubricant components in ordinary liquid lubricants represented by lubricating oil for internal combustion engines. Greases (semisolid lubricant) disclosed
in Patent Literatures 2 to 4 are considered not to suffer problems in practical use, since the inorganic solid lubricant component is held by a thickener in the greases. However, in view of improving microscopic uniformity, ease of mixing, and the like, it is preferable for a component incorporated in a grease composition to have solubility in the base oil.

[0008] A first object of the present invention is to provide a lubricant additive which has solubility in a base oil, does not include phosphorus or sulfur, and has a friction-reducing performance, and to provide a lubricating oil composition including the lubricant additive.

[0009] A second object of the present invention is to provide a lubricating oil composition which includes an oiliness agent based friction modifier and shows improved friction reducing performance under boundary lubricating conditions, with suppressed increase of phosphorus and sulfur content.

Solution to Problem

[0010] The inventor of the present invention has found that a compound having a BN six-membered ring as its basic skeleton, which is a smallest unit forming one layer of hexagonal boron nitride (h-BN), i.e. a borazine compound has both solubility and anti-friction performance, and improves the friction reducing performance of the lubricating oil composition containing an oiliness agent based friction modifier (ashless friction modifier) having a nitrogen atom in a polar group.

[0011] A first aspect of the present invention is a lubricant additive including a borazine compound represented by the following general formula (1), which achieves the first object.

$$\text{(In the formula (1), } R^1, R^3, \text{ and } R^5 \text{ are each independently hydrogen, a C1 to C30 hydrocarbyl group, or a C1 to C30 hydrocarbyl group comprising a heteroatom other than sulfur and phosphorus; and } R^2, R^4, \text{ and } R^6 \text{ are each independently hydrogen, a C1 to C30 hydrocarbyl group, or a C1 to C30 hydrocarbyl group including oxygen or boron or nitrogen).}
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[0012] In the present application, "or" means a logical sum unless otherwise noted. In the present invention, the "heteroatom" is interpreted in a broad sense, and means elements other than carbon and hydrogen. It should be noted that a noble gas is not included in the "heteroatom".

[0013] Regarding R^1, R^3, and R^5, a "C1 to C30 hydrocarbyl group including oxygen or boron or nitrogen" can be given as one preferable embodiment of the above-mentioned "C1 to C30 hydrocarbyl group including a heteroatom other than sulfur and phosphorus".

[0014] Regarding R^1, R^3, and R^5, a "C1 to C30 hydrocarbyl group including oxygen or boron" can be given as another preferable embodiment of the above-mentioned "C1 to C30 hydrocarbyl group including a heteroatom other than sulfur and phosphorus".

[0015] In one preferable embodiment of the lubricant additive according to the first aspect of the present invention, in the above formula (1), R^1, R^3, and R^5 are the same groups and R^2, R^4, and R^6 are the same groups. Such an embodiment allows easy production of the borazine compound.

[0016] A second aspect of the present invention is a lubricating oil composition including (A) a lubricating base oil, and (B) the lubricant additive according to the first aspect of the present invention, which achieves the first object.

[0017] A third aspect of the present invention is a lubricating oil composition including (A) the lubricating base oil, (B) the lubricant additive according to the first aspect of the present invention, and (C) a friction modifier represented by the following formula (2), which achieves the second object.
(In the formula (2), p is 0 or 1; q is 0 or 1; r is 0 or 1; R7 is a hydrocarbyl group having no less than 8 carbon atoms; and R8, R9 and R10 are each independently hydrogen or a C1 to C4 hydrocarbyl group.).

[0018] In one preferable embodiment of the lubricating oil composition according to the third aspect of the present invention, in the formula (2), at least R9 and R10 are hydrogen and where p and q are 1, R8, R9, and R10 are hydrogen. Such an embodiment makes it possible to increase adsorption ability to the friction surface, whereby it becomes easy to increase the friction-reducing effect.

[0019] One preferable embodiment of the lubricating oil composition according to the second or third aspect of the present invention further includes one or more selected from the group consisting of an ashless dispersant, an antioxidant, a friction modifier, a friction-reducing agent, a metallic detergent, a viscosity index improver, a pour point depressant, a corrosion inhibitor, an anti-rust agent, an anti-emulsifier, a metal deactivator, a defoamer, and a coloring agent.

[0020] The lubricating oil composition according to the second or third aspect of the present invention can be preferably used for lubrication of internal combustion engines.

Advantageous Effect of Invention

[0021] According to the first aspect of the present invention, it is possible to provide a lubricant additive having solubility in a base oil, not including phosphorus or sulfur, and having a friction-reducing performance.

[0022] According to the second aspect of the present invention, by containing the lubricant additive according to the first aspect of the present invention, it is possible to provide a lubricating oil composition which has improved friction-reducing performance, with suppressed increase of phosphorus and sulfur content.

[0023] According to the third aspect of the present invention, it is possible to provide a lubricating oil composition which contains an oiliness agent based friction modifier and which has further improved friction reducing performance under boundary lubricating conditions, with suppressed increase of phosphorus and sulfur content.

DESCRIPTION OF EMBODIMENTS

[0024] Hereinafter, the present invention will be described in detail. It should be noted that, unless otherwise noted, "A to B" regarding numerical values A and B means "A or more and B or less". In cases where the unit of the numerical value A is omitted, the unit given to the numerical value B is applied to the numerical value A.

<1. Lubricant additive>

[0025] The lubricant additive according to the first aspect of the present invention will be described.

(Borazine compound)

[0026] The lubricant additive of the present invention includes a borazine compound represented by the following general formula (1).
In the formula (1), R1, R3, and R5 that are substituents on nitrogen atoms are each independently hydrogen, a C1 to C30 hydrocarbyl group, or a C1 to C30 hydrocarbyl group including a heteroatom other than sulfur and phosphorus.

Here, examples of the C1 to C30 hydrocarbyl group include, specifically, alkyl group which may be cycloalkyl group or alkylcycloalkyl group, alkenyl group in which double bond(s) may be in any position), aryl group, alkyaryl group, arenylalkyl group, and the like.

Examples of the above cycloalkyl group include C5 to C7 cycloalkyl groups such as cyclopentyl group, cyclohexyl group, and cycloheptyl group. In the above alkylcycloalkyl group, alkyl substitution may be in any position of the cycloalkyl group.

Examples of the above aryl group include phenyl group, naphthyl group, and the like. In the above alkyaryl group and arylalkyl group, alkyl substitution may be in any position of the aryl group.

The above "C1 to C30 hydrocarbyl group including a heteroatom other than sulfur and phosphorus" means a C1 to C30 hydrocarbyl group functionalized to include a heteroatom other than sulfur and phosphorus. Examples of the hydrocarbyl group include the hydrocarbyl groups as above. As examples of the above "heteroatom other than sulfur and phosphorus", nonmetal typical elements other than these elements can be given. Among them, oxygen O, boron B, nitrogen N, silicon Si, and halogen (fluorine F, chlorine Cl, bromine Br, and iodine I) can be given as preferable examples, and a group containing one or more of these elements can preferably be employed. It should be noted that, as the halogen, F and Cl are preferable, and F is more preferable, in view of bond stability.

Specific examples of functionalization include functionalization by an ester bond (alkoxycarbonyl substitution or acyloxy substitution), functionalization by an acyl group, functionalization by a carboxy group or metal salt thereof, functionalization by an ether bond (alkoxy substitution), functionalization by a boryl group (e.g. dihydrocarbylboryl group), functionalization by a borate ester bond (e.g. dialkoxyboroxy substitution), functionalization by a borate ester bond (e.g. dialkoxyboroxy substitution), functionalization by an amino group (e.g. dihydrocarbylamino group), functionalization by an amide bond (-CO-N< bond) (aminocarbonyl substitution or acylamino substitution), functionalization by a silyl group (e.g. trihydrocarbylsilyl group), functionalization by a halogeno group (e.g. fluoro group), and the like.

As a preferable embodiment of the above "C1 to C30 hydrocarbyl group including a heteroatom other than sulfur and phosphorus", a "C1 to C30 hydrocarbyl group including oxygen or boron or nitrogen" can be given. The "C1 to C30 hydrocarbyl group including oxygen or boron or nitrogen" means a C1 to C30 hydrocarbyl group functionalized to include oxygen or boron or nitrogen. The above-described hydrocarbyl groups can be given as the hydrocarbyl group.

Specific examples of the functionalization in this embodiment include functionalization by an ester bond (alkoxycarbonyl substitution or acyloxy substitution), functionalization by an acyl group, functionalization by a carboxy group, functionalization by an ether bond (alkoxy substitution), functionalization by a boryl group (e.g. dihydrocarbylboryl group), functionalization by a borate ester bond (e.g. dialkoxyboroxy substitution), functionalization by an amino group (e.g. dihydrocarbylamino group), functionalization by an amide bond (-CO-N< bond) (aminocarbonyl substitution or acylamino substitution), and the like.

As another preferable embodiment of the above "C1 to C30 hydrocarbyl group including a heteroatom other than sulfur and phosphorus", a "C1 to C30 hydrocarbyl group including oxygen or boron" can be given. The "C1 to C30 hydrocarbyl group including oxygen or boron" means a C1 to C30 hydrocarbyl group functionalized to include oxygen or boron. The above-described hydrocarbyl groups can be given as the hydrocarbyl group.

Specific examples of the functionalization in this embodiment include functionalization by an ester bond (alkoxycarbonyl substitution or acyloxy substitution), functionalization by an acyl group, functionalization by a carboxy group, functionalization by an ether bond (alkoxy substitution), functionalization by a boryl group (e.g. dihydrocarbylboryl group), functionalization by a borate ester bond (e.g. dialkoxyboroxy substitution), and the like.

Regarding each of R1, R3, and R5 which is a substituent on nitrogen atom in the above general formula (1), in a case where it is a group other than hydrogen, its carbon number is preferably 3 or more, more preferably 6 or more, and preferably 24 or less, more preferably 18 or less, and still preferably 12 or less.

In the above general formula (1), R2, R3, and R5 that are substituents on boron atoms are each independently hydrogen, a C1 to C30 hydrocarbyl group, or a C1 to C30 hydrocarbyl group including oxygen or boron or nitrogen.

Regarding R2, R4, and R6, the C1 to C30 hydrocarbyl group is same as the C1 to C30 hydrocarbyl group described above regarding R1, R3, and R5.

The above "C1 to C30 hydrocarbyl group including oxygen or boron or nitrogen" means a C1 to C30 hydrocarbyl group functionalized to include oxygen or boron or nitrogen. The hydrocarbyl group described above can be given as the hydrocarbyl group. Examples of embodiments of the functionalization include functionalization by an ester bond (alkoxycarbonyl substitution or acyloxy substitution), functionalization by a carboxy group, functionalization by an ether bond (alkoxy substitution), functionalization by a boryl group (e.g. dihydrocarbylboryl group), functionalization by a borate ester bond (e.g. dialkoxyboroxy substitution), functionalization by an amino group (e.g. dihydrocarbylamino group), functionalization by an amide bond (-CO-N< bond) (aminocarbonyl substitution or acylamino substitution), and the like.

Regarding each of R2, R4, and R6 which is a substituent on boron atom in the above general formula (1), if it is a group other than hydrogen, the carbon number thereof is preferably 3 or more, more preferably 6 or more, and
preferably 24 or less, more preferably 18 or less, and still preferably 12 or less.

[0042] In view of reducing volatility of the borazine compound, preferably one or more of the six substituents R₁ to R₆ in the above general formula (1) are groups other than hydrogen, more preferably two or more are groups other than hydrogen, and especially preferably three or more are groups other than hydrogen.

[0043] In addition, in view of easy production of the borazine compound, preferably one or more of the substituents R₁ to R₆ in the above general formula (1) are groups other than hydrogen, more preferably two or more are groups other than hydrogen, and especially preferably three or more are groups other than hydrogen.

[0044] As the borazine compound satisfying these requirements, a borazine compound in which all of the substituents (R₁, R₃, and R₅) on nitrogen, or all of the substituents (R₂, R₄, and R₆) on boron, or both of them (all of R₁ to R₆) are groups other than hydrogen in the above general formula (1) is especially preferably employed.

[0045] In the lubricant additive of the present invention, one borazine compound represented by the above formula (1) may be used alone, or two or more borazine compound represented by the above formula (1) may be used in combination.

(Production of borazine compound)

[0046] The method of producing the borazine compound represented by the above formula (1) is not particularly limited, and a known synthetic method can be adequately employed. For the sake of ease of explanation, a case where R₁, R₃, and R₅ that are substituents on nitrogen atoms are the same groups, and R₂, R₄, and R₆ that are substituents on boron atoms are the same groups is mainly described as an example. In this case, the substitution pattern of the borazine compound represented by the above general formula (1) can be classified in the following patterns (I) to (VI).

(I) N-hydrogen or aliphatic substitution, B-hydrogen;
(II) N-hydrogen or aliphatic substitution, B-aliphatic substitution;
(III) N-hydrogen or aliphatic substitution, B-aromatic substitution;
(IV) N-aromatic substitution, B-hydrogen;
(V) N-aromatic substitution, B-aromatic substitution;
(VI) N-aromatic substitution, B-aliphatic substitution

Hereinafter, examples of the synthetic method will be described for each of the patterns (I) to (VI).

(Production of borazine compound: (I) N-hydrogen or aliphatic substitution, B-hydrogen)

[0047] In a case where R₁ = R₃ = R₅ = hydrogen or an aliphatic group, and R₂ = R₄ = R₆ = hydrogen in the general formula (1), for example as shown by the following formula (3), the borazine compound can be synthesized by a method of reacting an alkali borohydride ABH₄ (A is an alkali metal) such as sodium borohydride and an ammonium halide RNH₃X (X is a halogen; R may have a cyclic structure (e.g. a cycloalkyl group), may have a double bond, may be substituted by an aromatic group (e.g. an arylalkyl group), and may be hydrogen) such as alkylammonium chloride in a solvent. Details such as the reaction conditions in the method are disclosed for example in JP 2008-201729 A.

\[ \text{3 ABH}_4 + 3 \text{RNH}_3\text{X} \rightarrow \text{3 AX} + 9 \text{H}_2 \] (3)

[0048] For another example, as shown in the following formula (4), the borazine compound also can be synthesized by a method of reacting a borane (BH₃) complex such as borane-tetrahydrofuran complex or diborane (B₂H₆) and a nitrile RCN (R is an aliphatic group (which may have a cyclic structure, and may have a double bond) or an aromatic group) in a solvent. Details such as the reaction conditions in the method are disclosed for example in JP 2010-173945 A.
For another example, the borazine compound can be synthesized by a method of: synthesizing a trichloroborazine compound by reacting trichloroborane BCl₃ and an ammonium halide RNH₃X (X is a halogen; R may have a cyclic structure (e.g. a cycloalkyl group), may have a double bond, may be substituted by an aromatic group (e.g. an arylalkyl group), and may be hydrogen) such as alkylammonium chloride in a solvent as shown in the following formula (5); and thereafter reducing the trichloroborazine compound by sodium borohydride as shown in the following formula (6). Details of the reaction of the first step (formula (5)) such as the reaction conditions are disclosed for example in JP 2005-112723 A and JP 2005-104869 A. The reaction of the second step (formula (6)) is well known.

In a case where R₁ = R₃ = R₅ = hydrogen or an aliphatic group, and R₂ = R₄ = R₆ = an aliphatic group in the above general formula (1), for example, the borazine compound may be synthesized by a method of: synthesizing a trichloroborazine compound by reacting trichloroborane BCl₃ and an ammonium halide RNH₃X such as alkylammonium chloride in a solvent as already described referring to the formula (5); and thereafter reacting the trichloroborazine compound and an aliphatic Grignard reagent R’ MgX (X is a halogen; R’ may have a cyclic structure (e.g. a cycloalkyl group), may have a double bond, and may be substituted by an aromatic group (e.g. an arylalkyl group)) as shown in the following formula (7). Details such as the reaction conditions in the method are disclosed for example in JP 2005-053854 A, JP 2005-104869 A, and JP 2005-112723 A.
For another example, the borazine compound can be synthesized by a method of successively reacting the above-described borazine compound of (I) N-hydrogen or aliphatic substitution and B-hydrogen, and an alkene compound in two steps, under presence of a catalyst (e.g. RhCl (PPh₃)₃) (following formula (8)). Details such as the catalyst and the reaction conditions in the method are disclosed for example in JP 2010-037789 A.

(Production of borazine compound: (III) N-hydrogen or aliphatic substitution, B-aromatic substitution)

In a case where R₁ = R₃ = R₅ = hydrogen or an aliphatic group, and R₂ = R₄ = R₆ = an aromatic group in the above general formula (1), the borazine compound can be synthesized for example by a method of reacting the above-described borazine compound of (I) N-hydrogen or aliphatic substitution, B-hydrogen and an aryl Grignard reagent (following formula (9)). Details such as the reaction conditions in the method are disclosed for example in J. Am. Chem. Soc., 1959, 81, 582-586.

For another example, the borazine compound can be synthesized by a method of reacting the above-described borazine compound of (I) N-hydrogen or aliphatic substitution, B-hydrogen and an aryl halide ArX (X is a halogen; Ar is an aryl group which may be substituted by an alkyl group and the like), under presence of a catalyst (e.g. PdCl₂(PPh₃)₂) (following formula (10)). Details such as the catalyst and reaction conditions in the method are disclosed for example in JP 2010-280637 A.

(Production of borazine compound: (IV) N-aromatic substitution, B-hydrogen)

In a case where R₁ = R₃ = R₅ = an aromatic group, and R₂ = R₄ = R₆ = hydrogen in the above general formula (1), the borazine compound can be synthesized for example by a method of: synthesizing a trihaloborazine compound by reacting a trihaloborane such as trichloroborane BCl₃ and an arylamine ArNH₂ (Ar is an aryl group which may be substituted by an alkyl group and the like), under presence of a catalyst (e.g. PdCl₂(PPh₃)₂) (following formula (11)); and thereafter reducing the trihaloborazine compound by sodium borohydride as shown in the following formula (12). Details of the reaction of the first step (formula (11)) such as the reaction conditions are disclosed for example in JP 2005-170857 A. The reaction of the second step (formula (12)) is well known.
In a case where \( R_1 = R_3 = R_5 = \) an aromatic group, and \( R_2 = R_4 = R_6 = \) an aromatic group in the above general formula (1), the borazine compound can be synthesized for example by a method of: synthesizing a trihaloborazine compound by reacting a trihaloborane such as trichloroborane \( \text{BCl}_3 \) and an arylamine \( \text{ArNH}_2 \) as described above referring to the formula (11); and thereafter reacting the trihaloborazine compound and an organometallic compound formed by lithiation or transformation into a Grignard reagent of an aryl halide \( \text{Ar'Y} \) (\( Y \) is halogen; \( \text{Ar'} \) is an aryl group which may be substituted by an alkyl group and the like), as shown in the following formula (13). Details such as the reaction conditions in the method are disclosed for example in JP 2005-170857 A.

In a case where \( R_1 = R_3 = R_5 = \) an aromatic group, and \( R_2 = R_4 = R_6 = \) an aliphatic group in the above general formula (1), the borazine compound can be synthesized for example by a method of: synthesizing a trihaloborazine compound by reacting a trihaloborane such as trichloroborane \( \text{BCl}_3 \) and an arylamine \( \text{ArNH}_2 \) as described above referring to the formula (11); and thereafter reacting an aliphatic Grignard reagent \( \text{R'MgX} \) and the trihaloborazine compound (following formula (14)), as described above referring to the formula (7).
In the above explanation regarding the production method of the borazine compound, embodiments where substituents on nitrogen atoms R₁, R₃ and R₅ are the same groups, and substituents on boron atoms R₂, R₄, and R₆ are the same groups in the above general formula (1) have been explained as examples. However, the borazine compound which can be employed in the present invention is not limited to these embodiments. Borazine compounds having a substitution pattern in which substituents on nitrogen atoms R₁, R₃, and R₅ are different from each other, or in which substituents on boron atoms R₂, R₄, and R₆ are different from each other may also be employed, and such borazine compounds can also be synthesized. It is possible to synthesize the borazine compound having a substitution pattern in which substituents on nitrogen atoms R₁, R₃, and R₅ are different from each other, or the borazine compound having both substitution patterns, for example by: employing two or more ammonium salts or amines in combination as nitrogen sources of the borazine structure; employing two or more organometallic reagent for introducing substituents on boron atoms of the borazine structure in combination; adequately adjusting the stoichiometric relationship between the B-unsubstituted borazine and the Grignard reagent in the reaction of the formula (9); employing different alkene compounds in the first step and the second step in the reaction of the formula (8), and the like.

It is also possible to proceed the synthesis with the functional group of the substituent protected by an appropriate protecting group, and deprotect them after introducing all substituents. Introduction and deprotection of protective groups can be carried out by a known method. It is also possible to introduce all substituents and thereafter transform functional groups of the substituents to other functional groups by known synthetic methods.

The lubricating additive of the present invention has solubility in a base oil. Therefore, it can be incorporated in both of a lubricating oil (fluid lubricant) and a grease (semisolid lubricant), to improve friction-reducing effect of the lubricants. In a case where the lubricant additive is contained in a lubricating oil, the content thereof can be for example in a range of 0.01 to 5.0 mass%, as a content of the borazine compound represented by the above general formula (1) on the basis of the total amount of the lubricating oil composition as 100 mass%. In a case where the lubricant additive is contained in a grease, as the content thereof, for example a range of 0.1 to 10.0 mass% can be given as the content of the borazine compound represented by the above general formula (1) on the basis of the total amount of the grease composition as 100 mass%.

In addition, by incorporating the lubricant additive of the present invention in a lubricating oil (e.g. lubricating oil for internal combustion engines), it is possible to improve not only a friction-reducing property of the lubricating oil but also high-temperature detergency. Conventional friction-reducing agents used in lubricating oils do not have a high-temperature detergent property. Therefore, the lubricant additive of the present invention can be preferably used for lubrication of machines whose metal parts are exposed to a high temperature (e.g. internal combustion engines).

The principle of the lubricant additive of the present invention to increase the high-temperature detergency has not been fully clarified. However, the inventor of the present invention presumes as follows. That is, in a borazine ring, B atoms and N atoms are alternately arranged to form a 6-membered ring, and polarization of B-N bonds makes three partial positive charges and three partial negative charges form an electric multipole. This particular electric multipole structure shows a good affinity with a highly polar sludge. On the other hand, the borazine ring structure itself has no polarity as a whole. Therefore, the lubricant additive shows a good affinity with the base oil as well. As a result, the lubricant additive exhibits a deterging effect even under a high-temperature condition.

The lubricating oil composition according to the second aspect of the present invention will be described. The lubricating oil composition according to the second aspect of the present invention includes (A) a lubricating base oil, (B) the lubricant additive according to the first aspect of the present invention (hereinafter may be referred to as "borazine-based additive").

The lubricating base oil in the lubricating oil composition of the present invention is not particularly limited, and a mineral base oil or a synthetic base oil used for a general lubricating oil can be employed.

Specific examples of the mineral base oil include: a product made by purifying a lubricating oil fraction obtained by vacuum distillation of a residue of atmospheric distillation of a crude oil, by one or more treatments of solvent deasphalting, solvent extraction, hydrogenolysis, solvent dewaxing, hydrorefining, and the like; an wax-isomerized mineral
oil; a lubricating oil base oil produced by a method of isomerizing a GTL WAX (Gas To Liquid wax) produced for example by Fisher-Tropsch process, and the like. [0065] Examples of the synthetic base oil include: a poly α-olefin such as 1-octene oligomer and 1-decene oligomer, and hydrogenation product thereof; isobutene oligomer and hydrogenation product thereof; paraffin; alkylbenzene; alkyl-naphthalene; diesters such as ditridecyl glutarate, di(2-ethylhexyl) adipate, distearyldiphenyl ether, and polyalkylene glycol; dialkyldiphenyl ether. In addition, aromatic synthetic oils such as alkynaphthalene, alkylbenzene, and aromatic ester, and mixture thereof can be given as examples.

[0066] In the lubricating oil composition of the present invention, a mineral base oil, a synthetic base oil, or any mixture of two or more lubricating oils selected therefrom and the like may be employed as the lubricating base oil. One or more mineral base oil, one or more synthetic base oil, a mixture oil of one or more mineral base oil and one or more synthetic base oil, and the like can be given as examples.

[0067] Kinematic viscosity, NOACK evaporation loss, and viscosity index of the lubricating base oil in the lubricating oil composition of the present invention may be adequately determined depending on the purpose of use of the lubricating oil composition. For example, in a case where the lubricating oil composition is used for internal combustion engines, the kinematic viscosity of the lubricating base oil at 100 °C may be 3.0 to 16.3 mm²/s for example. Alternatively, for example in a case where the lubricating oil composition is used for transmissions, the kinematic viscosity of the lubricating base oil at 100 °C may be 3.5 to 25.0 mm²/s for example.

(B) Borazine-based additive

[0068] Details of the borazine-based additive have been already described above. The content of the borazine-based additive in the lubricating oil composition of the present invention is not particularly limited. As the content of the borazine composition represented by the above general formula (1), it is normally 0.01 mass% or more, preferably 0.05 mass% or more, and especially preferably 0.1 mass% or more, and normally 5.0 mass% or less, preferably 3.0 mass% or less, and especially preferably 1.0 mass% or less, on the basis of the total amount of the composition.

<3. Lubricating oil composition (third aspect of present invention)>

[0069] The lubricating oil composition according to the third aspect of the present invention will be described. The lubricating oil composition according to the third aspect of the present invention includes (A) the lubricating base oil, (B) the borazine-based additive, and (C) a friction modifier represented by the following general formula (2). The (A) lubricating base oil and the (B) borazine-base additive, and the contents thereof are same as in the lubricating oil composition according to the second aspect of the present invention.

(C) Friction modifier

[0070]

([0071] In the general formula (2), p is 0 or 1. When p = 0, the general formula (2) represents an amine compound.

[0072] When p = 0, q is 0 or 1. When p = 1 and q = 0, the general formula (2) represents an amide compound.

[0073] When p = 1 and q = 1, r is 0 or 1. When p = 1, q = 1, and r = 0, the general formula (2) represents a urea compound. When p = 1, q = 1, and r = 1, the general formula (2) represents a ureide compound.

[0074] In the general formula (2), R⁷ is a hydrocarbyl group having no less than 8 carbon atoms. The carbon number of R⁷ is preferably no less than 10, more preferably no less than 12; and preferably no more than 30, and more preferably no more than 24.

[0075] Specific examples of the hydrocarbyl group having no less than 8 carbon atoms include an alkyl group (which may be substituted by cycloalkyl group), alkylcycloalkyl group, alkenyl group (double bond(s) may be in any position).
aryl group, alkylaryl group, arylalkyl group, and the like.

[0076] Examples of the cycloalkyl group include a cycloalkyl group having 5 to 7 carbon atoms, such as a cyclopentyl group, cyclohexyl group, and cycloheptyl group. Alkyl substitution of the cycloalkyl group may be in any position.

[0077] Examples of the aryl group and alkylaryl group include: an alkylphenyl group such as a xylyl group, ethylphenyl group, propylphenyl group, butylphenyl group, pentylphenyl group, hexylphenyl group, heptylphenyl group, octylphenyl group, nonylphenyl group, decylphenyl group, undecylphenyl group, and dodecylphenyl group; and aryl group and alkylaryl group having no less than 8 carbon atoms such as a substituted or unsubstituted naphthyl group (preferably the alkyl group is a linear alkyl group; alkyl substitution of the aryl group may be in any position, and preferably in para position).

[0078] Examples of the arylalkyl group include an arylalkyl group having no less than 8 carbon atoms such as a phenylethyl group, phenylpropyl group, phenylbutyl group, phenylpentyl group, phenylhexyl group (preferably the alkyl group is a linear alkyl group; aryl substitution of the alkyl group may be in any position, and preferably in ω position).

[0079] The hydrocarbyl group of R7 is preferably an alkyl group or alkenyl group. When R7 is an alkenyl group, the alkyl groups between which double bonds are interposed are preferably linear alkyl groups.

[0080] When R7 is an alkyl group, R7 is preferably a linear alkyl group. In view of easy use under a low-temperature condition, it is more preferable that R7 is an alkyl group having a methyl group in α position. R7 being such an alkyl group makes it possible to lower the solidifying point compared with a case where R7 is a complete linear alkyl group.

[0081] In the general formula (2), R8, R9, and R10 are each independently hydrogen or a C1 to C4 hydrocarbyl group. Preferably, at least either one of R8 and R10 is hydrogen, and more preferably both R8 and R10 are hydrogen. When p = 1 and q = 1 (that is, when R8 exists), preferably R8 is hydrogen, and more preferably R8, R9, and R10 are each hydrogen. More hydrogen atoms being the groups other than R7 increase adsorbability to a friction surface, whereby it becomes easy to increase friction reducing effect.

[0082] When p = 0 in the general formula (2), the general formula (2) represents an amine compound. Such an amine compound can be adequately synthesized by a known method.

[0083] When p = 1 and q = 0 in the general formula (2), the general formula (2) represents an amide compound. Such an amide compound can be adequately synthesized by a known method such as a condensation reaction of a carboxylic acid and an amine.

[0084] When p = 1, q = 1, and r = 0 in the general formula (2), the general formula (2) represents a urea compound. A known synthetic method can be employed without particular limitations as the synthetic method of such a urea compound. For example, a synthetic method by a condensation reaction of an isocyanate compound and ammonia or an amine compound, as shown in the following formula (15) can be given.

\[
R^7 - \text{C} = \text{O} + \text{H} - \text{N} - \text{R}^9 \rightarrow \text{R}^7 - \text{C} = \text{O} - \text{N} - \text{R}^9, \quad (15)
\]

[0085] A known isocyanate compound can be used without particular limitations as the isocyanate compound in the above formula (15). Examples of the isocyanate compound which can be used in the reaction represented by the above formula (15) include an isocyanate compound in which R7 is a hydrocarbyl group (preferably an alkyl group or alkenyl group) having no less than 8 carbon atoms.

[0086] In addition, in the reaction represented by the above formula (15), a known primary or secondary amine compound or ammonia can be used as a nucleophilic reagent without particular limitations. Examples of the primary or secondary amine compound which can be used in the reaction represented by the above formula (15) include an amine compound having a hydrocarbyl group having 1 to 4 carbon atoms.

[0087] In addition, when p = 1, q = 1, and r = 1 in the general formula (2), the general formula (2) represents a ureide compound. A known synthetic method can be employed as the synthetic method of such a ureide compound, without particular limitations. For example, a synthetic method by a reaction of an acid chloride and urea or a urea compound represented by the following formula (16) can be given.

\[
R^7 - \text{N} - \text{C} = \text{O} - \text{N} - \text{R}^9 \rightarrow \text{R}^7 - \text{N} - \text{C} = \text{O} - \text{N} - \text{R}^9, \quad (16)
\]
A known acid chloride can be used as the acid chloride in the reaction represented by the above formula (16), without particular limitations. Examples of the acid chloride which can be used in the reaction represented by the above formula (16) include a carboxylic acid chloride in which R^7 is a hydrocarbyl group (preferably an alkyl group or alkenyl group) having no less than 8 carbon atoms.

A known urea compound can be used as the urea compound in the reaction represented by the above formula (16), without particular limitations. Examples of the urea compound which can be used in the reaction represented by the above formula (16) include urea, N-methylurea, N-ethylurea, N-tert-butylurea, N,N'-dimethylurea and the like. These urea compounds can be obtained by a known synthetic method such as a reaction of isocyanate and ammonia or an amine compound.

As the (C) component in the lubricating oil composition of the present invention, an amide compound (p = 1 and q = 0 in the general formula (2)) or a urea compound (p = 1, q = 1, and r = 0 in the general formula (2)) can be especially preferably used in view of better friction reducing effect under boundary lubricating conditions. It should be noted that, the inventor of the present invention presumes as follows regarding the reason why the lubricating oil composition of the present invention exhibits an improved friction reducing performance under boundary lubricating conditions. That is: an h-BN sheet-like film including boron and nitrogen is formed on a friction surface by the (B) component which is the borazine compound; the film is protected by the (C) component which is an ashless friction modifier having a nitrogen atom in a polar group interacting with the boron atoms of the film; and the ashless friction modifier ((C) component) itself contributes to the friction reduction effect, whereby the friction reducing effect is synergistically shown.

The content of the friction modifier represented by the above general formula (2) in the lubricating oil composition of the present invention is, on the basis of the total amount of the composition, normally 0.01 mass% or more, preferably 0.05 mass% or more, and especially preferably 0.1 mass% or more; and normally 10.0 mass% or less, more preferably 5.0 mass% or less, and especially preferably 3.0 mass% or less.

As the ashless dispersant, a known ashless dispersant can be used. In a case where an ashless dispersant is contained in the lubricating oil composition of the present invention, the content thereof is, on the basis of the total amount of the composition as 100 mass%, normally 0.01 mass% or more, preferably 0.1 mass% or more; and preferably 20 mass% or less, and preferably 10 mass% or less.

As the antioxidant, a known antioxidant can be used. In a case where an antioxidant is contained in the lubricating oil composition of the present invention, the content thereof is, on the basis of the total amount of the lubricating oil composition, normally 5.0 mass% or less, preferably 3.0 mass% or less; and preferably 0.1 mass% or more, and more preferably 0.5 mass% or more.

As the friction modifier other than the (B) component and (C) component, a friction-reducing agent, a metallic detergent, a viscosity index improver, a pour point depressant, a corrosion inhibitor, an anti-rust agent, a metal deactivator, a defoamer, and a coloring agent. These additives may be contained in the borazine-based additive together with the borazine compound represented by the general formula (1).
As the friction-reducing agent, a known friction-reducing agent can be used. For example, phosphorus compounds such as (mono, di, or tri-thio) phosphorous or phosphoric triesters and zinc dithiophosphate, and sulfur-containing compounds such as disulfides, olefin sulfides, sulfurized fatty oils, and dithiocarbamates can be given. In a case where these friction-reducing agents are contained in the lubricating oil composition of the present invention, the content thereof is, on the basis of the total amount of the lubricating oil composition, normally 0.005 mass% or more and 5 mass% or less.

As the metallic detergent, a known metallic detergent can be used. For example, alkali metal sulfonates, alkali earth metal sulfonates, alkali metal phenates, alkaline earth metal phenates, alkali metal salicylates, alkaline earth metal salicylates, and mixture thereof can be given. These metallic detergents may be overbased. In a case where these metallic detergents are contained in the lubricating oil composition of the present invention, the content thereof is not particularly limited. In a case where the lubricating oil composition is for internal combustion engines, the content thereof is, on the basis of the total amount of the lubricating oil composition, normally 0.01 mass% or more and 5 mass% or less in terms of metal element.

As the viscosity index improver, a known viscosity index improver can be used. For example, a so-called non-dispersive viscosity index improver such as a polymer or copolymer of monomers of one or two or more selected from various methacrylic acid esters and hydrogenated products thereof, a so-called dispersive viscosity index improver obtainable by copolymerizing various methacrylic acid esters including a nitrogen compound, a non-dispersive or dispersive ethylene-α-olefin copolymer and a hydrogenated product thereof, polyisobutylene and a hydrogenated product thereof, a hydrogenated product of styrene-diene copolymer, styrene-maleic anhydride ester copolymer, polyalkylstyrene, and the like can be given. The weight average molecular weight of the viscosity index improver is, for example in a case where a dispersive or non-dispersive polymetacrylate is used, normally 5,000 or more and 1,000,000 or less. For example in a case where polyisobutylene or a hydrogenated product thereof is used for an internal combustion engine, the number average molecular weight is normally 800 or more and 5,000 or less. For example in a case where the ethylene-α-olefin copolymer or a hydrogenated product thereof is used for an internal combustion engine, the number average molecular weight is 800 or more and 500,000 or less.

In a case where these viscosity index improvers are contained in the lubricating oil composition of the present invention, the content thereof is, on the basis of the total amount of the lubricating oil composition, normally 0.1 mass% or more and 20 mass% or less.

As the pour point depressant, a known pour point depressant such as a polymethacrylate-based polymer can be adequately used depending on the property of the lubricating base oil to be used. In a case where the pour point depressant is contained in the lubricating oil composition of the present invention, the content thereof is, on the basis of the total amount of the lubricating oil composition, normally 0.01 to 1 mass%.

As the corrosion inhibitor, a known corrosion inhibitor such as a benzotriazole-based compound, tolyltriazole-based compound, thiadiazole-based compound, and imidazole-based compound can be used. In a case where these corrosion inhibitors are contained in the lubricating oil composition of the present invention, the content thereof is, on the basis of the total amount of the lubricating oil composition, normally 0.005 mass% or more and 5 mass% or less.

As the anti-rust agent, a known anti-rust agent such as a petroleum sulfonate, alkylbenzene sulfonate, dinonylnaphthalene sulfonate, alkylsuccinic acid ester, and polyhydric alcohol ester can be used. In a case where these anti-rust agents are contained in the lubricating oil composition of the present invention, the content thereof is, on the basis of the total amount of the lubricating oil composition, normally 0.005 mass% or more and 5 mass% or less. In a case where polyisobutylene or a hydrogenated product thereof is used for an internal combustion engine, the number average molecular weight is normally 800 or more and 5,000 or less.

As the anti-emulsifier, a known anti-emulsifier such as a polyalkylene glycol nonionic surfactant such as polyoxyethylene alkyl ether, polyoxyethylene alkyl phenyl ether, and polyoxyethylene alkyl naphthyl ether can be used. In a case where these anti-emulsifiers are contained in the lubricating oil composition of the present invention, the content thereof is, on the basis of the total amount of the lubricating oil composition, normally 0.005 mass% or more and 5 mass% or less.

As the metal deactivator, a known metal deactivator such as imidazoline, pyrimidine derivative, alkythiadiazole, mercaptobenzothiazole, benzotriazole and derivatives thereof, 1,3,4-thiadiazole polsulfide, 1,3,4-thiadiazolyl-2, 5-bis-dialkyl dithiocarbamate, 2-(alkylthio)benzimidazole, and αβ-(o-carboxybenzyl)propionitrile can be used. In a case where these metal deactivators are contained in the lubricating oil composition of the present invention, the content thereof is, on the basis of the total amount of the lubricating oil composition, normally 0.005 mass% or more and 5 mass% or less.

As the defoamer, a known defoamer such as a silicone, fluoro silicone, and fluoroalkyl ether can be used. In a case where these defoamers are contained in the lubricating oil composition of the present invention, the content thereof is, on the basis of the total amount of the lubricating oil composition, normally 0.0005 mass% or more and 1 mass% or less.

As the coloring agent, a known coloring agent such as an azo compound can be used.

The lubricating oil composition according to the second aspect of the present invention has an improved friction-reducing performance by containing the (B) component (the borazine-based additive); therefore it can be preferably employed for lubrication of various machines. In addition, it has an improved high-temperature detergency as described above; therefore, it can be especially preferably used for lubrication of machines whose metal parts are exposed to a
high temperature (e.g. internal combustion engines). Specifically, it can be especially preferably used for lubrication of internal combustion engines in which phosphorus and sulfur content and the like that migrates into the exhaust gas need to be taken into consideration (e.g. internal combustion engine equipped with an exhaust gas processing apparatus).

By containing the borazine-based additive as the (B) component in addition to the (C) component which is the oiliness agent based friction modifier, the lubricating oil composition according to the third aspect of the present invention has a further improved friction reducing performance under boundary lubricating conditions, compared to a case in which the (C) component is contained alone in the lubricating oil composition. Therefore, the lubricating oil composition according to the third aspect of the present invention can be preferably employed for lubrication of various machines. Especially, since the (B) component does not contain sulfur or phosphorus, it is possible to improve the friction reducing performance under boundary lubricating conditions with suppressed increase of sulfur and phosphorus content in the lubricating oil composition. Therefore, it can be especially preferably used for lubrication of internal combustion engines, specifically for lubrication of internal combustion engines in which sulfur and phosphorus content and the like that migrates into the exhaust gas are desired to be suppressed (e.g. internal combustion engine equipped with an exhaust gas processing apparatus).

Examples

Hereinafter the present invention will be further specifically described with reference to Examples and Comparative Examples. However, the present invention is not limited to Examples.

<Example 1 and Comparative Examples 1 and 2>

As shown in Table 1, the lubricating composition according to the second aspect of the present invention (Example 1), and lubricating oil compositions for comparison (Comparative Examples 1 and 2) were each prepared. To all values of amount of components is applied a unit of mass% (on the basis of the total amount of composition).

(Evaluation of friction-reducing performance)

For each of the lubricating oil compositions of Example 1 and Comparative Examples 1 and 2, friction-reducing performance was evaluated by means of a ball-on-disk reciprocating friction test machine (SRV friction test machine manufactured by Optimol Instruments, ball: 12.7 mm of diameter; disk: 24 mm of diameter and 7 mm of thickness, material of the ball and disk corresponds to SUJ-2), under conditions of a temperature of 100°C, frequency of 50 Hz, load of 20N, amplitude of 1 mm, and examination time of 30 minutes. The value of load 20N corresponds to a boundary lubricating condition. The wear scar diameter of ball after the examination is also shown in Table 1.

As can be seen from Table 1, the lubricating oil composition of Example 1 which includes the borazine-based additive of the present invention was able to reduce the wear scar diameter by approximately 38%, compared to the lubricating oil of Comparative Example 1 consisting of only the base oil. In addition, compared to the lubricating oil composition of Comparative Example 2 consisting of the base oil and zinc dithiophosphate, the lubricating oil composition of Example 1 was able to reduce the wear scar diameter by approximately 8%.

From the above results, it was shown that the lubricant additive and lubricating oil composition of the present invention could improve friction-reducing performance.
As shown in Table 2, the lubricating oil composition according to the second aspect of the present invention (Example 2), and a lubricating oil composition for comparison (Comparative Example 3) were each prepared. To all values of amount of components is applied a unit of mass% (on the basis of the total amount of composition).

<table>
<thead>
<tr>
<th></th>
<th>Example 2</th>
<th>Comparative Example 3</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Base oil</strong></td>
<td>Group I base oil A (*1)</td>
<td>balance (*3)</td>
</tr>
<tr>
<td></td>
<td>Group I base oil B (*2)</td>
<td></td>
</tr>
<tr>
<td><strong>Additive</strong></td>
<td>2,4,6-Triphenylborazine (*4)</td>
<td>0.37</td>
</tr>
<tr>
<td></td>
<td>Viscosity index improver</td>
<td>5.3</td>
</tr>
<tr>
<td></td>
<td>Additive package (*5)</td>
<td>9.0</td>
</tr>
<tr>
<td><strong>Kinematic viscosity at 40°C (mm²/s)</strong></td>
<td>70.2</td>
<td>70.2</td>
</tr>
<tr>
<td><strong>Kinematic viscosity at 100°C (mm²/s)</strong></td>
<td>10.7</td>
<td>10.7</td>
</tr>
<tr>
<td><strong>Elemental analysis (mass%)</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>0.06</td>
<td>0.03</td>
</tr>
<tr>
<td>Ca</td>
<td>0.26</td>
<td>0.26</td>
</tr>
<tr>
<td>P</td>
<td>0.07</td>
<td>0.07</td>
</tr>
<tr>
<td>Zn</td>
<td>0.08</td>
<td>0.08</td>
</tr>
<tr>
<td>N</td>
<td>0.1</td>
<td>0.05</td>
</tr>
<tr>
<td>S</td>
<td>0.43</td>
<td>0.43</td>
</tr>
<tr>
<td><strong>HTT merit (280°C)</strong></td>
<td>8.5</td>
<td>7.5</td>
</tr>
<tr>
<td><strong>HTT merit (290°C)</strong></td>
<td>7.5</td>
<td>6.5</td>
</tr>
</tbody>
</table>

(*1) Solvent refined mineral oil, sulfur content: 0.12%, saturates content: 75%, viscosity index: 100
(*2) Solvent refined mineral oil, sulfur content: 0.58%, saturates content: 62%, viscosity index: 97
(*3) Base oil A / Base oil B = 73/27
(*4) R1=R2=R5=H and R2=R4=R6=Ph in the general formula (1)
(*5) Consisting of ZnDTP, metallic detergent, ashless dispersant, antioxidant, and pour point depressant

(Evaluation of high-temperature detergency)

For each of the lubricating oil compositions of Example 2 and Comparative Example 3, high-temperature detergency was evaluated by means of a hot tube test (conforming to JPI-5S-55 standard). Examinations were carried out at 280°C and 290°C. Results are also shown in Table 2.

As can be seen from Table 2, the lubricating oil composition of Example 2 containing the borazine-based additive of the present invention showed good hot tube merits at both examination temperatures of 280°C and 290°C, compared to the lubricating oil composition of the Comparative Example 3 which is same as the Example 2 except that the borazine-based additive of the present invention is not contained.

From these results, it was shown that the lubricant additive and lubricating oil composition according to the present invention could improve high-temperature detergency.

As shown in Table 3, lubricating oil compositions according to the third aspect of the present invention (Examples 3 and 4), lubricating oil compositions according to the second aspect of the present invention but not according to the third aspect of the present invention (Examples 5 and 6), and lubricating oil compositions for comparison (Comparative Examples 4 and 5) were each prepared. In Table 3, to all values of amount of components is applied a unit of mass% (on the basis of the total amount of composition).
<table>
<thead>
<tr>
<th>Base oil</th>
<th>Example 3</th>
<th>Example 4</th>
<th>Example 5</th>
<th>Example 6</th>
<th>Comparative Example 4</th>
<th>Comparative Example 5</th>
</tr>
</thead>
<tbody>
<tr>
<td>(A) PAO (*1)</td>
<td>balance</td>
<td>balance</td>
<td>balance</td>
<td>balance</td>
<td>balance</td>
<td>balance</td>
</tr>
<tr>
<td>(B) 2,4,6-triphenylborazine (*2)</td>
<td>0.37</td>
<td>0.37</td>
<td>0.37</td>
<td>0.37</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>(C1) oleylurea (*3)</td>
<td>0.55</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.55</td>
<td>-</td>
</tr>
<tr>
<td>(C2) oleylamide (*4)</td>
<td>-</td>
<td>1.00</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>1.00</td>
</tr>
<tr>
<td>(C3) glycerol monooleate</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Friction coefficient</td>
<td>0.109</td>
<td>0.114</td>
<td>0.224</td>
<td>0.176</td>
<td>0.146</td>
<td>0.164</td>
</tr>
</tbody>
</table>

(*1) Poly α-olefin base oil, kinematic viscosity at 100°C: 1.7 mm²/s
(*2) In the general formula (1), R₁=R³=R₅=H and R²=R⁴=R⁶=Ph
(*3) In the general formula (2), p=1, q=1, r=0, R⁷=-(CH₂)₇CH=CH(CH₂)₇CH₃, and R⁸=R⁹=R¹₀=H
(*4) In the general formula (2), p=1, q=0, R⁷=-(CH₂)₇CH=CH(CH₂)₇CH₃, and R⁹=R¹₀=H
For each of the lubricating oil compositions of Examples 3 to 6 and Comparative Examples 4 and 5, friction-reducing performance was evaluated by means of a ball-on-disk type reciprocating friction test machine (SRV friction test machine manufactured by Optimol Instruments, ball: 12.7 mm of diameter; disk: 24 mm of diameter and 7 mm of thickness, material of the ball and disk corresponds to SUJ-2). A running-in examination was carried out for 30 minutes under friction conditions of a temperature of 100 °C, frequency of 50 Hz, load of 20N, and amplitude of 1 mm; and thereafter, a measurement of friction coefficient was carried out under friction conditions of a temperature of 100°C, frequency of 10 Hz, load of 20N, and amplitude of 1 mm. The value of load 20N corresponds to a boundary lubricating condition. Measurement results of friction coefficient are also shown in Table 3.

As can be seen from Table 3, the lubricating oil compositions of Examples 3 and 4 were able to reduce the friction coefficient by approximately 49% or more, compared to the lubricating oil composition of Example 5 not containing the (C) component. Compared to the lubricating oil compositions of Example 6 containing glycerol monooleate which is an oiliness agent based friction modifier which does not fall under the (C) component, the lubricating oil compositions of Examples 3 and 4 were able to reduce the friction coefficient by approximately 35% or more. In addition, compared to the lubricating oil compositions of Comparative Examples 4 and 5 not containing the (B) component, the lubricating oil compositions of Example 3 and 4 were able to reduce the friction coefficient by approximately 22% or more.

From these results, it was shown that the lubricating oil composition according to the third aspect of the present invention could improve friction reducing performance under boundary lubricating conditions.

**Claims**

1. A lubricant additive comprising:
   a borazine compound represented by the following general formula (1):

   ![Borazine Compound](image)

   (In the formula (1), R1, R3, and R5 are each independently hydrogen, a C1 to C30 hydrocarbyl group, or a C1 to C30 hydrocarbyl group comprising a heteroatom other than sulfur and phosphorus; and R2, R4, and R6 are each independently hydrogen, a C1 to C30 hydrocarbyl group, or a C1 to C30 hydrocarbyl group comprising oxygen or boron or nitrogen.).

2. The lubricant additive according to claim 1, wherein in the formula (1) R1, R3, and R5 are each independently hydrogen, a C1 to C30 hydrocarbyl group, or a C1 to C30 hydrocarbyl group comprising a heteroatom other than sulfur and phosphorus; and R2, R4, and R6 are each independently hydrogen, a C1 to C30 hydrocarbyl group, or a C1 to C30 hydrocarbyl group comprising oxygen or boron or nitrogen.

3. The lubricant additive according to claim 1 or 2, wherein in the formula (1) R1, R3, and R5 are each independently hydrogen, a C1 to C30 hydrocarbyl group, or a C1 to C30 hydrocarbyl group comprising oxygen or boron or nitrogen.

4. The lubricant additive according to any one of claims 1 to 3, wherein in the formula (1), R1, R3, and R5 are the same groups; and R2, R4, and R6 are the same groups.

5. A lubricating oil composition comprising:
   (A) a lubricating base oil; and
   (B) the lubricant additive according to any one of claims 1 to 4.

6. The lubricating oil composition according to claim 5, further comprising:
(C) a friction modifier represented by the following general formula (2):

\[
R^7 \underset{O} \overset{N} \overset{\text{r}} \underset{\text{q}} \overset{N} \overset{R^8} \underset{R^9} \overset{R^{10}} \text{(2)}
\]

(In the formula (2), p is 0 or 1; q is 0 or 1; r is 0 or 1; \(R^7\) is a hydrocarbyl group having no less than 8 carbon atoms; and \(R^8, R^9\) and \(R^{10}\) are each independently hydrogen or a C1 to C4 hydrocarbyl group.)

7. The lubricating oil composition according to claim 6, wherein in the formula (2), at least \(R^8\) and \(R^{10}\) are hydrogen; and where p and q are 1, \(R^8, R^9,\) and \(R^{10}\) are hydrogen.

8. The lubricating oil composition according to any one of claims 5 to 7, further comprising:

one or more selected from the group consisting of an ashless dispersant, an antioxidant, a friction modifier, a friction-reducing agent, a metallic detergent, a viscosity index improver, a pour point depressant, a corrosion inhibitor, an anti-rust agent, an anti-emulsifier, a metal deactivator, a defoamer, and a coloring agent.

9. The lubricating oil composition according to any one of claims 5 to 8, which is a lubricating oil composition for internal combustion engines.
**INTERNATIONAL SEARCH REPORT**

**International application No.** PCT/JP2013/077121

A. **CLASSIFICATION OF SUBJECT MATTER**

C10M139/00(2006.01); C10M107/02(2006.01); C10M133/16(2006.01); C10N30/06(2006.01); C10N40/25(2006.01)

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

B. **FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

C10M139/00; C10M107/02; C10M133/16; C10N30/06; C10N40/25

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho 1922-1996  Jitsuyo Shinan Toroku Koho 1996-2013

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Electronic data base consulted during the international search (name of data base and; where practicable, search terms used)

CPlus/REGISTRY(STN), JSTPlus/JMEDPlus/JST7580(JDreamIII)

C. **DOCUMENTS CONSIDERED TO BE RELEVANT**

<table>
<thead>
<tr>
<th>Category*</th>
<th>Citation of document, with indication, where appropriate, of the relevant passages</th>
<th>Relevant to claim No.</th>
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<td>JP 40-20713 B1 (Farbenfabriken Bayer AG.), 14 September 1965 (14.09.1965), entire text (Family: none)</td>
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<td>04 November 1964 (04.11.1964), entire text &amp; DE 1163815 B</td>
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<td>GB 984806 A (FARBENFABRIKEN BAYER AG), 03 March 1965 (03.03.1965), entire text (Family: none)</td>
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X: Further documents are listed in the continuation of Box C.

See patent family annex.

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Date of the actual completion of the international search

20 December, 2013 (20.12.13)

Date of mailing of the international search report

07 January, 2014 (07.01.14)

Name and mailing address of the ISA/

Japanese Patent Office

Authorized officer

Facsimile No.

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<td>GB 1003090 A (FARBENFABRIKEN BAYER AG), 02 September 1965 (02.09.1965), entire text &amp; US 3255245 A &amp; US 3297750 A &amp; DE 1147944 B &amp; DE 1161890 B</td>
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REFERENCES CITED IN THE DESCRIPTION

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