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(54) LUBRICATING OIL COMPOSITION

(57) A lubricating oil composition including: a lubricant base oil having a kinematic viscosity at 40°C of 6.0 to 12.0 mm^2/s; (A) a succinimide dispersant having a polyisobutenyl group having a number average molecular weight of no less than 800, in an amount of no less than 80 mass ppm in terms of nitrogen and no more than 2.7 mass% as the entire compound; (B) a condensation reaction product of an alkyl- or alkenyl- succinic acid having a C8-30 alkyl or alkenyl group or anhydride thereof and a polyamine, or a modified product thereof, or any combination thereof, in an amount of 50 to 1300 mass ppm in terms of nitrogen, wherein the product of the weight average molecular weight of the component (A) and the amount of the component (A) as the entire compound (mass%) is no more than 16,000.

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Description

FIELD

[0001] The present invention relates to a lubricating oil composition, and more specifically, to a lubricating oil composition that can be preferably used for lubrication of automatic transmissions and/or electric motors.

BACKGROUND

[0002] One means of improving energy efficiency of gears such as transmissions and final drive gears is to use a less viscous lubricating oil. For example, a transmission, a final drive, or the like is equipped with a gear bearing mechanism. To use a less viscous lubricating oil for this is considered to reduce churning resistance and drag torque which are caused by the viscosity resistance of a lubricating oil to improve power transmission efficiency, which can result in improved fuel efficiency.

CITATION LIST

Patent Literature

[0003]

Patent Literature 1: JP 2009-249496 A
Patent Literature 2: JP 2014-159496 A
Patent Literature 3: JP 2016-003258 A
Patent Literature 4: JP 2016-020454 A
Patent Literature 5: WO 2020/095968 A1
Patent Literature 6: WO 2020/095969 A1
Patent Literature 7: WO 2020/095970 A1
Patent Literature 8: WO 2020/171188 A1

SUMMARY

Technical Problem

[0004] In recent years, an electric vehicle with an electric motor as a power source for running, and a hybrid vehicle with an electric motor and an internal combustion engine both as a power source for running have been attracting interest in view of energy efficiency and environmental compatibility. An electric motor generates heat during operation thereof, whereas including heat-sensitive components such as coils and magnets. A means of cooling an electric motor is, thus, provided for the aforementioned types of vehicles each with an electric motor as a power source for running. Known means of cooling an electric motor are air cooling, water cooling, and oil cooling. Among them, oil cooling system is to circulate oil in an electric motor to directly bring components in the electric motor which generates heat (such as coils, cores, and magnets) into contact with a coolant (oil), which can yield a high cooling effect. Oil (lubricating oil) is circulated in an electric motor under the oil cooling system, and thereby, the electric motor is cooled and lubricated at the same time. Electrical insulation is required of lubricating oils (electric motor oils) of electric motors.

[0005] Generally, different lubricating oils are used for lubrication of electric motors and lubrication of transmissions, respectively. If an electric motor and a transmission (gear mechanism) could be lubricated with the same lubricating oil, a lubricating oil circulation system could be simplified. Disadvantageously, a conventional transmission oil suffers insufficient electrical insulation for the use for lubrication of electric motors. In addition, a conventional electric motor oil suffers insufficient durability against oxidative deterioration for the use for lubrication of transmissions (gear mechanisms).

[0006] Oxidative deterioration is one factor in determination of a lifetime of lubricating oil that is to be exposed to high temperatures at a certain level or higher, such as transmission oils and electric motor oils. A highly polar component generated by oxidative deterioration of lubricating oil not only tends to deposit as an insoluble matter, but also can deteriorate the electrical insulation of the lubricating oil. The increase in acid number following the progress of the oxidation deterioration can lead to corrosion of metal members.

[0007] A detergent dispersant is an important component for alleviating the foregoing problems resulting from oxidative deterioration of lubricating oil to improve long drain performance of the lubricating oil. A detergent dispersant is a concept encompassing ashless dispersants and metallic detergents. A metallic detergent is an organic acid metal salt (such as alkaline earth metal salicylates, alkaline earth metal sulfonates, and alkaline earth metal phenates) that can form micelles

in oil, or a mixture of such a metal salt and a metal base (such as oxides and hydroxides). An ashless dispersant usually has, in a molecule thereof, a polar group for interacting with highly polar components (such as amino groups), and a long-chain alkyl or alkenyl group having sufficient lipophilicity (such as polyisobutenyl groups) for dispersing highly polar components in oil. Specific examples of compounds that are used as an ashless dispersant include: condensation reaction products of an alkyl or alkenyl succinic acid or anhydride thereof, and a polyamine; and Mannich reaction products of an alkylphenol or alkenylphenol, formaldehyde, and a polyamine. An ashless dispersant does not form micelles in oil. Thus, a longer-chain alkyl or alkenyl group than a lipophilic group of an organic acid constituting a metallic detergent is necessary for securing the lipophilicity necessary for functions as an ashless dispersant. As such an alkyl or alkenyl group, an alkyl or alkenyl group (polyisobutenyl group) derived from a polyolefin obtained by polymerization of an olefin such as isobutene is preferably used. Thus, an ashless dispersant usually has a larger molecular weight than a metallic detergent.

[0008] A higher content of a metallic detergent in a lubricating oil tends to lead remarkable deterioration of the electrical insulation of a fresh oil thereof. Therefore, a lower content of a metallic detergent in a lubricating oil is desirable in view of securing electrical insulation necessary for lubrication of electric motors. It is necessary to increase the content of an ashless dispersant in a lubricating oil comprising no or a less metallic detergent for alleviating the problems resulting from the oxidative deterioration of the lubricating oil to enhance the long drain performance thereof. Nevertheless, an ashless dispersant more easily increases the viscosity of a lubricating oil than a metallic detergent does, whereas not deteriorating the electrical insulation of a fresh oil as much as a metallic detergent does. Therefore, a lower content of an ashless dispersant is desirable in view of enhancing the energy saving performance of a lubricating oil.

[0009] An object of the present invention is to provide a lubricating oil composition that has electrical insulation required for lubrication of electric motors while being a less viscous lubricating oil composition having improved energy saving performance, and that can alleviate the problems resulting from oxidative deterioration in lubrication of automatic transmissions and lubrication of electric motors to improve long drain performance.

Solution to Problem

[0010] The present invention encompasses the following embodiments [1] to [10].

[1] A lubricating oil composition comprising:

(O) a lubricant base oil comprising at least one mineral base oil or at least one synthetic base oil or any combination thereof, and having a kinematic viscosity at 40°C of 6.0 to 12.0 mm^2/s;
(A) a condensation reaction product of a polyisobutenylsuccinic acid or anhydride thereof and a first polyamine, or a modified product thereof, or any combination thereof, in an amount of no less than 80 mass ppm in terms of nitrogen and no more than 2.7 mass% as an entire compound on the basis of the total mass of the composition, the polyisobutenylsuccinic acid having a polyisobutenyl group, the polyisobutenyl group having a number average molecular weight of no less than 800;
(B) a condensation reaction product of an alkyl- or alkenyl- succinic acid having a C8-30 alkyl or alkenyl group or anhydride thereof and a second polyamine, or a modified product thereof, or any combination thereof, in an amount of 50 to 1300 mass ppm in terms of nitrogen on the basis of the total mass of the composition,
wherein a product of a weight average molecular weight (unit: Da) of the component (A) and the amount of the component (A) as the entire compound (unit: mass%) is no more than 16,000.

[2] The lubricating oil composition according to [1], further comprising:
(C) at least one calcium sulfonate detergent overbased with calcium carbonate, or at least one calcium salicylate detergent overbased with calcium carbonate, or any combination thereof, in an amount of no less than 10 mass ppm and less than 100 mass ppm in terms of calcium on the basis of the total mass of the composition.
[3] The lubricating oil composition according to [1] or [2], further comprising:
(D) at least one amine antioxidant and at least one phenol antioxidant, in an amount of 0.1 to 3.0 mass% as a total amount thereof, on the basis of the total mass of the composition.
[4] The lubricating oil composition according to any one of [1] to [3], further comprising:
optionally (E) at least one phosphorus-containing compound, or at least one sulfur-containing compound comprising at least one sulfur atom having a formal oxidation number of no more than +II in a molecule thereof, or any combination thereof, in an amount of no more than 1000 mass ppm in terms of a total amount of phosphorus content and sulfur content on the basis of the total mass of the composition.
[5] The lubricating oil composition according to any one of [1] to [4], wherein a relation of the following equation (1) is met,

$$B \geq \max(50, f(A))$$

$$f(A) = 100 \times \left(-10.5 + 14.1 \times \left(\frac{A}{100} - 0.5\right)^{-0.3}\right) \quad \cdots (1)$$

wherein in the equation (1), A represents the amount (unit: mass ppm) of the component (A) in terms of nitrogen on the basis of the total mass of the composition; and B represents the amount (unit: mass ppm) of the component (B) in terms of nitrogen on the basis of the total mass of the composition.

[6] The lubricating oil composition according to any one of [1] to [5], further comprising:
optionally (F) at least one polyalkyl (meth)acrylate having a weight average molecular weight of more than 25,000, in an amount of no more than 5.0 mass% on the basis of the total mass of the composition.

[0011] In the present description, "(meth)acrylate" means "acrylate and/or methacrylate".

[7] The lubricating oil composition according to any one of [1] to [6], further comprising:
optionally, at least one polymer having a weight average molecular weight of no more than 25,000, in an amount of less than 0.1 mass% on the basis of the total mass of the composition.

[8] The lubricating oil composition according to any one of [1] to [7], which is used to lubricate an automatic transmission.

[9] The lubricating oil composition according to any one of [1] to [8], which is used to lubricate an electric motor.

[10] A method for lubricating an automatic transmission and an electric motor,

the automatic transmission and the electric motor being comprised in an automobile,
the method comprising:
supplying the lubricating oil composition as defined in any one of [1] to [9] to the automatic transmission of the automobile; and
supplying the lubricating oil composition to the electric motor of the automobile.

Advantageous Effects

[0012] The present invention can be provided with a lubricating oil composition that has electrical insulation required for lubrication of electric motors while being a less viscous lubricating oil composition having improved energy saving performance, and that can alleviate the problems resulting from oxidative deterioration in lubrication of automatic transmissions and lubrication of electric motors to improve extended drain performance.

Description of Embodiments

[0013] The present invention will be hereinafter described. In the present description, expression "A to B" concerning numerical values A and B shall be equivalent to "no less than A and no more than B" unless otherwise specified. In such expression, if a unit is added only to the numerical value B, the same unit shall be applied to the numerical value A. In the present description, the word "or" shall mean a logical sum unless otherwise specified. In the present description, expression "E_1 and/or E_2" concerning elements E_1 and E_2 shall be equivalent to "E_1, or E_2, or the combination thereof", and expression "E_1, ..., and/or E_N" concerning n elements E_1, ..., E_i, ..., E_N (where N is an integer of 3 or more) shall be equivalent to "E_1, ..., or Ei, ..., or E_N, or any combination thereof" (where i is a variable that takes any of integers satisfying $1 < i < N$). In the present description, "alkaline earth metal" encompasses magnesium.

[0014] In the present description, unless otherwise specified, the content of each of the elements of calcium, magnesium, zinc, phosphorus, sulfur, boron, barium, and molybdenum in an oil shall be measured conforming to JIS K0116 by inductively coupled plasma atomic emission spectrometry (intensity ratio method (internal standard method)), and the content of a nitrogen element in an oil shall be measured conforming to JIS K2609 by a chemiluminescence method. In the present description, "weight average molecular weight" and "number average molecular weight" mean a weight average molecular weight and a number average molecular weight measured by gel permeation chromatography (GPC) in terms of standard polystyrene, respectively. The measurement conditions for GPC are as follows.

[GPC Measurement Conditions]

Device: ACQUITY™ APC UV RI System manufactured by Waters Corporation
Column: two columns of ACQUITY™ APC XT900A manufactured by Waters Corporation (gel particle size: 2.5 μm, column size (inner diameter × length): 4.6 mm×150 mm); and one column of ACQUITY™ APC XT200A manufactured by Waters Corporation (gel particle size: 2.5 μm, column size (inner diameter × length): 4.6 mm×150 mm), connected in series in that order from the upstream side
Column temperature: 40°C
Sample solution: tetrahydrofuran solution of a sample with a concentration of 1.0 mass%
Solution injection volume: 20.0 μL
Eluent: tetrahydrofuran
Detector: differential refractometer
Standard material: standard polystyrene (Agilent EasiCal™ PS-1 manufactured by Agilent Technologies, Inc.) (8 points of molecular weights: 2698000, 597500, 290300, 133500, 70500, 30230, 9590 and 2970)

[0015] If the weight average molecular weight measured based on the foregoing conditions is less than 10000, the columns and the standard material are changed according to the following conditions, and then, the weight average molecular weight is measured again.

Column: one column of ACQUITY™ APC XT125A manufactured by Waters Corporation (gel particle size: 2.5 μm, column size (inner diameter × length): 4.6 mm×150 mm); and two columns of ACQUITY™ APC XT45A manufactured by Waters Corporation (gel particle size: 1.7 μm, column size (inner diameter × length): 4.6 mm×150 mm), connected in series in that order from the upstream side

Standard material: standard polystyrene (Agilent EasiCal™ PS-1 manufactured by Agilent Technologies, Inc.) (10 points of molecular weights: 30230, 9590, 2970, 890, 786, 682, 578, 474, 370 and 266)

<(O) Lubricant Base Oil>

[0016] A lubricating oil composition according to the present invention (hereinafter may be referred to as "lubricating oil composition" or "composition") comprises a lubricant base oil of a major amount, and at least one additive other than the base oil. In the lubricating oil composition according to the present invention, a lubricant base oil comprising at least one mineral base oil or at least one synthetic base oil or any combination thereof, and having a kinematic viscosity at 40°C of 6.0 to 12.0 mm^2/s (hereinafter may be referred to as "component (O)") is used as the lubricant base oil.

[0017] At least one mineral base oil, at least one synthetic base oil, or any mixed base oil thereof can be used as the lubricant base oil. In one embodiment, as the lubricant base oil, a Group I base oil of API base stock categories (hereinafter may be referred to as "API Group I base oil"), a Group II base oil thereof (hereinafter may be referred to as "API Group II base oil"), a Group III base oil thereof (hereinafter may be referred to as "API Group III base oil"), a Group IV base oil thereof (hereinafter may be referred to as "API Group IV base oil"), or a Group V base oil thereof (hereinafter may be referred to as "API Group V base oil"), or any mixed base oil thereof can be used. The API Group I base oil is a mineral base oil containing more than 0.03 mass% sulfur and/or less than 90 mass% saturates, and having a viscosity index of no less than 80 and less than 120. The API Group II base oil is a mineral base oil containing no more than 0.03 mass% sulfur and no less than 90 mass% saturates, and having a viscosity index of no less than 80 and less than 120. The API Group III base oil is a mineral base oil containing no more than 0.03 mass% sulfur and no less than 90 mass% saturates, and having a viscosity index of no less than 120. The API Group IV base oil is a poly-α-olefin base oil. The API Group V base oil is a base oil other than the Groups I to IV base oils, and preferred examples thereof include ester base oils. In the present description, the viscosity index means a viscosity index measured conforming to JIS K 2283-2000. In the present description, "sulfur content in the lubricant base oil" shall be measured conforming to JIS K 2541-2003. In the present description, "saturated content in the lubricant base oil" means a value measured conforming to ASTM D 2007-93.

[0018] In one embodiment, as the component (O), at least one API Group II base oil, at least one API Group III base oil, at least one API Group IV base oil, or at least one API Group V base oil, or any combination thereof can be preferably used.

[0019] Examples of the mineral base oil herein include paraffinic mineral oils, normal-paraffinic base oils, and isoparaffinic base oils which are refined with lubricating oil fractions that are obtained by atmospheric distillation and/or vacuum distillation of crude oil through one, or two or more in combination selected from refining processes such as solvent deasphalting, solvent extraction, hydrocracking, solvent dewaxing, catalytic dewaxing, hydrorefining, sulfuric acid washing, and white clay treatment, and any mixtures thereof. Normally, the API Group II base oil and the Group III base oil are produced via hydrocracking.

[0020] Preferred examples of the mineral base oil include base oils obtained by: refining a raw material base oil of any one of the following base oils (1) to (8) and/or lubricating oil fractions recovered from this raw material base oil by

a predetermined refining method; and recovering the refined lubricating oil fractions.

(1) a distillate obtained by atmospheric distillation of a paraffin base crude oil and/or a mixed base crude oil
(2) a distillate obtained by vacuum distillation of a reduced crude of a paraffin base crude oil and/or a mixed base crude oil (WVGO)
(3) a wax (such as slack wax) obtained through a lubricating oil dewaxing step, and/or a synthetic wax (such as FT wax and gas-to-liquid (GTL) wax) obtained through, for example, a Fischer-Tropsch (FT) process
(4) at least one base oil selected from the base oils (1) to (3), or a mixed oil of at least two selected from the base oils (1) to (3), or a mild hydrocracked oil or mixed oil thereof
(5) a mixed oil of at least two selected from the base oils (1) to (4)
(6) a deasphalted oil (DAO) of the base oil (1), (2), (3), (4) or (5)
(7) a mild hydrocracked oil (MHC) of the base oil (6)
(8) a mixed oil of at least two selected from the base oils (1) to (7)

[0021] Preferred examples of the above-described predetermined refining method include: hydrorefining such as hydrocracking and hydrofinishing; solvent refining such as furfural solvent extraction; dewaxing such as solvent dewaxing and catalytic dewaxing; white clay treatment using acid white clay, activated white clay, or the like; and chemical (acid or alkali) washing such as sulfuric acid washing and caustic soda washing. One of these refining methods may be used alone, or at least two of them may be used in combination. When at least two of the refining methods are used in combination, the order of using them is not specifically restricted, but can be suitably determined.

[0022] The following base oil (9) or (10) is especially preferable as the mineral base oil. The base oil (9) or (10) is obtained through a predetermined process on a base oil selected from the base oils (1) to (8), or on lubricating oil fractions recovered from the selected base oil.

[0023] (9) a hydrocracked base oil obtained by: hydrocracking a base oil selected from the base oils (1) to (8), or lubricating oil fractions recovered from the selected base oil; dewaxing, through a dewaxing process such as solvent dewaxing and catalytic dewaxing, the hydrocracked product, or lubricating oil fractions recovered therefrom by, for example, distillation; and optionally further distilling the dewaxed product

[0024] (10) a hydroisomerized base oil obtained by: hydroisomerizing a base oil selected from the base oils (1) to (8), or lubricating oil fractions recovered from the selected base oil; dewaxing, through a dewaxing process such as solvent dewaxing and catalytic dewaxing, the hydroisomerized product, or lubricating oil fractions recovered therefrom by, for example, distillation; and optionally further distilling the dewaxed product. A base oil produced via catalytic dewaxing as the dewaxing process is preferable.

[0025] A solvent refining process and/or hydrofinishing process may be further performed at a proper stage if necessary when the lubricant base oil (9) or (10) is obtained.

[0026] The catalyst used for the above-described hydrocracking or hydroisomerization is not specifically restricted. Preferably used is a hydrocracking catalyst that includes a metal having a hydrogenating ability (such as at least one of the group VIa and group VIII metals of the periodic table) and supported on a catalyst support including any of composite oxides having a cracking activity (such as silica-alumina, alumina-boria and silica-zirconia), and optionally further including a binder binding at least one of the composite oxides in combination; or a hydroisomerization catalyst that includes a metal having a hydrogenation ability, including at least one group VIII metal, and supported on a catalyst support including a zeolite (such as ZSM-5, zeolite beta, and SAPO-11). Such hydrocracking catalyst and hydroisomerization catalyst may be used in combination by stacking, mixing, or the like.

[0027] The reaction conditions upon the hydrocracking or hydroisomerization are not specifically restricted. Preferably, the hydrogen partial pressure is 0.1 to 20 MPa, the average reaction temperature is 150 to 450°C, LHSV is 0.1 to 3.0 hr^{-1}, and the hydrogen/oil ratio is 50 to 20000 scf/b.

[0028] The $\%C_P$ of the mineral base oil is preferably no less than 60, and more preferably no less than 65 in view of further improving the viscosity-temperature characteristics of the composition, and fuel efficiency; is preferably no more than 99, more preferably no more than 95, and further preferably no more than 94 in view of improving the solubility of the additive(s); and in one embodiment, can be 60 to 99, 60 to 95, 65 to 95, or 65 to 94.

[0029] The $\%C_A$ of the mineral base oil is preferably no more than 2, more preferably no more than 1, further preferably no more than 0.8, and especially preferably no more than 0.5 in view of further improving the viscosity-temperature characteristics of the composition, and fuel efficiency.

[0030] The $\%C_N$ of the mineral base oil is preferably no less than 1, and more preferably no less than 4 in view of improving the solubility of the additive(s); is preferably no more than 40, and more preferably no more than 35 in view of further improving the viscosity-temperature characteristics of the composition and fuel efficiency; and in one embodiment, can be 1 to 40, or 4 to 35.

[0031] In the present description, $\%C_P$, $\%C_N$ and $\%C_A$ mean a percentage of the paraffinic carbons to the total carbons, a percentage of the naphthenic carbons to the total carbons, and a percentage of the aromatic carbons to the total

carbons, respectively, obtained by the method conforming to ASTM D 3238-85 (ring analysis by the n-d-M method). That is, the foregoing preferred ranges of the $\%C_P$, $\%C_N$ and $\%C_A$ are based on values obtained according to this method. For example, the value of the $\%C_N$ obtained according to this method can be more than 0 even if the lubricant base oil has no naphthene content.

[0032] The saturated content in the mineral base oil is preferably no less than 90 mass%, more preferably no less than 95 mass%, and further preferably no less than 99 mass% on the basis of the total mass of the base oil in view of improving the viscosity-temperature characteristics of the composition. In the present description, the saturated content means a value measured conforming to ASTM D 2007-93.

[0033] For the separation method for the saturated content, any similar method that yields the same results can be used. Examples of such a method include the above-described method described in ASTM D 2007-93, the method described in ASTM D 2425-93, the method described in ASTM D 2549-91, methods using high performance liquid chromatography (HPLC), and improved methods thereof.

[0034] The aromatic content in the mineral base oil on the basis of the total mass of the base oil is preferably 0 to 10 mass%, more preferably 0 to 5 mass%, and especially preferably 0 to 1 mass%; and in one embodiment, can be no less than 0.1 mass%. The aromatic content no more than the foregoing upper limit can improve the low-temperature viscosity characteristics and the viscosity-temperature characteristics of the fresh oil, can further improve fuel efficiency, and can reduce the evaporation loss of the lubricating oil to reduce the consumption of the lubricating oil; and allows an additive to effectively work when the additive is incorporated to the lubricant base oil. The lubricant base oil may have no aromatic content, whereas the aromatic content no less than the foregoing lower limit can improve the solubility of the additive.

[0035] In the present description, the aromatic content means a value measured conforming to ASTM D 2007-93. Generally, the aromatic content includes alkylbenzenes and alkylnaphthalenes; anthracenes, phenanthrenes and alkylated compounds thereof; and further, compounds each having four or more fused benzene rings; and aromatic compounds each having a heteroatom, such as pyridines, quinolines, phenols, and naphthols.

[0036] Examples of the API Group IV base oil include oligomers and co-oligomers of a C2-32, preferably a C6-16 α-olefin, and hydrogenated products thereof: such as ethylene-propylene copolymers, polybutene, 1-octene oligomers, and 1-decene oligomers, and hydrogenated products thereof.

[0037] Preferred examples of the API Group V base oil include ester base oils such as: monoesters (such as butyl stearate, octyl laurate, and 2-ethylhexyl oleate); diesters (such as ditridecyl glutarate, bis(2-ethylhexyl) adipate, diisodecyl adipate, ditridecyl adipate, and bis(2-ethylhexyl) sebacate); polyvalent carboxylic acid esters (such as trimellitate esters); and polyol esters (such as trimethylolpropane caprylate, trimethylolpropane pelargonate, pentaerythritol 2-ethylhexanoate, and pentaerythritol pelargonate). Other examples of the API Group V base oil include aromatic synthetic base oils such as alkylbenzenes, alkylnaphthalenes, polyoxyalkylene glycol, dialkyl diphenyl ether, and polyphenyl ether.

[0038] The kinematic viscosity of the lubricant base oil (total base oil) at 40°C is no less than 6.0 mm^2/s, preferably no less than 6.5 mm^2/s, and more preferably no less than 7.0 mm^2/s in view of improving the electrical insulation of the fresh oil, and of sufficient oil film formation at a lubricating point to improve anti-wear performance; is no more than 12.0 mm^2/s in view of fuel efficiency, and the low-temperature viscosity characteristics of the lubricating oil composition; and in one embodiment, can be 6.0 to 12.0 mm^2/s, 6.5 to 12.0 mm^2/s, or 7.0 to 12.0 mm^2/s. In the present description, "kinematic viscosity at 40°C" means a kinematic viscosity at 40°C measured conforming to JIS K 2283-2000 by the use of an automated viscometer (trade name: "CAV-2100" manufactured by Cannon instrument company) as a measuring device.

[0039] The kinematic viscosity of the lubricant base oil (total base oil) at 100°C is preferably no less than 1.9mm^2/s, more preferably no less than 2.0 mm^2/s, and further preferably no less than 2.1 mm^2/s in view of further improving the electrical insulation of the fresh oil, and of sufficient oil film formation at a lubricating point to further improve anti-wear performance; is preferably no more than 3.5 mm^2/s, more preferably no more than 3.4 mm^2/s, and further preferably no more than 3.3 mm^2/s in view of further improving fuel efficiency; and in one embodiment, can be 1.9 to 3.5 mm^2/s, 2.0 to 3.4 mm^2/s, or 2.1 to 3.3 mm^2/s. In the present description, "kinematic viscosity at 100°C" means a kinematic viscosity at 100°C measured conforming to JIS K 2283-2000 by the use of an automated viscometer (trade name: "CAV-2100" manufactured by Cannon instrument company) as a measuring device.

[0040] The viscosity index of the lubricant base oil (total base oil) is preferably no less than 100, more preferably no less than 105, further preferably no less than 110, particularly preferably no less than 115, and most preferably no less than 120 in view of improving the viscosity-temperature characteristics of the composition, and of further improving fuel efficiency and anti-wear performance. In the present description, the viscosity index means a viscosity index measured conforming to JIS K 2283-2000 by the use of an automated viscometer (trade name: "CAV-2100" manufactured by Cannon instrument company) as a measuring device

[0041] The pour point of the lubricant base oil (total base oil) is preferably no more than -10°C, more preferably no more than -12.5°C, further preferably no more than -15°C, especially preferably no more than -17.5°C, and most preferably no more than -20.0°C in view of the low-temperature fluidity of the entire lubricating oil composition. In the present

description, the pour point means a pour point measured conforming to JIS K 2269-1987.

[0042] The sulfur content in the base oil depends on the sulfur content in the raw material thereof. For example, when a substantially sulfur-free raw material such as a synthetic wax component obtained through, for example, a Fischer-Tropsch reaction is used, a substantially sulfur-free base oil can be obtained; and when a sulfur-containing raw material such as slack wax obtained through the process of refining the base oil, and microwax obtained through a wax refining process is used, the sulfur content in the obtained base oil is normally no less than 100 mass ppm. The sulfur content in the lubricant base oil (total base oil) is normally no more than 0.03 mass%; and is preferably no more than 0.01 mass% in view of oxidation stability. In the present description, the sulfur content in the base oil means a sulfur content measured conforming to JIS K 2541-2003.

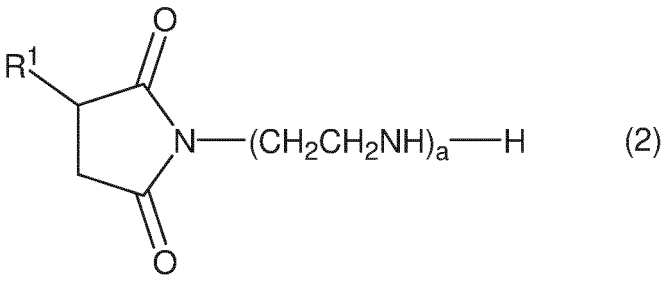
[0043] The lubricant base oil may comprise a single base oil component or may comprise a plurality of base oil components as long as the kinematic viscosity of the entire base oil (total base oil) at 40°C is 6.0 to 12.0 mm^2/s.

[0044] In one embodiment, the lubricant base oil can comprise at least one API Group II base oil, at least one API Group III base oil, or at least one API Group IV base oil, or any combination thereof in an amount of 80 to 100 mass%, 90 to 100 mass%, 90 to 99 mass%, or 95 to 99 mass% on the basis of the total mass of the base oil. In one embodiment, the lubricant base oil can comprise at least one API Group III base oil, or at least one API Group IV base oil, or any combination thereof in an amount of 80 to 100 mass%, 90 to 100 mass%, 90 to 99 mass%, or 95 to 99 mass% on the basis of the total mass of the base oil. The lubricant base oil may optionally comprise an API Group V base oil. In one embodiment, the content of at least one API Group V base oil in the lubricant base oil on the basis of the total mass of the base oil can be preferably 0 to 20 mass%, or 0 to 10 mass% in view of improving oxidation stability; and can be 1 to 10 mass%, or 1 to 5 mass% in view of improving anti-fatigue performance. The lubricant base oil may optionally comprise an API Group IV base oil. In one embodiment, the content of at least one API Group IV base oil in the lubricant base oil can be 0 to 60 mass%, 0 to 50 mass%, 1 to 60 mass%, or 1 to 50 mass% on the basis of the total mass of the base oil.

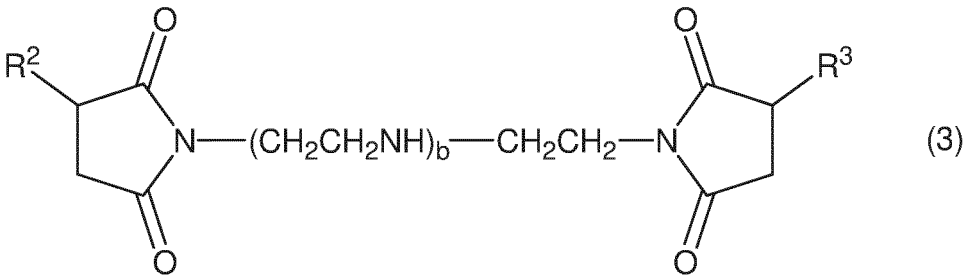
[0045] The content of the lubricant base oil (total base oil) in the lubricating oil composition on the basis of the total mass of the lubricating oil composition is no less than 60 mass%, and can be preferably 60 to 98.5 mass%, more preferably 70 to 98.5 mass%, and in one embodiment, 75 to 97 mass%.

<(A) First Succinimide Compound>

[0046] The lubricating oil composition according to the present invention comprises a condensation reaction product of a polyisobutenylsuccinic acid or anhydride thereof and a polyamine, or a modified product thereof, or any combination thereof (hereinafter may be referred to as "component (A)") in an amount of no less than 80 mass ppm in terms of nitrogen and no more than 2.7 mass% as the entire compound on the basis of the total mass of the composition. The polyisobutenylsuccinic acid has a polyisobutenyl group having a number average molecular weight of no less than 800. This condensation reaction product (condensation product) is polyisobutenylsuccinimide, and can be represented by the following general formula (2) or (3). Examples of the modified product will be described later.



(2)



(3)

[0047] In the general formula (2), R^1 is a polyisobutenyl group having a number average molecular weight of no less

than 800, and a represents an integer of 1 to 10, preferably 2 to 6. In one typical embodiment, the compound represented by the general formula (2) is obtainable as a mixture of compounds having different a's. In one embodiment, the carbon number of R^1 is preferably no less than 40, and more preferably no less than 60 in view of the solubility in the base oil; is preferably no more than 400, more preferably no more than 350, and further preferably no more than 250 in view of the low-temperature fluidity of the composition; and in one embodiment, can be 40 to 400, 60 to 350, or 60 to 250.

[0048] In the general formula (3), R^2 and R^3 are each independently a polyisobutenyl group having a number average molecular weight of no less than 800, and may be combination of different groups. In the general formula (3), b is an integer of 0 to 15, preferably 1 to 13, more preferably 1 to 11. In one typical embodiment, the compound represented by the general formula (3) is obtainable as a mixture of compounds having different b's. In one embodiment, the carbon number of each of R^2 and R^3 is preferably no less than 40, and more preferably no less than 60 in view of the solubility in the base oil; is preferably no more than 400, more preferably no more than 350, and further preferably no more than 250 in view of the low-temperature fluidity of the composition; and in one embodiment, can be 40 to 400, 60 to 350, or 60 to 250.

[0049] The polyisobutenyl groups (R^1 to R^3) in the component (A) are each a branched chain alkyl or alkenyl group derived from an oligomer of isobutene (polyisobutene). The polyisobutenylsuccinic anhydride herein can be obtained by, for example, reacting polyisobutene having a C=C double bond with maleic anhydride at 100 to 200°C. The polyisobutenyl group of the polyisobutenylsuccinic anhydride obtained by this reaction is an alkenyl group having a C=C double bond. This alkenylsuccinic anhydride is further subjected to a hydrogenation reaction, and thereby, the polyisobutenylsuccinic anhydride where the polyisobutenyl group is an alkyl group can be obtained.

[0050] The number average molecular weight of the polyisobutenyl group (any of R^1 to R^3) in the component (A) is no less than 800, and preferably no less than 900 in view of improving oil solubility; is preferably no more than 3500 in view of improving the low-temperature fluidity of the lubricating oil; and in one embodiment, can be 800 to 3500, or 900 to 3500. The number average molecular weight Mn^{PIB} of the polyisobutenyl group of the component (A) can be calculated from the number average molecular weight Mn^{SA} of the corresponding polyisobutenylsuccinic acid by the following equation (4):

$$(4): Mn^{PIB} = Mn^{SA} - 117.08$$

[0051] In the present description, the number average molecular weight of the polyisobutenylsuccinic acid means a number average molecular weight measured by gel permeation chromatography (GPC) in terms of standard polystyrene, and the measurement method thereof is as described above. When the component (A) is given, the number average molecular weight of the polyisobutenyl group in the component (A) can be determined by the steps of:

(1) reacting (for example, 100 mg of) the component (A) with water and a strong base (such as 5 mL of a 6 N sodium hydroxide solution) in an organic solvent (such as 0.5 mL of methanol) (for example, at 160°C for 5 hours), to hydrolyze the component (A) into a polyisobutenylsuccinic acid and a polyamine;
(2) adding a strong acid (such as 6 N hydrochloric acid) to the resultant mixture after the reaction of the step (1), to acidify the resultant mixture; and thereafter, performing extraction with a hydrophobic organic solvent (such as hexane and toluene);
(3) washing an organic phase obtained by the step (2) with an acidic aqueous solution (such as dilute hydrochloric acid), to remove the polyamine and a salt thereof (if any) from the organic phase, and thus to obtain an organic solvent solution of the polyisobutenylsuccinic acid;
(4) measuring, by GPC, the number average molecular weight Mn^{SA} of the polyisobutenylsuccinic acid obtained by the step (3); and
(5) calculating the number average molecular weight Mn^{PIB} of the polyisobutenyl group by the equation (4) from the number average molecular weight Mn^{SA} of the polyisobutenylsuccinic acid obtained by the step (4).

[0052] Polyisobutenylsuccinimide includes so-called monotype succinimide represented by the general formula (2) where only one terminal of the polyamine chain is imidated, and so-called bistype succinimide represented by the general formula (3) where both terminals of the polyamine chain are imidated. The lubricating oil composition may comprise either one of monotype and bistype succinimide, or may comprise both of them as a mixture. The content of bistype succinimide or any modified product thereof in the component (A) is preferably 50 to 100 mass%, and more preferably 70 to 100 mass% on the basis of the total mass of the component (A) (100 mass%).

[0053] As the component (A), the aforementioned condensation product may be used as it is (that is, non-modified succinimide), or a modified product (derivative) converted from the condensation product and described later may be used. The condensation product of the polyisobutenylsuccinic acid or anhydride thereof, and a polyamine may be bistype succinimide where both ends of the polyamine chain are imidated (see the general formula (3)), may be monotype

succinimide where only one end of the polyamine chain is imidated (see the general formula (2)), or may be a mixture thereof. Examples of a polyamine herein include polyethylenpolyamines having 3 to 17 nitrogen atoms such as diethylenetriamine, triethylenetetramine, tetraethylenepentamine, pentaethylenehexamine, hexaethyleneheptamine, heptaethyleneoctamine, octaethylenenonamine, nonaethylenedecamine, decaethyleneundecamine, undecaethylenedodecamine, dodecaethylenetridecamine, tridecaethylenetetradecamine, tetradecaethylenepentadecamine, pentadecaethylenehexadecamine, and hexadecaethyleneheptadecamine, and mixtures thereof. A polyamine raw material comprising at least one selected from them can be preferably used. In one embodiment, a polyamine raw material comprising at least one polyethylenpolyamine having 3 to 17, 3 to 15, or 3 to 13 nitrogen atoms can be preferably used. In another embodiment, a polyamine raw material comprising at least one polyethylenpolyamine having 3 to 11, or 3 to 7 nitrogen atoms can be preferably used. A commercially available polyethylenpolyamine is often a mixture of at least two polyethylenpolyamines having successive numbers of nitrogen atoms, respectively. Such a polyethylenpolyamine mixture can be also preferably used as the polyamine raw material for producing the component (A). The general formulae (2) and (3) each represent the structure of a condensation reaction product of a polyisobutenylsuccinic acid or anhydride thereof, and a linear polyethylenpolyamine. A commercially available polyethylenpolyamine having at least 4 nitrogen atoms may often comprise, as a structural isomer, a branched polyethylenpolyamine having the same number of nitrogen atoms, in addition to a linear polyethylenpolyamine. Branched and linear polyethylenpolyamines share a common feature that each pair of two adjacent amino groups are connected by an ethylene group. A linear polyethylenpolyamine having n nitrogen atoms (n is an integer of two or more) has two primary amino groups, and n-2 secondary amino groups, whereas a branched polyethylenpolyamine having k branches (k is an integer of 1 to n-3) and n nitrogen atoms has 2+k primary amino groups, n-2-2k secondary amino groups, and k tertiary amino groups. A polyethylenpolyamine mixture comprising such a branched structural isomer can be also preferably used as the polyamine raw material for producing the component (A). The component (A) also encompasses any condensation reaction product of such a branched polyethylenpolyamine and the polyisobutenylsuccinic acid or anhydrides thereof, and modified products thereof. The general formulae (2) and (3) each represent succinimide obtained by imidating one or two primary amino group(s). In the condensation reaction of a branched polyethylenpolyamine having k branches with the polyisobutenylsuccinic acid or anhydride thereof, 2+k primary amino groups can be imidated at the maximum. The component (A) also encompasses such a condensation reaction product (succinimide compound) obtained by imidating three or more primary amino groups, and modified products thereof. The polyamine raw material may optionally further comprise ethylenediamine. In view of improving the performance of the condensation product or modified product thereof as a dispersant, the content of ethylenediamine in the polyamine raw material is preferably 0 to 10 mass%, and more preferably 0 to 5 mass% on the basis of the total mass of the polyamine. Succinimide obtained as a condensation reaction product of the polyisobutenylsuccinic acid or anhydride thereof, and a mixture of at least two polyamines is a mixture comprising at least two compounds of the general formula (2) or (3) having different a's or b's. The condensation reaction of the polyisobutenylsuccinic acid or anhydride thereof with the polyamine can be performed in, for example, an organic solvent (such as toluene) that forms an azeotrope with water. That is, the condensation reaction product can be easily obtained by removing, by azeotropy with a solvent, water generated following the progress of the condensation reaction while refluxing and stirring a solution of the mixture of the polyisobutenylsuccinic acid or anhydride thereof with the polyamine. The reaction molar ratio of the polyisobutenylsuccinic acid or anhydride thereof with the polyamine in the condensation reaction can be, for example, polyisobutenylsuccinic acid or anhydride thereof:polyamine = 1:10 to 10:1, or 1:5 to 5:1.

[0054] The weight average molecular weight of the component (A) is preferably 1000 to 20000, more preferably 2000 to 20000, and further preferably 3000 to 15000; and in one embodiment, can be 4000 to 15000.

[0055] Examples of modified products (modified compounds, derivatives) of the polyisobutenylsuccinimide include (i) oxygen-containing organic compound-modified products, (ii) boric acid-modified products, (iii) phosphoric acid-modified products, (iv) sulfur-modified products, and (v) modified products of at least two of them in combination.

[0056] The (i) oxygen-containing organic compound-modified product is a modified compound where a part or all of the residual amino groups and/or imino groups is neutralized or amidated by making a C1-30 monocarboxylic acid such as fatty acids, a C2-30 polycarboxylic acid (such as ethanedioic acid, phthalic acid, trimellitic acid, and pyromellitic acid), an anhydride or ester thereof, a C2-6 alkylene oxide, or a hydroxy(poly)oxyalkylene carbonate react with the above-described polyisobutenylsuccinimide.

[0057] The (ii) boric acid-modified product is a modified compound where a part or all of the residual amino groups and/or imino groups is neutralized or amidated by making boric acid react with the above-described polyisobutenylsuccinimide.

[0058] The (iii) phosphoric acid-modified product is a modified compound where a part or all of the residual amino groups and/or imino groups is neutralized or amidated by making the above-described polyisobutenylsuccinimide react with phosphoric acid.

[0059] The (iv) sulfur-modified product is a modified compound obtained by making a sulfur compound react with the above-described polyisobutenylsuccinimide.

[0060] The (v) modified product of at least two of them in combination can be obtained by subjecting the above-

described polyisobutenylsuccinimide to at least two modifications selected from oxygen-containing organic compound modification, boron modification, phosphoric acid modification, and sulfur modification in combination.

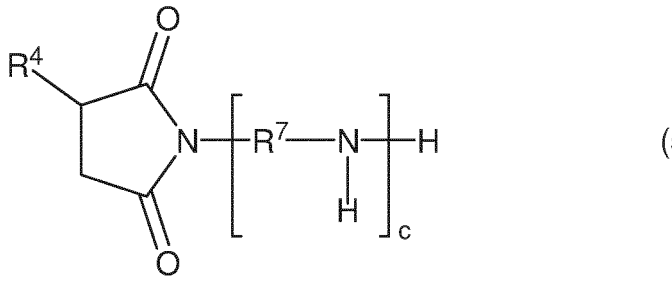
[0061] Among these modified products (derivatives) (i) to (v), a boric acid-modified compound, especially a boric acid-modified product of bistype polyisobutenylsuccinimide can be preferably used.

[0062] The content of the component (A) in the lubricating oil composition is no less than 80 mass ppm, and preferably no less than 100 mass ppm on the basis of the total mass of the composition in terms of nitrogen in view of improving the electrical insulation of the oxidatively deteriorated composition, and of suppressing the increase of the acid number of the oxidatively deteriorated composition; is no more than 2.7 mass%, and preferably no more than 2.5 mass% on the basis of the total mass of the composition as the entire compound in view of reducing the viscosity of the composition to improve fuel efficiency; and in one embodiment, can be no more than 2.3 mass%.

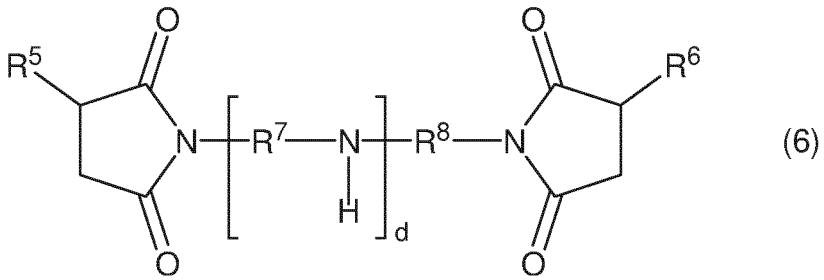
[0063] The product of the weight average molecular weight (unit: Da, that is, g/mol) of the component (A) and the amount of the component (A) in the lubricating oil composition as the entire compound (unit: mass%) is no more than 16,000, and is preferably no more than 15,000 in view of reducing the viscosity of the composition to improve fuel efficiency; and in one embodiment, can be no more than 14,000. When the component (A) comprises plural components, the weight average molecular weight (Da) as the entire component (A), and the amount (mass%) as the entire component (A) shall be used for the calculation of the product.

<(B) Second Succinimide Compound>

[0064] The lubricating oil composition according to the present invention comprises a condensation reaction product of an alkyl- or alkenyl- succinic acid having a C8-30 alkyl or alkenyl group or anhydride thereof and a polyamine, or a modified product thereof, or any combination thereof (hereinafter may be referred to as "component (B)"), in an amount of 50 to 1300 mass ppm in terms of nitrogen on the basis of the total mass of the composition. This condensation reaction product (condensation product) is alkyl or alkenyl succinimide, and can be represented by the following general formula (5) or (6).



(5)



(6)

In the general formulae (5) and (6), R^4 to R^6 each independently represent a C8-30, preferably a C12-24, and in one embodiment, a C12-18 alkyl or alkenyl group, and R^7 and R^8 each independently represent a C1-4, preferably a C2-3 alkylene group, and especially preferably an ethylene group. In the general formulae (5) and (6), c represents an integer of 1-7, preferably 1-6, more preferably 1-5, further preferably 1-4, and d represents an integer of 1-7, preferably 1-4, more preferably 1-3.

[0065] The component (B) is obtained through the reaction of an alkyl- or alkenyl- succinic acid having a C8-30, preferably a C12-28 alkyl or alkenyl group or anhydride thereof with a polyamine as a condensation reaction product (condensation product). As the component (B), this condensation product may be used as it is (that is, non-modified succinimide), or a modified product (derivative) converted from the condensation product and described later may be used. The condensation product of the alkyl- or alkenyl- succinic acid or anhydride thereof, and a polyamine may be bistype succinimide where both ends of the polyamine chain are imidated (see the general formula (6)), may be monotype succinimide where only one end of the polyamine chain is imidated (see the general formula (5)), or may be a mixture

thereof. Examples of a polyamine herein include polyethylenpolyamines having 3 to 9 nitrogen atoms such as diethylenetriamine, triethylenetetramine, tetraethylenepentamine, pentaethylenehexamine, hexaethyleneheptamine, heptaethyleneoctamine, and octaethylenenonamine, and ethylenediamine, and mixtures thereof. A polyamine raw material comprising at least one selected from them can be preferably used. In one embodiment, a polyamine raw material comprising at least one polyethylenpolyamine having 3 to 9, 3 to 6, or 3 to 5 nitrogen atoms can be preferably used. In another embodiment, a polyamine raw material comprising at least one polyethylenpolyamine having 3 to 8, 3 to 7, 3 to 6, or 3 to 5 nitrogen atoms, or ethylenediamine, or any combination thereof can be preferably used. A commercially available polyethylenpolyamine is often a mixture of at least two polyethylenpolyamines having successive numbers of nitrogen atoms, respectively. Such a polyethylenpolyamine mixture can be also preferably used as the polyamine raw material for producing the component (B). The general formulae (5) and (6) each represent the structure of a condensation reaction product of an alkyl- or alkenyl- succinic acid or anhydride thereof, and a linear polyethylenpolyamine. As descried above concerning the component (A), a commercially available polyethylenpolyamine having at least 4 nitrogen atoms can often comprise, as a structural isomer, a branched polyethylenpolyamine having the same number of nitrogen atoms, in addition to a linear polyethylenpolyamine. A polyethylenpolyamine mixture comprising such a branched structural isomer can be also preferably used as the polyamine raw material for producing the component (B). The component (B) also encompasses any condensation reaction product of such a branched polyethylenpolyamine and the alkyl- or alkenyl- succinic acid or anhydride thereof, and modified products thereof. The general formulae (5) and (6) each represent succinimide obtained by imidating one or two primary amino group(s). In the condensation reaction of a branched polyethylenpolyamine having k branches with the alkyl- or alkenyl- succinic acid or anhydride thereof, 2+k primary amino groups can be imidated at the maximum. The component (B) also encompasses such a condensation reaction product (succinimide compound) obtained by imidating three or more primary amino groups, and modified products thereof. The polyamine raw material may optionally comprise ethylenediamine. In view of enhancing the performance of the condensation product or modified product thereof as a friction modifier, and the effect of improving the resistance to oxidative deterioration, the content of ethylenediamine in the polyamine raw material is preferably 0 to 10 mass%, and more preferably 0 to 5 mass% on the basis of the total mass of the polyamine. Succinimide obtained as a condensation reaction product of the alkyl- or alkenylsuccinic acid or anhydride thereof and a mixture of at least two polyamines is a mixture comprising at least two compounds of the general formula (5) or (6) having different c's or d's. The condensation reaction of the alkyl- or alkenylsuccinic acid or anhydride thereof with the polyamine can be performed in, for example, an organic solvent (such as toluene) that forms an azeotrope with water. That is, the condensation reaction product can be easily obtained by removing, by azeotropy with a solvent, water generated following the progress of the condensation reaction while refluxing and stirring a solution of the mixture of the alkyl- or alkenyl- succinic acid or anhydride thereof with the polyamine. The reaction molar ratio of the alkyl- or alkenyl- succinic acid or anhydride thereof with the polyamine in the condensation reaction can be, for example, alkyl- or alkenyl- succinic acid or anhydride thereof:polyamine = 1:10 to 10:1, or 1:5 to 5:1.

[0066] Examples of modified products (derivatives) of succinimide compounds which can be used as the component (B) include modified products obtained by reacting the aforementioned succinimide compound (condensation reaction product) with at least one compound selected from boric acid, phosphoric acid, a C1-20 carboxylic acid, and a sulfur-containing compound. Among them, a boric acid-modified product can be preferably used.

[0067] A highly polar component such as the component (B) and metallic detergents easily deteriorates the electrical insulation of the fresh oil and the oxidatively deteriorated composition. However, the inventors of the present invention found that the use of the components (A) and (B) together in amounts within predetermined ranges each can suppress the increase of the acid number of the oxidatively deteriorated oil without significantly impairing the electrical insulation of the fresh oil and the oxidatively deteriorated composition while reducing the content of the component (A) to enhance energy saving performance.

[0068] The content of the component (B) in the lubricating oil composition on the basis of the total mass of the composition in terms of nitrogen is no less than 50 mass ppm in view of suppressing the increase of the acid number of the oxidatively deteriorated oil; and is no more than 1300 mass ppm in view of improving the electrical insulation of the oxidatively deteriorated oil.

[0069] Preferably, the relation of the following equation (1) is met in view of further suppressing the increase of the acid number of the oxidatively deteriorated oil without significantly impairing the electrical insulation of the fresh oil and the oxidatively deteriorated composition while reducing the content of the component (A) to enhance energy saving performance. In the following equation (1), the function max is a function to return the maximum value of an argument. That is, when f(A) > 50, max(50,f(A)) = f(A); when f(A) < 50, max(50,f(A)) = 50; and when f(A) = 50, max(50,f(A)) = 50.

$$B \geq \max(50, f(A))$$

$$f(A) = 100 \times \left(-10.5 + 14.1 \times \left(\frac{A}{100} - 0.5\right)^{-0.3}\right) \quad \cdots (1)$$

where A represents the amount (unit: mass ppm) of the component (A) in terms of nitrogen on the basis of the total mass of the composition, and B represents the amount (unit: mass ppm) of the component (B) in terms of nitrogen on the basis of the total mass of the composition.

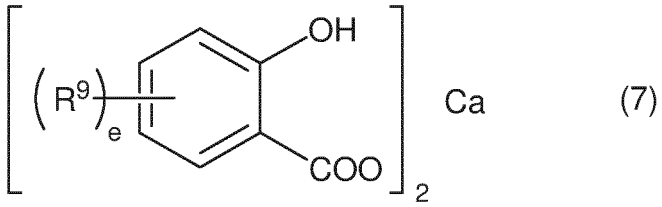
<(C) Calcium Detergent>

[0070] In one preferred embodiment, the lubricating oil composition can further comprise: at least one calcium sulfonate detergent overbased with calcium carbonate (hereinafter may be referred to as "component (C1)"), or at least one calcium salicylate detergent overbased with calcium carbonate (hereinafter may be referred to as "component (C2)"), or any combination thereof (hereinafter may be referred to as "component (C)"). The component (C) may comprise only the component (C1), or may comprise only the component (C2), or may comprise both the component (C1) and the component (C2).

[0071] A preferred example of the (C1) calcium sulfonate detergent overbased with calcium carbonate is an overbased salt of a calcium salt of an alkyl aromatic sulfonic acid obtained by sulfonation of an alkylaromatic. The weight average molecular weight of an alkylaromatic herein is preferably 300 to 1500, and more preferably 400 to 1300.

[0072] Examples of an alkyl aromatic sulfonic acid herein include what is called petroleum sulfonic acids and synthetic sulfonic acids. Examples of petroleum sulfonic acids herein include sulfonated products of alkylaromatics of lubricating oil fractions derived from mineral oil, and what is called mahogany acid that is a side product of white oil. One example of synthetic sulfonic acids herein is a sulfonated product of an alkylbenzene having a linear or branched alkyl group which is obtained by recovering side products in a manufacturing plant of an alkylbenzene which is to be a raw material of detergent or by alkylating benzene with a polyolefin. Another example of synthetic sulfonic acids herein is a sulfonated product of an alkylnaphthalene such as dinonylnaphthalene. For example, a fuming sulfuric acid or a sulfuric anhydride can be used as a sulfonating agent for sulfonating the foregoing alkylaromatics without particular restrictions.

[0073] The (C2) calcium salicylate detergent overbased with calcium carbonate is an overbased salt of a calcium salicylate. A preferred example of the calcium salicylate is a calcium salicylate represented by the following general formula (7).



(7)

[0074] In the formula (7), R^9 each independently represent a C14-30 alkyl or alkenyl group, and e represents 1 or 2, preferably 1. In the formula (7), when e = 2, R^9 may be combination of different groups.

[0075] The way of producing the calcium salicylate is not specifically limited, and any known way of producing a mono alkyl salicylate may be employed. For example, the calcium salicylate can be obtained by, for example: making a calcium base such as oxides and hydroxides of calcium react with, for example, a monoalkylsalicylic acid obtained by alkylating a phenol as a starting material with an olefin, and then carboxylating the resultant product with carbonic acid gas or the like, or with a monoalkylsalicylic acid obtained by alkylating a salicylic acid as a starting material with an equivalent of the olefin; or once converting the foregoing monoalkylsalicylic acid to an alkali metal salt such as a sodium salt and a potassium salt, and then performing transmetallation with a calcium salt.

[0076] The way of obtaining a calcium sulfonate or salicylate overbased with calcium carbonate is not specifically limited. For example, a calcium sulfonate or salicylate overbased with calcium carbonate can be obtained by reacting a calcium sulfonate or salicylate with a base such as calcium hydroxide in the presence of carbonic acid gas.

[0077] The base numbers of the component (C1) and the component (C2) are each preferably no less than 200 mgKOH/g, and more preferably no less than 250 mgKOH/g in view of improving anti-wear performance, anti-seizure performance, and the torque transmitting capacity of a wet clutch; is preferably no more than 600 mgKOH/g, and more preferably no more than 550 mgKOH/g from the same viewpoint; and in one embodiment, can be 200 to 600 mgKOH/g, or 250 to 550 mgKOH/g. When the component (C1) comprises two or more calcium sulfonate detergents overbased

with calcium carbonate, the base number of each of the calcium sulfonate detergents overbased with calcium carbonate is preferably within the foregoing range. Likewise, when the component (C2) comprises two or more calcium salicylate detergents overbased with calcium carbonate, the base number of each of the calcium salicylate detergents overbased with calcium carbonate is preferably within the foregoing range. In the present description, the base number means a base number measured conforming to JIS K2501 by the perchloric acid method. Generally, a metallic detergent is obtained by reaction in a diluent such as a solvent and a lubricant base oil. Therefore, a metallic detergent is on the market as diluted in a diluent such as a lubricant base oil. In the present description, the base number of a metallic detergent shall mean a base number of the metallic detergent with a diluent.

[0078] When the lubricating oil composition comprises the component (C), the content of the component (C) (the total content when the component (C) comprises two or more detergents) on the basis of the total mass of the composition in terms of calcium is preferably less than 100 mass ppm, is more preferably no more than 95 mass ppm or no more than 90 mass ppm, and in one embodiment, is no more than 80 mass ppm in view of further improving the electrical insulation of the fresh oil, and fuel efficiency, and of improving anti-fatigue performance; is preferably no less than 10 mass ppm, more preferably no less than 15 mass ppm, and in one embodiment, is no less than 20 mass ppm in view of improving anti-wear performance, anti-seizure performance, anti-fatigue performance, and the torque transmitting capacity of a wet clutch; and in one embodiment, can be no less than 10 mass ppm and less than 100 mass ppm, 10 to 95 mass ppm, 15 to 90 mass ppm, or 20 to 80 mass ppm.

[0079] Generally, in the field of lubricating oils, an organic acid metal base that can form micelles in base oil (such as alkali or alkaline earth metal alkylsalicylates, alkali or alkaline earth metal alkylbenzene sulfonates, and alkali or alkaline earth metal alkylphenates), or a mixture of such an organic acid metal base and a basic metal salt (such as a hydroxide, a carbonate or a borate of an alkali or alkaline earth metal that constitutes the foregoing organic acid metal base) is used as a metallic detergent. Such an organic acid usually has, in a molecule thereof, at least one polar group (such as a carboxy group, a sulfo group, and a phenolic hydroxy group) that has Broensted acidity and that can form a salt with a metal base (typically a metal oxide and/or metal hydroxide), and at least one lipophilic group such as linear or branched chain alkyl groups (for example, a linear or branched chain alkyl group having 6 or more carbons). A soap base of the metallic detergent means a conjugate base of an organic acid which constitutes the soap content of the metallic detergent (for example, alkylsalicylate anions in the salicylate detergent, alkylbenzenesulfonate anions in the sulfonate detergent, and alkylphenate anions in the phenate detergent).

[0080] The lubricating oil composition according to the present invention may optionally further comprise at least one metallic detergent other than the component (C). The total content of the entire metallic detergent including the component (C) in the lubricating oil composition on the basis of the total mass of the composition in terms of metal is preferably less than 100 mass ppm, more preferably no more than 95 mass ppm or no more than 90 mass ppm, and in one embodiment, is no more than 80 mass ppm in view of further improving the electrical insulation of the fresh oil, fuel efficiency, and anti-fatigue performance; is preferably no less than 10 mass ppm, and in one embodiment, is no less than 20 mass ppm in view of further improving anti-seizure performance, anti-fatigue performance, and the torque transmitting capacity of a wet clutch, and of further improving anti-wear performance; and in one embodiment, can be no less than 10 mass ppm and less than 100 mass ppm, 10 to 95 mass ppm, 15 to 90 mass ppm, or 20 to 80 mass ppm. In one embodiment, the lubricating oil composition according to the present invention can be a lubricating oil composition comprising no metallic detergent other than the component (C).

<(D) Antioxidant>

[0081] In one preferred embodiment, the lubricating oil composition can further comprise: at least one amine antioxidant (hereinafter may be referred to as "component (D1)"), and at least one phenol antioxidant (hereinafter may be referred to as "component (D2)") as an antioxidant (hereinafter may be referred to as "component (D)").

[0082] Examples of the component (D1) include aromatic amine antioxidants and hindered amine antioxidants. Examples of aromatic amine antioxidants herein include primary aromatic amine compounds such as alkylated-α-naphthylamine; and secondary aromatic amine compounds such as alkylated diphenylamine, phenyl-α-naphthylamine, alkylated phenyl-α-naphthylamine, and phenyl-β-naphthylamine. As the aromatic amine antioxidant, alkylated diphenylamine, or alkylated phenyl-α-naphthylamine, or combination thereof may be preferably used.

[0083] Examples of hindered amine antioxidants herein include compounds having a 2,2,6,6-tetraalkylpiperidine skeleton (2,2,6,6-tetraalkylpiperidine derivatives). As a 2,2,6,6-tetraalkylpiperidine derivative herein, a 2,2,6,6-tetraalkylpiperidine derivative having a substituent in 4-position is preferable. Two 2,2,6,6-tetraalkylpiperidine skeletons may be bonded with each other via a substituent in respective 4-positions thereof. The 2,2,6,6-tetraalkylpiperidine skeleton may have no substituent in N-position thereof, or may have a substituent of a C1-4 alkyl group in N-position thereof. The 2,2,6,6-tetraalkylpiperidine skeleton is preferably a 2,2,6,6-tetramethylpiperidine skeleton.

[0084] Examples of the substituent in 4-position of the 2,2,6,6-tetraalkylpiperidine skeleton include acyloxy group ($R^{10}COO$-), alkoxy group ($R^{10}O$-), alkylamino group ($R^{10}NH$-), and acylamino group ($R^{10}CONH$-). R^{10} is preferably a

C1-30, more preferably a C1-24, and further preferably a C1-20 hydrocarbon group. Examples of this hydrocarbon group include alkyl group, alkenyl group, cycloalkyl group, alkylcycloalkyl group, aryl group, alkylaryl group, and arylalkyl group.

[0085] When two 2,2,6,6-tetraalkylpiperidine skeletons are bonded with each other via a substituent in respective 4-positions thereof, examples of the substituent include hydrocarbylene bis(carbonyloxy) group (-OOC-R^{11}-COO-), hydrocarbylene diamino group (-HN-R^{11}-NH-), and hydrocarbylene bis(carbonylamino) group (-HNCO-R^{11}-CONH-). R^{11} is preferably a C1-30 hydrocarbylene group, and more preferably an alkylene group.

[0086] Acyloxy group is preferable as the substituent in 4-position of the 2,2,6,6-tetraalkylpiperidine skeleton. One example of a compound having acyloxy group in 4-position of the 2,2,6,6-tetraalkylpiperidine skeleton is an ester of 2,2,6,6-tetramethyl-4-piperidinol and a carboxylic acid. Examples of this carboxylic acid include C8-20 linear or branched chain aliphatic carboxylic acids.

[0087] Examples of the component (D2) (phenol antioxidant) include hindered phenol compounds and bisphenol compounds such as: 4,4'-methylenebis(2,6-ditert-butylphenol); 4,4'-bis(2,6-di-tert-butylphenol); 4,4'-bis(2-methyl-6-tert-tbutylphenol); 2,2'-methylenebis(4-ethyl-6-tert-butylphenol); 2,2'-methylenebis(4-methyl-6-tert-butylphenol); 4,4'-butylidenebis(3-methyl-6-tertbutylphenol); 4,4'-isopropylidenebis(2,6-di-tert-butylphenol); 2,2'-methylenebis(4-methyl-6-nonylphenol); 2,2'-isobutylidenebis(4,6-dimethylphenol); 2,2'-methylenebis(4-methyl-6-cyclohexylphenol); 2,6-di-tert-butyl-4-methylphenol; 2,6-di-tert-butyl-4-ethylphenol; 2,4-dimethyl-6-tertbutylphenol; 2,6-di-tert-butyl-4-(N,N'-dimethylaminomethyl)phenol; 4,4'-thiobis(2-methyl-6-tert-butylphenol); 4,4'-thiobis(3-methyl-6-tert-butylphenol); 2,2'-thiobis(4-methyl-6-tert-butylphenol); bis(3-methyl-4-hydroxy-5-tert-butylbenzyl) sulfide; bis(3,5-di-tert-butyl-4-hydroxybenzyl) sulfide; 3-(3,5-ditert-butyl-4-hydroxyphenyl)propionic acid esters; and 3-methyl-5-tert-butyl-4-hydroxyphenol fatty acid esters. Examples of 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid esters herein include octyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate; decyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate; dodecyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate; tetradecyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate; hexadecyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate; octadecyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate; pentaerythritol tetrakis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate]; and 2,2'-thiodiethylene bis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate].

[0088] When the lubricating oil composition comprises the component (D), the content of the component (D) as the total amount of the component (D1) and the component (D2) on the basis of the total mass of the composition is preferably no less than 0.1 mass%, more preferably no less than 0.2 mass%, and in one embodiment, no less than 0.3 mass% in view of further suppressing the increase of the acid number of the oxidatively deteriorated composition; is preferably no more than 3.0 mass%, more preferably no more than 2.5 mass%, and in one embodiment, no more than 2.3 mass% in view of further improving the electrical insulation of the fresh oil and the oxidatively deteriorated composition; and in one embodiment, can be 0.1 to 3.0 mass%, 0.2 to 2.5 mass%, or 0.3 to 2.3 mass%.

<(E) Phosphorus or Sulfur-Containing Additive>

[0089] In one preferred embodiment, the lubricating oil composition can further comprise: at least one phosphorus-containing compound (hereinafter may be referred to as a component (E1)), or at least one sulfur-containing compound comprising at least one sulfur atom having a formal oxidation number of no more than +II in a molecule thereof (hereinafter may be referred to as a component (E2)), or any combination thereof (hereinafter may be referred to as "component (E)").

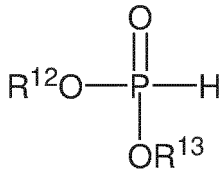
[0090] As the component (E1), a phosphorus-containing compound that functions as an anti-wear agent or extreme-pressure agent in lubricating oil can be used. As the component (E1), one phosphorus-containing compound may be used alone, or two or more phosphorus-containing compounds may be used in combination.

[0091] Examples of the component (E1) include phosphite esters, thiophosphite esters, dithiophosphite esters, trithiophosphite esters, phosphate esters, thiophosphate esters, dithiophosphate esters, trithiophosphate esters, amine salts thereof, and metal salts thereof.

[0092] The foregoing phosphorus-containing esters normally have a C2-30, preferably a C3-20 hydrocarbon group. Examples of C2-30 hydrocarbon groups herein include alkyl group, cycloalkyl group, alkyl-substituted cycloalkyl group, alkenyl group, aryl group, alkyl-substituted aryl group, and aryl-substituted alkyl group. These alkyl groups may be either linear or branched.

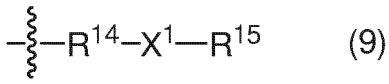
[0093] An example of salts of phosphorus-containing esters herein is a salt where part or all of the residual acidic hydrogen is neutralized which is obtained by making a metal base, or a nitrogen-containing compound such as ammonia, and an amine compound including only a C1-8 hydrocarbon group or a C1-8 hydroxy group-containing hydrocarbon group in a molecule thereof react with a phosphoric acid partial ester, a monothiophosphate partial ester, a dithiophosphate partial ester, a trithiophosphate partial ester, a phosphite partial ester, a thiophosphite partial ester, or a dithiophosphite partial ester.

[0094] As the component (E1), the phosphite ester compound represented by the following general formula (8) can be especially preferably used.



(8)

In the general formula (8), R^{12} and R^{13} are each independently a C1-18 linear chain hydrocarbon group, or a C4-20 group represented by the following general formula (9).



(9)

In the general formula (9), R^{14} is a C2-17 linear chain hydrocarbon group, is preferably an ethylene group or a propylene group, and in one embodiment, can be an ethylene group. In the general formula (9), R^{15} is a C2-17 linear chain hydrocarbon group, preferably a C2-16 linear chain hydrocarbon group, and especially preferably a C6-10 linear chain hydrocarbon group. X^1 is an oxygen atom or a sulfur atom, and preferably a sulfur atom. The carbon number of the group represented by the general formula (9) is preferably 5 to 20.

[0095] In the present description, "phosphorous acid" means H_3PO_3, which is an oxoacid of phosphorus having an oxidation number of +III. Normally, the phosphite ester compound represented by the general formula (8) has tautomerism. In the present description, any tautomers of the compound represented by the general formula (8) shall fall under the component (E1).

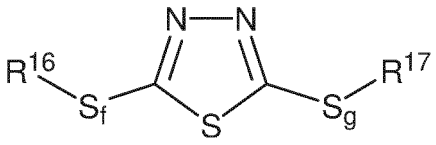
[0096] In one embodiment, preferred examples of R^{12} and R^{13} include C4-18 linear chain alkyl groups. Examples of linear chain alkyl groups include butyl group, pentyl group, hexyl group, heptyl group, octyl group, nonyl group, decyl group, undecyl group, dodecyl group, tridecyl group, tetradecyl group, pentadecyl group, hexadecyl group, heptadecyl group, and octadecyl group.

[0097] In one embodiment, preferred examples of R^{12} and R^{13} include 3-thiapentyl group, 3-thiahexyl group, 3-thiaheptyl group, 3-thiaoctyl group, 3-thianonyl group, 3-thiadecyl group, 3-thiaundecyl group, 4-thiahexyl group, 3-oxapentyl group, 3-oxahexyl group, 3-oxaheptyl group, 3-oxaoctyl group, 3-oxanonyl group, 3-oxadecyl group, 3-oxaundecyl group, 3-oxadodecyl group, 3-oxatridecyl group, 3-oxatetradecyl group, 3-oxapentadecyl group, 3-oxahexadecyl group, 3-oxaheptadecyl group, 3-oxaheptadecyl group, 3-oxanonadecyl group, 4-oxahexyl group, 4-oxaheptyl group, and 4-oxaoctyl group.

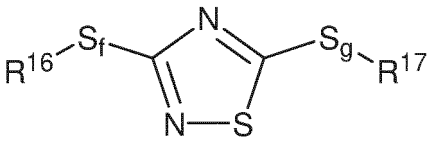
[0098] As the component (E2), a sulfur-containing compound that functions as an anti-wear agent or extreme-pressure agent in lubricating oil can be used. Such a sulfur-containing compound comprises at least one sulfur atom having a formal oxidation number of no more than +II in a molecule thereof. In this description, the formal oxidation number of a sulfur atom is determined based on the relation between the electronegativity of an atom bonded to this sulfur atom, and the electronegativity of the sulfur atom. That is, in the bond of a sulfur atom and an atom X, when the electronegativity of an element X is higher than the electronegativity of sulfur, all the electrons that are considered to be involved in the bond between both the atoms are given to the atom X. In contrast, in the bond of a sulfur atom and an atom X, when the electronegativity of an element X is lower than the electronegativity of sulfur, all the electrons that are considered to be involved in the bond between both the atoms are given to the sulfur atom. The bond of sulfur atoms each other does not change the oxidation number. In this description, the Allred-Rochow electronegativity shall be applied to the electronegativity of all the elements including sulfur.

[0099] Examples of the component (E2) include thiadiazole compounds, sulfurized fats, sulfurized fatty acids, sulfurized esters, sulfurized olefins, dihydrocarbyl (poly)sulfide, alkylthiocarbamoyl compounds, thiocarbamate compounds, thioterpene compounds, dialkyl thiodipropionate compounds, sulfurized mineral oils, zinc dithiocarbamate compounds, molybdenum dithiocarbamate compounds, and sulfolane compounds. As the component (E2), one sulfur-containing compound may be used alone, or two or more sulfur-containing compounds may be used in combination.

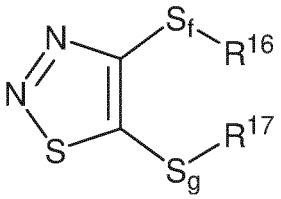
[0100] Preferred examples of thiadiazole compounds herein include the 1,3,4-thiadiazole compound represented by the following general formula (10), the 1,2,4-thiadiazole compound represented by the following general formula (11), and the 1,2,3-thiadiazole compound represented by the following general formula (12).



(10)



(11)



(12)

In the general formulae (10) to (12), R^{16} and R^{17} may be the same or different, and each independently represent a hydrogen atom or a C1-20 hydrocarbyl group; and f and g may be the same or different, and each independently represent an integer of 0 to 8.

[0101] Among the foregoing thiadiazole compounds, a thiadiazole compound represented by any of the general formulae (10) to (12), and having a hydrocarbyldithio group can be especially preferably used.

[0102] Sulfurized fats are products obtainable by reacting fat (such as lard oil, whale oil, vegetable oil, and fish oil) with sulfur or any sulfur-containing compound. The sulfur content in a sulfurized fat herein is not specifically limited, but usually 5 to 30 mass%.

[0103] As a sulfurized fatty acid herein, a product obtainable by sulfurizing an unsaturated fatty acid by any method can be used, and examples thereof include sulfurized oleic acid.

[0104] As a sulfurized ester herein, a product obtainable by sulfurizing an unsaturated fatty acid ester (such as a product obtained by reacting any alcohol with an unsaturated fatty acid (such as oleic acid, linoleic acid, or a fatty acid extracted from any of the foregoing animal and vegetable fats and oils) by any method can be used, and examples thereof include sulfurized methyl oleate, and sulfurized rice bran fatty acid octyl ester.

[0105] An example of sulfurized olefins herein is the compound represented by the following general formula (13). This compound can be obtained by reacting a C2-15 olefin, or a dimer, trimer, or tetramer thereof with a sulfurizing agent such as sulfur and sulfur chloride. Preferred examples of such an olefin include propylene, isobutene, and diisobutene.

$$R^{18}\text{-}S_h\text{-}R^{19} \qquad (13)$$

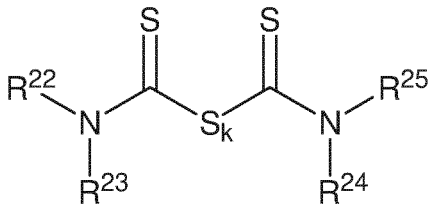
In the general formula (13), R^{18} represents a C2-15 alkenyl group, R^{19} represents a C2-15 alkyl or alkenyl group, and h represents an integer of 1 to 8.

[0106] Dihydrocarbyl (poly)sulfide herein is the compound represented by the following general formula (14). Here, when R^{15} and R^{16} are alkyl groups, the compound may be referred to as an alkyl sulfide.

$$R^{20}\text{-}S_i\text{-}R^{21} \qquad (14)$$

In the general formula (14), R^{20} and R^{21} may be the same or different, and each independently represent a C1-20 alkyl group (which may be a linear or branched chain, or may have a ring structure), a C6-20 aryl group, a C7-20 alkylaryl group, or a C7-20 arylalkyl group, and i represents an integer of 1 to 8.

[0107] An example of alkyl thiocarbamoyl compounds herein is the compound represented by the following general formula (15).

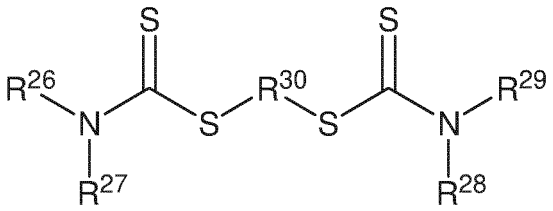


(15)

In the general formula (15), R^{22} to R^{25} may be the same or different, and each independently represent a C1-20 alkyl group, and k represents an integer of 1 to 8.

[0108] An example of alkyl thiocarbamate compounds herein is the compound represented by the following general

formula (16).



(16)

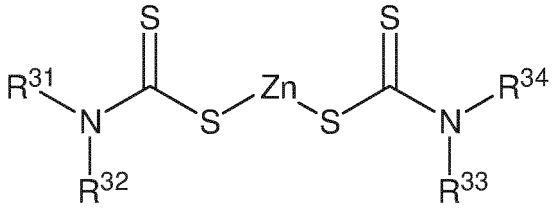
In the general formula (16), R^{26} to R^{29} may be the same or different, and each independently represent a C1-20 alkyl group, and R^{30} represents a C1-10 alkylene group.

[0109] An example of thioterpene compounds herein is a reaction product of phosphorus pentoxide and pinene.

[0110] Examples of dialkyl thiodipropionate compounds herein include dilauryl thiodipropionate, and distearyl thiodipropionate.

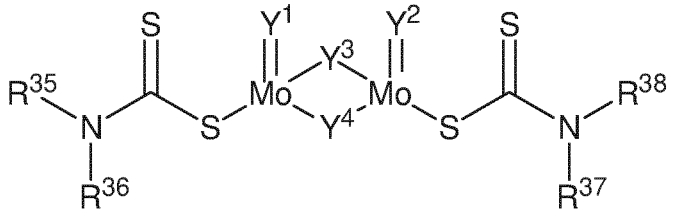
[0111] Sulfurized mineral oils are materials obtainable by dissolving elemental sulfur in mineral oil. The mineral oil used for a sulfurized mineral oil herein is not specifically limited, and examples thereof include paraffinic mineral oils and naphthenic mineral oils obtained by refining, by suitably combined known refining processes, lubricating oil fractions obtainable by topping and vacuum distillation of a crude oil. As elemental sulfur herein, elemental sulfur in any of a massive, powdery, or molten form may be used. The sulfur content in the sulfurized mineral oil is not specifically limited, and is normally 0.05 to 1.0 mass% on the basis of the total mass of the sulfurized mineral oil.

[0112] As a zinc dithiocarbamate compound herein, the compound represented by the following general formula (17) can be used. An example of a molybdenum dithiocarbamate compound herein is the compound represented by the following general formula (18).



(17)

In the general formula (17), R^{31} to R^{34} may be the same or different, and each independently represent a hydrocarbyl group having a carbon number of no less than 1.

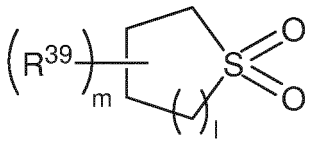


(18)

In the general formula (18), R^{35} to R^{38} may be the same or different, and each independently represent a hydrocarbyl group having a carbon number of no less than 1, and Y^1 to Y^4 each independently represent an oxygen atom or a sulfur atom.

[0113] The molybdenum dithiocarbamate compound of the general formula (18) is a binuclear complex having two molybdenum atoms in a molecule thereof. As a molybdenum dithiocarbamate compound herein, a mononuclear molybdenum complex having one molybdenum atom in a molecule thereof, or a polynuclear molybdenum complex having three or more molybdenum atoms in a molecule thereof can be also used.

[0114] As a sulfolane compound herein, for example, the compound represented by the following general formula (19) can be used.



(19)

In the general formula (19), l represents an integer of 1 or 2, m represents an integer of 0 or 1, and when m = 1, R^{39} represents a hydrocarbyl group having a carbon number of no less than 1.

[0115] The lubricating oil composition may optionally comprise the component (E). The content of the component (E) in the lubricating oil composition in terms of the total amount of the phosphorus content and the sulfur content on the basis of the total mass of the composition is preferably 0 to 1000 mass ppm, more preferably 0 to 900 mass ppm, and further preferably 0 to 800 mass ppm in view of further improving the electrical insulation of the fresh oil and the oxidatively deteriorated composition; and is preferably no less than 300 mass ppm, more preferably no less than 400 mass ppm, and further preferably no less than 500 mass ppm in view of improving anti-wear performance; and in one embodiment, can be 300 to 1000 mass ppm, 400 to 900 mass ppm, or 500 to 800 mass ppm. In the present description, any phosphorus-containing additive shall contribute to the content of the component (E1), and only an additive containing sulfur having a formal oxidation number of no more than +II but containing no phosphorus shall contribute to the content of the component (E2). When comprising both phosphorus, and sulfur having a formal oxidation number of no more than +II, the component (E1) shall contribute to both the phosphorus content and the sulfur content of the component (E). The component (E) is a compound comprising a phosphorus atom, or a sulfur atom having a formal oxidation number of no more than +II, whereas all the sulfur atoms having any formal oxidation numbers in the component (E) shall contribute to the sulfur content of the component (E) when the component (E) further comprises a sulfur atom having a formal oxidation number of no less than +III.

<(F) Poly(meth)acrylate>

[0116] In one embodiment, the lubricating oil composition according to the present invention can further comprise at least one polyalkyl (meth)acrylate having a weight average molecular weight of more than 25,000 (hereinafter may be referred to as "component (F)"). As the component (F), one polyalkyl (meth)acrylate may be used alone, or two or more polyalkyl (meth)acrylates may be used in combination. In the present description, "(meth)acrylate" means "acrylate and/or methacrylate".

[0117] As the component (F), a polyalkyl (meth)acrylate that is used as a viscosity index improver or a pour point depressant in lubricating oil, and has a weight average molecular weight of more than 25,000 can be used without particular limitations. As the component (F), either one of a non-dispersant poly(meth)acrylate and a dispersant poly(meth)acrylate may be used, or any combination thereof may be used. In view of improving anti-seizure performance, a non-dispersant poly(meth)acrylate is preferably used. In the present description, "dispersant poly(meth)acrylate" means a poly(meth)acrylate compound having a functional group including a nitrogen atom, and "non-dispersant poly(meth)acrylate" means a poly(meth)acrylate compound not having a functional group including a nitrogen atom.

[0118] The weight average molecular weight of the component (F) is preferably more than 25,000, and more preferably no less than 27,000 in view of improving anti-fatigue performance, and of further improving the electrical insulation of the fresh oil; is preferably no more than 100,000, and more preferably no more than 80,000 in view of improving anti-seizure performance; and in one embodiment, can be more than 25,000 and no more than 100,000, or 27,000 to 80,000.

[0119] The lubricating oil composition may optionally comprise the component (F). The content of the component (F) in the lubricating oil composition on the basis of the total mass of the composition is preferably 0 to 5.0 mass%, and more preferably 0 to 4.0 mass% in view of further improving fuel efficiency; is preferably no less than 0.01 mass%, and more preferably no less than 0.015 mass% in view of further improving the low-temperature fluidity of the fresh oil; and in one embodiment, can be 0.01 to 5.0 mass%, or 0.015 to 4.0 mass%.

<Other Additives>

[0120] In one embodiment, the lubricating oil composition can further comprise at least one additive selected from a friction modifier other than the component (B) or the component (E), a viscosity index improver other than the component (F), a pour point depressant other than the component (F), a corrosion inhibitor other than the component (E), an anti-rust agent, a metal deactivator other than the component (E), a defoaming agent, a demulsifier, and a coloring agent.

[0121] As a friction modifier other than the component (B) or the component (F), for example, at least one friction modifier selected from oil-soluble organic molybdenum compounds other than the component (E), and ashless friction modifiers other than the component (B) can be used. The lubricating oil composition may comprise no friction modifier. The content of the friction modifier in the lubricating oil composition is preferably 0 to 2 mass%, and more preferably 0

to 1 mass% on the basis of the total mass of the composition in view of further improving the electrical insulation of the fresh oil and the oxidatively deteriorated composition. The lower limit of this content is not particularly limited, and in one embodiment, can be no less than 0.005 mass%.

[0122] Examples of an oil-soluble organic molybdenum compound other than the component (E) include organic molybdenum compounds comprising no sulfur as a constituent element. Examples of organic molybdenum compounds comprising no sulfur as a constituent element include molybdenum-amine complexes, molybdenum-succinimide complexes, molybdenum salts of organic acid, and molybdenum salts of alcohol. The organic molybdenum compound herein may be a mononuclear molybdenum compound, or may be a polynuclear molybdenum compound such as binuclear molybdenum compounds and trinuclear molybdenum compounds.

[0123] The lubricating oil composition may optionally comprise a metal-containing additive (such as organic molybdenum compounds and zinc dialkyl dithiophosphate) other than the metallic detergent. The total content of metal elements in the lubricating oil composition is preferably less than 100 mass ppm in terms of metal on the basis of the total mass of the composition in view of further improving the electrical insulation of the fresh oil and the oxidatively deteriorated composition. In one embodiment, the total content of a metal-containing additive other than the component (C) in the lubricating oil composition is preferably 0 to 50 mass ppm, more preferably 0 to 30 mass ppm, and further preferably 0 to 10 mass ppm on the basis of the total mass of the composition in terms of metal.

[0124] As an ashless friction modifier other than the component (B), a known oiliness agent-based friction modifier can be used without any limitation. Examples of the ashless friction modifier include C6-50 compounds each including, in molecules thereof, at least one heteroatom selected from an oxygen atom, a nitrogen atom and a sulfur atom. More specifically, an ashless friction modifier such as aliphatic amine compounds, aliphatic imide compounds, fatty acid esters, fatty acid amides, fatty acid hydrazides, fatty acid metal salts, aliphatic alcohols, aliphatic ethers, and aliphatic urea compounds each having at least one C6-30, preferably C6-30 linear or branched chain alkyl or alkenyl group in molecules thereof can be preferably used.

[0125] As a viscosity index improver other than the component (F), a known viscosity index improver that is used in lubricating oil can be used without particular limitations. Examples of a viscosity index improver other than the component (F) include ethylene-α-olefin copolymers and hydrogenated products thereof, a copolymer of an α-olefin and an ester monomer having a polymerizable unsaturated bond, polyisobutylene and hydrogenated products thereof, hydrogenated products of styrene-diene copolymers, styrene-maleic anhydride/ester copolymers, and polyalkylstyrene. Among them, an ethylene-α-olefin copolymer or a hydrogenated product thereof, or combination thereof can be preferably used. The viscosity index improver may be either one of a dispersant and non-dispersant viscosity index improvers. In one embodiment, the weight average molecular weight of the viscosity index improver can be, for example, 3000 to 100,000. The lubricating oil composition may optionally comprise the viscosity index improver. The total content of the viscosity index improver in the lubricating oil composition is preferably 0 to 5.0 mass%, and more preferably 0 to 4.0 mass% on the basis of the total mass of the composition in view of further improving the electrical insulation of the oxidatively deteriorated composition. The lower limit of this total content is not particularly limited, and in one embodiment, can be no less than 0.1 mass%.

[0126] As a pour point depressant other than the component (F), a known pour point depressant such as an ethylene-vinyl acetate copolymer can be used according to the properties of the lubricant base oils to be used. The lubricating oil composition may optionally comprise a pour point depressant other than the component (F). The content of a pour point depressant other than the component (F) in the lubricating oil composition is preferably 0 to 1 mass%, and more preferably 0 to 0.8 mass% on the basis of the total mass of the composition in view of further improving the electrical insulation of the oxidatively deteriorated composition. The lower limit of this content is not particularly limited, and in one embodiment, can be no less than 0.015 mass%.

[0127] The lubricating oil composition may optionally comprise a polymer component other than the component (F) (such as viscosity index improvers and pour point depressants). The content of a polymer component having a weight average molecular weight of no more than 25,000 in the lubricating oil composition is preferably no less than 0 mass% and less than 0.1 mass%, more preferably 0 to 0.05 mass%, and especially preferably 0 to 0.01 mass% in view of further improving the electrical insulation of the oxidatively deteriorated composition.

[0128] A known corrosion inhibitor such as benzotriazole, tolyltriazole, and imidazole compounds can be used as a corrosion inhibitor other than the component (E). The lubricating oil composition may optionally comprise a corrosion inhibitor other than the component (E). The content of a corrosion inhibitor other than the component (E) in the lubricating oil composition is preferably 0 to 1 mass%, and more preferably 0 to 0.5 mass% on the basis of the total mass of the composition in view of further improving the electrical insulation of the fresh oil and the oxidatively deteriorated composition. The lower limit of this content is not particularly limited, and in one embodiment, can be no less than 0.01 mass%.

[0129] A known anti-rust agent such as petroleum sulfonate, alkylbenzenesulfonate, dinonylnaphthalenesulfonate, alkenylsuccinate esters, and polyol esters can be used as an anti-rust agent. The lubricating oil composition may optionally comprise this anti-rust agent. The content of the anti-rust agent in the lubricating oil composition is preferably 0 to 1 mass%, and more preferably 0 to 0.5 mass% on the basis of the total mass of the composition in view of further improving

the electrical insulation of the fresh oil and the oxidatively deteriorated composition. The lower limit of this content is not particularly limited, and in one embodiment, can be no less than 0.01 mass%.

[0130] A known metal deactivator such as imidazoline, pyrimidine derivatives, mercaptobenzothiazole, 2-(alkyldithio)benzimidazole, and β-(o-carboxybenzylthio)propionitrile can be used as a metal deactivator other than the component (E). The lubricating oil composition may optionally comprise a metal deactivator other than the component (E). The content of a metal deactivator other than the component (E) in the lubricating oil composition is preferably 0 to 1 mass%, and more preferably 0 to 0.5 mass% on the basis of the total mass of the composition in view of further improving the electrical insulation of the fresh oil and the oxidatively deteriorated composition. The lower limit of this content is not particularly limited, and in one embodiment, can be no less than 0.01 mass%.

[0131] A known defoaming agent such as silicones, fluorosilicones, and fluoroalkyl ethers can be used as a defoaming agent. The lubricating oil composition may optionally comprise this defoaming agent. The content of this defoaming agent in the lubricating oil composition is preferably 0 to 0.5 mass%, and more preferably 0 to 0.1 mass% in view of further improving the electrical insulation of the fresh oil and the oxidatively deteriorated composition. The lower limit of this content is not particularly limited, and in one embodiment, can be no less than 0.0001 mass%.

[0132] A known demulsifier such as polyalkylene glycol-based nonionic surfactants (such as polyoxyethylene alkyl ether, polyoxyethylene alkylphenyl ether, and polyoxyethylene alkylnaphthyl ether) can be used as a demulsifier. The lubricating oil composition may optionally comprise this demulsifier. The content of this demulsifier in the lubricating oil composition is preferably no more than 5 mass%, and more preferably no more than 3 mass% on the basis of the total mass of the composition in view of further improving the electrical insulation of the fresh oil and the oxidatively deteriorated composition. The lower limit of this content is not particularly limited, and in one embodiment, can be no less than 1 mass%.

[0133] As a coloring agent, for example, a known coloring agent such as azo compounds can be used.

<Lubricating Oil Composition>

[0134] The kinematic viscosity of the lubricating oil composition at 100°C is preferably no less than 1.8 mm^2/s, preferably no less than 2.0 mm^2/s, and more preferably no less than 2.2 mm^2/s, and in one embodiment, no less than 2.3 mm^2/s in view of further improving the electrical insulation of the fresh oil and the oxidatively deteriorated composition, and of sufficient oil film formation at a lubricating point to improve anti-wear performance; is preferably no more than 4.0 mm^2/s, and preferably no more than 3.8 mm^2/s in view of improving fuel efficiency; and in one embodiment, can be 1.8 to 4.0 mm^2/s, 2.0 to 4.0 mm^2/s, 2.2 to 4.0 mm^2/s, or 2.3 to 3.8 mm^2/s.

[0135] The kinematic viscosity of the lubricating oil composition at 40°C is preferably no less than 6.8 mm^2/s, and more preferably no less than 7.2 mm^2/s, and in one embodiment, no less than 8.0 mm^2/s in view of further improving the electrical insulation of the fresh oil and the oxidatively deteriorated composition, and anti-wear performance; is preferably no more than 14.5 mm^2/s, and more preferably no more than 13.7 mm^2/s, and in one embodiment, no more than 13.0 mm^2/s in view of further improving fuel efficiency; and in one embodiment, can be 6.8 to 14.5 mm^2/s, or 7.2 to 13.7 mm^2/s, or 8.0 to 13.0 mm^2/s.

[0136] Desirably, the kinematic viscosity of a lubricating oil composition is low in view of improving energy saving performance. Generally, however, addition of an additive to the base oil (component (O)) leads to an increase in kinematic viscosity of the entire lubricating oil composition. This means that the limit of the improvement in energy saving performance by the use of a less viscous lubricating oil is determined by the kinematic viscosity of the total base oil (component (O)). Therefore, desirably, the kinematic viscosity of the total base oil is lower in view of enhancing energy saving performance. In contrast, desirably, the kinematic viscosity of the total base oil is at a certain level or higher in view of improving anti-wear performance, oxidation stability, and the electrical insulation of the fresh oil and the oxidatively deteriorated composition. The viscosity increase effect of additives prevents a lubricating oil from being less viscous on condition that the kinematic viscosity of the total base oil is kept at a certain level or higher. The difference (KV_{40}^{Comp}-KV_{40}^{BO}) between the kinematic viscosity KV_{40}^{Comp} of the lubricating oil composition at 40°C and the kinematic viscosity KV_{40}^{BO} of the total base oil (component (O)) at 40°C, i.e., the viscosity increase effect of additives is preferably no more than 2.5 mm^2/s, and in one embodiment, is no more than 2.4 mm^2/s in view of further enhancing energy saving performance; and is preferably no less than 1.0 mm^2/s, is more preferably no less than 1.5 mm^2/s, and in one embodiment, is no less than 1.8 mm^2/s in view of further enhancing the durability against oxidative deterioration; and in one embodiment, can be 1.0 to 2.5 mm^2/s, 1.5 to 2.5 mm^2/s, or 1.8 to 2.4 mm^2/s.

[0137] The viscosity index of the lubricating oil composition is preferably no less than 100, and more preferably no less than 110 in view of further improving fuel efficiency and anti-wear performance; and in one embodiment, can be no less than 115, or no less than 120.

[0138] In one embodiment, the volume resistivity measured concerning the fresh oil of the lubricating oil composition at 80°C is preferably no less than 0.21×10^{10} Ω·cm. The upper limit of the volume resistivity of the fresh oil at 80°C is not particularly limited. In one embodiment, this volume resistivity can be 0.21×10^{10} to 0.60×10^{10} Ω·cm, or 0.21×10^{10} to 0.45×10^{10} Ω·cm. In the present description, the volume resistivity of the fresh oil shall be measured conforming to

the volume resistivity test specified in JIS C2101 at 80°C in oil temperature.

[0139] In one embodiment, the volume resistivity measured concerning the oxidatively deteriorated oil of the lubricating oil composition at 80°C is preferably no less than 0.10×10^{10} Ω·cm. The upper limit of the volume resistivity of the oxidatively deteriorated oil at 80°C is not particularly limited. In one embodiment, this volume resistivity can be 0.10×10^{10} to 0.40×10^{10} Ω·cm, or 0.10×10^{10} to 0.25×10^{10} Ω·cm. In the present description, the volume resistivity of the oxidatively deteriorated oil shall be measured conforming to the volume resistivity test specified in JIS C2101 at 80°C in oil temperature, and the oxidatively deteriorated oil is obtained by oxidation treatment on the fresh oil at 165°C for 150 hours by the ISOT method (Indiana Stirring Oxidation Test) specified in JIS K2514-1.

[0140] In one embodiment, the total content of a compound that has a non-phenolic OH group (which may be part of another functional group (such as carboxy group and phosphoric acid group)) or a salt thereof, >NH group, or - NH_2 group (hereinafter may be referred to as "O/N-based active hydrogen-containing group"), and that do not contribute to any of the contents of the metallic detergent (including metal salicylate detergents and metal sulfonate detergents such as the component (C), and metal phenate detergents), a phosphite diester compound having no O/N-based active hydrogen-containing group in the alcohol residue (such as the phosphite ester compound represented by the general formula (8) (component (E1)), the first succinimide compound (component (A)), the second succinimide compound (component (B)), the amine antioxidant or phenol antioxidant (component (D)), and the poly(meth)acrylate (such as the component (F)) can be preferably 0 to 500 mass ppm, in one embodiment, 0 to 300 mass ppm, and in another embodiment, 0 to 150 mass ppm on the basis of the total mass of the composition in terms of the total amount of an oxygen element and a nitrogen element in view of further improving the electrical insulation of the fresh oil and the oxidatively deteriorated oil. Examples of such an O/N-based active hydrogen compound include phosphoric acid and partial esters thereof, and salts thereof; phosphorous acid and partial esters thereof, and salts thereof (it is noted that phosphite diester having no foregoing O/N-based active hydrogen-containing group in the alcohol residue shall not fall under the O/N-based active hydrogen compound); nitrogen-containing oiliness agent-based friction modifiers having a N-H bond (such as primary fatty amines, secondary fatty amines, fatty acid primary amides, fatty acid secondary amides, aliphatic ureas having a N-H bond, and fatty acid hydrazides); nitrogen-containing oiliness agent-based friction modifiers having a hydroxy group (such as an amide of a fatty acid and a primary or secondary alkanolamine, and an amide of a primary or secondary fatty amine and an aliphatic hydroxy acid); nitrogen-containing oiliness agent-based friction modifiers having a carboxy group (which may be in a form of a salt) (such as N-acylated amino acids); oiliness agent-based friction modifiers having a hydroxy group (such as glycerol monooleate), and oiliness agent-based friction modifiers having a carboxy group (may be in a form of a salt) (such as fatty acids and fatty acid metal salts). When one O/N-based active hydrogen compound comprises both an oxygen element and a nitrogen element, both the amounts of the oxygen element and the nitrogen element which are derived from this compound shall contribute to the total content of the O/N-based active hydrogen compound (total amount of oxygen and nitrogen elements) irrespective of whether each oxygen atom of this compound is bonded to a hydrogen atom, and irrespective of whether each nitrogen atom of this compound is bonded to a hydrogen atom.

(Use)

[0141] The lubricating oil composition according to the present invention is less viscous but has electrical insulation required for lubrication of electric motors, and alleviates the problems resulting from oxidative deterioration in lubrication of automatic transmissions and lubrication of electric motors; thus, can be preferably used for lubrication of automatic transmissions as a lubricating oil with enhanced fuel efficiency, and can be also preferably used for lubrication and cooling of electric motors as a lubricating oil with enhanced energy saving performance. Both an automatic transmission and an electric motor can be also lubricated with the lubricating oil composition according to the present invention. Such a lubricating method can comprise: for example, supplying the lubricating oil composition according to the present invention to an automatic transmission of an automobile comprising the automatic transmission and an electric motor; and supplying the lubricating oil composition to the electric motor of the automobile.

Examples

[0142] Hereinafter, the present invention will be further specifically described based on examples and comparative examples. The present invention is not limited to these examples.

<Examples 1 to 20 and Comparative Examples 1 to 8>

[0143] As shown in tables 1 to 6, the lubricating oil compositions according to the present invention (examples 1 to 20), and lubricating oil compositions for comparison (comparative examples 1 to 8) were each prepared. In the items "base oil composition" in the tables, "mass%" means mass% on the basis of the total mass of the base oil (100 mass%);

and in the other items therein, "mass%" means mass% on the basis of the total mass of the lubricating oil composition (100 mass%). In the tables, "mass ppm" means mass ppm on the basis of the total mass of the lubricating oil composition, and the expression "mass ppm/X" for an element X means mass ppm as the amount of the element X on the basis of the total mass of the composition. The details of each of the components were as follows.

((O) Lubricant Base Oil)

[0144]

O-1: API Group III base oil (hydrocracked mineral base oil), kinematic viscosity (40°C): 7.0 mm^2/s, kinematic viscosity (100°C): 2.2 mm^2/s, viscosity index: 121, saturated content: 99.6, sulfur content: less than 1 mass ppm, $\%C_P$: 77.4, $\%C_N$: 22.0, $\%C_A$: 0.6
O-2: API Group III base oil (hydrocracked mineral base oil), kinematic viscosity (40°C): 19.2 mm^2/s, kinematic viscosity (100°C): 4.2 mm^2/s, viscosity index: 124, saturated content: 99.7, sulfur content: less than 1 mass ppm, $\%C_P$: 79.4, $\%C_N$: 20.6, $\%C_A$: 0.0
O-3: API Group III base oil (hydrocracked mineral base oil), kinematic viscosity (40°C): 18.2 mm^2/s, kinematic viscosity (100°C): 4.2 mm^2/s, viscosity index: 135, saturated content: 99.8, sulfur content: less than 1 mass ppm, $\%C_P$: 86.6, $\%C_N$: 13.4, $\%C_A$: 0.0
O-4: API Group III base oil (wax isomerized mineral base oil), kinematic viscosity (40°C): 9.2 mm^2/s, kinematic viscosity (100°C): 2.6 mm^2/s, viscosity index: 126, saturated content: 99.8, sulfur content: less than 1 mass ppm, $\%C_P$: 91.8, $\%C_N$: 8.2, $\%C_A$: 0.0
O-5: API Group IV base oil, kinematic viscosity (40°C): 18.4 mm^2/s, kinematic viscosity (100°C): 4.1 mm^2/s, viscosity index: 124
O-6: API Group V base oil (2-ethylhexyl oleate), kinematic viscosity (40°C): 8.2 mm^2/s, kinematic viscosity (100°C): 2.7 mm^2/s, viscosity index: 186

((A) First Succinimide Compound)

[0145]

A-1: condensation reaction product of a polyisobutenylsuccinic anhydride and a polyamine (non-modified polyisobutenylsuccinimide dispersant), weight average molecular weight: 5,200, N: 1.3 mass%, number average molecular weight of polyisobutenyl group: 1,600
A-2: boric acid-modified product of a condensation reaction product of a polyisobutenylsuccinic anhydride and a polyamine (boric acid-modified polyisobutenylsuccinimide dispersant), weight average molecular weight: 9,100, N: 0.73 mass%, B: 0.19 mass%, number average molecular weight of polyisobutenyl group: 2,500

((B) Second Succinimide Compound)

[0146] B-1: condensation reaction product of an octadecenyl succinic anhydride and a polyamine, N: 5.3 mass%

((C) Metallic Detergent)

[0147] C-1: calcium sulfonate overbased with calcium carbonate, base number: 300 mgKOH/g, Ca: 12.0 mass%

((D) Antioxidant)

[0148]

D-1: amine antioxidant (alkylated diphenylamine)
D-2: phenol antioxidant (octyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate)

((E) Phosphorus/Sulfur Additive)

[0149]

E-1: bis(3-thiaundecyl)phosphite, P: 7.3 mass%
E-2: thiadiazole compound having an alkyl group bonded to a thiadiazole ring via a disulfide bond (f, g = 2), represented

by any of the general formulae (10) to (12), S: 36 mass%

Table 1

Table 1		Examples				
		1	2	3	4	5
Base oil composition						
O-1	mass%		56	55	55	
O-2	mass%	19	41			
O-3	mass%			42		
O-4	mass%	81				85
O-5	mass%				45	15
O-6	mass%		3	3		
Viscosity properties of base oil						
kinematic viscosity (40°C)	mm^2/s	10.5	10.5	10.4	10.5	10.4
kinematic viscosity (100°C)	mm^2/s	2.9	2.9	2.9	2.9	2.9
(O) Total base oil	mass%	balance	balance	balance	balance	balance
(A) First succinimide						
A-1	mass% mass ppm/N	1.5 200	1.5 200	1.5 200	1.5 200	1.5 200
A-2	mass% mass ppm/N					
Mw × mass%		7800	7800	7800	7800	7800
(B) Second succinimide						
B-1	mass ppm/N	600	600	600	600	600
max(50, f(A))	mass ppm/N	199	199	199	199	199
(C) Ca detergent						
C-1	mass ppm/Ca	40	40	40	40	40
(D) Antioxidant						
D-1	mass%	0.3	0.3	0.3	0.3	0.3
D-2	mass%	0.2	0.2	0.2	0.2	0.2
(E) P/S additive						
E-1	mass ppm/P	150	150	150	150	150
E-2	mass ppm/S	260	260	260	260	260
P+S in total	mass ppm/P+S	721	721	721	721	721
Properties of composition						
kinematic viscosity (40°C)	mm^2/s	12.5	12.5	12.4	12.5	12.5
kinematic viscosity (100°C)	mm^2/s	3.3	3.3	3.3	3.3	3.2
viscosity index		139	138	142	138	140

(continued)

Table 1		Examples				
		1	2	3	4	5
viscosity increase caused by additives (40°C)	mm^2/s	2.0	2.0	2.0	2.0	2.1
Volume resistivity (80°C)						
fresh oil	$10^{10}\Omega \cdot cm$	0.37	0.36	0.37	0.37	0.37
oxidatively deteriorated oil	$10^{10}\Omega \cdot cm$	0.20	0.19	0.20	0.20	0.19
ISOT oxidation test						
increase in acid number	mgKOH/g	1.0	1.4	1.3	1.1	1.0

Table 2

Table 2		Examples				
		6	7	8	9	10
Base oil composition						
O-1	mass%	100				
O-2	mass%		19	19	19	19
O-3	mass%					
O-4	mass%		81	81	81	81
O-5	mass%					
O-6	mass%					
Viscosity properties of base oil						
kinematic viscosity (40°C)	mm^2/s	7.0	10.5	10.5	10.5	10.5
kinematic viscosity (100°C)	mm^2/s	2.2	2.9	2.9	2.9	2.9
(O) Total base oil	mass%	balance	balance	balance	balance	balance
(A) First succinimide						
A-1	mass%	1.5	2.1	1.1	1.1	0.8
	mass ppm/N	200	280	150	150	100
A-2	mass%					
	mass ppm/N					
Mw × mass%		7800	10920	5720	5720	4160
(B) Second succinimide						
B-1	mass ppm/N	600	50	600	400	800
max(50, f(A))	mass ppm/N	199	50	360	360	686
(C) Ca detergent						
C-1	mass ppm/Ca	40	40	40	40	40
(D) Antioxidant						
D-1	mass%	0.3	0.3	0.3	0.3	0.3
D-2	mass%	0.2	0.2	0.2	0.2	0.2

(continued)

Table 2		Examples				
		6	7	8	9	10
(E) P/S additive						
E-1	mass ppm/P	150	150	150	150	150
E-2	mass ppm/S	260	260	260	260	260
P+S in total	mass ppm/P+S	721	721	721	721	721
Properties of composition						
kinematic viscosity (40°C)	mm^2/s	8.8	12.6	12.4	12.4	12.4
kinematic viscosity (100°C)	mm^2/s	2.6	3.3	3.3	3.3	3.3
viscosity index		138	140	138	138	137
viscosity increase caused by additives (40°C)	mm^2/s	1.8	2.1	1.9	1.9	1.9
Volume resistivity (80°C)						
fresh oil	$10^{10}\Omega \cdot cm$	0.26	0.34	0.39	0.39	0.39
oxidatively deteriorated oil	$10^{10}\Omega \cdot cm$	0.16	0.21	0.12	0.11	0.11
ISOT oxidation test						
increase in acid number	mgKOH/g	1.3	1.4	1.2	1.4	1.4

Table 3

Table 3		Examples				
		11	12	13	14	15
Base oil composition						
O-1	mass%					
O-2	mass%	19	19	19	19	19
O-3	mass%					
O-4	mass%	81	81	81	81	81
O-5	mass%					
O-6	mass%					
Viscosity properties of base oil						
kinematic viscosity (40°C)	mm^2/s	10.5	10.5	10.5	10.5	10.5
kinematic viscosity (1 00°C)	mm^2/s	2.9	2.9	2.9	2.9	2.9
(O) Total base oil	mass%	balance	balance	balance	balance	balance
(A) First succinimide						
A-1	mass%	0.9	0.8	0.8	1.5	
	mass ppm/N	120	100	100	200	
A-2	mass%					
	mass ppm/N					1.5 110

(continued)

Table 3		Examples				
		11	12	13	14	15
Mw × mass%		4680	3900	3900	7800	13650
(B) Second succinimide						
B-1	mass ppm/N	500	1000	1300	600	600
max(50, f(A))	mass ppm/N	519	686	686	199	594
(C) Ca detergent						
C-1	mass ppm/Ca	40	40	40	40	40
(D) Antioxidant						
D-1	mass%	0.3	0.3	0.3	0.3	0.3
D-2	mass%	0.2	0.2	0.2	0.2	0.2
(E) P/S additive						
E-1	mass ppm/P	150	150	150	250	150
E-2	mass ppm/S	260	260	260	0	260
P+S in total	mass ppm/P+S	721	721	721	768	721
Properties of composition						
kinematic viscosity (40°C)	mm^2/s	12.3	12.4	12.5	12.5	12.9
kinematic viscosity (100°C)	mm^2/s	3.3	3.3	3.3	3.3	3.4
viscosity index		138	137	137	139	143
viscosity increase caused by additives (40°C)	mm^2/s	1.8	1.9	2.0	2.0	2.4
Volume resistivity (80°C)						
fresh oil	$10^{10}\Omega \cdot cm$	0.40	0.38	0.37	0.32	0.39
oxidatively deteriorated oil	$10^{10}\Omega \cdot cm$	0.10	0.11	0.12	0.13	0.14
ISOT oxidation test						
increase in acid number	mgKOH/g	1.4	1.3	1.3	1.4	1.3

Table 4

Table 4		Examples				
		16	17	18	19	20
Base oil composition						
O-1	mass%					
O-2	mass%	19	19	19	19	19
O-3	mass%					
O-4	mass%	81	81	81	81	81
O-5	mass%					

(continued)

Table 4		Examples				
		16	17	18	19	20
O-6	mass%					
Viscosity properties of base oil						
kinematic viscosity (40°C)	mm^2/s	10.5	10.5	10.5	10.5	10.5
kinematic viscosity (100°C)	mm^2/s	2.9	2.9	2.9	2.9	2.9
(O) Total base oil	mass%	balance	balance	balance	balance	balance
(A) First succinimide						
A-1	mass% mass ppm/N		1.9 250	0.9 120	1.5 200	1.5 200
A-2	mass% mass ppm/N	1.1 80				
Mw × mass%		10010	9880	4680	7800	7800
(B) Second succinimide						
B-1	mass ppm/N	1000	600	600	600	600
max(50, f(A))	mass ppm/N	973	95	519	199	199
(C) Ca detergent						
C-1	mass ppm/Ca	40	20	80	40	40
(D) Antioxidant						
D-1	mass%	0.3	0.3	0.3	0.5	1.1
D-2	mass%	0.2	0.2	0.2	0.4	1.1
(E) P/S additive						
E-1	mass ppm/P	150	150	150	150	150
E-2	mass ppm/S	260	260	260	260	260
P+S in total	mass ppm/P+S	721	721	721	721	721
Properties of composition						
kinematic viscosity (40°C)	mm^2/s	12.6	12.6	12.3	12.5	12.5
kinematic viscosity (100°C)	mm^2/s	3.3	3.3	3.3	3.3	3.3
viscosity index		139	139	138	139	139
viscosity increase caused by additives (40°C)	mm^2/s	2.1	2.1	1.8	2.0	2.0
Volume resistivity (80°C)						
fresh oil	$10^{10}\Omega \cdot cm$	0.41	0.38	0.35	0.31	0.23
oxidatively deteriorated oil	$10^{10}\Omega \cdot cm$	0.11	0.21	0.19	0.17	0.12
ISOT oxidation test						
increase in acid number	mgKOH/g	1.4	1.0	0.9	0.9	0.7

Table 5

Table 5		Comparative exmaples			
		1	2	3	4
Base oil composition O-1	mass%				
O-2	mass%	19	19	19	19
O-3	mass%				
O-4	mass%	81	81	81	81
O-5	mass%				
O-6	mass%				
Viscosity properties of base oil kinematic viscosity (40°C)	mm^2/s	10.5	10.5	10.5	10.5
kinematic viscosity (100°C)	mm^2/s	2.9	2.9	2.9	2.9
(O) Total base oil	mass%	balance	balance	balance	balance
(A) First succinimide A-1	mass% mass ppm/N	0.4 50	3.0 400		3.8 500
A-2	mass% mass ppm/N			2.0 150	
Mw × mass%		1950	15600	18200	19500
(B) Second succinimide B-1	mass ppm/N	1000	100	600	600
max(50, f(A))	mass ppm/N	50	50	360	50
(C) Ca detergent C-1	mass ppm/Ca	40	40	40	40
(D) Antioxidant D-1	mass%	0.3	0.3	0.3	0.3
D-2	mass%	0.2	0.2	0.2	0.2
(E) P/S additive E-1	mass ppm/P	150	150	150	150
E-2	mass ppm/S	260	260	260	260
P+S in total	mass ppm/P+S	721	721	721	721
Properties of composition kinematic viscosity (40°C)	mm^2/s	12.3	12.9	13.2	13.4
kinematic viscosity (100°C)	mm^2/s	3.2	3.4	3.5	3.5
viscosity index		135	144	145	142
viscosity increase caused by additives (40°C)	mm^2/s	1.8	2.4	2.7	2.9
Volume resistivity (80°C) fresh oil	$10^{10}\Omega \cdot cm$	0.43	0.27	0.36	0.21
oxidatively deteriorated oil	$10^{10}\Omega \cdot cm$	0.08	0.09	0.11	0.11
ISOT oxidation test					

(continued)

Table 5		Comparative exmaples			
		1	2	3	4
increase in acid number	mgKOH/g	2.0	1.2	1.2	0.3

Table 6

Table 6		Comparative exmaples			
		5	6	7	8
Base oil composition					
O-1	mass%				
O-2	mass%	19	19	19	19
O-3	mass%				
O-4	mass%	81	81	81	81
O-5	mass%				
O-6	mass%				
Viscosity properties of base oil					
kinematic viscosity (40°C)	mm^2/s	10.5	10.5	10.5	10.5
kinematic viscosity (100°C)	mm^2/s	2.9	2.9	2.9	2.9
(O) Total base oil	mass%	balance	balance	balance	balance
(A) First succinimide					
A-1	mass% mass ppm/N	1.5 200	0.8 100	2.1 280	3.1 410
A-2	mass% mass ppm/N				
Mw × mass%		7800	3900	10920	16120
(B) Second succinimide					
B-1	mass ppm/N	0	1500	0	0
max(50, f(A))	mass ppm/N	199	686	50	50
(C) Ca detergent					
C-1	mass ppm/Ca	40	40	120	90
(D) Antioxidant					
D-1	mass%	0.3	0.3	0.3	0.3
D-2	mass%	0.2	0.2	0.2	0.2
(E) P/S additive					
E-1	mass ppm/P	150	150	150	150
E-2	mass ppm/S	260	260	260	260
P+S in total	mass ppm/P+S	721	721	721	721
Properties of composition					
kinematic viscosity (40°C)	mm^2/s	12.4	12.5	12.8	13.1
kinematic viscosity (100°C)	mm^2/s	3.3	3.4	3.4	3.4
viscosity index		138	138	140	141

(continued)

Table 6		Comparative exmaples			
		5	6	7	8
viscosity increase caused by additives (40°C)	mm^2/s	1.9	2.0	2.3	2.6
Volume resistivity (80°C)					
fresh oil	$10^{10}\Omega \cdot cm$	0.39	0.37	0.16	0.21
oxidatively deteriorated oil	$10^{10}\Omega \cdot cm$	0.25	0.08	0.08	0.12
ISOT oxidation test					
increase in acid number	mgKOH/g	1.7	1.3	1.4	1.3

(Increase in Viscosity Resulting from Additives)

[0150] The increase (KV_{40}^{Comp}-KV_{40}^{BO}) of the kinematic viscosity of the composition at 40°C (KV_{40}^{Comp}) from the kinematic viscosity of the total base oil (component (O)) at 40°C (KV_{40}^{BO}) was calculated for each of the lubricating oil compositions, and thereby, the increase in viscosity resulting from the additives was evaluated. The results are shown in tables 1 to 6. A smaller value in the results means less deterioration of energy saving performance due to the viscosity increase effect of the additives. The increase in kinematic viscosity at 40°C resulting from the additives is preferably no more than 2.5 mm^2/s.

(ISOT Oxidation Test)

[0151] An oxidatively deteriorated oil of each of the lubricating oil compositions was obtained conforming to JIS K2514-1 by oxidation treatment on the fresh oil thereof at 165°C in oil temperature for 150 hours by the ISOT (Indiana Stirring Oxidation Test) method. The acid numbers of the fresh oil and the oxidatively deteriorated oil were measured conforming to JIS K2501-2003 by potentiometric titration, and the increase in acid number after the oxidative deterioration was evaluated. The results are shown in tables 1 to 6. In this test, a less increase of the acid number means better durability against the oxidative deterioration. The increase in acid number in this test is preferably no more than 1.5 mgKHO/g.

(Volume Resistivity)

[0152] The volume resistivity of the fresh oil, and the volume resistivity of the oxidatively deteriorated oil obtained by the foregoing ISOT oxidation test were measured for each of the lubricating oil compositions. The volume resistivity of the fresh oil, and the volume resistivity of the oxidatively deteriorated oil were each measured conforming to the volume resistivity test specified in JIS C2101 at 80°C in oil temperature. The results are shown in tables 1 to 6. In this test, higher volume resistivity means better electrical insulation. The volume resistivity of the fresh oil at 80°C in this test is preferably no less than $0.21 \times 10^{10}\ \Omega \cdot cm$. The volume resistivity of the oxidatively deteriorated oil at 80°C in this test is preferably no less than $0.10 \times 10^{10}\ \Omega \cdot cm$.

(Evaluation Results)

[0153] The lubricating oil compositions of examples 1 to 20 all showed good results of the increase in acid number of the oxidatively deteriorated oil, the electrical insulation of the fresh oil, and the electrical insulation of the oxidatively deteriorated composition while showing a less increase in viscosity resulting from the additives, and being less viscous.

[0154] The lubricating oil composition of comparative example 1, which comprised the component (A) in too small an amount (mass ppm) in terms of nitrogen, was inferior in the increase in acid number of the oxidatively deteriorated oil, and the electrical insulation of the oxidatively deteriorated composition.

[0155] The lubricating oil composition of comparative example 2, which comprised the component (A) in too large an amount (mass%) as the entire compound, was inferior in the electrical insulation of the oxidatively deteriorated composition.

[0156] The lubricating oil composition of comparative example 3, which comprised the component (A) such that the product of the weight average molecular weight of the component (A) and the amount (mass%) of the component (A) as the entire compound was too large, showed a more increase in viscosity resulting from the additives, and was inferior in energy saving performance.

[0157] The lubricating oil composition of comparative example 4, which comprised the component (A) such that the product of the weight average molecular weight of the component (A) and the amount (mass%) of the component (A) as the entire compound was too large, and that the amount (mass%) of the component (A) as the entire compound was also too large, showed a more increase in viscosity resulting from the additives, and was inferior in energy saving performance, and was also inferior in the electrical insulation of the fresh oil.

[0158] The lubricating oil composition of comparative example 5, which comprised no component (B), was inferior in the increase in acid number of the oxidatively deteriorated oil.

[0159] The lubricating oil composition of comparative example 6, which comprised the component (B) in too large an amount, was inferior in the electrical insulation of the oxidatively deteriorated composition.

[0160] The lubricating oil composition of comparative example 7, which comprised no component (B) but comprised the component (C), that is, the metallic detergent in a larger amount instead, so that the increase in acid number of the oxidatively deteriorated oil was suppressed, was inferior in the electrical insulation of the fresh oil and the oxidatively deteriorated oil.

[0161] The lubricating oil composition of comparative example 8, which comprised the component (C) in a smaller amount and the component (A) in a larger amount than those of comparative example 7, so that the increase in acid number of the oxidatively deteriorated oil was suppressed, was still inferior in the electrical insulation of the fresh oil, and showed a more increase in viscosity resulting from the additives and was also inferior in energy saving performance.

Claims

1. A lubricating oil composition comprising:

(O) a lubricant base oil comprising at least one mineral base oil or at least one synthetic base oil or any combination thereof, and having a kinematic viscosity at 40°C of 6.0 to 12.0 mm^2/s;
(A) a condensation reaction product of a polyisobutenylsuccinic acid or anhydride thereof and a first polyamine, or a modified product thereof, or any combination thereof, in an amount of no less than 80 mass ppm in terms of nitrogen and no more than 2.7 mass% as an entire compound on the basis of the total mass of the composition, the polyisobutenylsuccinic acid having a polyisobutenyl group, the polyisobutenyl group having a number average molecular weight of no less than 800;
(B) a condensation reaction product of an alkyl- or alkenyl- succinic acid having a C8-30 alkyl or alkenyl group or anhydride thereof and a second polyamine, or a modified product thereof, or any combination thereof, in an amount of 50 to 1300 mass ppm in terms of nitrogen on the basis of the total mass of the composition,
wherein a product of a weight average molecular weight (unit: Da) of the component (A) and the amount of the component (A) as the entire compound (unit: mass%) is no more than 16,000.

2. The lubricating oil composition according to claim 1, further comprising:
(C) at least one calcium sulfonate detergent overbased with calcium carbonate, or at least one calcium salicylate detergent overbased with calcium carbonate, or any combination thereof, in an amount of no less than 10 mass ppm and less than 100 mass ppm in terms of calcium on the basis of the total mass of the composition.

3. The lubricating oil composition according to claim 1 or 2, further comprising:
(D) at least one amine antioxidant and at least one phenol antioxidant, in an amount of 0.1 to 3.0 mass% as a total amount thereof, on the basis of the total mass of the composition.

4. The lubricating oil composition according to any one of claims 1 to 3, further comprising:
optionally (E) at least one phosphorus-containing compound, or at least one sulfur-containing compound comprising at least one sulfur atom having a formal oxidation number of no more than +II in a molecule thereof, or any combination thereof, in an amount of no more than 1000 mass ppm in terms of a total amount of phosphorus content and sulfur content on the basis of the total mass of the composition.

5. The lubricating oil composition according to any one of claims 1 to 4, wherein a relation of the following equation (1) is met,

$$B \geq \max(50, f(A))$$

$$f(A) = 100 \times \left(-10.5 + 14.1 \times \left(\frac{A}{100} - 0.5\right)^{-0.3}\right) \quad \cdots (1)$$

wherein in the equation (1), A represents the amount (unit: mass ppm) of the component (A) in terms of nitrogen on the basis of the total mass of the composition; and B represents the amount (unit: mass ppm) of the component (B) in terms of nitrogen on the basis of the total mass of the composition.

6. The lubricating oil composition according to any one of claims 1 to 5, further comprising:
optionally (F) at least one polyalkyl (meth)acrylate having a weight average molecular weight of more than 25,000, in an amount of no more than 5.0 mass% on the basis of the total mass of the composition.

7. The lubricating oil composition according to any one of claims 1 to 6, further comprising:
optionally, at least one polymer having a weight average molecular weight of no more than 25,000, in an amount of less than 0.1 mass% on the basis of the total mass of the composition.

8. The lubricating oil composition according to any one of claims 1 to 7, which is used to lubricate an automatic transmission.

9. The lubricating oil composition according to any one of claims 1 to 8, which is used to lubricate an electric motor.

10. A method for lubricating an automatic transmission and an electric motor,

the automatic transmission and the electric motor being comprised in an automobile,
the method comprising:

supplying the lubricating oil composition as defined in any one of claims 1 to 9 to the automatic transmission of the automobile; and
supplying the lubricating oil composition to the electric motor of the automobile.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2021/035044

A. CLASSIFICATION OF SUBJECT MATTER

C10M 171/02(2006.01)i; *C10N 10/04*(2006.01)n; *C10N 20/02*(2006.01)n; *C10N 20/04*(2006.01)n; *C10N 30/00*(2006.01)n; *C10N 30/10*(2006.01)n; *C10N 40/00*(2006.01)n; *C10N 40/04*(2006.01)n; ***C10M 101/02***(2006.01)i; ***C10M 129/10***(2006.01)i; ***C10M 133/12***(2006.01)i; ***C10M 133/16***(2006.01)i; ***C10M 133/56***(2006.01)i; ***C10M 135/00***(2006.01)i; ***C10M 137/00***(2006.01)i; ***C10M 145/14***(2006.01)i; ***C10M 159/22***(2006.01)i; ***C10M 159/24***(2006.01)i

FI: C10M171/02; C10M101/02; C10M133/56; C10M133/16; C10M159/24; C10M159/22; C10M133/12; C10M129/10; C10M137/00; C10M135/00; C10M145/14; C10N20:02; C10N20:04; C10N10:04; C10N40:04; C10N40:00 D; C10N30:00 Z; C10N30:10

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)

C10M171/02; C10N10/04; C10N20/02; C10N20/04; C10N30/00; C10N30/10; C10N40/00; C10N40/04; C10M101/02; C10M129/10; C10M133/12; C10M133/16; C10M133/56; C10M135/00; C10M137/00; C10M145/14; C10M159/22; C10M159/24

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

Published examined utility model applications of Japan 1922-1996
Published unexamined utility model applications of Japan 1971-2021
Registered utility model specifications of Japan 1996-2021
Published registered utility model applications of Japan 1994-2021

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2020/095968 A1 (JXTG NIPPON OIL & ENERGY CORP) 14 May 2020 (2020-05-14) paragraphs [0040]-[0048], [0050]-[0060], [0080]-[0085], examples 25, 26, claims	1-10
A	JP 2003-113391 A (NIPPON OIL CORP) 18 April 2003 (2003-04-18) entire text	1-10
A	US 2018/0371357 A1 (THE LUBRIZOL CORPORATION) 27 December 2018 (2018-12-27) entire text	1-10
A	JP 9-202890 A (TONEN CORP) 05 August 1997 (1997-08-05) entire text	1-10

☐ 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	Date of mailing of the international search report
07 December 2021	**14 December 2021**
Name and mailing address of the ISA/JP **Japan Patent Office (ISA/JP)** **3-4-3 Kasumigaseki, Chiyoda-ku, Tokyo 100-8915** **Japan**	Authorized officer Telephone No.

Form PCT/ISA/210 (second sheet) (January 2015)

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/JP2021/035044

Patent document cited in search report	Publication date (day/month/year)	Patent family member(s)	Publication date (day/month/year)
WO 2020/095968 A1	14 May 2020	WO 2020/095969 A1	
		WO 2020/095970 A1	
JP 2003-113391 A	18 April 2003	US 2004/0192562 A1 entire text	
		WO 2003/031544 A1	
US 2018/0371357 A1	27 December 2018	WO 2017/105747 A1	
		EP 3390594 A	
		CN 108473904 A	
		CA 3008675 A1	
JP 9-202890 A	05 August 1997	EP 889112 A1 entire text	
		WO 1998/017747 A1	

Form PCT/ISA/210 (patent family annex) (January 2015)

REFERENCES CITED IN THE DESCRIPTION

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Patent documents cited in the description

- JP 2009249496 A **[0003]**
- JP 2014159496 A **[0003]**
- JP 2016003258 A **[0003]**
- JP 2016020454 A **[0003]**
- WO 2020095968 A1 **[0003]**
- WO 2020095969 A1 **[0003]**
- WO 2020095970 A1 **[0003]**
- WO 2020171188 A1 **[0003]**