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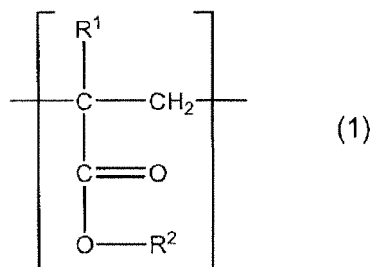
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(54) **ENGINE OIL COMPOSITION**

(57) An engine oil composition comprising: a lubricating base oil having a kinematic viscosity at 100°C of 1 to 5 mm²/s; a poly(meth)acrylate viscosity index improver containing a structural unit represented by the formula (1) in a proportion of 30 to 90 mol% and having a PSSI in an ultrasonic shear test of 15 or less; and a friction modifier, the engine oil composition has a kinematic viscosity at 100°C of 4 to 8 mm²/s, and an HTHS viscosity at 150°C of less than 2.6 mPa·s: [wherein R¹ represents hydrogen or a methyl group; and R² represents a linear or branched hydrocarbon group having a carbon number of 6 or less]:



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Description**Technical Field**

5 [0001] The present invention relates to an engine oil composition.

Background Art

10 [0002] In the related art, lubricating oils are used for internal combustion engines, transmissions, and other machine apparatuses to smooth their action. Particularly in lubricating oil for internal combustion engines (engine oil), high performance is required with higher performance of the internal combustion engines, higher outputs, severer operation conditions, and the like. Accordingly, to meet such required performances, various additives such as anti-wear agents, metallic detergents, ash-free dispersants, and antioxidants are blended with the conventional engine oil (for example, see Patent Literatures 1 to 3 below). Moreover, the fuel efficiency required for the lubricating oil is increasing recently.
15 For this reason, examinations have been conducted into ways of application of high viscosity index base oil and application of a variety of friction modifiers (for example, see Patent Literature 4 below).

Citation List20 **Patent Literature****[0003]**

25 Patent Literature 1: Japanese Patent Application Laid-Open No. 2001-279287
Patent Literature 2: Japanese Patent Application Laid-Open No. 2002-129182
Patent Literature 3: Japanese Patent Application Laid-Open No. 08-302378
Patent Literature 4: Japanese Patent Application Laid-Open No. 06-306384

Summary of Invention

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Technical Problem

[0004] However, it cannot be said that the conventional lubricating oils are not sufficient in fuel efficiency.

35 [0005] For example, as a standard method for fuel efficiency, a reduction in the kinematic viscosity of the lubricating oil and an increase in the viscosity index (multi-grading by a combination of low viscosity base oils with viscosity index improvers), and blending of friction reducing agents are known. In the case of a reduction in viscosity, the lubrication performance is reduced under severe lubrication conditions (under high temperature high shear conditions) due to a reduction of the viscosity of the lubricating oil or the base oil that forms the lubricating oil, occurrence of failures such as wear, burning, and fatigue breaking is concerned. Moreover, for blending of friction reducing agents, ash-free friction
40 modifiers and molybdenum friction modifiers are known, but there is demand for an oil with fuel efficiency superior to that of standard oils having these friction reducing agents.

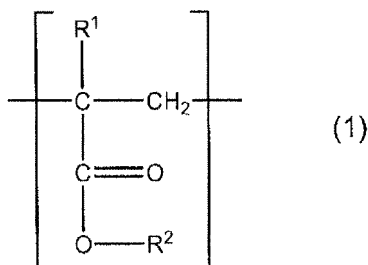
[0006] To prevent deficits in the reduction in viscosity and give fuel efficiency while maintaining durability, it is effective to increase an HTHS viscosity at 150°C ("HTHS viscosity" is also referred to as "high temperature high shear viscosity") while reducing a kinematic viscosity at 40°C, a kinematic viscosity at 100°C, and an HTHS viscosity at 100°C; however,
45 it is very difficult for the conventional lubricating oil to satisfy these requirements all.

[0007] Because of recent progress in the engine technique, the HTHS viscosity at 150°C can be further reduced while maintaining the durability of the engine, and the kinematic viscosity can be reduced more significantly than ever. For further improvement in fuel efficiency, an engine oil having an HTHS viscosity at 150°C of lower than 2.6 mPa·s, which is the lower limit of the HTHS viscosity at 150°C of an SAE OW-20 engine oil is developed and being used. However,
50 because for the engine oil having an HTHS viscosity at 150°C of lower than 2.6 mPa·s, the lubrication state of the engine oil to be used is severer than ever, it is important to maintain the HTHS viscosity at 150°C reduced by usage at a constant level or more. Moreover, it is important to reduce the kinematic viscosity as much as possible while reducing the coefficient of friction in a boundary lubrication region.

55 [0008] The present invention has been made in consideration of such circumstances, and an object of the present invention is to provide an engine oil composition excellent in fuel efficiency in which in an engine oil having an HTHS viscosity at 150°C of less than 2.6 mPa·s, a kinematic viscosity at 40°C and a kinematic viscosity at 100°C may be sufficiently reduced from the beginning of usage after usage for a long time, and an increase in a coefficient of friction in a boundary lubrication region may be sufficiently suppressed.

Solution to Problem

[0009] To solve the above problems, the present invention provides an engine oil composition including a lubricating base oil having a kinematic viscosity at 100°C of 1 to 5 mm²/s, a poly(meth)acrylate viscosity index improver (A) containing a structural unit represented by the following formula (1) in a proportion of 30 to 90 mol% and having a PSSI in an ultrasonic shear test of 20 or less, and a friction modifier (B), wherein the engine oil composition has a kinematic viscosity at 100°C of 4 to 8 mm²/s, and an HTHS viscosity at 150°C of less than 2.6 mPa·s.



wherein R¹ represents hydrogen or a methyl group; and R² represents a linear or branched hydrocarbon group having a carbon number of 6 or less.

[0010] It is preferable that the viscosity index improver (A) be a viscosity index improver having a PSSI of 10 or less and a ratio of the molecular weight to the PSSI (Mw/PSSI) of 1×10^4 or more.

[0011] It is preferable that the friction modifier (B) be an organic molybdenum friction modifier.

[0012] Here, the PSSI in the ultrasonic shear test in the present invention designates the reduction rate in the viscosity of the viscosity index improver evaluated according to JASO M347-95 (automatic transmission oil shear stability test method) under the condition where only the volume of a sample is increased. More specifically, it means a permanent shear stability index of a polymer (Permanent Shear Stability Index) obtained by adjusting an output with Standard oil A specified in the ASTM test method, performing a shear test at an amplitude of 28 μm, a vibration number of 10 KHz, an irradiation time of 10 minutes, and the volume of the sample of 60 mL, and calculating the measured kinematic viscosity. The PSSI is calculated from $((V1 - V2)/V1 \times 100)$ (%) based on the viscosity (V1) at 100°C per amount of the viscosity index improver to be added which is measured before the shear test and the viscosity (V2) at 100°C per amount of the viscosity index improver to be added which is measured after the shear test.

Advantageous Effects of Invention

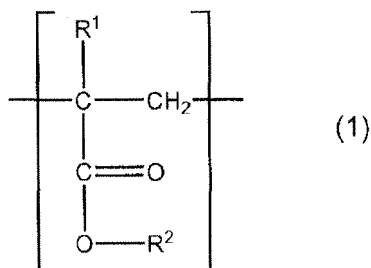
[0013] As above, according to the present invention, an engine oil composition excellent in fuel efficiency in which in an engine oil having an HTHS viscosity at 150°C of less than 2.6 mPa·s, the kinematic viscosity at 40°C and the kinematic viscosity at 100°C may be sufficiently reduced from the beginning of usage after usage for a long time, and an increase in the coefficient of friction in the boundary lubrication region may be sufficiently suppressed can be provided.

[0014] Moreover, the engine oil composition according to the present invention may be suitably used not only for gasoline engines, diesel engines, gas engines, and the like for bicycles, automobiles, power generation, cogeneration, and the like but also for a variety of engines using fuels having a sulfur content of 50 mass ppm or less. The lubricating oil composition according to the present invention is also useful for a variety of engines for ships and outboard motors.

Description of Embodiments

[0015] Hereinafter, suitable Embodiment of the present invention will be described in detail.

[0016] The engine oil composition according to the present embodiment includes a lubricating base oil having a kinematic viscosity at 100°C of 1 to 5 mm²/s, a poly(meth)acrylate viscosity index improver (A) containing one or two more of structural unit represented by the following formula (1) in a proportion of 30 to 90 mol% and having a PSSI in an ultrasonic shear test of 15 or less, and a friction modifier (B).



[wherein R¹ represents hydrogen or a methyl group; and R² represents a linear or branched hydrocarbon group having a carbon number of 6 or less].

[0017] In the engine oil composition according to the present embodiment, a lubricating base oil having a kinematic viscosity at 100°C of 1 to 5 mm²/s (hereinafter referred to as a "lubricating base oil according to the present embodiment") is used.

[0018] Examples of the lubricating base oil according to the present embodiment include those having a kinematic viscosity at 100°C of 1 to 5 mm²/s among paraffin mineral oils, normal paraffin base oils, and isoparaffin base oils obtainable by refining a lubricating oil fraction obtainable by normal pressure distillation and/or reduced pressure distillation of a crude oil using one or two or more refining treatments selected from solvent deasphalting, solvent extraction, hydrocracking, solvent dewaxing, catalytic dewaxing, hydrorefining, sulfuric acid washing, clay treatment, and the like.

[0019] Preferable examples of the lubricating base oil according to the present embodiment may include base oils obtainable by using the base oils (1) to (8) shown below as the raw material, and refining the raw material oil and/or a lubricating oil fraction recovered from the raw material oil by a predetermined refining method to recover a lubricating oil fraction:

- (1) Distilled oil obtainable by normal pressure distillation of a paraffinic crude oil and/or a mixed-base crude oil,
- (2) Distilled oil obtainable by reduced pressure distillation of a paraffinic crude oil and/or a normal pressure distillation residue oil of a mixed-base crude oil (WVGO),
- (3) Waxes obtainable by a lubricating oil dewaxing step (such as slack wax) and/or synthetic waxes obtainable by a gas-to-liquid (GTL) process or the like (such as Fischer-Tropsch wax and GTL wax),
- (4) One or two or more mixed oils selected from the base oils (1) to (3) and/or a mild hydrocracked oil of the mixed oil,
- (5) Two or more mixed oils selected from the base oils (1) to (4),
- (6) Deasphalted oil (DAO) of the base oil (1), (2), (3), (4) or (5),
- (7) Mild hydrocracked oil (MHC) of the base oil (6), and
- (8) Mixed oil of two or more selected from the base oils (1) to (7).

[0020] For the predetermined refining method, hydrorefining such as hydrocracking and hydrofinishing; solvent refining such as furfural solvent extraction; dewaxing such as solvent dewaxing and catalytic dewaxing; white clay refining with acid clay or activated clay; and chemical (acid or alkali) washing such as sulfuric acid washing and sodium hydroxide washing are preferable. In the present embodiment, among these refining methods, one method may be used alone, or two or more may be used in combination. If two or more refining methods are combined, the order is not particularly limited, and can be properly selected.

[0021] Furthermore, for the lubricating base oil according to the present embodiment, a base oil selected from the base oils (1) to (8) or the following base oil (9) or (10) obtainable by performing a predetermined treatment on the lubricating oil fraction recovered from the base oil is particularly preferable:

- (9) Hydrocracking base oils obtainable by hydrocracking the base oil selected from the base oils (1) to (8) or the lubricating oil fraction recovered from the base oil, and performing a dewaxing treatment such as solvent dewaxing and catalytic dewaxing on the product or the lubricating oil fraction recovered from the product by distillation or the like, or performing distillation after the dewaxing treatment, and
- (10) Hydrogenation isomerized base oils obtainable by hydrogenation isomerizing the base oil selected from the base oils (1) to (8) or the lubricating oil fraction recovered from the base oil, and performing a dewaxing treatment such as solvent dewaxing and catalytic dewaxing on the product or the lubricating oil fraction recovered from the product by distillation or the like, or performing distillation after the dewaxing treatment.

[0022] In production of the lubricating base oil (9) or (10), a solvent refining treatment and/or a hydrofinishing step may be included as preferably steps when necessary.

[0023] The catalyst used in the hydrocracking and hydrogenation isomerization is not particularly limited, and hydro-

cracking catalysts in which a carrier is a composite oxide having cracking activity (such as silica alumina, alumina boria, and silica zirconia) or one or more of the composite oxides in combination bounded by a binder, and a metal having a hydrogenation activity (such as one or more metals in Groups VIa and VIII in the periodic table) is carried to the carrier; or hydrogenation isomerization catalysts in which a metal containing at least one metal among metals in Group VIII and having a hydrogenation activity is carried to a carrier containing zeolite (such as ZSM-5, zeolite beta, and SAPO-11) are preferably used. The hydrocracking catalyst and the hydrogenation isomerization catalyst may be used in combination by stacking or mixing.

[0024] The reaction conditions in hydrocracking and hydrogenation isomerization are not particularly limited, and a hydrogen partial pressure of 0.1 to 20 MPa, an average reaction temperature of 150 to 450°C, an LHSV of 0.1 to 3.0 hr⁻¹, and the hydrogen/oil ratio of 50 to 20000 scf/b are preferred.

[0025] The kinematic viscosity at 100°C of the lubricating base oil according to the present embodiment needs to be 5 mm²/s or less, preferably 4.5 mm²/s or less, more preferably 4 mm²/s or less, still more preferably 3.8 mm²/s or less, particularly preferably 3.7 mm²/s or less, and most preferably 3.6 mm²/s or less. The kinematic viscosity at 100°C needs to be 1 mm²/s or more. It is preferable that the kinematic viscosity at 100°C be 1.5 mm²/s or more. The kinematic viscosity at 100°C is more preferably 2 mm²/s or more, still more preferably 2.5 mm²/s or more, and particularly preferably 3 mm²/s or more. The kinematic viscosity at 100°C here designates the kinematic viscosity at 100°C specified in ASTM D-445. In the case where the kinematic viscosity at 100°C of the lubricating base oil exceeds 5 mm²/s, low temperature viscosity properties may be reduced, and sufficient fuel efficiency may not be obtained. At a kinematic viscosity of 1 mm²/s or less, formation of an oil film in a lubrication place is insufficient; for this reason, lubrication is inferior, and the evaporation loss of the lubricating oil composition may be increased.

[0026] The kinematic viscosity at 40°C of the lubricating base oil according to the present embodiment is preferably 40 mm²/s or less, more preferably 30 mm²/s or less, still more preferably 25 mm²/s or less, particularly preferably 20 mm²/s or less, and most preferably 17 mm²/s or less. The kinematic viscosity at 40°C is preferably 6.0 mm²/s or more, more preferably 8.0 mm²/s or more, still more preferably 10 mm²/s or more, particularly preferably 12 mm²/s or more, and most preferably 14 mm²/s or more. In the case where the kinematic viscosity at 40°C of the lubricating base oil exceeds 40 mm²/s, low temperature viscosity properties may be reduced, and sufficient fuel efficiency may not be obtained. At a kinematic viscosity of 6.0 mm²/s or less, formation of an oil film in a lubrication place is insufficient; for this reason, lubrication is inferior, and the evaporation loss of the lubricating oil composition may be increased.

[0027] It is preferable that the viscosity index of the lubricating base oil according to the present embodiment be 100 or more. The viscosity index is more preferably 105 or more, still more preferably 110 or more, particularly preferably 115 or more, and most preferably 120 or more. If the viscosity index is less than 100, not only viscosity-temperature properties, heat and oxidation stability, and anti-volatilization are reduced, but also the coefficient of friction tends to be increased; and resistance against wear tends to be reduced.

[0028] The viscosity index in the present invention means the viscosity index measured according to JIS K 2283-1993.

[0029] It is preferable that the lubricating base oil in the engine oil composition according to the present embodiment be a mixture of a first lubricating base oil component having a kinematic viscosity at 100°C of 3.5 mm²/s or more and having a viscosity index of 120 or more and a second lubricating base oil component having a kinematic viscosity at 100°C of less than 3.5 mm²/s. By using the mixture of the first lubricating base oil component and the second lubricating base oil component, high viscosity temperature properties may be given to improve fuel efficiency more significantly.

[0030] The density (ρ_{15}) at 15°C of the first lubricating base oil component used in the engine oil composition according to the present embodiment is preferably 0.860 or less, more preferably 0.850 or less, still more preferably 0.840 or less, and particularly preferably 0.822 or less.

[0031] The density at 15°C in the present invention means a density measured at 15°C according to JIS K 2249-1995.

[0032] The pour point of the first lubricating base oil component used in the engine oil composition according to the present embodiment is preferably -10°C or less, more preferably -12.5°C or less, still more preferably -15°C or less, and particularly preferably -20°C or less. If the pour point exceeds the upper limit value, the fluidity at a low temperature of the entire lubricating oil using the lubricating base oil tends to be reduced. The pour point in the present invention means the pour point measured according to JIS K 2269-1987.

[0033] It is preferable that the kinematic viscosity at 100°C of the first lubricating base oil component used in the engine oil composition according to the present embodiment be 5 mm²/s or less. The kinematic viscosity is more preferably 4.5 mm²/s or less, still more preferably 4.0 mm²/s or less, and particularly preferably 3.9 mm²/s or less. It is preferable that the kinematic viscosity at 100°C be 3.5 mm²/s or more. The kinematic viscosity is more preferably 3.6 mm²/s or more, still more preferably 3.7 mm²/s or more, and particularly preferably 3.8 mm²/s or more. If the kinematic viscosity at 100°C exceeds 5 mm²/s, low temperature viscosity properties may be reduced, and sufficient fuel efficiency may not be obtained. At a kinematic viscosity less than 3.5 mm²/s, formation of an oil film in a lubrication place is insufficient; for this reason, lubrication is inferior, and the evaporation loss of the engine oil composition may be increased.

[0034] The kinematic viscosity at 40°C of the first lubricating base oil component used in the engine oil composition according to the present embodiment is preferably 40 mm²/s or less, more preferably 30 mm²/s or less, still more

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preferably 25 mm²/s or less, particularly preferably 20 mm²/s or less, and most preferably 17 mm²/s or less. The kinematic viscosity at 40°C is preferably 6.0 mm²/s or more, more preferably 8.0 mm²/s or more, still more preferably 10 mm²/s or more, particularly preferably 12 mm²/s or more, and most preferably 14 mm²/s or more. In the case where the kinematic viscosity at 40°C exceeds 40 mm²/s, low temperature viscosity properties may be reduced, and sufficient fuel efficiency may not be obtained. At a kinematic viscosity of 6.0 mm²/s or less, formation of an oil film in a lubrication place is insufficient; for this reason, lubrication is inferior, and the evaporation loss of the lubricating oil composition may be increased.

[0035] It is preferable that the viscosity index of the first lubricating base oil component used in the engine oil composition according to the present embodiment be 100 or more. The viscosity index is more preferably 110 or more, still more preferably 120 or more, particularly preferably 130 or more, and most preferably 140 or more. The viscosity index is preferably 170 or less, more preferably 160 or less, still more preferably 155 or less, and particularly preferably 150 or less. If the viscosity index is less than 100, not only viscosity-temperature properties, heat and oxidation stability, and anti-volatilization are reduced, but also the coefficient of friction tends to be increased; and resistance against wear tends to be reduced. If the viscosity index exceeds 170, low temperature viscosity tends to be increased to reduce the fuel efficiency at low oil temperatures. Moreover, startability tends to be reduced.

[0036] The density (ρ_{15}) at 15°C of the second lubricating base oil component used in the engine oil composition according to the present embodiment is preferably 0.860 or less, more preferably 0.850 or less, still more preferably 0.840 or less, and particularly preferably 0.835 or less.

[0037] The pour point of the second lubricating base oil component used in the engine oil composition according to the present embodiment is preferably -10°C or less, more preferably -12.5°C or less, still more preferably -15°C or less, and particularly preferably -20°C or less. If the pour point exceeds the upper limit value, the fluidity at a low temperature of the entire lubricating oil using the lubricating base oil tends to be reduced.

[0038] It is preferable that the kinematic viscosity at 100°C of the second lubricating base oil component used in the engine oil composition according to the present embodiment be less than 3.5 mm²/s. The kinematic viscosity is more preferably 3.4 mm²/s or less, and still more preferably 3.3 mm²/s or less. It is preferable that the kinematic viscosity at 100°C be 2 mm²/s or more, and the kinematic viscosity is more preferably 2.5 mm²/s or more, and still more preferably 3.0 mm²/s or more. If the kinematic viscosity at 100°C exceeds 3.5 mm²/s, low temperature viscosity properties may be reduced, and sufficient fuel efficiency may not be obtained. At a kinematic viscosity less than 2 mm²/s, formation of an oil film in a lubrication place is insufficient; for this reason, lubrication is inferior, and the evaporation loss of the lubricating oil composition may be increased.

[0039] The kinematic viscosity at 40°C of the second lubricating base oil component used in the engine oil composition according to the present embodiment is preferably 20 mm²/s or less, more preferably 18 mm²/s or less, still more preferably 16 mm²/s or less, and particularly preferably 14 mm²/s or less. The kinematic viscosity at 40°C is preferably 6.0 mm²/s or more, more preferably 8.0 mm²/s or more, still more preferably 10 mm²/s or more, particularly preferably 12 mm²/s or more, and most preferably 13 mm²/s or more. If the kinematic viscosity at 40°C exceeds 20 mm²/s, low temperature viscosity properties may be reduced, and sufficient fuel efficiency may not be obtained. At a kinematic viscosity of 6.0 mm²/s or less, formation of an oil film in a lubrication place is insufficient; for this reason, lubrication is inferior, and the evaporation loss of the lubricating oil composition may be increased.

[0040] It is preferable that the viscosity index of the second lubricating base oil component used in the engine oil composition according to the present embodiment be 100 or more. The viscosity index is more preferably 105 or more, and still more preferably 110 or more. The viscosity index is preferably 160 or less, more preferably 150 or less, still more preferably 140 or less, and particularly preferably 135 or less. If the viscosity index is less than 100, not only viscosity-temperature properties, heat and oxidation stability, and anti-volatilization are reduced, but also the coefficient of friction tends to be increased. Moreover, resistance against wear tends to be reduced. If the viscosity index exceeds 160, low temperature viscosity tends to be increased to reduce the fuel efficiency at low oil temperatures. Moreover, startability tends to be reduced.

[0041] The sulfur content of the lubricating base oil used in the present embodiment depends on the sulfur content of the raw material. For example, in the case where a raw material containing substantially no sulfur as a synthetic wax component obtainable by the Fischer-Tropsch reaction or the like is used, the lubricating base oil containing substantially no sulfur may be obtainable. In the case where a raw material containing sulfur such as slack wax produced in the refining process of the lubricating base oil and microcrystalline wax produced in the wax refining process thereof is used, the sulfur content of the lubricating base oil to be obtainable is usually 100 mass ppm or more. In the lubricating base oil according to the present embodiment, from the viewpoint of further improvement in heat and oxidation stability and a reduction in sulfur, it is preferable that the sulfur content be 100 mass ppm or less, it is more preferable that the sulfur content be 50 mass ppm or less, it is still more preferable that the sulfur content be 10 mass ppm or less, and it is particularly preferable that the sulfur content be 5 mass ppm or less.

[0042] The nitrogen content of the lubricating base oil used in the present embodiment is preferably 7 mass ppm or less, more preferably 5 mass ppm or less, and still more preferably 3 mass ppm or less. If the nitrogen content exceeds

5 mass ppm, heat and oxidation stability tends to be reduced. The nitrogen content in the present invention means the nitrogen content measured according to JIS K 2609-1990.

[0043] It is preferable that %C_p of the lubricating base oil used in the present embodiment be 70 or more. %C_p is preferably 80 or more, more preferably 85 or more, still more preferably 87 or more, and particularly preferably 90 or more. %C_p is preferably 99.9 or less, more preferably 98 or less, still more preferably 96 or less, and particularly preferably 94 or less. If %C_p of the lubricating base oil is less than the lower limit value, viscosity-temperature properties, heat and oxidation stability, and friction properties tend to be reduced; furthermore, if an additive is blended with the lubricating base oil, the effect of the additive tends to be reduced. If %C_p of the lubricating base oil exceeds the upper limit value, the solubility of the additive tends to be reduced.

[0044] It is preferable that %C_A of the lubricating base oil used in the present embodiment be 2 or less. %C_A is more preferably 1 or less, still more preferably 0.8 or less, and particularly preferably 0.5 or less. If %C_A of the lubricating base oil exceeds the upper limit value, viscosity-temperature properties, heat and oxidation stability, and fuel efficiency tend to be reduced.

[0045] It is preferable that %C_N of the lubricating base oil used in the present embodiment be 30 or less. %C_N is preferably 25 or less, more preferably 20 or less, still more preferably 15 or less, and particularly preferably 10 or less. %C_N is preferably 1 or more, more preferably 3 or more, still more preferably 5 or more, and particularly preferably 6 or more. If %C_N of the lubricating base oil exceeds the upper limit value, viscosity-temperature properties, heat and oxidation stability, and friction properties tend to be reduced. If %C_N is less than the lower limit value, the solubility of the additive tends to be reduced.

[0046] %C_p, %C_N, and %C_A in the present invention mean the percentage of the number of paraffin carbon atoms to the total number of carbon atoms, the percentage of the number of naphthene carbon atoms to the total number of carbon atoms, and the percentage of the number of aromatic carbon atoms to the total number of carbon atoms, respectively, which are determined by the method (n-d-M ring analysis) according to ASTM D 3238-85. Namely, preferable ranges of %C_p, %C_N, and %C_A above described are based on the values determined by the method above; for example, even in a lubricating base oil containing no naphthene content, %C_N determined by the method may indicate a value more than zero.

[0047] The content of the saturates in the lubricating base oil used in the present embodiment is preferably 90% by mass or more, preferably 95% by mass or more, more preferably 99% by mass or more based on the total amount of the lubricating base oil; the proportion of the cyclic saturates in the saturates is preferably 40% by mass or less, preferably 35% by mass or less, preferably 30% by mass or less, more preferably 25% by mass or less, and still more preferably 21% by mass or less. The proportion of the cyclic saturates in the saturates is preferably 5% by mass or more, and more preferably 10% by mass or more. If the content of the saturates and the proportion of the cyclic saturates in the saturates each satisfy the conditions above, viscosity-temperature properties and heat and oxidation stability may be improved; if an additive is blended with the lubricating base oil, the function of the additive may be demonstrated at a higher level while the additive is sufficiently stably dissolved in the lubricating base oil. Furthermore, according to the present embodiment, the friction properties of the lubricating base oil itself may be improved; as a result, an improvement in a friction reducing effect and thus an improvement in energy saving properties may be attained.

[0048] The saturates in the present invention are measured by the method described in ASTM D 2007-93 above.

[0049] In the method for separating the saturates or in composition analysis of the cyclic saturates, a noncyclic saturates, or the like, similar methods that give similar results can be used. Examples of the methods, besides the method above, can include the method described in ASTM D 2425-93, the method described in ASTM D 2549-91, the method by a high performance liquid chromatography (HPLC), or the modified methods thereof.

[0050] The aromatic content of the lubricating base oil used in the present embodiment is preferably 5% by mass or less, more preferably 4% by mass or less, still more preferably 2% by mass or less, and particularly preferably 1% by mass or less based on the total amount of the lubricating base oil, and is preferably 0.1% by mass or more, and more preferably 0.2% by mass or more based on the total amount of the lubricating base oil. If the content of the aromatics exceeds the upper limit value, viscosity-temperature properties, heat and oxidation stability, friction properties, anti-volatilization, and low temperature viscosity properties tend to be reduced; if an additive is blended with the lubricating base oil, the effect of the additive tends to be reduced. The lubricating base oil according to the present embodiment may be a lubricating base oil containing no aromatics, but the content of the aromatics in the range of the lower limit value or more can further enhance the solubility of the additive.

[0051] The aromatic content in the present invention means the value measured according to ASTM D 2007-93. The aromatics usually include alkylbenzene and alkylnaphthalene; anthracene, phenanthrene, and alkylated products thereof; compounds in which four or more benzene rings are condensed; and aromatic compounds having a heteroatom such as pyridines, quinolines, phenols, and naphthols.

[0052] A synthetic base oil may be used as the lubricating base oil according to the present embodiment. Examples of the synthetic base oil are base oils having a kinematic viscosity at 100°C of 1 to 20 mm²/s. The examples include poly- α -olefins or hydrides thereof, isobutene oligomers or hydrides thereof, isoparaffin, alkylbenzene, alkylnaphthalene,

diesters (such as ditridecyl glutarate, di-2-ethylhexyl adipate, diisodecyl adipate, ditridecyl adipate, and di-2-ethylhexyl-
 icebacate), polyol esters (such as trimethylolpropane caprylate, trimethylolpropane pelargonate, pentaerythritol 2-ethyl-
 hexanoate, and pentaerythritol pelargonate), polyoxyalkylene glycol, dialkyldiphenyl ether, polyphenyl ether; among
 these, poly- α -olefins are preferable. Examples of poly- α -olefins typically include oligomers or co-oligomers of α -olefins
 having a carbon number of 2 to 32, preferably a carbon number of 6 to 16 (such as 1-octene oligomer, decene oligomer,
 and ethylene-propylene co-oligomer) and hydrides thereof.

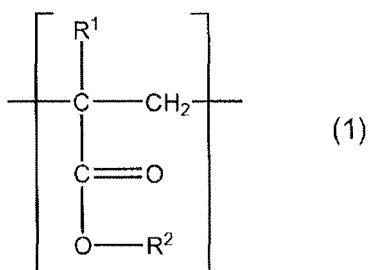
[0053] The method for producing poly- α -olefin is not particularly limited, and examples thereof include a method for
 polymerizing α -olefin in the presence of a polymerization catalyst containing a complex of aluminum trichloride or boron
 trifluoride, water, an alcohol (such as ethanol, propanol, and butanol), and carboxylic acid or an ester thereof, such as
 a Friedel-Crafts catalyst.

[0054] In the engine oil composition according to the present embodiment, the lubricating base oil according to the
 present embodiment may be used alone, or the lubricating base oil according to the present embodiment may be used
 in combination with one or two or more of other base oils. In the case where the lubricating base oil according to the
 present embodiment is used in combination with the other base oil(s), it is preferable that the proportion of the lubricating
 base oil according to the present embodiment in the mixed base oils be 30% by mass or more, it is more preferable that
 the proportion of the lubricating base oil according to the present embodiment in the mixed base oils be 50% by mass
 or more, and it is still more preferable that the proportion of the lubricating base oil according to the present embodiment
 in the mixed base oils be 70% by mass or more.

[0055] The other base oils used in combination with the lubricating base oil according to the present embodiment is
 not particularly limited, and examples of a mineral oil-based base oil include solvent refined mineral oils, hydrocracked
 mineral oils, hydrotreated mineral oils, and solvent dewaxed base oils whose kinematic viscosity at 100°C is 5 mm²/s
 or more and 100 mm²/s or less.

[0056] Examples of the other synthetic base oils used in combination with the lubricating base oil according to the
 present embodiment include the synthetic base oils described above whose kinematic viscosity at 100°C is out of the
 range of 1 to 5 mm²/s.

[0057] Moreover, the engine oil composition according to the present embodiment contains a poly(meth)acrylate
 viscosity index improver (A) (hereinafter referred to as "viscosity index improver according to the present embodiment"
 for convenience) containing one or two or more of the structural units represented by the following formula (1) in a
 proportion of 30 to 90 mol% and having the PSSI in the ultrasonic shear test of 15 or less. Thereby, the fuel efficiency,
 the maintenance of fuel efficiency, and the durability may be enhanced more significantly compared to the case where
 the present configuration is not included. The compound has any form as long as the compound satisfies the condition
 that the compound is a poly(meth)acrylate viscosity index improver having the structural unit represented by the following
 formula (1) in the proportion of 30 to 90 mol% and having the PSSI in the ultrasonic shear test of 15 or less. Specific
 examples of the compound can include non-dispersive or dispersive poly(meth)acrylate viscosity index improvers,
 (meth)acrylate-olefin copolymers, or a mixture thereof.



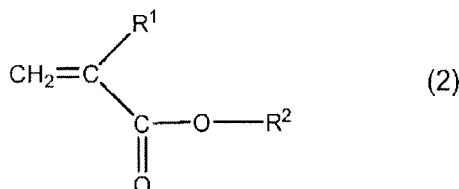
[wherein R¹ represents hydrogen or a methyl group; and R² represents a linear or branched hydrocarbon group having
 a carbon number of 6 or less].

[0058] R² in the structural unit represented by the formula (1), as described above, is a linear or branched hydrocarbon
 group having a carbon number of 6 or less and may be one hydrocarbon group or a mixture of two or more thereof; R²
 is preferably a linear or branched hydrocarbon having a carbon number of 4 or less, still more preferably a linear or
 branched hydrocarbon having a carbon number of 3 or less, and more preferably a hydrocarbon group having a carbon
 number of 2 or less.

[0059] In the viscosity index improver according to the present embodiment, the proportion of the (meth)acrylate
 structural unit represented by the formula (1) in a polymer is 30 to 90 mol% as described above, preferably 80 mol% or
 less, more preferably 70 mol% or less, still more preferably 65 mol% or less, and particularly preferably 60 mol% or less.
 The proportion is preferably 32 mol% or more, more preferably 35 mol% or more, and still more preferably 40 mol% or
 more. At a proportion more than 90 mol%, the solubility in the base oil, the effect of improving viscosity temperature

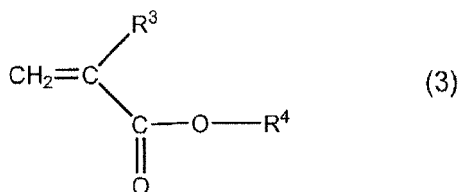
properties, and low temperature viscosity properties may be inferior; at a proportion less than 20 mol%, the effect of improving viscosity temperature properties may be inferior.

[0060] Besides the (meth)acrylate structural unit represented by the formula (1), the viscosity index improver according to the present embodiment may be a copolymer having any (meth)acrylate structural unit. Such a copolymer may be obtainable by copolymerizing one or two or more of monomers represented by the following formula (2) (hereinafter referred to as "Monomer (M-1)") and a monomer other than Monomer (M-1).



[wherein R¹ represents a hydrogen atom or a methyl group; and R² represents a linear or branched hydrocarbon group having a carbon number of 6 or less].

[0061] Any monomer can be used in combination with Monomer (M-1), and a monomer represented by the following formula (3) (hereinafter referred to as "Monomer (M-2)") is suitable, for example. A copolymer of Monomer (M-1) and Monomer (M-2) is a non-dispersive poly(meth)acrylate viscosity index improver.

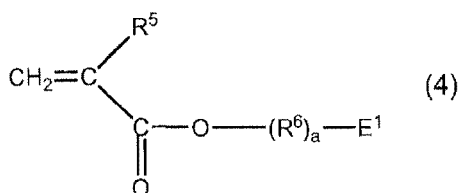


[wherein R³ represents a hydrogen atom or a methyl group; and R⁴ represents a linear or branched hydrocarbon group having a carbon number of 7 or more].

[0062] R⁴ in the structural unit represented by the formula (3) is a linear or branched hydrocarbon group having a carbon number of 7 or more, preferably a linear or branched hydrocarbon having a carbon number of 10 or more, more preferably a linear or branched hydrocarbon having a carbon number of 15 or more, and still more preferably a branched hydrocarbon group having a carbon number of 18 or more. The upper limit of the hydrocarbon group represented as R⁴ is not particularly limited, and it is preferable that it be a linear or branched hydrocarbon group having a carbon number of 2000 or less. It is more preferably a linear or branched hydrocarbon having a carbon number of 500 or less, still more preferably a linear or branched hydrocarbon having a carbon number of 100 or less, particularly preferably a branched hydrocarbon having a carbon number of 50 or less, and most preferably a branched hydrocarbon having a carbon number of 25 or less.

[0063] In the viscosity index improver according to the present embodiment, the (meth)acrylate structural unit represented by the formula (3) in the polymer may be one or two or more mixtures, and it is preferable that the proportion be 0.5 to 70 mol%, the proportion is more preferably 60 mol% or less, still more preferably 50 mol% or less, particularly preferably 40 mol% or less, and most preferably 30 mol% or less. The proportion is preferably 1 mol% or more, more preferably 3 mol% or more, still more preferably 5 mol% or more, and particularly preferably 10 mol% or more. At a proportion more than 70 mol%, the effect of improving viscosity temperature properties and low temperature viscosity properties may be inferior; at a proportion less than 0.5 mol%, the effect of improving viscosity temperature properties may be inferior.

[0064] It is suitable that the other monomers used in combination with Monomer (M-1) be one or two or more selected from the monomer represented by the following formula (4) (hereinafter referred to as "Monomer (M-3)") and the monomer represented by the following formula (5) (hereinafter referred to as "Monomer (M-4)"). A copolymer of Monomers (M-1) and (M-3) and/or (M-4) is the so-called dispersive poly(meth)acrylate viscosity index improver. The dispersive poly(meth)acrylate viscosity index improver may further contain Monomer (M-2) as the constitutional monomer.



[wherein R⁵ represents a hydrogen atom or a methyl group, R⁶ represents an alkylene group having a carbon number of 1 to 18, E¹ represents an amine residue or heterocycle residue having a nitrogen number of 1 to 2 and an oxygen number of 0 to 2, and a represents 0 or 1].

[0065] Examples of the alkylene group having a carbon number of 1 to 18 represented by R⁶ specifically may include an ethylene group, a propylene group, a butylene group, a pentylene group, a hexylene group, a heptylene group, an octylene group, a nonylene group, a decylene group, an undecylene group, a dodecylene group, a tridecylene group, a tetradecylene group, a pentadecylene group, a hexadecylene group, a heptadecylene group, and an octadecylene group (these alkylene groups may be linear or branched).

[0066] Examples of the group represented by E¹ specifically can include a dimethylamino group, a diethylamino group, a dipropylamino group, a dibutylamino group, an anilino group, a toluidino group, a xylidino group, an acetylamino group, a benzoylamino group, a morpholino group, a pyrrolyl group, a pyrrolino group, a pyridyl group, a methylpyridyl group, a pyrrolidinyl group, a piperidinyl group, a quinonyl group, a pyrrolidonyl group, a pyrrolidono group, an imidazolino group, and a pyrazino group.



[wherein R⁷ represents a hydrogen atom or a hydrocarbon group, and E² represents a hydrocarbon group or an amine residue or heterocycle residue having a nitrogen number of 1 to 2 and an oxygen number of 0 to 2].

[0067] Examples of the group represented by E² specifically can include a dimethylamino group, a diethylamino group, a dipropylamino group, a dibutylamino group, an anilino group, a toluidino group, a xylidino group, an acetylamino group, a benzoylamino group, a morpholino group, a pyrrolyl group, a pyrrolino group, a pyridyl group, a methylpyridyl group, a pyrrolidinyl group, a piperidinyl group, a quinonyl group, a pyrrolidonyl group, a pyrrolidono group, an imidazolino group, and a pyrazino group.

[0068] Preferable examples of Monomers (M-3) and (M-4) specifically may include dimethylaminomethyl methacrylate, diethylaminomethyl methacrylate, dimethylaminoethyl methacrylate, diethylaminoethyl methacrylate, 2-methyl-5-vinyl pyridine, morpholinomethyl methacrylate, morpholinoethyl methacrylate, N-vinylpyrrolidone, and mixtures thereof.

[0069] The copolymerization molar ratio of a copolymer of Monomer (M-1) and Monomers (M-2) to (M-4) is not particularly limited, and Monomer (M-1): Monomers (M-2) to (M-4) = 20:80 to 90:10 or so is preferable, and the ratio is more preferably 30:70 to 80:20, and still more preferably 40:60 to 70:30.

[0070] The method for producing the viscosity index improver according to the present embodiment is not particularly limited, and examples thereof include a method in which using a control radical polymerization process, an alkyl methacrylate serving as an arm portion (polymerization chain of alkyl methacrylate) is polymerized, and the polyalkyl methacrylate is then reacted with a polyfunctional compound having two or more ethylenic unsaturated double bonds.

[0071] The control radical polymerization process involves an atom transfer radical polymerization (ATRP) process, a reversible addition-fragmentation chain transfer (RAFT) process, or a nitroxide mediated polymerization process.

[0072] The discussion on the polymer mechanism of the ATRP polymerization is shown in Matyjaszewski et al., Reaction scheme 11.1, p. 524; Reaction scheme 11.4, p. 566; Reaction scheme 11, 7, p. 571; Reaction scheme 11.8, p. 572; and Reaction scheme 11.9, p. 575.

[0073] The discussion on the polymer mechanism of the RAFT polymerization is shown in Matyjaszewski et al., section 12.4.4, pp. 664 to 665.

[0074] The detailed description of nitroxide mediated polymerization (Chapter 10, pp. 463 to 522), ATRP (Chapter 11, pp. 523 to 628), and RAFT (Chapter 12, pp. 629 to 690) are shown in "Handbook of Radical Polymerization" (Krzysztof Matyjaszewski and Thomas P. Davis, copyright 2002, published by John Wiley and Sons Inc. (hereinafter referred to as "Matyjaszewski et al.")).

[0075] The synthesis can be performed as a batch operation, a semi-batch operation, a continuous step, a feed step, or a bulk step. The synthesis may be performed in an emulsion, a solution, or a suspension.

[0076] In the synthesis, by changing the amounts of an initiator and the polyfunctional compound having two or more ethylenic unsaturated double bonds to be used, the average molecular weight of the polymethacrylate or viscosity index improver to be produced may be adjusted.

[0077] The reaction rate to the viscosity index improver using the synthesized arm portion is 70% or more, preferably 80% or more, and more preferably 85% or more based on the amount of the polymer reacted to become the viscosity index improver. If the reaction rate is low, the arm portion remains, and the molecular weight cannot be increased.

[0078] The PSSI in the ultrasonic shear test (shear stability index) of the viscosity index improver according to the present embodiment is 15 or less, preferably 12 or less, more preferably 10 or less, and still more preferably 5 or less. If the PSSI in the ultrasonic shear test exceeds 15, shear stability is poor, and initial viscosity temperature properties and fuel efficiency are reduced in order to keep the kinematic viscosity and the HTHS viscosity after usage at a constant level or more.

[0079] The PSSI (Permanent Shear Stability Index) of the viscosity index improver according to the present embodiment in a diesel injector method is preferably 20 or less, more preferably 15 or less, still more preferably 10 or less, particularly preferably 5 or less, and most preferably 3 or less. If the PSSI exceeds 20, shear stability is poor, and to keep the kinematic viscosity and the HTHS viscosity after usage at a constant level or more, the initial fuel efficiency may be reduced.

[0080] The "PSSI in the diesel injector method" here means the permanent shear stability index of the polymer (Permanent Shear Stability Index) calculated based on the data measured by ASTM D 6278-02 (Test Method for Shear Stability of Polymer Containing Fluids Using a European Diesel Injector Apparatus) in accordance with ASTM D 6022-01 (Standard Practice for Calculation of Permanent Shear Stability Index).

[0081] It is preferable that the weight average molecular weight (M_w) of the viscosity index improver according to the present embodiment be 100,000 or more, and the weight average molecular weight is more preferably 200,000 or more, still more preferably 300,000 or more, and particularly preferably 400,000 or more. It is preferable that the weight average molecular weight be 1,000,000 or less, and the weight average molecular weight is more preferably 900,000 or less, still more preferably 700,000 or less, and particularly preferably 600,000 or less. If the weight average molecular weight is less than 100,000, the effect of improving a viscosity index when the viscosity index improver is dissolved in the lubricating base oil is small; not only fuel efficiency and low temperature viscosity properties are inferior but also cost may increase. If the weight average molecular weight exceeds 1,000,000, the effect of increasing viscosity is excessively increased; not only fuel efficiency and low temperature viscosity properties are inferior, but also shear stability, the solubility in the lubricating base oil, and storage stability are reduced.

[0082] It is preferable that the ratio of the weight average molecular weight of the viscosity index improver according to the present embodiment to the PSSI in the ultrasonic shear test (M_w /PSSI) be 1.0×10^4 or more, and the ratio is more preferably 2.0×10^4 or more, still more preferably 5.0×10^4 or more, and particularly preferably 8.0×10^4 or more. If M_w /PSSI is less than 1.0×10^4 , fuel efficiency and low temperature startability, namely, viscosity temperature properties and low temperature viscosity properties may be reduced.

[0083] It is preferable that the ratio of the weight average molecular weight to the PSSI in the diesel injector method of the viscosity index improver according to the present embodiment (M_w /PSSI) be 1.0×10^4 or more, and the ratio is more preferably 2.0×10^4 or more, still more preferably 5.0×10^4 or more, and particularly preferably 8.0×10^4 or more. If M_w /PSSI is less than 1.0×10^4 , fuel efficiency and low temperature startability, namely, viscosity temperature properties and low temperature viscosity properties may be reduced.

[0084] It is preferable that the ratio (M_w/M_n) of the weight average molecular weight (M_w) to the number average molecular weight (M_n) of the viscosity index improver according to the present embodiment be 5.0 or less, and the ratio is more preferably 4.0 or less, still more preferably 3.5 or less, particularly preferably 3.0 or less, and most preferably 2.0 or less. It is preferable that M_w/M_n be 1.0 or more, and the ratio is more preferably 1.1 or more, and still more preferably 1.2 or more. If M_w/M_n is 4.0 or more or 1.0 or less, solubility and the effect of improving viscosity temperature properties may be reduced so that sufficient storage stability and fuel efficiency cannot be maintained.

[0085] The hydrocarbon main chain ratio of the viscosity index improver according to the present embodiment is preferably 0.3 or less, more preferably 0.2 or less, more preferably 0.18 or less, still more preferably 0.16 or less, still more preferably 0.14 or less, particularly preferably 0.10 or less, and most preferably 0.05 or less. The hydrocarbon main chain ratio is preferably 0.005 or more, more preferably 0.01 or more, and still more preferably 0.02 or more. If the hydrocarbon main chain ratio exceeds 0.3, shear stability is reduced, and viscosity temperature properties and fuel efficiency may be reduced. If the hydrocarbon main chain ratio is less than 0.01, the solubility in the base oil is reduced, and viscosity temperature properties and fuel efficiency may be reduced.

[0086] The "hydrocarbon main chain ratio" in the present invention means the proportion of the carbon number of the polymethacrylic acid main chain of the total number of carbon atoms of the molecule (the ratio, that is, the carbon number of the poly(meth)acrylic acid main chain/the total number of carbon atoms in the molecule).

[0087] Because usually the poly(meth)acrylate viscosity index improver is a mixture of a plurality of polymers having different structures or molecular weights, the proportion is calculated as an average value of the poly(meth)acrylate

viscosity index improver. If two or more poly(meth)acrylic acid chains are present in the molecule, a longer chain of these poly(meth)acrylic acid chains is the "poly(meth)acrylic acid main chain."

[0088] For the poly(meth)acrylate viscosity index improver having a star structure (structure in which a plurality of arm portions as the poly(meth)acrylic acid chain is connected to a core portion, also referred to as a "star-like structure"), the influences of the core portion is small, and the core portion is excluded from the calculation. Usually the molecular weight of each arm portion is substantially equal, and the weight average molecular weight of the arm portion measured by GPC analysis (standard substance: polystyrene) is used in calculation of the carbon number of the arm portion in the main chain.

[0089] Specifically, first, from the weight average molecular weight measured by GPC analysis (standard substance: polystyrene), the blending ratio of monomers or the weight average molecular weight of the arm portion measured by GPC analysis (standard substance: polystyrene) and the blending ratio of monomers, the average polymerization number (A1) of each monomer in the molecule is calculated. From A1, the total carbon number (B1) and the carbon number (C1) of the polymethacrylic acid main chain in one molecule are calculated, and C1/B1 is calculated. C1/B1 is the hydrocarbon main chain ratio.

[0090] For the poly(meth)acrylate viscosity index improver having a star structure, the number of arm portions (D) defined by the number average molecular weight of the poly(meth)acrylate viscosity index improver/the number average molecular weight of the arm portion is calculated, and C1/(B1 × D) is calculated. C1/(B1 × D) is the hydrocarbon main chain ratio of the poly(meth)acrylate viscosity index improver having a star structure.

[0091] The content of the viscosity index improver according to the present embodiment is preferably 0.1 to 50% by mass, more preferably 0.5 to 40% by mass, still more preferably 1 to 30% by mass, particularly preferably 5 to 20% by mass based on the total amount of the composition. If the content of the viscosity index improver is less than 0.1% by mass, the effect of improving the viscosity index and the effect of reducing the viscosity of a product are reduced, and fuel efficiency may not be improved. At a content more than 50% by mass, the cost of the product is significantly increased, and the viscosity of the base oil needs to be reduced; for this reason, the lubrication performance under severe lubrication conditions (high temperature high shear conditions) may be reduced, causing deficits such as wear, seizure, and fatigue breaking.

[0092] Besides the viscosity index improver according to the present embodiment, the engine oil composition according to the present embodiment may further contain a non-dispersive or dispersive poly(meth)acrylate, a non-dispersive or dispersive ethylene- α -olefin copolymer or a hydride thereof, polyisobutylene or a hydride thereof, a styrene-diene hydrogenation copolymer, a styrene-maleic anhydride ester copolymer, polyalkylstyrene and the like usually used.

[0093] It is preferable that the content of the viscosity index improver in the engine oil composition according to the present embodiment be 0.1 to 50% by mass based on the total amount of the composition, and the content is preferably 0.5 to 20% by mass, more preferably 1.0 to 15% by mass, and still more preferably 1.5 to 12% by mass based on the total amount of the composition. If the content is less than 0.1% by mass, low temperature properties may be insufficient; if the content exceeds 50% by mass, the shear stability of the composition may be reduced.

[0094] The engine oil composition according to the present embodiment contains a friction modifier (B). Thereby, fuel efficiency performance may be increased compared to the case where the present configuration is not included. Examples of the friction modifier (B) include one or more friction modifiers selected from organic molybdenum compounds and ash-free friction modifiers.

[0095] Examples of organic molybdenum compounds used in the present embodiment may include organic molybdenum compounds containing sulfur such as molybdenum dithiophosphate, molybdenum dithiocarbamate (MoDTC); complexes of molybdenum compounds (such as molybdenum oxides such as molybdenum dioxide and molybdenum trioxide; molybdic acids such as orth-molybdic acid, para-molybdic acid, and (poly)molybdic acid sulfide; metal salts thereof; molybdates such as ammonium salts thereof; molybdenum sulfides such as molybdenum disulfide, molybdenum trisulfide, molybdenum pentasulfide, and molybdenum polysulfide; molybdenum sulfide acid and metal salts or amine salts thereof; and molybdenum halides such as molybdenum chloride) with sulfur-containing organic compounds (such as alkyl(thio)xanthate, thiadiazole, mercaptothiadiazole, thiocarbonate, tetrahydrocarbylthiuram disulfide, bis(di(thio)hydracarbonyldithiophosphonate) disulfide, organic

[0096] (poly)sulfides, and sulfurized esters), or other organic compounds; or complexes of sulfur-containing molybdenum compounds such as molybdenum sulfide and molybdenum sulfide acid with alkenyl succinimides.

[0097] For the organic molybdenum compound, organic molybdenum compounds containing no sulfur as a constitutional element may be used. Examples of the organic molybdenum compounds containing no sulfur as a constitutional element specifically include molybdenum-amine complexes, molybdenum-succinimide complexes, molybdenum salts of organic acids, and molybdenum salts of alcohols; among these, molybdenum-amine complexes, molybdenum salts of organic acids, and molybdenum salts of alcohols are preferable.

[0098] In the engine oil composition according to the present embodiment, if the organic molybdenum compound is used, its content is not particularly limited; the content is preferably 0.001% by mass or more, more preferably 0.005% by mass or more, still more preferably 0.01% by mass or more, and particularly preferably 0.03% by mass or more, and

preferably 0.2% by mass or less, more preferably 0.1% by mass or less, still more preferably 0.08% by mass or less, and particularly preferably 0.06% by mass or less based on the total amount of the lubricating oil composition in terms of the molybdenum element. If the content is less than 0.001% by mass, the effect of reducing friction by addition thereof tends to be insufficient, and the fuel efficiency and heat and oxidation stability of the lubricating oil composition tend to be insufficient. If the content exceeds 0.2% by mass, the effect corresponding to the content is not obtained, and the storage stability of the lubricating oil composition tends to be reduced.

[0099] As the ash-free friction modifier, any compound usually used as a friction modifier for a lubricating oil may be used, and examples thereof include compounds having a carbon number of 6 to 50 and containing one or two or more hetero elements selected from an oxygen atom, a nitrogen atom, and a sulfur atom in the molecule. More specifically, examples thereof include ash-free friction modifiers such as amine compounds, fatty acid esters, fatty acid amides, fatty acids, aliphatic alcohols, aliphatic ethers, urea compounds, and hydrazide compounds having at least one alkyl group or alkenyl group having a carbon number of 6 to 30, particularly linear alkyl group having a carbon number of 6 to 30, a linear alkenyl group, a branched alkyl group, and a branched alkenyl group in the molecule.

[0100] The content of the ash-free friction modifier in the engine oil composition according to the present embodiment is preferably 0.01% by mass or more, more preferably 0.1 % by mass or more, and still more preferably 0.3% by mass or more, and preferably 3% by mass or less, more preferably 2% by mass or less, and still more preferably 1% by mass or less based on the total amount of the lubricating oil composition. If the content of the ash-free friction modifier is less than 0.01% by mass, the effect of reducing friction by addition thereof tends to be insufficient; at a content more than 3% by mass, the effect such as anti-wear additives is readily inhibited, or the solubility of the additive tends to be reduced.

[0101] In the present embodiment, it is preferable that the friction modifier (B) be an organic molybdenum friction modifier, it is more preferable that the friction modifier (B) be an organic molybdenum compound containing sulfur, and it is still more preferable that the friction modifier (B) be molybdenum dithiocarbamate.

[0102] To further improve the performance, any additives usually used in the lubricating oil may be contained in the engine oil composition according to the present embodiment according to the purpose. Examples of such additives may include additives such as a metallic detergent besides the first and the second perbasic metal salts, an ash-free dispersant, an anti-wear agent (or extreme-pressure agent), an antioxidant, a corrosion inhibitor, a rust inhibitor, an antiemulsifier, a metal deactivator, and an antifoaming agent.

[0103] If these additives are contained in the engine oil composition according to the present embodiment, it is preferable that the content of each additive be 0.01 to 10% by mass based on the total amount of the lubricating oil composition.

[0104] It is preferable that the kinematic viscosity at 100°C of the engine oil composition according to the present embodiment be 4 to 12 mm²/s, and kinematic viscosity is preferably 9.0 mm²/s or less, more preferably 8.0 mm²/s or less, still more preferably 7.0 mm²/s or less, and particularly preferably 6.8 mm²/s or less. The kinematic viscosity at 100°C of the engine oil composition according to the present embodiment is preferably 4.5 mm²/s or more, more preferably 5.0 mm²/s or more, still more preferably 5.5 mm²/s or more, particularly preferably 6.0 mm²/s or more. The kinematic viscosity at 100°C here designates a kinematic viscosity at 100°C specified in ASTM D-445. If the kinematic viscosity at 100°C is less than 4 mm²/s, insufficient lubrication may be caused; at a kinematic viscosity more than 12 mm²/s, necessary low temperature viscosity and sufficient fuel efficiency performance may not be obtainable.

[0105] It is preferable that the kinematic viscosity at 40°C of the engine oil composition according to the present embodiment be 4 to 50 mm²/s, and the kinematic viscosity is preferably 40 mm²/s or less, more preferably 35 mm²/s or less, particularly preferably 30 mm²/s or less, and most preferably 28 mm²/s or less. The kinematic viscosity at 40°C of the engine oil composition according to the present embodiment is preferably 15 mm²/s or more, more preferably 18 mm²/s or more, still more preferably 20 mm²/s or more, particularly preferably 22 mm²/s or more, and most preferably 25 mm²/s or more. The kinematic viscosity at 40°C here designates a kinematic viscosity at 40°C specified in ASTM D-445. If the kinematic viscosity at 40°C is less than 4 mm²/s, insufficient lubrication may be caused; at a kinematic viscosity more than 50 mm²/s, necessary low temperature viscosity and sufficient fuel efficiency performance may not be obtainable.

[0106] It is preferable that the viscosity index of the engine oil composition according to the present embodiment be in the range of 140 to 400, and the viscosity index is preferably 180 or more, more preferably 190 or more, still more preferably 200 or more, particularly preferably 210 or more, and most preferably 215 or more. If the viscosity index of the lubricating oil composition is less than 140, it may be difficult to improve fuel efficiency while maintaining the HTHS viscosity at 150°C, and to reduce the low temperature viscosity at -35°C. If the viscosity index of the lubricating oil composition exceeds 400 and more, evaporation properties may be reduced, and deficits due to insufficient solubility of the additive and matching properties with a seal material may be caused.

[0107] It is preferable that the HTHS viscosity at 100°C of the engine oil composition according to the present embodiment be 5.5 mPa·s or less, and the HTHS viscosity is more preferably 5.0 mPa·s or less, still more preferably 4.7 mPa·s or less, particularly preferably 4.5 mPa·s or less, and most preferably 4.4 mPa·s or less. The HTHS viscosity is preferably 3.0 mPa·s or more, still more preferably 3.5 mPa·s or more, particularly preferably 4.0 mPa·s or more, and most preferably 4.1 mPa·s or more. The HTHS viscosity at 100°C in the present invention designates a high temperature high shear

viscosity at 100°C specified in ASTM D4683. If the HTHS viscosity at 100°C is less than 3.0 mPa·s, insufficient lubrication may be caused; at an HTHS viscosity more than 5.5 mPa·s, necessary low temperature viscosity and sufficient fuel efficiency performance may not be obtainable.

[0108] The HTHS viscosity at 150°C of the engine oil composition according to the present embodiment is less than 2.6 mPa·s, and the HTHS viscosity is more preferably 2.5 mPa·s or less, still more preferably 2.45 mPa·s or less, and particularly preferably 2.4 mPa·s or less. The HTHS viscosity is preferably 2.0 mPa·s or more, more preferably 2.1 mPa·s or more, still more preferably 2.2 mPa·s or more, and particularly preferably 2.3 mPa·s or more. The HTHS viscosity at 150°C here designates a high temperature high shear viscosity at 150°C specified in ASTM D4683. If the HTHS viscosity at 150°C is less than 2.0 mPa·s, insufficient lubrication may be caused; at an HTHS viscosity more than 2.6 mPa·s, sufficient fuel efficiency performance may not be obtainable.

[0109] It is preferable that the ratio of the HTHS viscosity at 150°C to HTHS viscosity at 100°C of the engine oil composition according to the present embodiment (HTHS viscosity at 150°C/HTHS viscosity at 100°C) be 0.50 or more, and the ratio is more preferably 0.52 or more, still more preferably 0.53, and particularly preferably 0.54 or more. If the ratio is less than 0.50, necessary low temperature viscosity and sufficient fuel efficiency performance may not be obtainable.

[0110] The engine oil composition according to the present embodiment may sufficiently reduce the kinematic viscosity at 40°C, kinematic viscosity at 100°C and HTHS viscosity at 100°C in an engine oil having an HTHS viscosity at 150°C of less than 2.6 mPa·s, may sufficiently suppress an increase in the coefficient of friction in the boundary lubrication region, and has high fuel efficiency. The lubricating oil composition according to the present embodiment having such high properties may be suitably used as energy saving engine oils such as energy saving gasoline engine oils and energy saving diesel engine oils.

Examples

[0111] Hereinafter, the present invention will be more specifically described based on Examples and Comparative Example, but the present invention will not be limited to Examples below.

(Synthesis Example 1: synthesis of Non-dispersive PMA viscosity index improver A-1)

<Synthesis of arm molecule>

[0112] To a 300 ml 5-necked separable flask having an anchor metallic stirring blade (with a vacuum seal), a Dimroth condenser, a 3-necked cock for introducing nitrogen, and a sample introduction port mounted thereon, 25.2 parts by mass of methyl methacrylate, 36.5 parts by mass of methacrylate in which R⁴ in the formula (3) was an alkyl group having a carbon number of 18, and 120 parts by mass of a hydrocarbon solvent (SAE10) as a solvent were placed, and a uniform solution was prepared under stirring. The solution was cooled to 0°C with an ice bath, and vacuum degassing/nitrogen purging of the reaction system was performed 5 times using a diaphragm pump. Under a nitrogen stream, 0.27 parts by mass of azobisisobutyronitrile (AIBN) as a radical initiator, 0.013 parts by mass of 1,4-cyclohexadiene, and 0.11 parts by mass of iodine were charged from the sample introduction port; then, polymerization was performed under a nitrogen atmosphere at a solution temperature of 80°C for 12 hours to prepare an arm molecule solution.

[0113] As a result of GPC analysis (standard substance: polystyrene), the weight average molecular weight of the obtained arm molecule was 87400, the number average molecular weight (Mn) was 62000, and the degree of dispersion (Mw/Mn) was 1.41.

<Synthesis of star polymer>

[0114] To the arm solution, 0.07 parts by mass of azobisisobutyronitrile (AIBN) and 2.14 parts by mass of ethylene glycol dimethacrylate were added, and polymerization was performed under a nitrogen atmosphere at a solution temperature of 80°C for 12 hours to prepare a solution of a target star polymer (hereinafter referred to as "Non-dispersive PMA viscosity index improver A-1").

[0115] As a result of GPC analysis (standard substance: polystyrene), the weight average molecular weight (Mw) of the obtained Non-dispersive PMA viscosity index improver A-1 was 570000, the number average molecular weight (Mn) was 470000, the degree of dispersion (Mw/Mn) was 1.23, PSSI was 3.8, and Mw/PSSI was 1.5×10^5 . The arm conversion rate of the Non-dispersive PMA viscosity index improver A-1 was 64% by mass, the average number of arms was 8, and the hydrocarbon main chain ratio was 0.025.

[0116] Here, the arm conversion rate and the average number of arms are values calculated based on the following expressions, respectively.

arm conversion rate = GPC area of star polymer / (GPC area of
star polymer + GPC area of remaining arm molecule) × 100

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average number of arms = Mn of star polymer / Mn of arm
molecule (rounded to whole numbers)

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[0117] The weight average molecular weight and the number average molecular weight are the weight average molecular weight and the number average molecular weight in terms of polystyrene measured by using an HLC-8220 GPC apparatus made by Tosoh Corporation having 3 TSKgel Super MultiPore HZ-M columns made by Tosoh Corporation (4.6 mm ID × 15 cm) in series and tetrahydrofuran as a solvent at a temperature of 40°C, a flow rate of 0.35 mL/min, a sample concentration of 1% by mass, an amount of sample injection of 5 µL with a detector reference refractive index meter (RI).

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(Synthesis Example 2: synthesis of Non-dispersive PMA viscosity index improver A-2)

[0118] A star polymer (hereinafter referred to as "Non-dispersive PMA viscosity index improver A-2") was synthesized in the same manner as in Synthesis Example 1 except that instead of the arm molecule solution in Synthesis Example 1, an arm molecule solution containing an arm molecule including 70 mol% methyl methacrylate, and 30 mol% methacrylate in which R⁴ in the formula (4) was an alkyl group having 18 carbon atoms (weight average molecular weight: 49600, number average molecular weight (Mn): 40000, degree of dispersion (Mw/Mn): 1.24) was used.

[0119] The Mw of the obtained non-dispersive PMA viscosity index improver A-2 was 370000, Mn was 320000, Mw/Mn was 1.15, PSSI was 4.5, Mw/PSSI was 8.2×10^4 , and the hydrocarbon main chain ratio was 0.025.

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(Synthesis Example 3: synthesis of Non-dispersive PMA viscosity index improver A-3)

[0120] A star polymer (hereinafter referred to as "Non-dispersive PMA viscosity index improver A-2") was synthesized in the same manner as in Synthesis Example 1 except that instead of the arm molecule solution in Synthesis Example 1, an arm molecule solution containing an arm molecule including 70 mol% methyl methacrylate and 30 mol% methacrylate in which R⁴ in the formula (4) was an alkyl group having 18 carbon atoms (weight average molecular weight: 54000, number average molecular weight (Mn): 42000, degree of dispersion (Mw/Mn): 1.29) was used.

[0121] The Mw of Non-dispersive PMA viscosity index improver A-2 obtained was 490000, Mn was 410000, Mw/Mn was 1.19, PSSI was 2.2, Mw/PSSI was 2.2×10^5 , and the hydrocarbon main chain ratio was 0.020.

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(Examples 1 to 5, Comparative Examples 1 to 4)

[0122] In Examples 1 to 5 and Comparative Examples 1 to 5, base oils and additives shown below were used to prepare engine oil compositions having compositions shown in Table 2. The properties of Base oils O-1, O-2, and O-3 are shown in Table 1.

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(Base oils)

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[0123]

O-1 (Base oil 1): mineral oil prepared by hydrocracking /hydrogenation isomerization of a n-paraffin containing oil

O-2 (Base oil 2): hydrocracked mineral oil

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O-3 (Base oil 3): hydrocracked mineral oil

(Additives)

[0124]

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A-1: non-dispersive PMA viscosity index improver prepared in Synthesis Example 1 (copolymer prepared by reacting 70 mol% methyl methacrylate, 30 mol% methacrylate in which R⁴ in the formula (3) was an alkyl group having a carbon number of 18, a small amount of a polymerization initiator, and ethylene glycol dimethacrylate. Mw = 570000,

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Mn = 470000, Mw/Mn = 1.23, PSSI in the ultrasonic shear test = 10.8, Mw/PSSI = 5.3×10^4 , PSSI in the diesel injector method = 3.8, hydrocarbon main chain ratio = 0.025)

A-2: non-dispersive PMA viscosity index improver prepared in Synthesis Example 2 (copolymer prepared by reacting 70 mol% methyl methacrylate, 30 mol% methacrylate in which R⁴ in the formula (3) was an alkyl group having a carbon number of 18, a small amount of a polymerization initiator, and ethylene glycol dimethacrylate. Mw = 370000, Mn = 320000, Mw/Mn = 1.15, PSSI in the ultrasonic shear test = 4.5, Mw/PSSI = 8.2×10^4 , PSSI in the diesel injector method= 2.2, hydrocarbon main chain ratio = 0.025)

A-3: non-dispersive PMA viscosity index improver prepared in Synthesis Example 3 (copolymer prepared by reacting 70 mol% methyl methacrylate, 30 mol% methacrylate in which R⁴ in the formula (3) was an alkyl group having a carbon number of 18, a small amount of a polymerization initiator, and ethylene glycol dimethacrylate. Mw = 490000, Mn = 410000, Mw/Mn = 1.19, PSSI in the ultrasonic shear test = 6.7, Mw/PSSI = 7.3×10^4 , hydrocarbon main chain ratio = 0.020)

a-1: dispersive PMA viscosity index improver (copolymer prepared by reacting 20 mol% methyl methacrylate, 80 mol% methacrylate in which R⁴ in the formula (3) was an alkyl group having a carbon number of 12 to 15, and small amounts of a dispersive group and a polymerization initiator. Mw = 300000, Mn = 70000, Mw/Mn = 4.0, PSSI in the ultrasonic shear test = 43, Mw/PSSI = 7.0×10^3 , hydrocarbon main chain ratio = 0.13)

a-2: dispersive PMA viscosity index improver (copolymer prepared by reacting 20 mol% methyl methacrylate, 80 mol% methacrylate in which R⁴ in the formula (3) was an alkyl group having a carbon number of 12 to 15, and small amounts of a dispersive group and a polymerization initiator. Mw = 80000, Mn = 30000, Mw/Mn = 2.7, PSSI in the ultrasonic shear test = 18, Mw/PSSI = 4.4×10^3 , hydrocarbon main chain ratio = 0.13)

a-3: non-dispersive PMA viscosity index improver (copolymer prepared by reacting 30 mol% methyl methacrylate, 70 mol% methacrylate in which R⁴ in the formula (3) was an alkyl group having a carbon number of 12 to 15, and a small amount of a polymerization initiator. Mw = 400000, Mn = 110000, Mw/Mn = 3.9, PSSI in the ultrasonic shear test = 49, Mw/PSSI = 8.2×10^3 , hydrocarbon main chain ratio = 0.16)

B-1: MoDTC (alkyl group chain length: C8/C13, Mo content: 10 mass%, sulfur content: 11 mass%)

B-2: glycerol monooleate

C-1: other additives (such as a succinimide dispersant, ZnDTP, an antioxidant, an anti-wear agent, a pour-point depressant, and an antifoaming agent).

[Table 1]

		O-1	O-2	O-3
		Base oil 1	Base oil 2	Base oil 3
Density (15°C)	g/cm ³	0.820	0.835	0.8320
Kinematic viscosity (40°C)	mm ² /s	15.8	20.0	13.5
(100°C)	mm ² /s	3.85	4.29	3.27
Viscosity index		141	123	112
Pour point	°C	-22.5	-17.5	-22.5
Aniline point	°C	119	116	109
Iodine number		0.06	0.05	5.38
Sulfur content	massppm	<1	<1	<1
Nitrogen content	massppm	<3	<3	<3
n-d-M analysis	%CP	93.3	80.7	72.6
	%CN	6.7	19.3	23.4
	%CA	0	0	0
Separation by chromatography mass%	Saturate content	99.6	99.7	99.6
	Aromatic content	0.2	0.2	0.3
	Resin content	0.1	0.1	0.1
	Recover rate	99.9	100	100
Paraffin content based on saturate content	mass%	87.1	53.8	50.7
Naphthene content based on saturate content	mass%	12.9	46.2	49.3

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[Evaluation of lubricating oil composition]

[0125] In the engine oil compositions in Examples 1 to 5 and Comparative Examples 1 to 4, the kinematic viscosity at 40°C or 100°C, the viscosity index, the HTHS viscosity at 100°C or 150°C, and the HTHS viscosity after the ultrasonic shear test were measured. The obtained results are shown in Table 2.

(1) Kinematic viscosity: ASTM D-445

(2) Viscosity index: JIS K 2283-1993

(3) HTHS viscosity: ASTM D-4683

(4) Ultrasonic shear test: according to JASO M347-95, an output was adjusted with Standard oil A specified in the ASTM test method, and a shear test was performed at an amplitude of 28 μm , the vibration number of 10 KHz, an irradiation time of 10 minutes, and a sample volume of 60 mL. The PSSI was calculated from $((VI - V2)/V1 \times 100)$ (%) based on the viscosity (V1) at 100°C per amount of the viscosity index improver to be added which was measured before the shear test and the viscosity (V2) at 100°C per amount of the viscosity index improver to be added which was measured after the shear test.

(5) Valve train motoring friction test: using an apparatus that could measure the friction torque of a pair of a cam and a tappet of a valve train in a direct-acting 4-cylinder engine, a friction torque at an oil temperature of 100°C and the number of rotation of 350 rpm was measured. A motoring friction improving rate where Comparative Example 5 was used as a standard oil was calculated.

[Table 2]

Base oil	Based on total amount of base oil		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	% by mass	50									
O-2	Base oil 2	% by mass		50	50	50	50	50	50	50	50	50
O-3	Base oil 3	% by mass	50	50	50	50	50	50	50	50	50	50
	Viscosity of base oil (40°C)	mm ² /s	14.7	16.2	16.2	16.2	16.2	16.2	16.2	16.2	16.2	16.2
	Viscosity of base oil (100°C)		3.6	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7
	Viscosity index of base oil		124	117	117	117	117	117	117	117	117	117
Additives	Based on total amount of composition											
A-1	Viscosity index improver 1	% by mass	9.3	8.5			8.8				11.3	
A-2	Viscosity index improver 2	% by mass			9.7							
A-3	Viscosity index improver 3	% by mass				9.5						
a-1	Viscosity index improver 4	% by mass						5.5				5.5

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(continued)

Base oil	Based on total amount of base oil		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
a-2	Viscosity index improver 5	% by mass							5.9			
a-3	Viscosity index improver 6	% by mass								5.8		
B-1	MoDTC	% by mass	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	0.7	
B-2	Ester	% by mass					0.5					
C-1	Other additives	% by mass	10	10	10	10	10	10	10	10	10	10
Results of evaluation												
Kinematic viscosity		40°C	25.2	26.9	26.7	27.7	26.4	35.9	32.3	35.4	30.4	36.2
		100°C	6.3	6.4	6.2	6.4	6.3	8.5	7.0	8.4	7.4	8.6
Viscosity index			216	202	195	195	203	225	185	226	224	228
HTHS viscosity		100°C	4.30	4.37	4.57	4.49	4.35	5.00	4.95	5.51	4.79	4.97
HTHS viscosity		150°C	2.34	2.35	2.33	2.33	2.35	2.48	2.36	2.50	2.70	2.48
HTHS viscosity (100°C)/HTHS viscosity (150°C)			0.54	0.54	0.51	0.52	0.54	0.50	0.48	0.45	0.56	0.50
HTHS viscosity after ultrasonic shear		150°C	2.30	2.30	2.30	2.30	2.30	2.30	2.30	2.30	-	230

[0126] As shown in Table 2, the engine oil compositions in Examples 1 to 5 including all the components (A) and (B), whose kinematic viscosity at 100°C is 4 to 8 mm²/s, and whose HTHS viscosity at 150°C is adjusted to be less than 2.6 mPa·s show that kinematic viscosity and HTHS viscosity at 100°C are lower and fuel efficiency is higher than those of the engine oil compositions in Comparative Examples 1 to 3 in which HTHS viscosities at 150°C after the ultrasonic shear test are substantially the same and which do not include the component (A) and the engine oil composition whose HTHS viscosity at 150°C is adjusted to be 2.7 mPa·s. The motoring friction improving rate of the engine oil composition in Comparative Example 5 not containing the component (B) is significantly poor.

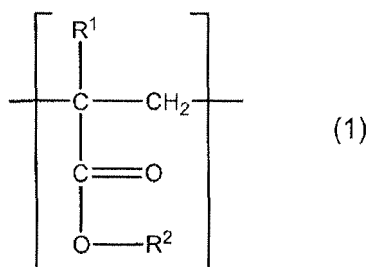
Claims

1. An engine oil composition comprising:

a lubricating base oil having a kinematic viscosity at 100°C of 1 to 5 mm²/s;

a poly(meth)acrylate viscosity index improver containing a structural unit represented by the following formula (1) in a proportion of 30 to 90 mol% and having a PSSI in an ultrasonic shear test of 15 or less; and a friction modifier,

wherein the engine oil composition has a kinematic viscosity at 100°C of 4 to 8 mm²/s, and an HTHS viscosity at 150°C of less than 2.6 mPa·s:



wherein R¹ represents hydrogen or a methyl group; and R² represents a linear or branched hydrocarbon group having a carbon number of 6 or less.

2. The engine oil composition according to claim 1, wherein the viscosity index improver is a viscosity index improver having a PSSI in the ultrasonic shear test of 10 or less and a ratio of a molecular weight to the PSSI (Mw/PSSI) of 1×10^4 or more.

3. The engine oil composition according to claim 1 or 2, wherein the friction modifier is an organic molybdenum friction modifier.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2013/070096

A. CLASSIFICATION OF SUBJECT MATTER

C10M145/14(2006.01)i, C10M161/00(2006.01)i, C10M139/00(2006.01)n,
C10N10/12(2006.01)n, C10N20/02(2006.01)n, C10N20/04(2006.01)n, C10N30/00
(2006.01)n, C10N30/02(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)
C10M101/00-177/00, 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

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X	WO 2009/072524 A1 (Nippon Oil Corp.), 11 June 2009 (11.06.2009), claims; examples; comparative examples & JP 2009-161612 A & JP 2009-167278 A & JP 2009-155639 A & US 2011/0003725 A1 & EP 2241611 A1 & EP 2474601 A1 & EP 2484746 A1 & CN 101883840 A & CN 103013634 A	1-3

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

* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
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"O" document referring to an oral disclosure, use, exhibition or other means	
"P" document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search
17 October, 2013 (17.10.13)

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29 October, 2013 (29.10.13)

Name and mailing address of the ISA/
Japanese Patent Office

Authorized officer

Facsimile No.

Telephone No.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2013/070096

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
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