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(54) **LUBRICANT OIL COMPOSITION FOR INTERNAL COMBUSTION ENGINE**

(57) The present invention provides a lubricating oil composition for an internal combustion engine that is excellent in functions as an engine oil for an internal combustion engine employing heat management and in particular fuel saving properties and detergency, comprising (A) a base oil having 100°C kinematic viscosity of 3.0 to 5.0 mm<sup>2</sup>/s and (B) a boronated succinimide in an amount of 0.007 percent by mass or more as boron and in an amount of 5 percent by mass or less as the succinimide

ashless dispersant, on the basis of the total mass of the composition, (C) a phenol-based antioxidant in an amount of 0.5 percent by mass or more and (D) a viscosity index improver having a ratio of the weight average molecular weight and PSSI of  $1.2 \times 10^4$  or greater in an amount of 0.1 to 5 percent by mass, the composition having a 150°C HTHS viscosity of 2.0 to 2.8 mPa·s, a 100°C HTHS viscosity of 4.8 mPa·s or lower and a viscosity index of 180 or greater.

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**Description****Technical Field**

5 [0001] The present invention relates to lubricating oil compositions for internal combustion engines (hereinafter also referred to as "engine oil"). More specifically, the present invention relates to an engine oil with excellent fuel saving properties, suitable for diesel engines.

**Background Art**

10 [0002] Internal combustion engines have been required to further improve the fuel saving properties for recent environmental measures such as control of CO<sub>2</sub> emissions.

[0003] An improvement in fuel economy by a lubricating oil has been carried out by reducing the working viscosity to reduce the viscous resistance (for example, see Patent Literature 1). However, there exists a certain viscosity necessary for lubrication of an internal combustion engine that limits the improvement. Internal combustion engine oils have been used as hydraulic pressure sources for driving valves and thus need to have a certain degree of viscosity to maintain the hydraulic pressure. Also for this reason, the engine oils have a limitation to the reduction of the viscosity. In order to overcome these limitations, heat management for an internal combustion engine is being introduced. For example, the required viscosity of an internal combustion engine oil can be further reduced by lowering the maximum working temperature thereof or by improving pumps of an internal combustion engine to decrease the hydraulic pressure required for the pumps, depending on the purposes or use conditions of the internal combustion engine. Whereby, further fuel saving can be achieved.

**Citation List****Patent Literature**

[0004] Patent Literature 1: Japanese Patent Application Laid-Open Publication No. 2010-31082

**Summary of Invention****Technical Problem**

35 [0005] Conventional engine oils comprise a lubricating base oil blended with a viscosity index improver, a detergent dispersant, a friction modifier and the like to fulfill their required properties. However, the engine oils has caused a problem that with the conventional blend balance, they cannot be reduced in viscosity sufficiently as engines oils for an internal combustion engine employing the above-described heat management. That is, if the necessary additives are used in their conventional amount ratios, the viscosity increase caused by the additives is significant, and thus the base oil viscosity must be significantly reduced to reduce the viscosity of the lubricating oil composition. Such significant reduction of the base oil viscosity causes an increase in engine oil consumption due to an increase in evaporation loss and also leads to failure to secure the viscosity necessary for a high shear speed region and thus increases the risk of defective lubrication.

40 [0006] The present invention has been made in view of the above-described current situations and has an object to provides a lubricating oil composition for an internal combustion engine that is excellent in functions as an engine oil for an internal combustion engine employing heat management and in particular fuel saving properties and detergency.

**Solution to Problem**

50 [0007] The present invention has been completed as the results of extensive studies to achieve the above object.

[0008] That is, the present invention relates to a lubricating oil composition for an internal combustion engine comprising (A) a base oil having 100°C kinematic viscosity of 3.0 to 5.0 mm<sup>2</sup>/s and (B) a boronated succinimide in an amount of 0.007 percent by mass or more as boron and in an amount of 5 percent by mass or less as the succinimide ashless dispersant, on the basis of the total mass of the composition, (C) a phenol-based antioxidant in an amount of 0.5 percent by mass or more and (D) a viscosity index improver having a ratio of the weight average molecular weight and PSSI of 1.2 x 10<sup>4</sup> or greater in an amount of 0.1 to 5 percent by mass, the composition having a 150°C HTHS viscosity of 2.0 to 2.8 mPa·s, a 100°C HTHS viscosity of 4.8 mPa·s or lower and a viscosity index of 180 or greater.

55 [0009] The present invention also relates to the foregoing lubricating oil composition for an internal combustion engine wherein the ratio of the boronated succinimide to the non-boronated succinimide is from 1.0 to 3.0 by weight.

**[0010]** The present invention also relates to the foregoing lubricating oil composition for an internal combustion engine wherein the ratio of the total content of the boronated succinimide, Component (B) and a non-boronated succinimide to the content of the viscosity index improver, Component (D) is 6 or less.

## Advantageous Effect of Invention

**[0011]** According to the present invention, a lubricating oil composition for an internal combustion engine is provided, which is excellent in functions as an engine oil for an internal combustion engine employing heat management and in particular fuel saving properties and detergency.

**[0012]** The lubricating oil composition of the present invention is suitably used in gasoline engines, diesel engines and gas engines for two- and four-wheeled vehicles, power generators and cogenerations and further not only those using fuel with a sulfur content of 50 ppm by mass or less but also various engines of ships and outboard motors.

## Description of Embodiments

**[0013]** The present invention will be described in detail.

**[0014]** The lubricating base oil of the lubricating oil composition of the present invention may be a mineral base oil or a synthetic base oil.

**[0015]** Examples of the mineral lubricating base oils include those having a 100°C kinematic viscosity satisfying the above-described requirement selected from: paraffinic mineral base oils which can be produced by subjecting a lubricating oil fraction produced by atmospheric- and/or vacuum-distillation of crude oil, to any one of or any suitable combination of refining processes selected from solvent deasphalting, solvent extraction, hydrocracking, solvent dewaxing, catalytic dewaxing, hydrotreating, sulfuric acid treatment, and clay treatment; n-paraffinic base oils; and iso-paraffinic base oils.

**[0016]** Examples of preferred lubricating base oils include base oils produced using the following base oils (1) to (8) as a feedstock by refining the feedstock and/or a lubricating oil fraction recovered therefrom in a given refining process and recovering a lubricating oil fraction:

- (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 dewaxing of lubricating oil (slack wax) and/or a synthetic wax produced through a gas to liquid (GTL) process (Fischer-Tropsch wax, GTL wax) ;
- (4) a mixed oil of one or more types selected from base oils (1) to (3) and/or an oil produced by mild-hydrocracking the mixed oil;
- (5) a mixed oil of two or more types selected from base oils (1) to (4) above;
- (6) a deasphalted oil (DAO) produced by deasphalting base oil (1), (2) (3), (4) or (5);
- (7) an oil produced by mild-hydrocracking (MHC) base oil (6); and
- (8) a mixed oil of two or more types selected from base oils (1) to (7) above.

**[0017]** The above-mentioned given refining process is preferably hydro-refining such as hydrocracking or hydrofinishing, solvent refining such as furfural extraction, dewaxing such as solvent dewaxing and catalytic dewaxing, clay refining with acidic clay or active clay or 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.

**[0018]** The lubricating base oil used in the present invention is particularly preferably the following base oil (9) or (10) produced by subjecting a base oil selected from the above-described base oils (1) to (8) or a lubricating oil fraction recovered therefrom to a specific treatment:

- (9) a hydrocracked base oil produced by hydrocracking a base oil selected from base oils (1) to (8) or a lubricating oil fraction recovered from the base oil, and subjecting the resulting product or a lubricating oil fraction recovered therefrom by distillation, to a dewaxing treatment such as solvent or catalytic dewaxing, optionally followed by distillation; or
- (10) a hydroisomerized base oil produced by hydroisomerizing a base oil selected from base oils (1) to (8) or a lubricating oil fraction recovered from the base oil, and subjecting the resulting product or a lubricating oil fraction recovered therefrom by distillation, to a dewaxing treatment such as solvent or catalytic dewaxing, optionally followed by distillation.

**[0019]** If necessary, a solvent refining process and/or a hydrofinishing process may be carried out at appropriate timing

upon production of lubricating base oil (9) or (10).

**[0020]** No particular limitation is imposed on the catalyst used in the above-described hydrocracking and hydroisomerizing. However, the catalyst is preferably a hydrocracking catalyst comprising any one of complex oxides having cracking activity (for example, silica-alumina, alumina boria, or silica zirconia) or one or more types of such complex oxides bound with a binder, used as a support and a metal with hydrogenation capability (for example, one or more types of metals of Groups VIa and VIII of the periodic table) supported on the support, or a hydroisomerizing catalyst comprising a support containing zeolite (for example, ZSM-5, zeolite beta, or SAPO-11) and a metal with hydrogenation capability, containing at least one or more types of metals of Group VIII of the periodic table and supported on the support. The hydrocracking and hydroisomerizing catalysts may be laminated or mixed so as to be used in combination.

**[0021]** No particular limitation is imposed on the conditions under which the hydrocracking and hydroisomerizing are carried out. Preferably, the hydrogen partial pressure is from 0.1 to 20 MPa, the average reaction temperature is from 150 to 450°C, the LHSV is from 0.1 to 3.0 hr<sup>-1</sup>, and the hydrogen/oil ratio is from 50 to 20000 scf/b.

**[0022]** The 100°C kinematic viscosity of the lubricating base oil of the present invention is necessarily 5.0 mm<sup>2</sup>/s or lower, preferably 4.5 mm<sup>2</sup>/s or lower, particularly preferably 4.2 mm<sup>2</sup>/s or lower. Whilst, the kinematic viscosity is necessarily 3.0 mm<sup>2</sup>/s or higher, preferably 3.4 mm<sup>2</sup>/s or higher, more preferably 3.7 mm<sup>2</sup>/s or higher.

**[0023]** The 100°C kinematic viscosity used herein refers to the 100°C kinematic viscosity determined in accordance with ASTM D-445.

**[0024]** If the 100°C kinematic viscosity of the lubricating base oil exceeds 5 mm<sup>2</sup>/s, the resulting composition would be degraded in low temperature viscosity characteristics and may not obtain sufficiently improved fuel saving properties. If the 100°C kinematic viscosity is lower than 3.0 mm<sup>2</sup>/s, the resulting lubricating oil composition would be poor in lubricity due to its insufficient oil film formation at lubricating sites and would be large in evaporation loss of the composition.

**[0025]** The viscosity index of the lubricating base oil of the present invention is preferably 120 or greater, more preferably 125 or greater, more preferably 130 or greater, most preferably 140 or greater. Whilst, the viscosity index is preferably 160 or less.

**[0026]** A viscosity index of less than 120 would not only cause the viscosity-temperature characteristics, thermal/oxidation stability, anti-evaporation properties to degrade but also cause the friction coefficient to increase and likely cause the friction coefficient to increase and cause the antiwear properties to degrade. A viscosity index of greater than 160 would tend to degrade the low temperature viscosity characteristics.

**[0027]** The viscosity index referred herein denotes the viscosity index measured in accordance with JIS K 2283-1993.

**[0028]** The sulfur content of the lubricating base oil used in the present invention depends on the sulfur content of the raw material thereof. For example, when a raw material containing substantially no sulfur such as a synthetic wax component produced by Fischer-Tropsch reaction is used, a lubricating base oil containing substantially no sulfur can be produced.

**[0029]** The lubricating base oil used in the present invention contains sulfur in an amount of preferably 10 mass ppm or less, more preferably 5 mass ppm or less, and particularly preferably contains no sulfur with the objective of further improving thermal/oxidation stability and lowering the sulfur content.

**[0030]** The %C<sub>P</sub> of the lubricating base oil used in the present invention is preferably 70 or greater, more preferably 80 or greater, more preferably 85 or greater, most preferably 90 or greater and preferably 95 or less.

**[0031]** If the %C<sub>P</sub> of the lubricating base oil is less than 70, the resulting composition would tend to be degraded in viscosity-temperature characteristics, thermal/oxidation stability and friction characteristics and when blended with additives, would tend to degrade the efficacy thereof. The %C<sub>P</sub> is preferably 95% or less because the %CN of the base oil is preferably on the order of 5% in view of solubility of additives.

**[0032]** The %C<sub>A</sub> of the lubricating base oil used in the present invention is preferably 2 or less, more preferably 1 or less, more preferably 0.8 or less, particularly preferably 0.5 or less, most preferably 0. If the %C<sub>A</sub> of the lubricating base oil exceeds 2, the resulting composition would tend to be degraded in viscosity-temperature characteristics, thermal/oxidation stability and fuel saving properties.

**[0033]** The %C<sub>P</sub> and %C<sub>A</sub> referred in the present invention denote the percentage of paraffin carbon number in the total carbon number and the percentage of the aromatic carbon number in the total carbon number, respectively, determined by a method (n-d-M ring analysis) in accordance with ASTM D 3238-85.

**[0034]** Examples of the synthetic base oil include poly- $\alpha$ -olefins and hydrogenated compounds thereof; isobutene oligomers and hydrogenated compounds thereof; isoparaffins; alkylbenzenes; and alkylnaphthalenes, among which poly- $\alpha$ -olefins are preferable.

**[0035]** 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, decene oligomer, oligomers or cooligomers of 1-dodecene, and hydrogenated compounds thereof.

**[0036]** These synthetic base oils may be those that are commercially available if their viscosity is within the same range as the aforesaid mineral base oils. These synthetic base oils may be each used alone or in combination with the above-described mineral base oils. No particular limitation is imposed on the mix ratio of these bases oils.

[0037] The engine oil of the present invention contains a boronated succinimide as Component (B).

[0038] In the present invention, the boronated succinimide is preferably mixed with a non-boronated succinimide. The non-boronated succinimide denotes succinimide before being boronated.

[0039] Examples of the succinimide include succinimides having at least one alkyl or alkenyl group having 40 to 400, preferably 60 to 350 carbon atoms per molecule or derivatives thereof. If the carbon number of the alkyl or alkenyl group is fewer than 40, Component (B) would tend to be degraded in solubility in the lubricating base oil. Whereas, if the carbon number of the alkyl or alkenyl group is more than 400, the resulting lubricating oil composition would be degraded in low-temperature fluidity. The alkyl or alkenyl group may be straight-chain or branched but is preferably a branched alkyl or alkenyl group derived from oligomers of olefins such as propylene, 1-butene or isobutylene or a cooligomer of ethylene and propylene. The succinimide is preferably a mono- or bis-succinimide.

[0040] No particular limitation is imposed on the method for producing the succinimide. For example, a method may be used, wherein an alkyl or alkenyl succinimide produced by reacting a compound having an alkyl or alkenyl group having 40 to 400 carbon atoms with maleic anhydride at a temperature of 100 to 200°C is reacted with a polyamine. Examples of the polyamine include diethylene triamine, triethylene tetramine, tetraethylene pentamine, and pentaethylene hexamine.

[0041] Boronation is generally carried out by allowing succinimide to react with boric acid to neutralize the whole or part of the remaining amino and/or imino groups.

[0042] Examples of a method of producing a boronated succinimide are those disclosed in Japanese Patent Publication Nos. 42-8013 and 42-8014 and Japanese Laid-Open Patent Publication Nos. 51-52381 and 51-130408. More specifically, a boronated succinimide may be produced by mixing polyamine and polybutenylsuccinic acid (anhydride) with a boron compound such as boric acid, a boric acid ester, or a borate in a solvent including alcohols, organic solvent such as hexane or xylene, or a light fraction lubricating base oil and by heating the mixture under appropriate conditions. The boron content of the boron acid-modified succinimide produced in this manner is generally from 0.1 to 45 percent by mass.

[0043] No particular limitation is imposed on the boron content of the boronated succinimide used in the present invention, which is, however, usually from 0.1 to 3 percent by mass, preferably 0.2 percent by mass or more, more preferably 0.3 percent by mass or more, more preferably 0.5 percent by mass or more. The boron content is preferably 2 percent by mass or less, more preferably 1.5 percent by mass or less, more preferably 1 percent by mass or less.

[0044] The boronated succinimide is preferably a boronated succinimide, particularly desirously a boron-containing bis-type succinimide, with a boron content within the above-described range. If the boron content is more than 3 percent by mass, not only concerns about stability are arisen, but also concerns about influences on an exhaust-gas after-treatment system would be arisen accompanied with an increase in sulfated ash content due to the too much boron in the composition. If the boron content is less than 0.1 percent by mass, effects of addition of the boronated succinimide cannot be expected.

[0045] In the present invention, the boronated succinimide is preferably used in the form of a mixture with a non-boronated succinimide. The ratio of the above-described boronated succinimide and the non-boronated succinimide (boronated succinimide/non-boronated succinimide) is preferably within the range of 1.0 to 3.0, more preferably 1.2 or greater and preferably 2.6 or less, more preferably 2.0 or less, more preferably 1.5 or less by weight ratio.

[0046] If the ratio of the boronated succinimide and the non-boronated succinimide exceeds 3.0, not only concerns about stability are arisen, but also concerns about influences on an exhaust-gas after-treatment system would be arisen accompanied with an increase in sulfated ash content due to the too much boron in the composition. Whilst, the ratio is less than 1.0, effects of addition of the boronated succinimide cannot be expected.

[0047] The reason why the mixture is preferably used in combination is that the boronated succinimide alone results in an unstable boronated compound, which would often precipitate and the mixture is excellent in the balance of detergency.

[0048] The content of Component (B) of the lubricating oil composition for an internal combustion engine according to the present invention is necessarily 0.007 percent by mass or more, preferably 0.01 percent by mass or more and 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 as boron on the basis of the total mass of the lubricating oil composition for an internal combustion engine.

[0049] This is because if the content is less than 0.007 percent by mass or less, effects by boron cannot be expected and if the content exceeds 0.1 percent by mass, the composition would lack in stability.

[0050] In the present invention, the total content of the boronated succinimide and the non-boronated succinimide is 5 percent by mass or less on the basis of the total mass of the composition.

[0051] The molecular weight of Component (B) is determined by the carbon number of alkyl or alkenyl group and structure of the polyamine but is preferably 2500 or greater, more preferably 3000 or greater, more preferably 3500 or greater. Whilst, the molecular weight is preferably 10000 or less, more preferably 7000 or less, more preferably 5000 or less. If the molecular weight is less than 2500, the resulting composition would be less in detergency effect. Whilst, if the molecular weight exceeds 10000, the resulting composition would be deteriorated in low temperature viscosity.

[0052] The boronated succinimide and non-boronated succinimide are generally provided in a state where they are

dissolved in a solvent equivalent to a lubricating base oil for manufacturing reasons. The content referred in the present invention denotes the net content of the compound excluding the solvent.

**[0053]** Therefore, for example, when succinimide dissolved in a solvent is used, the effective concentration of the succinimide is calculated and then succinimide is added in such an amount to be the net amount.

**[0054]** For example, the effective concentration may be calculated in the following manner. Into a 50 ml volume sack-like rubber made from natural rubber is accurately weighed out 1 to 2 grams of an additive solution containing the succinimide. An upper portion of the rubber is tied with thread or the like so that the content does not spill out from the rubber sack. The rubber sack is then placed in a Soxhlet extractor into which a piece of filter paper has been put to extract the solvent using heptane as an extract solvent at 50°C for 24 hours. After completion of the extraction, the rubber sack containing the sample is allowed to stand at room temperature for 24 hours and measured. The sample remaining in the rubber sack is the succinimide and the effective concentration can be calculated from the weight of the sample when placed initially in the rubber sack.

**[0055]** The engine oil of the present invention contains a phenol-based antioxidant as Component (C).

**[0056]** Examples of the phenol-based ashless antioxidant include: phenol-based antioxidants containing no sulfur as a constitutional element such as 4,4'-methylenebis(2,6-di-tert-butylphenol), 4,4'-bis(2,6-di-tert-butylphenol), 4,4'-bis(2-methyl-6-tert-butylphenol), 2,2'-methylenebis(4-ethyl-6-tert-butylphenol), 2,2'-methylenebis(4-methyl-6-tert-butylphenol), 4,4'-butylidenebis(3-methyl-6-tert-butylphenol), 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-tert-butylphenol, 2,6-di-tert-butyl-4-hydroxyphenylpropionate, tridecyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate, pentaerythrityl-tetraakis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate], octadecyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate, octyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate, and octyl-3-(3-methyl-5-tert-butyl-4-hydroxyphenyl) propionate; phenol-based ashless antioxidants containing sulfur as a constitutional element such as 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, and 2,2'-thio-diethylenebis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate]; and mixtures thereof.

**[0057]** Among these, preferred examples include hydroxyphenyl-substituted ester-based antioxidants that are esters of hydroxyphenyl group-substituted fatty acids and alcohols having 4 to 12 carbon atoms such as octyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate and octyl-3-(3-methyl-5-tert-butyl-4-hydroxyphenyl) propionate and bisphenol-based ashless antioxidants. More preferred examples include hydroxyphenyl-substituted ester-based antioxidants. Phenolic compounds with a molecular weight of 240 or greater are preferable because they are high in decomposition temperature and thus can exhibit their effects under higher temperature conditions.

**[0058]** The engine oil may further contain an amine-based ashless antioxidant. Examples of the amine-based ashless antioxidant include phenyl- $\alpha$ -naphthylamine, alkylphenyl- $\alpha$ -naphthylamines and dialkyldiphenylamines.

**[0059]** The engine oil of the present invention contains a viscosity index improver having a weight-average molecular weight and PSSI ratio of  $1.2 \times 10^4$  or greater as Component (D).

**[0060]** The weight-average molecular weight (MW) of the viscosity index improver used in the present invention is preferably 600,000 or less, more preferably 500,000 or less, more preferably 460,000 or less. The Mw is preferably 10,000 or greater, more preferably 50,000 or greater, more preferably 100,000 or greater, particularly preferably 200,000 or greater.

**[0061]** If the viscosity index improver has a weight average molecular weight of less than 10,000, it would be less effective in viscosity index enhancement when it is dissolved in a lubricating base oil and the resulting composition would not only be poor in fuel saving properties and low temperature viscosity characteristics but also be high in production cost. If the viscosity index improver has a weight-average molecular weight of greater than 600,000, it would exert the viscosity increasing effect too much and thus the resulting composition would not only be poor in fuel saving properties and low temperature viscosity characteristics but also be degraded in shear stability, solubility in a lubricating base oil and storage stability.

**[0062]** The viscosity index improver used in the present invention has a PSSI (permanent shear stability index) of necessarily 20 or less, more preferably 17 or less, more preferably 16 or less, particularly preferably 15 or less. If the PSSI exceeds 20, the resulting composition would be degraded in shear stability and thus needed to be enhanced in initial kinematic viscosity, possibly resulting in degraded fuel saving properties. If the PSSI is less than 1, the viscosity index improver would be less effective in viscosity index enhancement when it is dissolved in a lubricating base oil and thus the resulting composition would not only be poor fuel saving properties and low temperature viscosity characteristics but also increased in production cost. The PSSI is, therefore, preferably 1 or greater.

**[0063]** The ratio of the weight-average molecular weight and PSSI (MW/PSSI) of the viscosity index improver used in the present invention is necessarily  $1.2 \times 10^4$  or greater, preferably  $1.5 \times 10^4$  or greater, more preferably  $2.0 \times 10^4$  or greater. If the MW/PSSI is less than  $1.2 \times 10^4$ , the resulting composition would be degraded in fuel saving properties

and low temperature startability, i.e., viscosity temperature characteristics and low temperature viscosity characteristics.

**[0064]** Specific examples of the compound include non-dispersant type or dispersant type ester group-containing viscosity index improvers, non-dispersant type or dispersant type poly(meth)acrylate viscosity index improvers, styrene-diene hydrogenated copolymers, non-dispersant type or dispersant type ethylene- $\alpha$ -olefin copolymers or hydrogenated compounds thereof, polyisobutylene and hydrogenated compounds thereof, styrene-maleic anhydride ester copolymer, polyalkylstyrenes, (meth)acrylate-olefin copolymers and mixtures thereof.

**[0065]** The content of Component (D) in the lubricating oil composition of the present invention is preferably from 0.1 to 5 percent by mass, more preferably 0.5 percent by mass or more, more preferably from 1.0 percent by mass or more on the basis of the total mass of the composition. The content of Component (D) is preferably 3 percent by mass or less, more preferably 2 percent by mass or less. If the content is less than 0.1 percent by mass, the resulting composition would be insufficient in low temperature characteristics. If the content exceeds 5 percent by mass, the resulting composition would be degraded in shear stability.

**[0066]** As with the above-described succinimide, a viscosity index improver is generally provided in a state where it is dissolved in a solvent equivalent to a lubricating base oil, the content of the viscosity index improver referred herein denote the net content excluding the solvent.

**[0067]** In the present invention, the ratio of the total content of the boronated succinimide, Component (B) and the non-boronated succinimide to the content of the viscosity index improver, Component (D) is 6 or less.

**[0068]** That is, this indicates that there is a certain limit to the ratio of the total content of the boronated succinimide, Component (B) and the non-boronated succinimide to the content of the viscosity index improver, Component (D). This is because although both Component (D) and Component (B) involve an effect on the increase of the composition viscosity, Component (B) in particular involve an effect largely on the increase of the low temperature viscosity and thus it is necessary to restrain Component (B) from contributing to the viscosity increase rate of the composition.

**[0069]** Therefore, the ratio of the total content of the boronated succinimide, Component (B) and the non-boronated succinimide to the content of the viscosity index improver, Component (D) is 6 or less, preferably 5 or less, more preferably 4 or less, more preferably 3.5 or less, most preferably 3 or less.

**[0070]** The lubricating oil composition for an internal combustion engines of the present invention may be blended with any additives that have been generally used in a lubricating oil depending on the purposes in order to further enhance the properties. Examples of such additives include metallic detergents, friction modifiers, ashless dispersants other than Component (B), antiwear agents (or extreme pressure additives), antioxidants other than Component (C), corrosion inhibitors, rust inhibitors, demulsifiers, metal deactivators, and anti-foaming agents.

**[0071]** Examples of the metallic detergent include normal salts and/or basic salts of alkali metal/alkaline earth metal sulfonates, alkali metal/alkaline earth metal phenates, and alkali metal/alkaline earth metal salicylates. Examples of the alkali metal include sodium and potassium. Examples of the alkaline earth metal include magnesium, calcium and barium. Preferred are magnesium and calcium, and particularly preferred is calcium.

**[0072]** Examples of the friction modifier include any compounds that are usually used as a friction modifier for lubricating oils, for example organic molybdenum compounds and ashless friction modifiers.

**[0073]** Examples of the organic molybdenum compound include molybdenum dithiocarbamate, molybdenum dihydrophosphate, molybdenum-amine complex, molybdenum-succinimide complex, molybdenum salts of organic acids, and molybdenum salts of alcohols.

**[0074]** Examples of the ashless friction modifier include ashless friction modifiers such as amine compounds, fatty acid esters, fatty acid amides, fatty acids, aliphatic alcohols, and aliphatic ethers, each having at least one alkyl or alkenyl group having 6 to 30 carbon atoms, in particular straight-chain alkyl or alkenyl group having 6 to 30 carbon atoms per molecule. Alternative examples include various ashless friction modifiers as exemplified in International Publication No. 2005/037967 Pamphlet.

**[0075]** In the present invention, most preferred is molybdenum dithiocarbamate because it can mostly reduce friction.

**[0076]** The antiwear agent (or extreme pressure additive) may be any antiwear agents or extreme pressure additives that are used for lubricating oil. For example, sulfuric-, phosphoric- and sulfuric-phosphoric extreme pressure additives may be used. Specific examples include zinc dialkyldithiophosphate (ZnDTP), phosphorus acid esters, thiophosphorus acid esters, dithiophosphorus acid esters, trithiophosphorus acid esters, phosphoric acid esters, thiophosphoric acid esters, dithiophosphoric acid esters, trithiophosphoric acid esters, amine salts, metal salts or derivatives thereof, dithiocarbamates, zinc dithiocarbamates, disulfides, polysulfides, sulfurized olefins and sulfurized fats and oils. Among these antiwear agents, preferred are sulfuric extreme pressure additives, and particularly preferred are zinc dialkyldithiophosphate.

**[0077]** Examples of the corrosion inhibitor include benzotriazole-, tolyltriazole-, thiadiazole-, and imidazole-types compounds.

**[0078]** Examples of the rust inhibitor include petroleum sulfonates, alkylbenzene sulfonates, dinonylnaphthalene sulfonates, alkenyl succinic acid esters, and polyhydric alcohol esters.

**[0079]** Examples of the demulsifier include polyalkylene glycol-based non-ionic surfactants such as polyoxyethylene-

alkyl ethers, polyoxyethylenealkylphenyl ethers, and polyoxyethylenealkylnaphthyl ethers.

**[0080]** Examples of the metal deactivator include imidazolines, pyrimidine derivatives, alkylthiadiazoles, mercapto-benzothiazoles, benzotriazoles and derivatives thereof, 1,3,4-thiadiazolepolysulfide, 1,3,4-thiadiazolyl-2,5-bisdi-alkyldithiocarbamate, 2-(alkyldithio)benzimidazole, and  $\beta$ -(o-carboxybenzylthio)propionitrile.

**[0081]** Examples of the anti-foaming agent include silicone oil with a 25°C kinematic viscosity of 1000 to 100,000 mm<sup>2</sup>/s, alkenylsuccinic acid derivatives, esters of polyhydroxy aliphatic alcohols and long-chain fatty acids, aromatic amine salts of methylsalicylate and o-hydroxybenzyl alcohol.

**[0082]** When these additives are contained in the lubricating oil composition for an internal combustion engine of the present invention, they are contained in an amount of 0.01 to 10 percent by mass on the total composition mass basis.

**[0083]** The 150°C HTHS viscosity of the lubricating oil composition for an internal combustion engine of the present invention is 2.8 mPa·s or lower, preferably 2.6 mPa·s or lower, more preferably 2.4 mPa·s or lower and 2.0 mPa·s or higher, preferably 2.1 mPa·s or higher, more preferably 2.2 mPa·s or higher.

**[0084]** If the 150°C HTHS viscosity exceeds 2.8 mPa·s, the composition may not obtain sufficient fuel saving properties. If it is lower than 2.0 mPa·s, the composition would lack lubricity.

**[0085]** The 150°C HTHS viscosity referred herein denotes the high temperature high shear viscosity at 100°C defined in accordance with ASTM D4683.

**[0086]** The 100°C HTHS viscosity of lubricating oil composition for an internal combustion engine of the present invention is 4.8 mPa·s or lower, preferably 4.7 mPa·s or lower, more preferably 4.6 mPa·s or lower, particularly preferably 4.5 mPa·s or lower.

**[0087]** If the 100°C HTHS viscosity exceeds 4.8 mPa·s, the resulting composition would not obtain sufficient fuel saving properties. If the 100°C HTHS viscosity is lower than 3.9 mPa·s, it could cause the engine hydraulic pressure to be insufficient and thus is preferably 3.9 mPa·s or higher.

**[0088]** The 100°C HTHS viscosity referred herein denotes the high temperature high shear viscosity at 100°C defined in accordance with ASTM D6616.

**[0089]** The HTHS viscosity (150°C)/HTHS viscosity (100°C) is preferably 0.45 or higher, more preferably 0.047 or higher, more preferably 0.049 or higher, most preferably 0.51 or higher. This is because if the HTHS viscosity (100°C) is lower than the HTHS viscosity (150°C), the composition would be excellent in fuel saving properties.

**[0090]** The 100°C kinematic viscosity of the lubricating oil composition for an internal combustion engine of the present invention is preferably 8 mm<sup>2</sup>/s or lower, more preferably 7.5 mm<sup>2</sup>/s or lower, more preferably 7 mm<sup>2</sup>/s or lower, most preferably 6.8 mm<sup>2</sup>/s or lower. The 100°C kinematic viscosity of the lubricating oil composition for an internal combustion engine of the present invention is preferably 4 mm<sup>2</sup>/s or higher, more preferably 5 mm<sup>2</sup>/s or higher, more preferably 6 mm<sup>2</sup>/s or higher, most preferably 6.3 mm<sup>2</sup>/s or higher. The 100°C kinematic viscosity used herein refers to the 100°C kinematic viscosity determined in accordance with ASTM D-445. If the 100°C kinematic viscosity is lower than 4 mm<sup>2</sup>/s, the resulting lubricating oil composition would lack lubricity. If the 100°C kinematic viscosity exceeds 8 mm<sup>2</sup>/s, the resulting composition would not obtain the required low temperature viscosity and sufficient fuel saving properties.

**[0091]** The viscosity index of the lubricating oil composition for an internal combustion engine of the present invention is 180 or greater, more preferably 190 or greater, more preferably 200 or greater, particularly preferably 210 or greater, most preferably 220 or greater. If the lubricating oil composition of the present invention has a viscosity index of less than 180, it would be difficult to improve the fuel saving properties while maintaining the 150°C HTHS viscosity and to reduce the low temperature viscosity at -35°C. If the viscosity index of the lubricating oil composition of the present invention is greater than 300, the resulting composition would be degraded in evaporability and cause malfunctions caused by the lack of solubility of additives and the incompatibility with seal materials and the viscosity index is, therefore, preferably 300 or less.

#### Examples

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

**[0093]** (Examples 1 to 5, Comparative Examples 1 to 5)

**[0094]** Lubricating oil compositions of the present invention (Example 1 to 5) and those for comparison (Comparative Examples 1 to 5) were each prepared to carry out a hot tube test. The results are set forth in Table 1 below.

**[0095]** The test was carried out in accordance with JPI 5S-55-99 under conditions where the amount of samples was 10 g, the test temperature was 300°C, and the test time was 16 hours.



[Table 1]

Base Oil	Base oil total mass basis	Example 1	Example 2	Example 3	Example 4	Example 5	Comparative Example 1	Comparative Example 2	Comparative Example 3	Comparative Example 4	Comparative Example 5
O-1	base oil 1 mass%	100	90			90	100	90	90	90	90
O-2	base oil 2 mass%		10			10		10	10	10	10
O-3	base oil 3 mass%			65							
O-4	base oil 4 mass%			35							
O-5	base oil 5 mass%				100						
	base oil viscosity (40°C) mm <sup>2</sup> /s	15.6	16.8	15.6	17.1	16.8	15.6	16.8	16.8	16.8	16.8
	base oil viscosity (100°C)	3.8	4.0	3.7	3.9	4.0	3.8	4.0	4.0	4.0	4.0
	base oil viscosity index	142	141	126	126	141	142	141	141	141	141
Additives	Composition total mass basis										
A-1	mass%	1.8	1.8	1.8	1.8	0.9	3.6	3.6	3.6	1.8	1.8
A-2	mass%	2.3	2.3	2.3	2.3	2.3	2.3	2.3	0.0	1.1	2.3
A-3	boron content mass%	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.000	0.005	0.011
A-4	mass%	0.8	0.8	0.8	0.8	0.8	0.3	0.3	0.8	0.8	0.3
B-1	mass%	1.4	1.3	1.3	1.5	1.5	0.8	0.7	0.7	1.4	1.3
	mass%	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4
C-1	other additives mass%	5	5	5	5	5	5	5	5	5	5
boronated (A-2)+non-boronated (A-1)	mass%	4.1	4.1	4.1	4.1	3.2	5.9	5.9	3.6	2.9	4.1

(continued)

Base Oil	Base oil total mass basis	Example 1	Example 2	Example 3	Example 4	Example 5	Comparative Example 1	Comparative Example 2	Comparative Example 3	Comparative Example 4	Comparative Example 5
boronated (A-2)/non-boronated (A-1)		1.3	1.3	1.3	1.3	2.5	0.6	0.6	0.0	0.6	1.3
succinimide amount (A-1+A-2)/VM amount (A-4)		3.0	3.3	3.3	2.7	2.2	7.8	8.7	5.3	2.1	3.3
Evaluation results											
kinematic viscosity	40°C mm <sup>2</sup> /s	26.0	27.4	28.0	28.8	25.1	28.0	29.1	27.6	25.5	25.9
	100°C mm <sup>2</sup> /s	6.6	6.6	6.7	6.7	6.5	6.5	6.5	6.5	6.5	6.6
viscosity index		231	211	209	202	229	196	190	205	225	210
HTHS viscosity	100°C mPa·s	4.5	4.6	4.6	4.6	4.5	5.0	5.2	4.7	4.5	4.6
HTHS viscosity	150°C mPa·s	2.3	2.3	2.3	2.3	2.3	2.3	2.3	2.3	2.3	2.3
HTHS viscosity (150°C)/HTHS viscosity (100°C)		0.51	0.50	0.50	0.50	0.51	0.46	0.44	0.49	0.51	0.50
HTT (test temperature) 300°C	grade	5	5	5	5	4	5	5	3	2	3
O-1 40°C: 15.6 mm <sup>2</sup> /s, 100°C: 3.8mm <sup>2</sup> /s, VI: 142, sulfur content: <10 ppm, %CP: 92.5, %CN: 7.5, %CA: 0 O-2 40°C: 36.1 mm <sup>2</sup> /s, 100°C: 6.4mm <sup>2</sup> /s, VI: 131, sulfur content: <10 ppm, %CP: 80.6, %CN: 19.4, %CA: 0 O-3 40°C: 19.6 mm <sup>2</sup> /s, 100°C: 4.2mm <sup>2</sup> /s, VI: 122, sulfur content: <10ppm, %CP: 80.7, %CN: 19.3, %CA: 0 O-4 40°C: 13.5 mm <sup>2</sup> /s, 100°C: 3.2 mm <sup>2</sup> /s, VI: 112, sulfur content: <10ppm, %CP: 72.6, %CN: 27.4, %CA: 0 O-5 40°C: 17.3 mm <sup>2</sup> /s, 100°C: 3.9 mm <sup>2</sup> /s, VI: 126, sulfur content <10 ppm, %CP: 91.4, %CN: 8.6, %CA: 0 A-1 polybutenyl succinimide, average molecular weight: 9000 A-2 boron-modified polybutenyl succinimide, average molecular weight: 4000 B: 0.5 mass% A-3 phenol-based antioxidant (hindered phenol) A-4 non-dispersant type polymethacrylate, weight average molecular weight: 450,000, PSSI: 5 B-1 MoDTC C-1 ZnDTP: 1 mass%, amine-based anti-oxidant: 1.3 mass%, Ca salicylate: 2.5 mass%, dimethylsilicone: 0.002 mass%											

**[0096]** As set forth in Table 1, the lubricating oil compositions of Examples 1 to 5 containing all Components (A) to (D) are higher in 100°C HTHS viscosity and thus better in fuel saving properties than the lubricating oil compositions of Comparative Examples 1 and 2 not containing Component (B). The compositions of the present invention are higher in grade of HTT test and thus excellent in detergency comparing with those of Comparative Example 3 to 5 not containing Components (B) or (C).

## Claims

1. A lubricating oil composition for an internal combustion engine comprising (A) a base oil having 100°C kinematic viscosity of 3.0 to 5.0 mm<sup>2</sup>/s and (B) a boronated succinimide in an amount of 0.007 percent by mass or more as boron and in an amount of 5 percent by mass or less as the succinimide ashless dispersant, on the basis of the total mass of the composition, (C) a phenol-based antioxidant in an amount of 0.5 percent by mass or more and (D) a viscosity index improver having a ratio of the weight average molecular weight and PSSI of  $1.2 \times 10^4$  or greater in an amount of 0.1 to 5 percent by mass, the composition having a 150°C HTHS viscosity of 2.0 to 2.8 mPa·s, a 100°C HTHS viscosity of 4.8 mPa·s or lower and a viscosity index of 180 or greater.
2. The lubricating oil composition for an internal combustion engine according to claim 1 wherein the ratio of the boronated succinimide to a non-boronated succinimide is from 1.0 to 3.0 by weight.
3. The lubricating oil composition for an internal combustion engine according to claim 1 or 2 wherein the ratio of the total content of the boronated succinimide, Component (B) and the non-boronated succinimide to the content of the viscosity index improver, Component (D) is 6 or less.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2013/068093

## A. CLASSIFICATION OF SUBJECT MATTER

C10M169/04(2006.01)i, C10M129/10(2006.01)i, C10M133/56(2006.01)i,  
C10M139/00(2006.01)i, C10N20/02(2006.01)n, C10N20/04(2006.01)n, C10N30/04  
(2006.01)n, C10N30/06(2006.01)n, C10N40/25(2006.01)n

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

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C10M169/04, C10M129/10, C10M133/56, C10M139/00, C10N20/02, C10N20/04,  
C10N30/04, C10N30/06, C10N40/25

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

Jitsuyo Shinan Koho 1922-1996 Jitsuyo Shinan Toroku Koho 1996-2013  
Kokai Jitsuyo Shinan Koho 1971-2013 Toroku Jitsuyo Shinan Koho 1994-2013

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.
Y	JP 2009-167278 A (Nippon Oil Corp.), 30 July 2009 (30.07.2009), paragraphs [0003] to [0115]; examples & US 2011/0003725 A1 & EP 2241611 A1 & WO 2009/072524 A1 & CN 101883840 A	1-3
Y	JP 2008-239762 A (Nippon Oil Corp.), 09 October 2008 (09.10.2008), paragraphs [0119] to [0121] & WO 2008/123227 A1 & CN 101646758 A	1-3
Y	JP 2009-127029 A (Cosmo Oil Lubricants Co., Ltd.), 11 June 2009 (11.06.2009), paragraph [0039] (Family: none)	1-3

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

\* Special categories of cited documents:

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Date of the actual completion of the international search  
02 September, 2013 (02.09.13)

Date of mailing of the international search report  
10 September, 2013 (10.09.13)

Name and mailing address of the ISA/  
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## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2013/068093

5	C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
	Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
10	A	JP 2011-140573 A (JX Nippon Oil & Energy Corp.), 21 July 2011 (21.07.2011), paragraphs [0004] to [0116] & US 2012/0283159 A1 & EP 2522709 A1 & WO 2011/083601 A1 & CN 102712869 A	1-3
15	A	JP 2009-235258 A (Nippon Oil Corp.), 15 October 2009 (15.10.2009), paragraphs [0006] to [0093] & WO 2009/118984 A1	1-3
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**REFERENCES CITED IN THE DESCRIPTION**

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- JP 51052381 A [0042]
- JP 51130408 A [0042]
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