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#### (54)LUBRICATING OIL COMPOSITION

(57)The present invention provides a lubricating oil composition which can meet the demand for such fuel saving performance as exceeds the fuel saving performance of a conventional low viscosity lubricating oil composition. The lubricating oil composition comprises (A) a lubricating base oil having a  $%C_A$  of 2 or less and adjusted to have a 100°C kinematic viscosity of 1.5 to 4.5 mm<sup>2</sup>/s and (B) a viscosity index improver comprising (B1) a poly (meth)acrylate having a weight-average molecular weight of 50,000 or less in an amount of 1 to 10 percent by mass and (B2) a poly(meth)acrylate having a weightaverage molecular weight of 100,000 to 250,000 in an amount of 0.1 to 5 percent by mass on the total composition mass basis, the composition having a 80°C highshear viscosity (Vs: mPa·s) and a 80°C kinematic viscosity (Vk: mm<sup>2</sup>/s) ratio (Vs/Vk) of less than 1 and a coefficient of traction of 0.02 or less at 40°C, an average speed of 3.0 m/s, a slip ratio of 10%, and a contact pressure of 0.4 GPa.

#### Description

#### **Technical Field**

**[0001]** The present invention relates to lubricating oil compositions, more specifically to those having an enhanced fuel saving performance, suitable for automobile automatic transmissions, manual transmissions, and continuously variable transmissions.

#### **Background Art**

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**[0002]** Recently, energy saving in automobiles sand construction or agricultural machinery, i.e., fuel saving has become an urgent need in order to deal with environmental issues such as reduction in carbon dioxide emissions, and units such as engines, transmissions, final reduction gears, compressors, or hydraulic power units have been strongly demanded to contribute to energy saving. Consequently, the lubricating oils used in these units are required to be reduced in stir resistance and frictional resistance more than before.

**[0003]** Reduction of the viscosity of a lubricating oil can be pointed out as an effective energy saving means. For example, an automobile automatic transmission or continuously variable transmission has a torque converter, a wet clutch, a gear bearing mechanism, an oil pump and a hydraulic control system while a manual transmission or final reduction gear unit has a gear bearing mechanism. Reduction of the viscosity of a lubricating oil to be used in such transmissions can reduce the stir and frictional resistances in the torque converter, wet clutch, gear bearing mechanism and oil pump and thus enhance the power transmission efficiency, resulting in an improvement in the fuel efficiency of an automobile.

**[0004]** However, as described above, the lubricating oil used in these transmissions is also used as a medium for a hydraulic control system and thus would have problems that it would fail to generate sufficient hydraulic pressure due to the leakage from the oil pump or control valve if it is excessively reduced in viscosity. The lubricating oil used in the above devices until the working life thereof comes an end is thus required to maintain a certain viscosity therefor.

[0005] Conventionally, an automatic transmission oil has been provided, which is low in viscosity as the whole composition to enhance fuel efficiency but increased in base oil viscosity to retain shear stability, lubricating oil life and the like and also increased in high temperature high shear (HTHS) viscosity and thus also oil film retaining properties to obtain excellent anti-wear properties, anti-pitching properties, shear stability and low temperature viscosity characteristics (see, for example, Patent Literature 1 below). However, the oil has been unable to meet the recent further fuel saving demand.

### **Citation List**

#### **Patent Literature**

[0006] Patent Literature 1: Japanese Patent Application Publication 2009-096925

#### 40 Summary of Invention

### **Technical Problem**

**[0007]** The present invention has been made in view of these circumstances and has an object to provide a lubricating oil composition accomplishing a further excellent fuel saving performance while maintaining viscosity with means exceeding the conventional concept.

#### **Solution to Problem**

**[0008]** The inventors of the present invention have focused on a lubricating base oil and a polymer in order to solve the above problems and as the result of study thereof have accomplished the present invention on the basis of the finding that the above problems was able to be solved with a lubricating oil composition comprising a selected specific base oil and a selected specific poly(meth)acrylate-based additive.

**[0009]** That is, the present invention relates to a lubricating oil composition comprising (A) a lubricating base oil having a  ${}^{\circ}$ C<sub>A</sub> of 2 or less and adjusted to have a 100°C kinematic viscosity of 1.5 to 4.5 mm<sup>2</sup>/s and (B) a viscosity index improver comprising (B1) a poly(meth)acrylate having a weight-average molecular weight of 50,000 or less in an amount of 1 to 10 percent by mass and (B2) a poly(meth)acrylate having a weight-average molecular weight of 100, 000 to 250,000 in an amount of 0.1 to 5 percent by mass on the total composition mass basis, the composition having a 80°C

high-shear viscosity (Vs: mPa·s) and a 80°C kinematic viscosity (Vk: mm²/s) ratio (Vs/Vk) of less than 1 and a coefficient of traction of 0.02 or less at 40°C, an average speed of 3.0 m/s, a slip ratio of 10%, and a contact pressure of 0.4 GPa. **[0010]** The present invention also relates to the aforesaid lubricating oil composition wherein the 80°C viscosity reduction rate 8 hours after a sonic shear test is 8% or less.

**[0011]** The present invention also relates to the aforesaid lubricating oil composition wherein it is used for a mechanism containing an oil pump as a structural element.

[0012] The present invention also relates to the aforesaid lubricating oil composition wherein it is used for a continuously variable transmission.

#### Advantageous Effects of Invention

**[0013]** The lubricating oil composition of the present invention can reduce resistance to lubrication more than the conventional oils and thus are suitably used in for example automobile manual transmissions, automatic transmissions, continuously variable transmissions or final reduction gears, in particularly suitably continuously variable transmissions so that the composition can contribute to an enhancement in the fuel efficiency of an automobile.

#### **Description of Embodiment**

**[0014]** The present invention will be described in detail below.

**[0015]** The lubricating base oil referred to as (A) used in the present invention is a lubricating base oil having a %C<sub>A</sub> of 2 or less and adjusted to have a 100°C kinematic viscosity of 1.5 to 4.5 mm<sup>2</sup>/s and may be a mineral lubricating base oil, a synthetic lubricating base oil or a mixture thereof.

**[0016]** Examples of the mineral lubricating base oil which may be used in the present invention include paraffinic or naphthenic mineral base oils which can be produced by subjecting a lubricating oil fraction produced by atmospheric-or vacuum-distillation of a crude oil, to any one of or any suitable combination of refining processes selected from solvent deasphalting, solvent extraction, hydrocracking, solvent dewaxing, catalytic dewaxing, hydrorefining, sulfuric acid treatment, and clay treatment; n-paraffins; and iso-paraffins. These base oils may be used alone or in combination at an arbitrary ratio.

[0017] Examples of preferred mineral lubricating base oils include the following base oils:

(1) a distillate oil produced by atmospheric distillation of a paraffin base crude oil and/or a mixed base crude oil;

- (2) a whole vacuum gas oil (WVGO) produced by vacuum distillation of the topped crude of a paraffin base crude oil and/or a mixed base crude oil;
- (3) a wax produced by a lubricating oil dewaxing process and/or a Fischer-Tropsch wax produced by a GTL process;
- (4) an oil produced by mild-hydrocracking (MHC) one or more oils selected from oils of (1) to (3) above;
- (5) a mixed oil of two or more oils selected from (1) to (4) above;
- (6) a deasphalted oil (DAO) produced by deasphalting an oil of (1), (2) (3), (4) or (5);
- (7) an oil produced by mild-hydrocracking (MHC) an oil of (6); and
- (8) a lubricating oil produced by subjecting a mixed oil of two or more oils selected from (1) to (7) used as a feed stock and/or a lubricating oil fraction recovered therefrom to a normal refining process and further recovering a lubricating oil fraction from the refined product.

**[0018]** No particular limitation is imposed on the normal refining process used herein. Therefore, there may be used any refining process having been conventionally used upon production of a lubricating base oil. Examples of the normal refining process include (a) hydro-refining processes such as hydrocracking and hydrofinishing, (b) solvent refining such as furfural extraction, (c) dewaxing such as solvent dewaxing and catalytic dewaxing, (d) clay refining with acidic clay or active clay and (e) chemical (acid or alkali) refining such as sulfuric acid treatment and sodium hydroxide treatment. In the present invention, any one or more of these refining processes may be used in any combination and order.

**[0019]** The mineral lubricating base oil used in the present invention is particularly preferably a base oil produced by further subjecting a base oil selected from (1) to (8) described above to the following treatments.

**[0020]** That is, preferred are a hydrocracked mineral oil and/or wax-isomerized isoparaffinic base oil produced by hydrocracking or wax-isomerizing a base oil selected from (1) to (8) described above as it is or a lubricating fraction recovered therefrom and subjecting the resulting product as it is or a lubricating fraction recovered therefrom to dewaxing such as solvent dewaxing or catalytic dewaxing, followed by solvent refining or followed by solvent refining and then dewaxing such as solvent dewaxing or catalytic dewaxing. The hydrocracked mineral oil and/or wax-isomerized isoparaffinic base oil are used in an amount of preferably 30 percent by mass or more, more preferably 50 percent by mass or more, and particularly preferably 70 percent by mass or more, on the total base oil mass basis.

[0021] Examples of synthetic lubricating base oils which may be used in the present invention include poly-α-olefins

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and hydrogenated compounds thereof; isobutene oligomers and hydrogenated compounds thereof; isoparaffins; alkyl-benzenes; alkylnaphthalenes; diesters such as ditridecyl glutarate, di-2-ethylhexyl adipate, diisodecyl adipate, ditridecyl adipate and di-2-ethylhexyl sebacate; polyol esters such as trimethylolpropane caprylate, trimethylolpropane pelargonate, pentaerythritol 2-ethylhexanoate and pentaerythritol pelargonate; polyoxyalkylene glycols; dialkyldiphenyl ethers; and polyphenyl ethers.

**[0022]** Preferred synthetic lubricating base oils are poly- $\alpha$ -olefins. Typical examples of poly- $\alpha$ -olefins include oligomers or cooligomers of  $\alpha$ -olefins having 2 to 32, preferably 6 to 16 carbon atoms, such as 1-octene oligomer, 1-decene oligomer, ethylene-propylene cooligomer, and hydrogenated compounds thereof.

**[0023]** No particular limitation is imposed on the method of producing poly- $\alpha$ -olefins. For example, poly- $\alpha$ -olefins may be produced by polymerizing  $\alpha$ -olefins in the presence of a polymerization catalyst such as a Friedel-Crafts catalyst containing aluminum trichloride, boron trifluoride or a complex of boron trifluoride with water, an alcohol such as ethanol, propanol and butanol, a carboxylic acid or an ester (for example, ethyl acetate and ethyl proplonate).

**[0024]** Component (A) used in the present invention may be a mixture of two or more types of mineral base oils or two or more types of synthetic base oils or a mixture of mineral base oils and synthetic base oils. The mix ratio of two or more base oils in such mixtures may be arbitrarily selected.

**[0025]** The lubricating base oil referred to as Component (A) used in the transmission lubricating oil composition of the present invention is a lubricating base oil adjusted so that the 100°C kinematic viscosity is from 1.5 to 4.5 mm<sup>2</sup>/s. **[0026]** Component (A) is preferably one or more types selected from the following (A-a) to (A-c):

(A-a) a mineral base oil having a  $100^{\circ}$ C kinematic viscosity of 1.5 to lower than 3.5 mm<sup>2</sup>/s, preferably 1.9 to 3.2 mm<sup>2</sup>/s; (A-b) a mineral base oil having a  $100^{\circ}$ C kinematic viscosity of 3.5 to lower than 7 mm<sup>2</sup>/s, preferably 3.6 to 4.5 mm<sup>2</sup>/s; and

(A-c) a poly  $\alpha$ -olefin base oil having a 100°C kinematic viscosity of 1.5 to lower than 7 mm<sup>2</sup>/s, preferably 3.8 to 4.5 mm<sup>2</sup>/s.

**[0027]** Lubricating base oils (A-a) to (A-b) have a  ${}^{\circ}C_A$  of 2 or less, preferably 1 or less, more preferably 0.5 or less, particularly preferably substantially 0. Lubricating oil (A-c) has a  ${}^{\circ}C_A$  of substantially 0. The use of lubricating base oil (A) having a  ${}^{\circ}C_A$  of 2 or less renders it possible to produce a lubricating oil composition with an excellent oxidation stability and a coefficient of traction of 0.02 or less that is required by the present invention.

[0028] The  $%C_A$  used herein denotes the percentage of the aromatic carbon number in the total carbon number, determined in accordance with ASTM D 3238-85.

**[0029]** No particular limitation is imposed on the viscosity index of lubricating base oils (A-a) to (A-c), which is, however, preferably 80 or greater, more preferably 100 or greater, particularly preferably 120 or greater and usually 200 or less, preferably 160 or less. The use of a lubricating base oil having a viscosity index of greater than 80 renders it possible to produce a composition exhibiting excellent viscosity characteristics from low temperatures to high temperatures and also having a low coefficient of traction. The use of a lubricating base oil having a too high viscosity index results in a too much normal paraffins in the resulting composition and also deteriorates the low temperature fluidity thereof.

**[0030]** No particular limitation is imposed on the sulfur content of lubricating base oils (A-a) to (A-b) used in the present invention, which is, however, 0.1 percent by mass or less, more preferably 0.05 percent by mass or less, more preferably 0.02 percent by mass or less, particularly preferably 0.01 percent by mass or less, most preferably 0.005 percent by mass or less. The sulfur content of lubricating base oil (A-c) is substantially 0%. Reduction of the sulfur content of Component (A) renders it possible to produce a composition having a more excellent oxidation stability.

[0031] In the present invention, any one or more of the above-described base oils (A-a) to (A-c) may be used. Above all, preferably (A-a) and (A-b) and/or (A-c) are used in combination. When Component (A-a) and/or Component (A-b) and Component (A-c) are used in combination, the content of Component (A-c) is preferably 1 to 50 percent by mass, more preferably 3 to 20 percent by mass, more preferably 3 to 10 percent by mass on the total base oil mass basis. In particular, blending of Component (A-c) in an amount of on the order of 3 to 10 percent by mass renders it possible to produce a composition exhibiting excellent effects in fatigue life, low temperature characteristics, and oxidation stability at a low cost.

[0032] Lubricating base oil (A) used in the present invention has a 100°C kinematic viscosity of 1.5 to 4.5 mm²/s, preferably 2.8 to 4.0 mm²/s, particularly preferably 3.6 to 3.9 mm²/s. The use of a lubricating base oil with a 100°C kinematic viscosity of 4.5 mm²/s or lower renders it possible to produce a lubricating oil composition with a smaller frictional resistance at lubricating sites because its fluid resistance is small and thus with excellent low temperature viscosity (for example, the -40°C Brookfield viscosity is 20,000 Pa·s or less). The use of a lubricating base oil with a 100°C kinematic viscosity of 1.5 mm²/s or higher renders it possible to produce a lubricating oil composition which is sufficient in oil film formation and thus more excellent in lubricity and less in evaporation loss of the base oil under elevated temperature conditions.

 $\textbf{[0033]} \quad \text{Lubricating base oil (A) used in the present invention has a \% C_{A} of 2 or less, preferably 1 or less, more pre$ 

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0.5 or less, particularly preferably substantially 0. The use of the lubricating base oil (A) with a  ${}^{\circ}$ C<sub>A</sub> of 2 or less renders it possible to produce a composition with more excellent oxidation stability and a coefficient of traction 0.02 or less, which is required by the present invention.

**[0034]** No particular limitation is imposed on the viscosity index of lubricating base oil (A) used in the present invention, which is, however, preferably 80 or greater, more preferably 100 or greater, particularly preferably 120 or greater. The use of a lubricating base oil with a viscosity index of 80 or greater renders it possible to produce a composition with excellent viscosity characteristics from low temperatures to high temperatures and also a lower coefficient of traction.

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**[0035]** No particular limitation is imposed on the sulfur content of lubricating base oil (A) used in the present invention, which is, however, preferably 0.1 percent by mass or less, more preferably 0.05 percent by mass or less, more preferably 0.02 percent by mass or less, particular preferably 0.01 percent by mass or less, most preferably 0.005 percent by mass or less. Decrease of the sulfur content of Component (A) results in a composition with an excellent oxidation stability.

[0036] Lubricating base oil (A) used in the present invention is as described above, but when the resulting lubricating oil composition has a problem in providing fatigue life, this base oil may be mixed with a solvent-refined base oil having a kinematic viscosity of 20 mm $^2$ /s to 50 mm $^2$ /s to an extent that the mixed lubricating base oil (A) is adjusted to have a %C<sub>A</sub> of 2 or less and a 100°C kinematic viscosity of 1.5 to 4.5 mm $^2$ /s and the resulting lubricating oil composition has coefficient of traction of 0.02 or less at 40°C, an average speed of 3.0 m/s, a slip ratio of 10%, and a contact pressure of 0.4 GPa.

[0037] The solvent-refined base oil with a kinematic viscosity of 20 mm $^2$ /s to 50 mm $^2$ /s is preferably such an oil with a sulfur content of 0.3 to 0.7 percent by mass and a  $^{8}$ C $_{A}$  of 5 or greater and 9 or less.

**[0038]** The viscosity index improver (Component (B)) contained in the lubricating oil composition of the present invention is preferably a poly(meth)acrylate-based additive substantially containing a structural unit derived from a monomer represented by formula (1) below.

$$CH_2 = C$$

$$C \longrightarrow C \longrightarrow R^2$$

$$C \longrightarrow C$$

$$C \longrightarrow$$

[0039] In formula (1),  $R^1$  is hydrogen or methyl, preferably methyl, and  $R^2$  is a hydrocarbon group having 1 to 30 carbon atoms.

**[0040]** Specific examples of the hydrocarbon group having 1 to 30 carbon atoms include alkyl groups having 1 to 30 carbon atoms, such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, straight-chain or branched pentyl, straight-chain or branched hexyl, straight-chain or branched octyl, straight-chain or branched octyl, straight-chain or branched undecyl, straight-chain or branched dodecyl, straight-chain or branched tetradecyl, straight-chain or branched pentadecyl, straight-chain or branched hexadecyl, straight-chain or branched hexadecyl, straight-chain or branched nonadecyl, straight-chain or branched henceicosyl, straight-chain or branched henceicosyl, straight-chain or branched henceicosyl, straight-chain or branched tetracosyl groups.

[0041] Component (B) used in the present invention may contain a structural unit derived from a monomer represented by formula (2) or (3) below.

**[0042]** In formula (2), R<sup>3</sup> is hydrogen or methyl, R<sup>4</sup> is an alkylene group having 1 to 30 carbon atoms, E<sup>1</sup> is an amine residue or heterocyclic residue having 1 or 2 nitrogen atoms and 0 to 2 oxygen atoms, and a is an integer of 0 or 1.

$$CH_2 = C R^5$$

$$E^2$$
(3)

**[0043]** In formula (3), R<sup>5</sup> is hydrogen or methyl, and E<sup>2</sup> is an amine residue or heterocyclic residue having 1 or 2 nitrogen atoms and 0 to 2 oxygen atoms.

**[0044]** Specific examples of the groups represented by E<sup>1</sup> and E<sup>2</sup> include dimethylamino, diethylamino, dipropylamino, dibutylamino, anilino, toluidino, acetylamino, benzoilamino, morpholino, pyrrolyl, pyrrolino, pyridyl, methylpyridyl, pyrolidinyl, piperidinyl, quinonyl, pyrrolidonyl, pyrrolidono, imidazolino and pyrazino groups.

**[0045]** Preferable examples include dimethylaminomethyl methacrylate, diethylaminomethyl methacrylate, dimethylaminoethyl methacrylate, diethylaminoethyl methacrylate, 2-methyl-5-vinyl pyridine, morpholinomethyl methacrylate, morpholinoethyl methacrylate, N-vinyl pyrrolidone and mixtures thereof.

**[0046]** Specific examples of Component (B) include copolymers of monomers (Ba) to (Bd) represented by formula (1) and polar group-containing monomers (Be) represented by formula (2) and/or (3) used if necessary:

- (Ba) (meth)acrylates wherein R<sup>2</sup> is an alkyl group of 1 to 4 carbon atoms;
- (Bb) (meth)acrylate wherein R<sup>2</sup> is an alkyl group of 5 to 10 carbon atoms;
- (Bc) (meth)acrylates wherein R<sup>2</sup> is an alkyl group of 12 to 18 carbon atoms;
- (Bd) (meth)acrylate wherein R<sup>2</sup> is an alkyl group of 20 or more carbon atoms; and
- (Be) polar group-containing monomers.

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[0047] The structural ratio of monomers (Ba) to (Be) are preferably as follows on the total monomer mass basis:

Component (Ba): preferably 10 to 60 percent by mass, more preferably 20 to 50 percent by mass,

Component (Bb): preferably 0 to 50 percent by mass, more preferably 0 to 20 percent by mass

Component (Bc): preferably 10 to 60 percent by mass, more preferably 20 to 40 percent by mass,

Component (Bd): preferably 1 to 20 percent by mass, more preferably 5 to 10 percent by mass

Component (Be): preferably 0 to 20 percent by mass, more preferably 0 to 10 percent by mass, particularly preferably 0 to 5 percent by mass.

**[0048]** Blending of a poly(meth)acrylate-based viscosity index improver with this formulation can improve the low temperature viscosity characteristics of the resulting composition and extend the fatigue life thereof at the same time.

**[0049]** No particular limitation is imposed on the method for producing the above-described poly(meth)acrylate. For example, it can be easily produced by the radical-solution polymerization of a mixture of monomers (Ba) to (Be) in the presence of a polymerization initiator such as benzoyl peroxide.

[0050] Component (B) used in the present invention is a viscosity index improver having two types of weight-average molecular weights, i.e., comprising (B1) a poly(meth)acrylate having a weight-average molecular weight of 50,000 or less in an amount of 1 to 10 percent by mass and (B2) a poly(meth)acrylate having a weight-average molecular weight of 100,000 to 250,000 in an amount of 0.1 to 5 percent by mass, on the total composition mass basis.

[0051] The weight-average molecular weight of Component (B1) is 50, 000 or less, preferably 40,000 or less, more preferably 30,000 or less and 5,000 or more, preferably 10,000 or more, more preferably 15,000 or more. Component (B1) with a weight-average molecular weight of more than 50,000 leads to a too low shear stability in combination with Component (B2), resulting in a lubricating oil composition which would fail to maintain the required viscosity. Component (B1) with a weight-average molecular weight of less than 5, 000 causes the high-shear viscosity to increase and thus fails to satisfy the 80°C high-shear viscosity (Vs: mPa·s) and 80°C kinematic viscosity (Vk: mm²/s) ratio (Vs/Vk) which is less than 1.

**[0052]** When the TBS viscometer is used to measure the high-shear viscosity, the ratio (Vs/Vk) of the high-shear viscosity (Vs: mPa·s) measured at a shear speed of 10<sup>-6</sup>/s, which is equal to 1 is the case where the molecular weight exceeds 50,000, and thus the value of less than 1 referred herein is a theoretical value.

[0053] The weight-average molecular weight of Component (B2) is 250,000 or less, preferably 200,000 or less, more preferably 170,000 or less and 100,000 or more, preferably 120,000 or more, more preferably 150,000 or more. Component (B2) with a weight-average molecular weight of more than 250,000 leads to a too low shear stability in combination with Component (B1), resulting in a lubricating oil composition which would fail to maintain the required viscosity. Component (B2) with a weight-average molecular weight of less than 100,000 is unlikely to reduce the high-shear viscosity

sufficiently, and thus cannot satisfy the 80°C high-shear viscosity (Vs: mPa·s) and 80°C kinematic viscosity (Vk: mm²/s) ratio (Vs/Vk) which is less than 1.

[0054] The weight-average molecular weight used herein denotes a weight-average molecular weight on polystyrene basis determined with a differential refractive index detector (RI) at a temperature of  $23^{\circ}$ C, a flow rate of 1 mL/min, a sample concentration of 1 percent by mass, and a sample injection amount of 75  $\mu$ L, using 150-C ALC/GPC manufactured by Waters having two columns GMHHR-M (7.8 mm IDx30 cm) equipped in series therein and tetrahydrofuran as a solvent. [0055] Component (B1) used in the present invention is added in an amount of 10 percent by mass or less, preferably 9 percent by mass or less, more preferably 8 percent by mass or less and 1 percent by mass or more, preferably 2 percent by mass or more, more preferably 4 percent by mass or more preferably 5 percent by mass or more on the total composition mass basis. Addition of Component (B1) in an amount of more than 10 percent by mass increases the high-shear viscosity too much while addition of Component (B1) in an amount of less than 1 percent by mass fails to obtain a sufficient composition viscosity.

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[0056] Component (B2) used in the present invention is added in an amount of 5 percent by mass or less, preferably 4 percent by mass or less, more preferably 3 percent by mass or less and 0.1 percent by mass or more, preferably 0.5 percent by mass or more, more preferably 1 percent by mass or more on the total composition mass basis. Addition of Component (B2) in an amount of more than 5 percent by mass results in a lubricating oil composition which is reduced in viscosity caused by shear and thus fails to obtain the necessary viscosity for extending the working life of machines. On the other hand, addition of Component (B2) in an amount of less than 0.1 percent by mass fails to obtain a sufficient composition viscosity.

**[0057]** The amount of Component (B) that is a poly(meth)acrylate-based additive in the lubricating oil composition of the present invention is as described above but is preferably such an amount that the resulting lubricating oil composition has a 80°C kinematic viscosity of 5 to 10 mm<sup>2</sup>/s, preferably 6 to 9 mm<sup>2</sup>/s and a viscosity index of 120 to 270, preferably 150 to 250, more preferably 170 to 220.

**[0058]** In the lubricating oil composition of the present invention, the 80°C high-shear viscosity (Vs: mPa·s) and 80°C kinematic viscosity (Vk: mm²/s) ratio (Vs/Vk) is necessarily less than 1. The high-shear viscosity referred herein is determined in accordance with ASTM D-4683 and is also referred to as "HTHS viscosity". In the present invention, the 80°C shear viscosity is measured at  $1\times10^6$  s<sup>-1</sup>.

[0059] A Vs/Vk of less than 1 means that the viscosity of the composition is reduced when high shear is applied thereto, and this is a phenomenon that could occur in a lubricating oil composition having a viscosity increased with a viscosity index improver because the shear speed is increased under conditions where sliding occurs through an extremely thin oil film, specifically conditions of lubrication through an oil film of 100  $\mu$ m or thinner, for example conditions of lubricating bearings, clutches or gear tooth surfaces. The present invention aims at securing a fuel saving performance by setting the level of reduction in viscosity to less than 1 so as to decrease the resistance caused by the reduction of viscosity occurring under high shear conditions. The viscosity when a high shear is applied only refers to a fluid lubricating state and thus no contact between individual pieces occurs.

**[0060]** Therefore, lower this value is, more improved the fuel saving performance is, but there is a certain limit to this value because if the value is too low, a problem in lubricity would arise. The lower limit of the value is 0.7, preferably 0.8 or greater, more preferably 0.85 or greater.

**[0061]** The lubricating oil composition of the present invention has a coefficient of traction of 0.02 or less, an average speed of 3.0 m/s, a slip ratio of 10%, and a contact pressure of 0.4 GPa, at 40°C.

**[0062]** This coefficient of traction is measured with a steel ball disk. While a disk with a radius of 13 cm is rotated at 286.7 rpm, a load of 20 N is applied on a ball with a radius of 1.27 cm placed on a position 10 cm apart radially from the center of the disk to measure the rotation torque applied to the ball at 40°C, an average speed of 3.0 m/s, a slip ratio of 10%, and a contact pressure of 0.4 GPa.

**[0063]** This condition does not reach what is called the complete elastohydrodynamic lubrication condition and thus is still in an intermediate region between the fluid lubrication condition and the elastohydrodynamic lubrication condition. Conventionally, the coefficient of traction of a lubricating oil composition is measured at a high contact pressure, specifically a contact pressure of greater than 1 GPa, and a composition with a higher oil film formability, i.e., coefficient of traction under such a high contact pressure condition is likely to form an oil film and has been evaluated as having an excellent lubricity under sever conditions.

**[0064]** However, the condition of measuring the coefficient of traction in the present invention is that for measuring it under an intermediate surface pressure condition that is 0.4 GPa as described above and thus can be regarded as a typical condition for parts where the coefficient of traction is involved with the resistance to lubrication among the parts of a machine to be lubricated except for ball bearings or roller bearings. Therefore, reduction of the coefficient of traction under the above-described conditions reduces the resistance relating to the coefficient of traction under lubricating conditions in a machine. That is, since the coefficient of traction at 40°C, an average speed of 3.0 m/s, a slip ratio of 10% and a contact pressure of 0.4 GPa is set to 0.02 or less, the composition of the present invention can secure a fuel saving effect.

**[0065]** Under these conditions, the coefficient of traction is 0.02 or less and is better as it is low but as described above, is preferably 0.005 or greater to secure the lubricity for ball bearings or roller bearings under a higher contact pressure condition.

[0066] The lubricating oil composition of the present invention has a viscosity reduction rate of preferably 8% or less at 80°C 8 hours after a sonic shear stability test. The sonic shear stability test referred herein is determined by a method prescribed in JASO M 347.

**[0067]** As described above, the lubricating oil composition of the present invention is also used as a medium for a hydraulic system, and if reduced in viscosity causes the system to have problems such as failure to produce sufficient pressure, due to leakage of the oil from the oil pump or control valve. Thus, the lubricating oil to be used until the working life of a system ends needs to maintain the viscosity as required for this purpose.

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**[0068]** The lubricating oil composition of the present invention needs to maintain a sufficient viscosity even if applied with repetitive shear force. What is meant by that the lubricating oil composition of the present invention has a viscosity reduction rate of 8% or less at 80°C 8 hours after a sonic shear stability test is the value to ensure that the composition has such a viscosity. The viscosity reduction rate is preferably 7% or less, more preferably 6% or less.

**[0069]** The lubricating oil composition of the present invention can be applicable to various devices such as manual transmissions, automatic transmissions, continuously variable transmissions, final reduction gears, and engines of automobiles, as well as agricultural machines and constructing machines. The lubricating oil composition is most suitably used in a continuously variable transmission. This is because this transmission has many parts likely subjected to shear and high contact pressure and the composition can exhibit its performances most effectively for the transmission.

**[0070]** If necessary, the lubricating oil composition of the present invention may be blended with various additives such as viscosity index improvers, extreme pressure additives, dispersants, metallic detergents, friction modifiers, anti-oxidants, corrosion inhibitors, rust inhibitors, demulsifiers, metal deactivators, pour point depressants, seal swelling agents, anti-foaming agents, and dyes, alone or in combination in order to further enhance the properties of the composition or impart the composition with properties required for a lubricating oil.

**[0071]** Examples of the viscosity index improvers include those other than Component (B) that are the above-described poly(meth)acrylates, such as non-dispersant or dispersant type ethylene- $\alpha$ -olefin copolymers and hydrogenated compounds thereof; polyisobutylene and hydrogenated compounds thereof; styrene-diene hydrogenated copolymers; styrene-maleic anhydride ester copolymers; polyalkylstyrenes; and copolymers of (meth)acrylate monomers represented by formula (1) and unsaturated monomers such as ethylene/propylene/styrene/maleic anhydride.

**[0072]** When the lubricating oil composition of the present invention is blended with a viscosity index improver (excluding Component (B)), no particular limitation is imposed on the content thereof if the resulting composition meets the requirements regarding the 100°C kinematic viscosity and viscosity index. The content is usually from 0.1 to 15 percent by mass, preferably 0.5 to 5 percent by mass on the total composition mass basis.

[0073] The extreme pressure additive is preferably blended, which comprises at least one type of phosphorus extreme pressure additive selected from phosphorous acid, phosphorus acid monoesters, phosphorus acid diesters, phosphorus acid triesters, and salts thereof; at least one type of sulfur extreme pressure additive selected from sulfurized fats and oils, sulfurized olefins, dihydrocarbyl polysulfides, dithiocarbamates, thiaziazoles, and benzothiazoles; and/or at least one type of phosphorus-sulfur extreme pressure additive selected from thiophosphorus acid, thiophosphorus acid monoesters, thiophosphorus acid diesters, thiophosphorus acid triesters, dithiophosphorus acid, trithiophosphorus acid, trithiophosphorus acid, trithiophosphorus acid, trithiophosphorus acid monoesters, trithiophosphorus acid diesters, trithiophosphorus acid triesters, and salts thereof.

**[0074]** The dispersant may be an ashless dispersant such as succinimide, benzylamine, polyamines, each having a hydrocarbon group having 40 to 400 carbon atoms, and/or a boron compound derivative thereof.

**[0075]** Any one or more compounds selected from these dispersants may be contained in an amount of usually from 0.01 to 15 percent by mass, preferably from 0.1 to 8 percent by mass on the total composition mass basis.

**[0076]** Examples of the metallic detergent include those such as alkaline earth metal sulfonates, alkaline earth metal phenates, and alkaline earth metal salicylates.

**[0077]** Any one or more compounds selected from these metallic detergents may be contained in an amount of usually 0.01 to 10 percent by mass, preferably 0.1 to 5 percent by mass on the total composition mass basis.

**[0078]** The friction modifier may be any compound that has been generally used as a friction modifiers for lubricating oils. Specific examples include amine compounds, imide compounds, fatty acid esters, fatty acid amides, and fatty acid metal salts, each having per molecule at least one alkyl or alkenyl group having 6 to 30 carbon atoms, particularly a straight-chain alkyl or alkenyl group having 6 to 30 carbon atoms.

[0079] Any one or more compounds selected from these friction modifiers may be contained in an amount of usually 0.01 to 5.0 percent by mass, preferably 0.03 to 3.0 percent by mass on the total composition mass basis.

[0080] The anti-oxidant may be any anti-oxidant that has been usually used in lubricating oils, such as phenol- or amine-based compounds.

[0081] Specific examples of the anti-oxidant include alkylphenols such as 2-6-di-tert-butyl-4-methylphenol; bisphenols

such as methylene-4,4-bisphenol(2,6-di-tert-butyl-4-methylphenol); naphthylamines such as phenyl- $\alpha$ -naphthylamine; dialkyldiphenylamines; zinc dialkyldithiophosphoric acids such as di-2-ethylhexyldithiophosphoric acid; and esters of (3,5-di-tert-butyl-4-hydroxyphenyl)fatty acid (propionic acid) or (3-methyl-5-tert-butyl-4-hydroxyphenyl)fatty acid (propionic acid) with a monohydric or polyhydric alcohol such as methanol, octanol, octadecanol, 1,6-hexanediol, neopentyl glycol, thiodiethylene glycol, triethylene glycol and pentaerythritol.

**[0082]** Any one or more of compounds selected from these compounds may be contained in any amount, which is, however, usually from 0.01 to 5 percent by mass, preferably from 0.1 to 3 percent by mass on the total composition mass basis.

[0083] Examples of the corrosion inhibitor include benzotriazole-, tolyltriazole-, thiadiazole-, and imidazole-types compounds.

**[0084]** Examples of the rust inhibitor include petroleum sulfonates, alkylbenzene sulfonates, dinonylnaphthalene sulfonates, alkenyl succinic acid esters, and polyhydric alcohol esters.

**[0085]** Examples of the demulsifier include polyalkylene glycol-based non-ionic surfactants such as polyoxyethylenealkyl ethers, polyoxyethylenealkylphenyl ethers, and polyoxyethylenealkylnaphthyl ethers.

[0086] Examples of the metal deactivator include imidazolines, pyrimidine derivatives, alkylthiadiazoles, mercaptobenzothiazoles, benzotriazoles and derivatives thereof, 1,3,4-thiadiazolepolysulfide, 1,3,4-thiadiazolyl-2,5-bisdialkyldithiocarbamate, 2-(alkyldithio)benzoimidazole, and  $\beta$ -(o-carboxybenzylthio)propionitrile.

**[0087]** The pour point depressant may be any of the known pour point depressants selected depending on the type of lubricating base oil but are preferably poly(meth)acrylates having a weight average molecular weight of preferably 20, 000 to 500,000, more preferably 50,000 to 300,000, particularly preferably 80,000 to 200,000.

**[0088]** The anti-foaming agent may be any compound that has been usually used as an anti-foaming agent for lubricating oils. Examples of such an anti-foaming agent include silicones such as dimethylsilicone and fluorosilicone. Any one or more of compounds selected from these compounds may be contained in any amount.

**[0089]** The seal swelling agent may be any compound that has been usually used as a seal swelling agent for lubricating oils. Examples of such a seal swelling agents include ester-, sulfur- and aromatic-based seal swelling agents.

**[0090]** The dye may be any compound that has been usually used and may be blended in any amount. The amount is usually from 0.001 to 1.0 percent by mass on the total composition mass basis.

**[0091]** When these additives are blended with the lubricating oil composition of the present invention, the corrosion inhibitor, rust inhibitor, and anti-foaming agent are each contained in an amount of 0.005 to 5 percent by mass, the pour point depressant and metal deactivator are each contained in an amount of 0.005 to 2 percent by mass, the seal swelling agent is contained in an amount of 0.01 to 5 percent by mass, and the anti-foaming agent is contained in an amount of 0.0005 to 1 percent by mass, all on the total composition mass basis.

### **Examples**

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**[0092]** The present invention will be described more specifically with reference to the following Examples and Comparative Examples but not limited thereto.

(Examples 1 and 2 and Comparative Examples 1 to 5)

**[0093]** Lubricating oil compositions for transmissions according to the present invention (Examples 1 and 2) were prepared in accordance with the formulations set forth in Table 1. The performances of each composition were evaluated with the following tests. The results are set forth in Table 1.

**[0094]** Also, lubricating oil composition for transmissions for comparison (Comparative Examples 1 to 5) were prepared in accordance with the formulations set forth in Table 1. The performances of each composition were also evaluated with the following tests. The results are also set forth in Table 1.

[Pump Test Power Consumption Reduction Rate]

[0095] A lubricating oil is circulated in a test apparatus including a pressure valve and an oil tank at a constant pump revolution number so as to measure the power consumed by the pump. The apparatus has a structure in which the lubricating oil is pumped out from the oil tank and returned through the pressure valve to the tank. The pressure valve is adjustably loaded during the circulation, and the oil tank has a heater and thus can adjust the lubricating oil temperature to any temperature. In these examples, the lubricating oil compositions were compared in terms of power consumption where the lubricating oil temperature and load were set to 80°C and 13 MPa, respectively.

5		Comparative Example 5		20		50	30		6.5				6		27.3	9.6	153	,	8.3 8.3	3	0.85	7.3	8.6
10		Comparative Example 4		65		35			4			1.2	6		24.9	8.6	189		7.3	15	0.81	5.7	7.0
15		Comparative Example 3		65		35					9		6		24.7	8.6	195		7.4	14	0.81	5.9	7.3
20 25		Comparative Example 2		65		35				6.2			ō		24.9	8.6	189	,	7.8	6	0.81	6.9	8.5
30	[Table 1]	Comparative Example 1		65		35			6.5				ō		25.0	8.6	185	,	8.3	3	0.81	7.0	8.6
35		Example 2	80		20				4		7		Ō		23.9	8.6	209	,	8.2	5	0.78	6.2	7.9
40		Example 1		65		35			4		8		o o		24.9	8.6	189	,	8.2	5	0.81	6.4	7.9
45			in mass%	mass%	mass%	mass%	mass%	mass%	mass%		Mm <sup>2</sup> /s	mm²/s		ć	mm <sup>2</sup> /s		g/cm <sup>2</sup>	mPa·s	mm <sup>2</sup> /s				
50		Total Base Oil Mass Basis	Base Oil 1	Base Oil 2	Base Oil 3	Base Oil 4	Base Oil 5	Total Composition Mass Basis	Viscosity Index Improver 1	Viscosity Index Improver 2	Viscosity Index Improver 3	Viscosity Index Improver 4	Additive Package	valuation	Kinematic Viscosity (40°C)	Kinematic Viscosity (80°C) Vk	lex	ShearTest (Sonic 8h) Kinematic Viscosity after Shear		Viscosity Reduction Rate	c)	sity (80°C)	Vs (unit coversion form density data)
55		Base Oil	0-1	0-2	0-3	4-0	0.5	Additives	A-1	A-2	A-3	A-4	B-1	Results of Evaluation	Kinematic Vi	Kinematic Vi	Viscosity Index	ShearTest (Sonic 8h) Kinematic Viscosity a	Test	Viscosity Re	Density (80°C)	HTHS Viscosity (80°C)	Vs (unit cove data)

5		Comparative Example 5	1.00	0.032	-1.8	ased anti-foaming
10		Comparative Example 4	0.81	0.020	1.7	rraction: 0.008. traction: 0.015. traction: 0.017. traction: 0.077. traction: 0.070. 00) s%, mass%, silicone-b
15		Comparative Example 3	0.85	0.020	4.	om, Coefficient of 1 pm, Coefficient of pm, Coefficient of pm, Coefficient of pm, Coefficient of PMA, Mw80,000) he PMA, Mw400,0 i-oxidant 3.0 mass welling agent 1.0.
20 25		Comparative Example 2	0.99	0.020	0.0	r Content: <10 pp fur Content: <10 pt ar Content: <10 pt ar Content: <10 pt fur Content: <10 pt on-dispersanttype ion-dispersant typ to 5 mass%, ant mass%, oil seal s
30	(continued)	Comparative Example 1	1.00	0.020	0.0	Base Oil 1 %CP: 92.5,%CA: 0, Kinematic Viscosity (100°C): 3.9mm²/s. Viscosity Index: 143, Sulfur Content: <10 ppm, Coefficient of traction: 0.008.  Base Oil 2 %CP: 78.5, %CA: 0, Kinematic Viscosity (100°C): 4.2 mm²/s, Viscosity Index: 124, Sulfur Content: <10 ppm, Coefficient of traction: 0.007.  Base Oil 3 %CP: 92.4, %CA: 0. Kinematic Viscosity (100°C): 2.7mm²/s, Viscosity Index: 128, Sulfur Content: <10 ppm, Coefficient of traction: 0.007.  Base Oil 4 %CP: 68.7, %CA: 0, Kinematic Viscosity (100°C): 2.4mm²/s, Viscosity Index: 43, Sulfur Content: <10 ppm, Coefficient of traction: 0.017.  Base Oil 5 %CP: 45.4, %CA: 9.5. Kinematic Viscosity (100°C): 5.8mm²/s, Viscosity Index: 43, Sulfur Content: <10 ppm, Coefficient of traction: 0.070.  Viscosity Index Improver 1 (Non-dispersant type PMA, Mw30,000), Viscosity Index Improver 2 (Non-dispersant type PMA, Mw400,000)  Viscosity Index Improver 3 (Non-dispersant type PMA, Mw150,000), Viscosity Index Improver 4 (Non-dispersant type PMA, Mw150,000), Viscosity Index Improver 4 (Non-dispersant type PMA, Mw150,000), Viscosity Index Improver 5 (mass%, anti-oxidant 3.0 mass%, anti-oxidant 4.0 mass%, metal deactivator 0.2 mass%, oil seal swelling agent 1.0 mass%, silicone-based anti-foaming agent (slight amount) are contained
35		Example 2	0.92	0.010	3.5	Jamm <sup>2</sup> /s. Viscosi 2 mm <sup>2</sup> /s, Viscos 7 mm <sup>2</sup> /s, Viscos 4 mm <sup>2</sup> /s, Viscos 5.8 mm <sup>2</sup> /s, Visco 00), Viscosity In 000), Viscosity In 10.3 mass%, C
40		Example 1	0.92	0.020	0.8	sity (100°C): 3.5 sity (100°C): 4.5 sity (100°C): 2.5 sity (100°C): 2.5 sity (100°C): 2.5 sity (100°C): 3.6 PMA, Mw30,0 s.7 PMA, Mw150,1 anti-wear ager modified mixture
45		Ξ			er %	Kinematic Visco Kinematic Visco Kinematic Visco Kinematic Visco S. Kinematic Visco -dispersant type dispersant type cicl ester-basec nodified /boron-ned
50		Total Base Oil Mass Basis		action	Pump Test Reduction rate of power consumption 80°C, 13 Mpa. Comparison with Comparative Example 1	Base Oil 1 %CP: 92.5, %CA: 0, Kinematic Viscosity (100°C) Base Oil 2 %CP: 78.5, %CA: 0, Kinematic Viscosity (100°C) Base Oil 3 %CP: 92.4, %CA: 0. Kinematic Viscosity (100°C) Base Oil 4 %CP: 68.7, %CA: 0, Kinematic Viscosity (100°C) Base Oil 5 %CP: 45.4, %CA: 9.5. Kinematic Viscosity (100°C) Viscosity Index Improver 1 (Non-dispersant type PMA, Mw3 Viscosity Index Improver 3 (Non-dispersant type PMA, Mw4 Additive Package: phosphorus acid ester-based anti-wear ashless dispersant (non-boron-modified horn-modified mix agent (slight amount) are contained
55		Base Oil	Vs/Vk	Coefficient of traction	Pump Test Reduction rate of pow consumption 80°C,13 Mpa. Comparison with Comparative Example 1	Base Oil 1 %CF Base Oil 2 %CF Base Oil 3 %CF Base Oil 4 %CF Base Oil 5 %CF Viscosity Index Viscosity Index Additive Packag ashless dispersagent (slight am

[0096] As apparent from the results set forth in Table 1, the lubricating oil compositions of Examples 1 and 2 have a Vs/Vk of less than 1 and a coefficient of traction of 0.02 or less. The compositions have a viscosity reduction rate of 8% or less at 80°C 8 hours after an sonic shear test. Whereas, the composition of Comparative Example 1 fails to have a Vs/Vk of less than 1, the compositions of Comparative Examples 2 to 4 have a viscosity reduction rate of greater than 8%, and the composition of Comparative Example 5 fails to have a Vs/Vk of less than 1 and have a greater coefficient of traction that is 0.032.

[Industrial Applicability]

[0097] The lubricating oil composition of the present invention has an excellent fuel saving performance and is advantageously used for manual transmissions, automatic transmissions, continuously variable transmissions and final reduction gears.

#### 15 Claims

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- 1. A lubricating oil composition comprising:
  - (A) a lubricating base oil having a  ${}^{\circ}C_A$  of 2 or less and adjusted to have a 100 ${}^{\circ}C$  kinematic viscosity of 1.5 to 4.5 mm<sup>2</sup>/s; and
  - (B) a viscosity index improver comprising (B1) a poly(meth)acrylate having a weight-average molecular weight of 50,000 or less in an amount of 1 to 10 percent by mass and (B2) a poly(meth)acrylate having a weight-average molecular weight of 100,000 to 250,000 in an amount of 0.1 to 5 percent by mass on the total composition mass basis,

the composition having a 80°C high-shear viscosity (Vs: mPa·s) and a 80°C kinematic viscosity (Vk: mm²/s) ratio (Vs/Vk) of less than 1 and a coefficient of traction of 0.02 or less at 40°C, an average speed of 3.0 m/s, a slip ratio of 10%, and a contact pressure of 0.4 GPa.

- 30 2. The lubricating oil composition according to claim 1 wherein the 80°C viscosity reduction rate 8 hours after a sonic shear test is 8% or less.
  - 3. The lubricating oil composition according to claim 1 or 2, wherein it is used for a mechanism containing an oil pump as a structural element.
  - 4. The lubricating oil composition according to claim 1 or 2 wherein it is used for a continuously variable transmission.

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## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2011/070813

## A. CLASSIFICATION OF SUBJECT MATTER

C10M145/14(2006.01)i, C10N20/02(2006.01)n, C10N20/04(2006.01)n, C10N30/06(2006.01)n, C10N40/04(2006.01)n

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) C10M145/14, C10N20/02, C10N20/04, C10N30/06, C10N40/04

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Jitsuyo Shinan Koho 1922–1996 Jitsuyo Shinan Toroku Koho 1996–2011 Kokai Jitsuyo Shinan Koho 1971–2011 Toroku Jitsuyo Shinan Koho 1994–2011

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 Y	WO 2006/043709 A1 (Nippon Oil Corp.), 27 April 2006 (27.04.2006), claims; page 1, lines 1 to 19; page 6, lines 19 to 22; example 14	1,3-4 2-4
	& CN 101065469 A	
Y A	JP 2004-155924 A (Tonen General Sekiyu Kabushiki Kaisha et al.), 03 June 2004 (03.06.2004), claims; paragraphs [0001], [0035] (Family: none)	2-4

× Fur	rther documents are listed in the continuation of Box C.		See patent family annex.
"A" docu	cial categories of cited documents: ument defining the general state of the art which is not considered e of particular relevance		later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"L" docu cited spec "O" docu "P" docu	ier application or patent but published on or after the international g date ument which may throw doubts on priority claim(s) or which is d to establish the publication date of another citation or other cital reason (as specified) ument referring to an oral disclosure, use, exhibition or other means ument published prior to the international filing date but later than priority date claimed	"Y"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art document member of the same patent family
	ne actual completion of the international search November, 2011 (22.11.11)	Date	of mailing of the international search report 06 December, 2011 (06.12.11)
	d mailing address of the ISA/ panese Patent Office	Auth	orized officer
Facsimile	No.	Teler	phone No.

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# INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2011/070813

	a). DOCUMENTS CONSIDERED TO BE RELEVANT	1
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No
А	WO 2007/123266 Al (Nippon Oil Corp.), 01 November 2007 (01.11.2007), claims; examples 1 to 4 & CN 101437927 A & EP 2009083 Al & JP 2007-284635 A & US 2009/0131291 Al	1-4
А	WO 2009/125551 A1 (Nippon Oil Corp.), 15 October 2009 (15.10.2009), claims; examples 1 to 4 & CN 102037107 A	1-4
A	WO 2007/001000 Al (Nippon Oil Corp.), 04 January 2007 (04.01.2007), claims; examples 1 to 5 & CN 101213277 A & EP 1908816 Al & JP 2008-308697 A & JP 2009-173948 A & US 2010/0144571 Al	1-4

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## REFERENCES CITED IN THE DESCRIPTION

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## Patent documents cited in the description

• JP 2009096925 A [0006]