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(54) **LUBRICATING OIL COMPOSITION FOR INTERNAL-COMBUSTION ENGINE, AND METHOD FOR REDUCING FRICTION IN GASOLINE ENGINE**

(57) Provided is a lubricating oil composition for internal combustion engines capable of exhibit a sufficient friction-reducing effect from a low-temperature range assuming engine starting to a practical temperature range of 80°C or higher. The lubricating oil composition for in-

ternal combustion engines contains a surfactant having an alkylene oxide as the constituent unit and having an HLB value of 7 or more and less than 15, and a lubricant base oil.

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Description

Technical Field

5 **[0001]** The present invention relates to a lubricating oil composition for an internal combustion engine.

Background Art

10 **[0002]** Recently, for reducing energy loss and carbon dioxide emission in driving automobiles, development of fuel-saving performance of automobiles is under investigation. As a means for improving fuel-saving performance of automobiles, car body weight reduction is now under way, but lubricating oil is also desired to contribute toward fuel-saving performance. Consequently, improvement of further friction reduction by lubricating oil is being investigated.

[0003] Methods for friction reduction by lubricating oil have been proposed in PTLs 1 to 5.

[0004]

15 PTL 1 proposes use of a molybdenum compound such as MoDTC that is a typical one as a method for friction reduction, as a friction reducer.

PTLs 2 and 3 propose use of a boron-containing compound prepared by heating and stirring an organic compound having a hydroxyl group and an amino group, and a boric acid or a boric acid derivative in a high-temperature environment, as a friction reducer.

20 PTL 4 proposes a lubricating oil composition prepared by blending a nonionic surfactant having an HLB value of 15 or more, into a lubricant base oil.

PTL 5 proposes a lubricating oil composition for use in a friction-type driving force transmitting device, containing a specific amine compound.

Citation List

Patent Literature

30 **[0005]**

PTL 1: JP 2015-010177 A

PTL 2: JP 2014-118558 A

PTL 3: WO2014/098161

35 PTL 4: JP 2002-294270 A

PTL 5: WO2011/062282

Disclosure of Invention

Technical Problem

40 **[0006]** Heretofore, regarding the fuel-saving performance of engine oil, the fuel economy performance mainly in a temperature range of 80 to 100°C or so is generally targeted, assuming after the end of warming-up operation of engines. Recently, however, fuel-saving performance in a low-temperature range of 25 to 60°C or so has become required, assuming at a time of engine starting.

45 **[0007]** However, mere addition of a molybdenum-based friction reducer as in PTL 1 could not realize a friction-reducing effect in a low-temperature range at a time of engine starting or the like, and therefore fuel-saving performance could not be sufficiently improved.

50 **[0008]** The friction reducer and the lubricating oil composition proposed in PTLs 2 to 5 are silent in friction reduction in a low-temperature range.

[0009] On the other hand, as a method of realizing a friction-reducing effect in a low temperature range, heretofore, an ash-free friction reducer such as a glycerin monooleate or the like has been used. However, such an ash-free friction reducer could not exhibit a friction-reducing effect in a practical temperature range of 80°C or higher.

55 **[0010]** At present, a lubricating oil composition for internal combustion engines is required to have a high friction-reducing effect from a low-temperature range at engine starting to a practical temperature range of 80°C or higher, and to have a low-ash content. Merely combining a molybdenum-based friction reducer and an ash-free friction reducer could hardly satisfy the requirements.

[0011] Given the situation, a lubricating oil composition using an ash-free friction reducer that can realize a sufficient

friction-reducing effect from a low-temperature range at engine starting to even a practical temperature range of 80°C or higher is desired.

[0012] An object of the present invention is to provide a lubricating oil composition for internal combustion engines that can realize a sufficient friction-reducing effect not only in a low-temperature range assuming engine starting but also in a practical temperature range of 80°C or higher.

Solution to Problem

[0013] For solving the above-mentioned problems, an aspect of the present invention provides a lubricating oil composition for internal combustion engines, which contains a surfactant having an alkylene oxide as the constituent unit and having an HLB value of 7 or more and less than 15, and a lubricant base oil.

Advantageous Effects of Invention

[0014] The lubricating oil composition for internal combustion engines of the present invention can better a friction-reducing effect from a low-temperature range assuming engine starting to a practical temperature range of 80°C or higher, and eventually can better fuel-saving performance.

Description of Embodiments

[0015] Embodiments of the present invention are described below.

[Lubricating Oil Composition for Internal Combustion Engine]

[0016] The lubricating oil composition for internal combustion engines of this embodiment contains a surfactant having an alkylene oxide as the constituent unit and having an HLB value of 7 or more and less than 15, and a lubricant base oil.

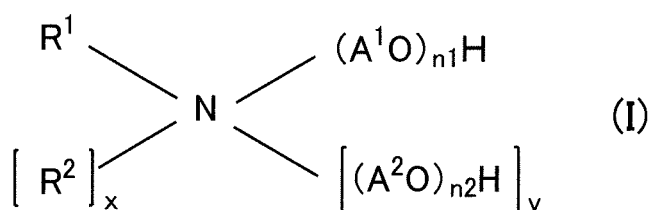
<Surfactant>

[0017] The lubricating oil composition for internal combustion engines of this embodiment contains a surfactant having an alkylene oxide as the constituent unit and having an HLB value of 7 or more and less than 15.

[0018] A surfactant having an HLB value falling within the above range but not having an alkylene oxide as the constituent unit tends to result in insufficient friction reduction. A surfactant having an alkylene oxide as the constituent unit but having an HLB value of less than 7 poorly adsorbs to a metal surface and is therefore poor in friction reduction. A surfactant having an alkylene oxide as the constituent unit but having an HLB value or more than 15 poorly dissolves in a lubricant base oil and is therefore extremely difficult to use.

[0019] As the above surfactant, various kinds of surfactants can be used, but from the viewpoint of friction reduction owing to adsorption to metal surfaces, an amine compound, an amide compound and the like in which an alkylene oxide bonds to the nitrogen atom are preferred. Among these, an amine compound is preferred, and among amine compounds, a tertiary amine is preferred.

[0020] The surfactant of a tertiary amine compound includes compounds represented by the following general formula (I). The compounds represented by the general formula (I) are favorable in the point that the compounds have the above-mentioned effect and the ash content therein is 0% by mass:



wherein R^1 and R^2 each independently represent an alkyl group having 4 to 18 carbon atoms, or an alkenyl group having 4 to 18 carbon atoms, wherein x represents 0 or 1, when $x = 0$, y is 1, and when $x = 1$, y is 0, wherein A^1O and A^2O each independently represent an oxyalkylene group having 2 to 4 carbon atoms, and wherein n_1 and n_2 each indicate an average addition molar number of the oxyalkylene group, and each independently represent an integer of 1 to 13, and $n_1 + n_2$ is 5 to 14.

[0021] The alkyl group and the alkenyl group for R^1 and R^2 may be linear, branched or cyclic, but is preferably linear.

R¹ and R² each are preferably an alkenyl group. When x = 0, the alkyl group and the alkenyl group for R¹ each preferably have 12 to 18 carbon atoms. When x = 1, the alkyl group and the alkenyl group for R¹ and R² each preferably have 4 to 16 carbon atoms.

[0022] The oxyalkylene group for A¹O and A²O preferably has 2 to 3 carbon atoms, more preferably 2 carbon atoms.

[0023] Preferably, n₁ and n₂ each are independently an integer of 2 to 10, more preferably 3 to 7. n₁+n₂ is preferably 8 to 12, more preferably 9 to 11.

[0024] Regarding (A¹O)_{n1} and (A²O)_{n2}, oxyalkylene groups having a different number of carbon atoms may bond to each other randomly or in blocks. For example, (A¹O)_{n1} and (A²O)_{n2} may be those of ethylene oxide (EO) groups and propylene oxide (PO) groups bonding to each other randomly or in blocks.

[0025] In the case where a tertiary amine of the general formula (I) is used as the surfactant, those of the same kind or those of different kinds as combined may be used. The same kind means that R¹, R² and others in the above general formula (I) are all the same. Different kinds mean that one or more of R¹, R² and others in the above general formula (I) differs from each other.

[0026] In the case where different kinds are mixed, it is desirable that they contain large quantities of preferred embodiments. For example, the proportion of the mass of the tertiary amine of the general formula (I) where R¹ and R² each an alkenyl group to the total mass of the tertiary amine [mass of the tertiary amine of the general formula (I) where R¹ and R² each are an alkenyl group/total mass of the tertiary amine of the general formula (I)] is preferably 60% by mass or more, more preferably 70% by mass or more, even more preferably 80% by mass or more.

[0027] From the viewpoint of stability of the effect, tertiary amines of the general formula (I) where R¹ all has the same number of carbon atoms are preferably used. In the case where the tertiary amine of the general formula (I) contains R², those where additionally R² all has the same number of carbon atoms are preferably used.

[0028] As the surfactant, a polyoxyalkylene fatty acid ester is also preferred.

[0029] The number of carbon atoms constituting the oxyalkylene group of the polyoxyalkylene fatty acid ester is preferably 2 to 4, more preferably 2 to 3, even more preferably 2. Regarding the bonding mode of the oxyalkylene group, oxyalkylene groups differ in point of the number of constituent carbon atoms may bond to each other randomly or in blocks. Preferably, the average addition molar number of the oxyalkylene groups is an integer of 2 to 10, more preferably 3 to 7.

[0030] The number of carbon atoms of constituting the constituent unit derived from the fatty acid of the polyoxyalkylene fatty acid ester is preferably 8 to 28, more preferably 14 to 22, even more preferably 16 to 20.

[0031] The polyoxyalkylene fatty acid ester of the type includes a polyoxyethylene oleate, a polyoxyethylene stearate, etc.

[0032] The molecular weight of the above-mentioned surfactant is, from the viewpoint of reducing friction and making friction reduction consistent with detergency, preferably in a range of 350 to 950 g/mol, more preferably in a range of 440 to 940 g/mol.

[0033] In this embodiment, the molecular weight of the surfactant is one measured on a mass spectrum according to liquid chromatography mass spectrometry (LC/MS). Specifically, a range in which a peak of mass/charge ratio (m/z) of the surfactant appears is considered as a range of the molecular weight (g/mol) of the surfactant.

[0034] Also preferably, the surfactant contains 0% by mass of ash.

[0035] In this embodiment, the surfactant is contained in the lubricating oil composition for internal combustion engines preferably in an amount of 0.01 to 2.0% by mass, more preferably 0.1 to 1.5% by mass, even more preferably 0.2 to 1.0% by mass.

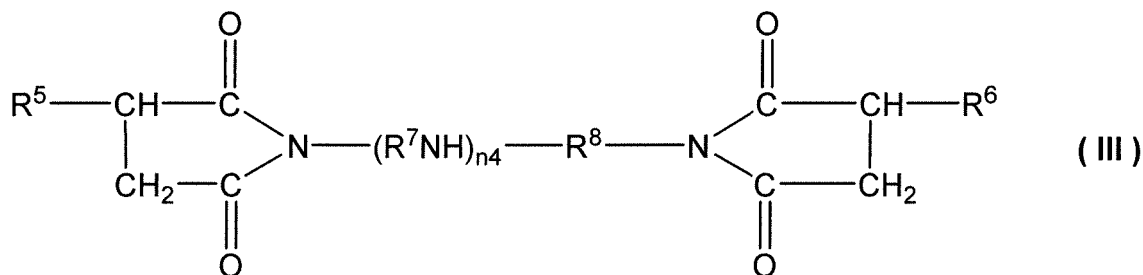
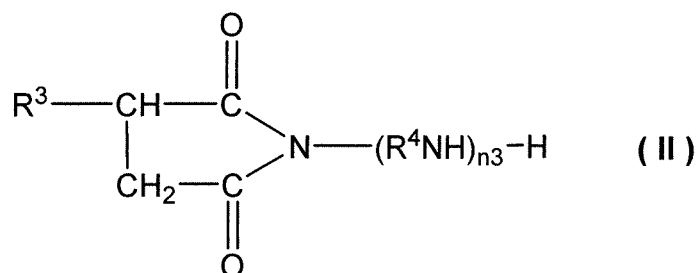
[0036] When the content of the surfactant is 0.01% by mass or more, friction can be reduced from a low-temperature range assuming engine starting to a practical temperature range of 80°C or higher. When the content of the surfactant is 2.0% by mass or less, reduction in detergency can be readily prevented from lowering while maintaining friction reduction.

<Boron-Modified Succinimide>

[0037] Preferably, the lubricating oil composition for internal combustion engines of this embodiment further contains a boron-modified succinimide.

[0038] Containing a boron-modified succinimide along with the above-mentioned surfactant, the composition can reduce friction more efficiently from a low-temperature range assuming engine starting to a practical temperature range of 80°C or higher and can make a detergency of the composition good.

[0039] The boron-modified succinimide includes those prepared by boronating an alkenyl or alkylsuccinic acid monoimide or an alkenyl or alkylsuccinic acid bisimide. The alkenyl or alkylsuccinic acid monoimide includes compounds represented by the following general formula (II). The alkenyl or alkylsuccinic acid bisimide includes compounds represented by the following general formula (III).



[0040] In the formulae (II) and (III), R^3 , R^5 and R^6 each represent an alkenyl group or an alkyl group, and the weight-average molecular weight thereof is preferably 500 to 3,000, more preferably 1,000 to 3,000.

[0041] When the weight-average molecular weight of R^3 , R^5 and R^6 is 500 or more, the solubility of the compound in a lubricant base oil is good. When 3,000 or less, the compound is expected to adequately exhibit the effect thereof. R^5 and R^6 may be the same or different.

[0042] R^4 , R^7 and R^8 each represent an alkylene group having 2 to 5 carbon atoms, and R^7 and R^8 may be the same or different. n_3 represents an integer of 1 to 10, n_4 represents 0 or an integer of 1 to 10. Here, n_3 is preferably 2 to 5, more preferably 2 to 4. When n_3 is 2 or more, the compound is expected to exhibit more easily the effect of the boron-modified succinimide. When n_3 is 5 or less, the solubility of the compound in a lubricant base oil is bettered more.

[0043] In the general formula (III), n_4 is preferably 1 to 6, more preferably 2 to 6. When n_4 is 1 or more, the compound is expected to adequately exhibit the effect of thereof. When n_4 is 6 or less, the solubility of the compound in a lubricant base oil is bettered more.

[0044] The alkenyl group includes a polybutenyl group, a polyisobutenyl group, an ethylene-propylene copolymer. The alkyl group includes those prepared by hydrogenating these. A polybutenyl group or a polyisobutenyl group is a preferred alkenyl group. The polybutenyl group is preferably one prepared through polymerization of a mixture of 1-butene and isobutene, or polymerization of high-purity isobutene. Specific examples of a preferred alkyl group include those prepared by hydrogenating a polybutenyl group or a polyisobutenyl group.

[0045] The boron-modified succinimide may be obtained, for example, by reacting a polyolefin and a maleic anhydride to give an alkenylsuccinic anhydride (A), separately reacting a polyamine and a boron compound to give an intermediate (B), and reacting the alkenylsuccinic anhydride (A) and the intermediate (B) for imidation. The monoimide or the bisimide may be produced by varying the ratio of the alkenylsuccinic anhydride or the alkylsuccinic anhydride to the polyamine.

[0046] The boron-modified succinimide may also be produced by treating an alkenyl or alkylsuccinic acid monoimide or an alkenyl or alkylsuccinic acid bisimide not containing boron, with a boron compound.

[0047] As the olefin monomer to form the above-mentioned polyolefin, one alone or two or more kinds of α -olefins having 2 to 8 carbon atoms may be used either singly or as combined. A mixture of isobutene and 1-butene is preferably used.

[0048] On the other hand, the polyamine includes simple diamines such as ethylenediamine, propylenediamine, butylenediamine, pentylenediamine, etc.; polyalkylenepolyamines such as diethylenetriamine, triethylenetetramine, tetraethylenepentamine, pentaethylenhexamine, di(methylethylene)triamine, dibutylenetriamine, tributylenetetramine, pentapentylenhexamine, etc.; piperazine derivatives such as aminoethylpiperazine, etc.

[0049] The boron compound includes boric acid, boric acid salts, boric acid esters, etc.

[0050] The boric acid includes orthoboric acid, metaboric acid, paraboric acid, etc. The boric acid salt includes ammonium borates such as ammonium metaborate, ammonium tetraborate, ammonium pentaborate, ammonium octaborate, etc. The boric acid ester includes monomethyl borate, dimethyl borate, trimethyl borate, monoethyl borate, diethyl borate, triethyl borate, monopropyl borate, dipropyl borate, tripropyl borate, monobutyl borate, dibutyl borate, tributyl borate, etc.

[0051] The ratio of the boron atom amount to the nitrogen atom amount contained in the boron-modified succinimide (B/N ratio) is, from the viewpoint of friction reduction, preferably 0.6 or more on a mass standard, more preferably 0.7

or more, even more preferably 0.8 or more. The B/N ratio is, though not specifically limited thereto, preferably 2.0 or less, more preferably 1.5 or less, even more preferably 1.3 or less.

[0052] Preferably, from the viewpoint of friction reduction, the boron-modified succinimide contains a large amount of a 3-coordinated boron-modified succinimide, and specifically contains a 3-coordinated boron-modified succinimide in a ratio by mol of 0.50 or more relative to the total amount of the 3-coordinated and 4-coordinated boron-modified succinimides, more preferably in a ratio by mol of 0.60 or more, even more preferably 0.65 or more.

[0053] The ratio of the 3-coordinated boron-modified succinimide and the 4-coordinated boron-modified succinimide may be measured, for example, through ^{11}B -NMR based on $\text{BF}_3\cdot\text{OEt}_2$ standard (0 ppm). In the ^{11}B -NMR, a peak for the 3-coordinated boron-modified succinimide appears at 5 to 25 ppm, and a peak for the 4-coordinated boron-modified succinimide appears at -10 to 5 ppm, and therefore, by calculating the integrated value of each peak, the above-mentioned ratio can be calculated.

[0054] In this embodiment, the content of the boron-modified succinimide is preferably 0.1 to 15.0% by mass in the lubricating oil composition for internal combustion engines, more preferably 0.2 to 10.0% by mass, even more preferably 0.5 to 5.0% by mass, still more preferably 0.5 to 2.0% by mass. When the content of the boron-modified succinimide falls within the above range, the lubricating oil composition can reduce friction more efficiently from a low-temperature range assuming engine starting to a practical temperature range of 80°C or higher and can better detergency.

[0055] In this embodiment, the boron atom-equivalent content of the boron-modified succinimide is preferably 0.2% by mass or less in the lubricating oil composition for internal combustion engines, more preferably 0.001 to 0.05% by mass, even more preferably 0.005 to 0.03% by mass. When the boron atom-equivalent content of the boron-modified succinimide falls within the above range, the lubricating oil composition can reduce friction more efficiently from a low-temperature range assuming engine starting to a practical temperature range of 80°C or higher and can better detergency.

[0056] In this embodiment, the ratio by mass of the content of the surfactant to the content of the boron-modified succinimide (content of boron-modified succinimide/content of surfactant) is preferably 100 or less, more preferably 20 or less, even more preferably 5 or less. When the ratio by mass falls within the above range, the lubricating oil composition can reduce friction more efficiently from a low-temperature range assuming engine starting to a practical temperature range of 80°C or higher and can better detergency.

[0057] In this embodiment, the ratio by mass of the content of the surfactant to the content of the boron atom-equivalent content of the boron-modified succinimide (boron atom-equivalent content of boron-modified succinimide/content of surfactant) is preferably 1 or less, more preferably 0.2 or less, even more preferably 0.05 or less. When the ratio by mass falls within the above range, the lubricating oil composition can reduce friction more efficiently from a low-temperature range assuming engine starting to a practical temperature range of 80°C or higher and can satisfy more readily both friction reduction and detergency.

<Poly(meth) Acrylate>

[0058] Preferably, the lubricating oil composition for internal combustion engines of this embodiment further contains a poly(meth)acrylate as a viscosity index improver.

[0059] By containing a poly(meth)acrylate, the lubricating oil composition can improve fuel-saving performance more efficiently in addition to the effect of improving fuel-saving performance owing to friction reduction by the surfactant and the boron-modified succinimide therein.

[0060] The monomer to constitute the poly(meth)acrylate is an alkyl (meth)acrylate and is preferably an alkyl (meth)acrylate having a linear alkyl group with 1 to 18 carbon atoms or a branched alkyl group with 3 to 34 carbon atoms.

[0061] Preferred examples of the monomer to constitute alkyl (meth)acrylate include methyl (meth)acrylate, ethyl (meth)acrylate, propyl (meth)acrylate, butyl (meth)acrylate, pentyl (meth)acrylate, hexyl (meth)acrylate, hexyl (meth)acrylate, heptyl (meth)acrylate, octyl (meth)acrylate, nonyl (meth)acrylate, decyl (meth)acrylate, dodecyl (meth)acrylate, tetra(meth)acrylate, hexa(meth)acrylate, octadecyl (meth)acrylate, etc. Two or more kinds of these monomers may be used to give a copolymer. The alkyl group of these monomers may be linear or branched.

[0062] The alkyl (meth)acrylate having a branched alkyl group with 3 to 34 carbon atoms includes isopropyl (meth)acrylate, 2-ethylhexyl (meth)acrylate, 3,5,5-trimethylhexyl (meth)acrylate, 2-butyloctyl (meth)acrylate, 2-hexyldecyl (meth)acrylate, 2-octyldodecyl (meth)acrylate, 2-decyltetradecyl (meth)acrylate, 2-dodecylhexadecyl (meth)acrylate, 2-tetradecyloctadecyl (meth)acrylate.

[0063] The poly(meth)acrylate is preferably one having a weight-average molecular weight of 100,000 to 600,000, more preferably 15,000 to 300,000.

[0064] In this embodiment, "weight-average molecular weight" is meant to indicate a polystyrene-equivalent molecular weight measured through gel permeation chromatography (GPC).

[0065] SSI of the poly(meth)acrylate is preferably 30% or less, more preferably 1 to 28%. When the weight-average molecular weight thereof falls within the above range, the poly(meth)acrylate may have SSI of 30% or less.

[0066] Here, SSI means a shear stability index, and indicates the decomposition resistance of the poly(meth)acrylate.

Having a larger SSI value, the polymer is more unstable against shearing and is decomposed more readily.

[Math. 1]

$$SSI = \frac{Kv_0 - Kv_1}{Kv_0 - Kv_{oil}} \times 100$$

[0067] SSI is to indicate viscosity reduction by polymer-derived shearing as percentage, and is calculated according to the above-mentioned calculation formula. In the formula, Kv_0 is a value of 100°C kinematic viscosity of a mixture prepared by adding a poly(meth)acrylate to a base oil. Kv_1 is a value of 100°C kinematic viscosity of a mixture prepared by adding a poly(meth)acrylate to a base oil after the mixture has made to pass through a high-shear Bosch diesel injector for 30 cycles according to a process of ASTM D6278. Kv_{oil} is a value of 100°C kinematic viscosity of a base oil. As the base oil, Group-II base oil having a 100°C kinematic viscosity of 5.35 mm²/s and a viscosity index of 105 is used.

[0068] The content of the poly(meth)acrylate is, from the viewpoint of fuel-saving performance, preferably 1 to 15% by mass in the lubricating oil composition for internal combustion engines, more preferably 2 to 10% by mass, even more preferably 3 to 8% by mass.

<Molybdenum Compound>

[0069] From the viewpoint of friction reduction, preferably, the lubricating oil composition for internal combustion engines of this embodiment further contains a molybdenum compound.

[0070] The molybdenum compound includes MoDTC (molybdenum dialkyldithiocarbamate), MoDTP (molybdenum dialkyldithiophosphate), etc.

[0071] The content of the molybdenum compound is preferably 2.0% or less by mass in the lubricating oil composition for internal combustion engines, more preferably 0.1 to 1.0% by mass.

<Lubricant Base Oil>

[0072] The lubricant base oil includes a mineral oil and/or a synthetic oil.

[0073] The mineral oil includes a paraffin-base mineral oil, an intermediate-base mineral oil, a naphthene-base mineral oil and the like produced through ordinary purification such as solvent purification, hydrogenation purification, etc.; a wax-isomerized oil produced through isomerization of a wax such as a wax (gas-to-liquid gas) produced according to a Fischer Tropsch process or the like, a mineral oil wax, etc.

[0074] The synthetic oil includes a hydrocarbon synthetic oil, an ether synthetic oil, etc. The hydrocarbon synthetic oil includes α -olefin oligomers or hydrides thereof such as a polybutene, a polyisobutylene, a 1-octene oligomer, a 1-decene oligomer, an ethylene-propylene copolymer, etc.; alkylbenzenes, alkyl naphthalenes, etc. The ether synthetic oil includes polyoxyalkylene glycols, polyphenyl ethers, etc.

[0075] The lubricant base oil may be a single system using one kind of the above-mentioned mineral oils and synthetic oils, or may also be a mixed system, such as a mixture prepared by mixing two or more kinds of mineral oils, a mixture prepared by mixing two or more kinds of synthetic oils, or a mixture prepared by mixing one or more kinds of mineral oils and one or more kinds of synthetic oils.

[0076] In particular, as the lubricant base oil, use of one or more kinds selected from mineral oils and synthetic oils classified in Group 3 or Group 4 in base oil classification by American Petroleum Institute is preferred.

[0077] The content of the lubricant base oil is preferably 70% by mass or more and less than 100% by mass in the lubricating oil composition for internal combustion engines, more preferably 75% by mass or more and 95% by mass or less, even more preferably 80% by mass or more and 90% by mass or less.

<Additive>

[0078] The lubricating oil composition for internal combustion engines of this embodiment may contain additives such as a metallic detergent, an antioxidant, an anti-wear agent, etc.

[0079] The content of these additives is each preferably 0.01 to 5% by mass relative to the total amount of the lubricating oil composition for internal combustion engines.

<Physical Properties of Lubricating Oil Composition for Internal Combustion Engine>

[0080] The lubricating oil composition for internal combustion engines of this embodiment is, from the viewpoint of

friction reduction from a low-temperature range assuming engine starting to a practical temperature range of 80°C or higher, preferably such that the 40°C kinematic viscosity, the 100°C kinematic viscosity and the 150°C HTHS viscosity thereof each fall within the following range.

[0081] The 40°C kinematic viscosity is preferably 20 to 40 mm²/s, more preferably 30 to 35 mm²/s.

[0082] The 100°C kinematic viscosity is preferably 3.0 to 12.5 mm²/s, more preferably 4.0 to 9.3 mm²/s.

[0083] The 150°C HTHS viscosity is preferably 1.4 to 2.9 mPa·s, more preferably 1.7 to 2.9 mPa·s.

[0084] The kinematic viscosity is measured according to JIS K2283. The HTHS viscosity is measured according to ASTM D4683 using a TBS viscometer (tapered bearing simulator viscometer), at an oil temperature of 100°C, at a shear rate of 10⁶/s, at a rotation number (motor) of 3000 rpm, and with a distance (rotor-stator distance) of 3 μm.

<Use of Lubricating Oil Composition for Internal Combustion Engine>

[0085] The lubricating oil composition for internal combustion engines of this embodiment can be favorably used for various internal combustion engines for four-wheel cars, two-wheel cars, etc. Among internal combustion engines, the composition is especially favorably used for gasoline engines.

[Friction Reducing Method for Internal Combustion Engine]

[0086] The friction reducing method for internal combustion engines of this embodiment includes adding the lubricating oil composition for internal combustion engines of this embodiment mentioned above, to an internal combustion engine.

[0087] According to the friction reducing method for internal combustion engines of this embodiment, the friction-reducing effect can be bettered from a low-temperature range assuming engine starting to a practical temperature range of 80°C or higher, and eventually fuel-saving performance can be thereby bettered. In the case of a gasoline engine as an internal combustion engine, the above-mentioned effects can be especially favorably bettered.

Examples

[0088] Next, the present embodiment is described in more detail with reference Examples.

1. Preparation of lubricating oil composition for internal combustion engines

[0089] Lubricating oil compositions for internal combustion engines of Examples, Comparative Examples and Reference Example were prepared according to the compositional ratio shown in Table 1. The lubricating oil compositions for internal combustion engines of Examples, Comparative Examples and Reference Examples were all so controlled as to have an HTHS viscosity at 150°C of 2.6 mPa·s.

2. Measurement and Evaluation

[0090] The lubricating oil compositions for internal combustion engines of Examples, Comparative Examples and Reference Example were measured and evaluated as follows. The results are shown in Table 1.

2-1. Kinematic Viscosity

[0091] According to the description of the main text of the present invention, the 40°C kinematic viscosity and 100°C kinematic viscosity of each lubricating oil composition for internal combustion engines were measured.

2-2. Friction Coefficient

[0092] The friction coefficient of each lubricating oil composition for internal combustion engines was measured under the condition mentioned below.

Tester: MTM (Mini Traction Machine) tester, manufactured by PCS Instruments Corporation

Test Piece: Standard test piece

Rubbing Time: 2 hours

Oil Temperature: 80°C

Load: 10 N

Measuring Speed: 1.2 m/s

Table 1

	Example	Comparative Example				Reference Example
		1	2	3	4	
Lubricant Base Oil	balance	balance	balance	balance	balance	balance
Surfactant A	0.50	-	0.50	-	-	-
Surfactant B	-	0.50	-	-	-	-
Surfactant C	-	-	-	0.50	-	-
Surfactant D	-	-	-	-	0.50	-
Surfactant E	-	-	-	-	-	-
Boron-Modified Succinimide	-	-	1.00	-	-	-
MoDTC	-	-	-	-	-	0.70
Polymethacrylate	5.50	5.50	5.20	5.50	5.20	5.50
Metallic Detergent	2.40	2.40	2.40	2.40	2.40	2.40
Antioxidant	0.50	0.50	0.80	0.50	0.80	0.50
Package Additive	5.60	5.60	5.60	5.60	5.60	5.60
Other Additive	0.20	0.20	0.25	0.20	0.25	0.20
40°C Kinematic Viscosity (mm ² /s)	31.56	31.78	33.69	31.70	33.32	33.73
100°C Kinematic Viscosity (mm ² /s)	7.64	7.71	7.85	7.67	7.81	7.89
Friction Coefficient	0.0162	0.0196	0.0145	0.0178	0.0229	0.0270

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[0093] The materials in Table 1 are as follows.

<Lubricant Base Oil>

5 **[0094]** Mineral oil having 100°C kinematic viscosity of 4.07 mm²/s, viscosity index: 131, %C_A: -0.4, %C_N: 12.8, %C_P: 87.6

<Surfactant A>

10 **[0095]** Mixture of 69% by mass of tertiary amine of general formula (I) (x = 0, n₁+n₂ = 10, number of carbon atoms in A¹O and A²O: 2, R¹: oleyl group) and 31% by mass of tertiary amine of general formula (I) (x = 0, n₁+n₂ = 10, number of carbon atoms in A¹O and A²O: 2, R¹: stearyl group) (mass ratio was measured through liquid chromatography mass spectrometry). HLB value: 13.2. Peak appearing position in mass spectrum in liquid chromatography mass spectrometry: 440 to 940 m/z (nearly equal to molecular weight range: 440 to 940 g/mol).

15 **[0096]** The condition for liquid chromatography mass spectrometry is as follows.

<Condition for Liquid Chromatography Mass Spectrometry>

[0097]

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Detector: Photodiode array detector, evaporative light scattering detector

Column: Inertsil ODS (3.0 × 150 mm, 3 μm)

Mobile Phase:

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A) MeCN/(0.1% formic acid + 0.1% ammonium formate) = 80/20

B) THF

Mass Spectrometry: Iontrap MS manufactured by Thermo Fisher Scientific Corporation

Ion Source: Heated ESI positive, negative

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m/z Range: 150 to 1000

<Surfactant B>

35 **[0098]** Mixture of 85% by mass of tertiary amine of general formula (I) (x = 0, n₁+n₂ = 7, number of carbon atoms in A¹O and A²O: 2, R¹: oleyl group) and 15% by mass of tertiary amine of general formula (I) (x = 0, n₁+n₂ = 10, number of carbon atoms in A¹O and A²O: 2, R¹: stearyl group) (mass ratio was measured through liquid chromatography mass spectrometry). HLB value: 11.7. Peak appearing position in mass spectrum in liquid chromatography mass spectrometry: 400 to 850 m/z (nearly equal to molecular weight range: 400 to 850 g/mol).

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<Surfactant C>

[0099] Polyoxyalkylene fatty acid ester (HLB value: 11.1, number of carbon atoms of oxyalkylene group: 2, average addition molar number of oxyalkylene group: 5, number of carbon atoms of constituent unit derived from fatty acid: 18)

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<Surfactant D>

[0100] Trade name: Ethomeen O/12, manufactured by Lion Akzo Corporation, substance name: polyoxyethylene oleylamine (tertiary amine of general formula (I) where x = 0, n₁+n₂ = 2, number of carbon atoms in A¹O and A²O: 2), HLB value 6.5, weight-average molecular weight: 356

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<Surfactant E>

[0101] Trade name: INFINEUM-C 9440, manufactured by Infineum Corporation, substance name: glycerol monooleate

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<Boron-Modified Succinimide>

[0102] Boron-modified polybutenylsuccinic acid bisimide, ratio of 3-coordination to 4-coordination (integral value of peaks of 3-coordination/integral value of peaks of 4-coordination + integral value of peaks of 3-coordination): 0.67, boron

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atom amount/nitrogen atom amount: 1.1, boron content: 1.30% by mass, nitrogen content: 1.23% by mass.

<MoDTC>

5 **[0103]** MoDTC having an Mo content of 0.07% by mass

<Polymethacrylate>

10 **[0104]** Weight-average molecular weight: 230,000, SSI: 25.2%

<Metallic Detergent>

[0105] Calcium salicylate, calcium content: 7.8% by mass, overbased, base number 224 mg-KOH/g

15 <Antioxidant>

[0106] Phenol-based antioxidant, trade name: IRGANOX-L 135, manufactured by BASF Corporation

<Package Additive>

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[0107] Package additive containing ZnDTP, high-molecular bisimide, amine antioxidant

<Other Additive>

25 **[0108]** Metal deactivator, pour point depressant

[0109] From the results in Table 1, it is confirmed that the lubricating oil compositions for internal combustion engines of Examples 1 to 4 are excellent in the friction-reducing effect in a practical temperature range of 80°C or higher though using an ash-free friction reducer. In particular, it is confirmed that the lubricating oil composition for internal combustion engines of Example 3 using both a surfactant having an alkylene oxide as the constituent unit and having an HLB value of 7 or more and less than 15, and a boron-modified succinimide is extremely excellent in the above-mentioned effect.

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2-3. Detergency

35 **[0110]** The lubricating oil compositions for internal combustion engines of Examples 1 and 3, Comparative Examples 1 to 3 and Reference Example were further evaluated in point of detergency.

<Evaluation Score>

40 **[0111]** The test temperature was set at 300°C, and the other conditions followed JPI-5S-55-99. According to JPI-5S-55-99, the glass tube after the test was scored from point 0 (black) to point 10 (colorless) at intervals of 0.5, and evaluated in 21 ranks. A larger number means better detergency.

<Adhesion Amount>

45 **[0112]** In the above score test, the mass of lacquer adhering to the glass tube after the test was measured. A smaller adhesion amount means better detergency.

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Table 2

Detergency	Hot Tube Score	Example 1	Example 3	Comparative Example 1	Comparative Example 2	Comparative Example 3	Reference Example
	Hot Tube Amount of Adhered Substance (g)	0.5	8.0	0.0	0.0	8.0	0.0
		44.2	0.3	38.8	38.2	0.7	121.5

From the results in Table 2, it is confirmed that the lubricating oil composition for internal combustion engines of Example 3 using both a surfactant having an alkylene oxide as the constituent unit and having an HLB value of 7 or more and less than 15, and a boron-modified succinimide can further better detergency.

5 Industrial Applicability

10 **[0113]** Taking advantage of the characteristics thereof of reducing friction and bettering fuel-saving performance from a low-temperature range assuming engine starting to a practical temperature range of 80°C or higher, the lubricating oil composition for internal combustion engines of this embodiment can be favorably used for various internal combustion engines for four-wheel cars, two-wheel cars, etc. Among internal combustion engines, the composition is especially favorably used for gasoline engines.

15 Claims

1. A lubricating oil composition for internal combustion engines, comprising a surfactant having an alkylene oxide as the constituent unit and having an HLB value of 7 or more and less than 15, and a lubricant base oil.
2. The lubricating oil composition for internal combustion engines according to claim 1, wherein the surfactant has a molecular weight in a range of 350 to 950 g/mol.
3. The lubricating oil composition for internal combustion engines according to claim 1 or 2, wherein the surfactant is an amine compound.
- 25 4. The lubricating oil composition for internal combustion engines according to claim 3, wherein the amine compound is a tertiary amine.
5. The lubricating oil composition for internal combustion engines according to claim 1 or 2, wherein the surfactant is a polyoxyalkylene fatty acid ester.
- 30 6. The lubricating oil composition for internal combustion engines according to any one of claims 1 to 5, wherein the content of the surfactant in the lubricating oil composition is 0.01 to 2.0% by mass.
7. The lubricating oil composition for internal combustion engines according to any one of claims 1 to 6, further comprising a boron-modified succinimide.
- 35 8. The lubricating oil composition for internal combustion engines according to claim 7, wherein the boron atom-equivalent content of the boron-modified succinimide in the lubricating oil composition is 0.2% by mass or less.
- 40 9. The lubricating oil composition for internal combustion engines according to claim 7 or 8, wherein the ratio by mass of the content of the boron-modified succinimide to the content of the surfactant (content of boron-modified succinimide/content of surfactant) is 100 or less.
- 45 10. The lubricating oil composition for internal combustion engines according to any one of claims 7 to 9, wherein the ratio by mass of the boron atom-equivalent content of the boron-modified succinimide to the content of the surfactant (boron atom-equivalent content of boron-modified succinimide/content of surfactant) is 20 or less.
11. The lubricating oil composition for internal combustion engines according to any one of claims 1 to 10, further comprising a poly(meth)acrylate.
- 50 12. The lubricating oil composition for internal combustion engines according to any one of claims 1 to 11, wherein the lubricant base oil is one or more selected from mineral oils and synthetic oils classified in Group 3 and Group 4 in base oil classification by American Petroleum Institute.
- 55 13. The lubricating oil composition for internal combustion engines according to any one of claims 1 to 12, which has a kinematic viscosity at 100°C of 3.8 to 12.5 mm²/s.
14. The lubricating oil composition for internal combustion engines according to any one of claims 1 to 13, which is used

for gasoline engines.

15. A friction reducing method for gasoline engines, comprising adding the lubricating oil composition for internal combustion engines of any one of claims 1 to 13 to a gasoline engine.

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INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2016/057465

A. CLASSIFICATION OF SUBJECT MATTER

C10M169/04(2006.01)i, C10M101/02(2006.01)i, C10M107/02(2006.01)i,
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C10N20/00
(2006.01)n, C10N20/02(2006.01)n, C10N20/04(2006.01)n, C10N30/06(2006.01)n,

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C10M169/04, C10M101/02, C10M107/02, C10M133/04, C10M139/00, C10M145/14,
C10N20/00, C10N20/02, C10N20/04, 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 1996-2016
Kokai Jitsuyo Shinan Koho 1971-2016 Toroku Jitsuyo Shinan Koho 1994-2016

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

Japio-GPG/FX & keyword: lubrication, alkylene oxide, internal combustion
engine and related terms,

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	JP 2010-209182 A (Cosmo Oil Lubricants Co., Ltd.), 24 September 2010 (24.09.2010), claims; paragraphs [0001] to [0058]; examples (Family: none)	1-2, 6-13 14-15
X	JP 2013-194097 A (Idemitsu Kosan Co., Ltd.), 30 September 2013 (30.09.2013), claims; paragraphs [0001] to [0059]; examples & US 2015/0080277 A1 claims; paragraphs [0001] to [0149] & WO 2013/137477 A1 & EP 2826845 A1 & CN 104169402 A & KR 10-2014-0135177 A	1-4, 6-15



Further documents are listed in the continuation of Box C.



See patent family annex.

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"&" document member of the same patent family

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INTERNATIONAL SEARCH REPORT

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C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 2004-353456 A (Nissan Motor Co., Ltd.), 16 December 2004 (16.12.2004), claims; paragraphs [0001] to [0051]; examples & US 2004/0241448 A1 claims; paragraphs [0001] to [0119] & EP 1482190 A2 & CN 1573143 A	1, 3-4, 6-15
X Y	JP 2001-139978 A (Tonen Corp.), 22 May 2001 (22.05.2001), claims; paragraphs [0001] to [0042]; examples (Family: none)	1-2, 5-13 14-15
X	JP 2014-227424 A (Showa Shell Sekiyu Kabushiki Kaisha), 08 December 2014 (08.12.2014), paragraphs [0001] to [0085]; comparative examples 8 to 11 & WO 2014/184362 A1	1-2, 5-6, 11-15
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Y	JP 2008-056876 A (Idemitsu Kosan Co., Ltd.), 13 March 2008 (13.03.2008), paragraphs [0001] to [0008] & US 2009/0203561 A1 paragraphs [0001] to [0022] & WO 2008/029756 A1 & EP 2060619 A1 & KR 10-2009-0046817 A & CN 101511983 A & RU 2009112387 A	14-15
P, X	JP 5807833 B1 (Turbulence Ltd.), 10 November 2015 (10.11.2015), claims; paragraphs [0001] to [0099]; examples (Family: none)	1-3, 5-6, 13-15
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INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2016/057465

Continuation of A. CLASSIFICATION OF SUBJECT MATTER
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C10N40/25(2006.01)n

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

Continuation of B. FIELDS SEARCHED

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

JDreamIII & keyword: JUNKATSU, ARUKIREN'OKISAIDO, NAINEN KIKAN (in Japanese) and related terms

REFERENCES CITED IN THE DESCRIPTION

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