

(19)



Europäisches Patentamt

European Patent Office

Office européen des brevets



(11)

EP 1 559 772 A1

(12)

EUROPEAN PATENT APPLICATION

published in accordance with Art. 158(3) EPC

(43) Date of publication:

03.08.2005 Bulletin 2005/31

(21) Application number: **03770108.3**

(22) Date of filing: **31.10.2003**

(51) Int Cl.7: **C10M 161/00**

// (C10M161/00, 133:00,
143:00, 145:10, 149:00,
C10N20:02, 20:04, 30:06,
40:04)

(86) International application number:

PCT/JP2003/014032

(87) International publication number:

WO 2004/041977 (21.05.2004 Gazette 2004/21)

(84) Designated Contracting States:

**AT BE BG CH CY CZ DE DK EE ES FI FR GB GR
HU IE IT LI LU MC NL PT RO SE SI SK TR**

Designated Extension States:

AL LT LV MK

(30) Priority: **05.11.2002 JP 2002321754**

(71) Applicant: **Nippon Oil Corporation**
Tokyo 105-8412 (JP)

(72) Inventors:

- **ARIMOTO, Naozumi,**
c/o NIPPON OIL CORPORATION
Yokohama-shi, Kanagawa 231-0815 (JP)
- **OKAWA, Tetsuo,**
c/o NIPPON OIL CORPORATION
Yokohama-shi, Kanagawa 231-0815 (JP)

(74) Representative: **HOFFMANN EITLE**
Patent- und Rechtsanwälte
Arabellastrasse 4
81925 München (DE)

(54) **LUBRICATING OIL**

(57) The present invention provides a lubricating oil composition which has a low viscosity and is capable of maintaining an anti-wear characteristic at the initial use thereof and after the long-term use thereof, in particular, a lubricating oil composition suitable for automatic transmissions or continuously variable transmissions by incorporating, into (A) a lubricant base oil comprising a mineral oil, a synthetic oil or a mixture thereof, (B) a nitrogen-containing compound having at least one alkyl group or alkenyl group having a number-average mo-

lecular weight of 900 or more and/or a derivative thereof in an amount of 0.01 to 0.20% by mass in terms of the content of nitrogen of the total of the composition, and (C) a viscosity index improver having a weight-average molecular weight of 40,000 or less, so as to set the viscosity index of the composition to 160 or more and set the kinematic viscosity of the composition into the range of 20 to 30 mm²/s at 40°C.

EP 1 559 772 A1

Description

Technical Field

[0001] The present invention relates to a lubricating oil composition, specifically, a lubricating oil composition which has a low viscosity and a good anti-wear characteristic, in particular, which is capable of maintaining an anti-wear characteristic for a long term.

Background Art

[0002] In recent years, abatement of emission of carbon dioxide gas has been becoming a pressing need in light of an environmental problem, and an improvement in the fuel efficiency of automobiles has been becoming a more important theme than in the past. It can be considered that the following is effective for means for improving the fuel efficiency of automobiles: a decrease in stirring resistance by making the viscosity of a used lubricating oil low and a decrease in friction in boundary lubricating range by using a friction decreasing agent. This is producing an advantageous effect.

[0003] About low-viscosity lubricating oil for internal combustion engines, suggested is a fuel consumption saving engine oil wherein a viscosity index improver having a high molecular weight is incorporated, or wherein a friction decreasing agent such as an organic molybdenum compound is further used (see, for example, Japanese Patent Application Laid-Open (JP-A) No. 2001-181664). In automatic transmissions composed of a torque converter, a wet clutch, a gear shaft bearing mechanism, an oil pump, and an oil pressure control mechanism, it is necessary to optimize the frictional property of the wet clutch; therefore, it is considered that a friction decreasing agent such as an organic-molybdenum compound is not usually used and it is effective to make the viscosity of the lubricating oil therein low. Thus, an improvement in fuel efficiency is expected by decreasing the stirring resistance of the torque converter or the oil pump. However, the decrease in the viscosity of the lubricating oil directly causes a deterioration in the anti-wear characteristic thereof. It is therefore difficult to make the improvement in fuel efficiency by the decrease in the viscosity and the anti-wear characteristic compatible. Thus, the anti-wear characteristic is necessarily given priority.

[0004] As a conventional oil for automobile transmissions which is capable of maintaining various performances, such as transmission driving property, for a long term, disclosed is an oil wherein one or more lubricant base oils of a synthetic oil type and/or mineral oil type, an anti-wear agent, an extreme-pressure agent, a metal detergent, an ashless dispersant, a friction modifier and others are optimally blended (see, for example, JP-A Nos. 3-39399, 7-268375, and 2000-63869). A viscosity index improver is generally selected from improvers having an average molecular weight of 1,000 to 1,000,000. As a lubricating oil into which a viscosity index improver having a low molecular weight is incorporated, there is disclosed, for example, an automatic transmission oil composition using the following together: 1 to 15% by weight of a monoolefin polymer or copolymer having a number-average molecular weight of 1,000 to 10,000, and 1 to 5% by weight of a copolymer of an ester made from a saturated aliphatic monohydric alcohol having an average molecular weight of 5,000 to 50,000 and methacrylic acid (see JP-A No. 61-9497); a semi-synthetic automatic transmission oil composition comprising 3 to 5% by weight of an ethylene/ α -olefin copolymer having a number-average molecular weight of 26,000 to 40,000 (JP-A No. 1-168798); or a semi-synthetic automatic transmission oil composition comprising 27 to 34% by weight of a polymethacrylate having a number-average molecular weight of 19,000 to 23,000 (see JP-A No. 1-271494); or the like.

[0005] However, none of these compositions are compositions for improving fuel efficiency. Accordingly, the kinematic viscosity thereof is high and investigation is not made at all about effect on the anti-wear characteristic of the compositions at the initial use stage thereof or after long-term use thereof, the effect being the most important theme in the case that the viscosity of the lubricating oil therein is made low. Additionally, the optimization of a viscosity index improver and other additives therein is not sufficiently investigated. Furthermore, the kinematic viscosity of automatic transmission oils which are commercially available at present is generally in the range of 30 to 50 mm²/s at 40°C. It is therefore considered that there is not substantially any low-viscosity transmission oil wherein a fuel efficiency improvement and an anti-wear characteristic are compatible.

Disclosure of the Invention

[0006] Thus, the present invention has been made in light of such a situation, and an object of the present invention is to provide a lubricating oil composition having a low viscosity and further having a sufficient anti-wear characteristic at the initial use stage thereof and after long-term use thereof, in particular, a low-viscosity lubricating oil composition suitable for automatic transmissions or continuously variable transmissions.

[0007] The inventors have made eager investigations for solving the above-mentioned problems to find out that the problems can be solved by incorporating, into a lubricant base oil, a specific nitrogen-containing compound and a

viscosity index improver having a specific molecular weight optimally. Thus, the present invention has been made.

[0008] Accordingly, the present invention is a lubricating oil composition which is obtained by incorporating, into (A) a lubricant base oil comprising a mineral oil, a synthetic oil or a mixture thereof, (B) a nitrogen-containing compound having at least one alkyl group or alkenyl group having a number-average molecular weight of 900 or more and/or a derivative thereof in an amount of 0.01 to 0.20% by mass in terms of the content of nitrogen of the total of the composition, and (C) a viscosity index improver having a weight-average molecular weight of 40, 000 or less, so as to set the viscosity index of the composition to 160 or more and set the kinematic viscosity of the composition into the range of 20 to 30 mm²/s at 40°C.

[0009] It is preferable that the (B) component is a nitrogen-containing compound having two alkyl groups or alkenyl groups having a number-average molecular weight of 1200 or more and/or a derivative thereof.

[0010] It is also preferable that the lubricating oil composition of the invention essentially comprises, as the (B) component, a boron-modified compound of a nitrogen-containing compound having at least one alkyl group or alkenyl group having a number-average molecular weight of 900 or more in an amount of 0.002% or more by mass in terms of the content of boron of the total of the composition.

[0011] It is also preferable that the boron-modified compound is a boron-modified compound of a nitrogen-containing compound having at least one alkyl group or alkenyl group having a number-average molecular weight of 1200 or more.

[0012] It is also preferable that the boron-modified compound is a boron-modified compound of a nitrogen-containing compound having two alkyl groups or alkenyl groups having a number-average molecular weight of 1200 or more.

[0013] It is also preferable that the boron-modified compound is a boron-modified compound of a nitrogen-containing compound having two alkyl groups or alkenyl groups having a number-average molecular weight of 1700 or more.

[0014] It is also preferable that the ratio by mass of boron to nitrogen (B/N ratio) in the boron-modified compound is from 0.01 to 3.

[0015] It is also preferable that the (B) component comprises a boron-modified compound of a bis type succinimide having a poly (iso) butenyl group having a number-average molecular weight of 2000 or more.

[0016] It is also preferable that the (B) component comprises both of a bis type succinimide having a poly (iso) butenyl group having a number-average molecular weight of 900 or more and less than 2000 and boron-modified compounds a bis type succinimide having a poly(iso)butenyl group having a number-average molecular weight of 2000 or more.

[0017] It is also preferable that the lubricating oil composition of the invention is used in an automatic transmission or a continuously variable transmission.

[0018] The invention is also a method for maintaining the anti-wear characteristic of an automatic transmission or a continuously variable transmission by use of the above-mentioned low viscosity lubricating oil composition.

Best Modes for Carrying Out the Invention

[0019] The present invention is described in detail hereinafter.

[0020] The (A) component in the invention is a lubricant base oil comprising a mineral oil, a synthetic oil, or a mixture thereof, and is not particularly limited. Any oil that is usually used as a base oil of a lubricating oil composition can be used whether the oil is a mineral oil or a synthetic oil.

[0021] Examples of the mineral oil include paraffin type and naphthene type mineral lubricant base oils obtained by refining lubricating oil fractions yielded by distilling crude oil under normal pressure or under reduced pressure by an appropriate combination from solvent deasphalting, solvent extraction, hydrocracking, solvent dewaxing, catalytic dewaxing, hydrotreating, sulfuric acid washing, clay treatment and other refining treatments; and normal paraffins.

[0022] Examples of the synthetic base oil include poly- α -olefins (such as polybutene, 1-octene oligomer, 1-decene oligomer, and ethylene/propylene oligomer) and hydrogenated products thereof, isobutene oligomer and hydrogenated products thereof, iso-paraffin, alkylbenzene, alkylnaphthalene, diesters (such as ditridecyl glutarate, di-2-ethylhexyl adipate, diisodecyl adipate, ditridecyl adipate and di-2-ethylhexyl sebacate), polyol esters (such as trimethylolpropane caprilate, trimethylolpropane pelargonate, pentaerythritol 2-ethyl hexanoate, and pentaerythritol pelargonate), polyoxyalkylene glycol, dialkyldiphenyl ether, and polyphenyl ether.

[0023] The lubricant base oil of the invention may be a mixture of two or more mineral base oils or two or more synthetic base oils, or a mixture of one or more mineral base oils and one or more synthetic base oils. The blend ratio between the two or more base oils in the mixture can be selected at will.

[0024] About these lubricant base oils in the invention, the kinematic viscosity thereof is not particularly limited, and is desirably as follows: the lower limit of the kinematic viscosity is 1 mm²/s, preferably 2 mm²/s at 100°C, and the upper limit of the kinematic viscosity is 5 mm²/s, preferably 4 mm²/s, particularly preferably 3.5mm²/s. When the kinematic of the lubricant base oil is set to 1 mm²/s or more at 100°C, it is possible to obtain a lubricating oil composition which has a sufficient oil-film forming power and a good lubricity and is smaller in evaporation loss of the base oil at high temperature conditions. On the other hand, when the kinematic viscosity is set to 5 mm²/s or less at 100°C, it becomes possible to obtain a lubricating oil composition having a smaller friction resistance at lubrication spots since the fluid

resistance becomes small.

[0025] About these lubricant base oils in the invention, the viscosity index thereof is not particularly limited. It is desired that the viscosity index is 80 or more, preferably 90 or more, particularly preferably 110 or more. When the viscosity index is set to 80 or more, a composition good in wear prevention after the use thereof for a long term can be obtained.

[0026] The (B) component of the invention correspond to nitrogen-containing compounds having at least one alkyl group or alkenyl group having a number-average molecular weight of 900 or more and/or derivatives thereof. One or more selected therefrom at will can be incorporated.

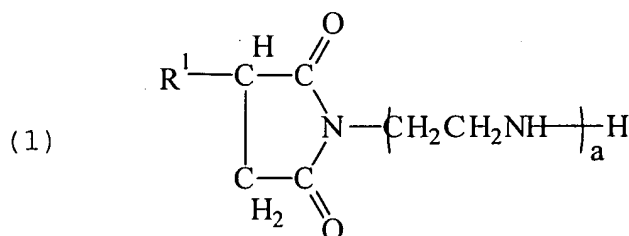
[0027] Specifically, an example of the (B) component may be one or more compounds selected from:

(B-1) a succinimide having at least one alkyl or alkenyl group having a number-average molecular weight of 900 or more, or a derivative thereof;

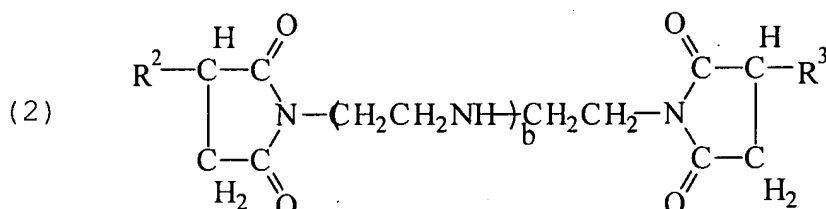
(B-2) a benzyl amine having at least one alkyl or alkenyl group having a number-average molecular weight of 900 or more, or a derivative thereof; and

(B-3) a polyamine having at least one alkyl or alkenyl group having a number-average molecular weight of 900 or more, or a derivative thereof.

[0028] More specific examples of the (B-1) succinimide include compounds represented by the following formula (1) or (2):



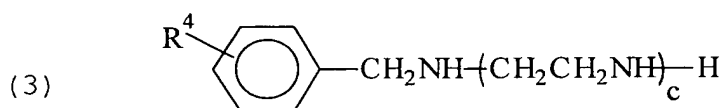
wherein R¹ represents an alkyl or alkenyl group having a number-average molecular weight of 900 or more, and a represents an integer of 1 to 5, preferably 2 to 4, and



wherein R² and R³ each independently represent an alkyl or alkenyl group having a number-average molecular weight of 900 or more, and b represents an integer of 0 to 4, preferably 1 to 3.

[0029] Succinimide is classified into the so-called mono type succinimide as represented by the formula (1), wherein succinic anhydride is added to one end of a polyamine at the time of imidation, and the so-called bis type succinimide as represented by the formula (2), wherein succinic anhydrides are added to both ends of a polyamine. As the (B-1) component, both thereof can be used. Preferable is the bis type succinimide, that is, succinimide having two alkyl or alkenyl groups having a number-average molecular weight of 900 or more since the composition easily maintains an anti-wear characteristic at the initial use thereof and after the long-term use thereof.

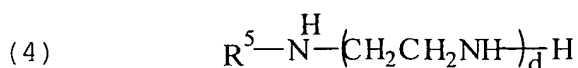
[0030] More specific examples of the (B-2) benzylamine include compounds represented by the following formula (3):



wherein R⁴ represents an alkyl or alkenyl group having a number-average molecular weight of 900 or more, and c represents an integer of 1 to 5, preferably 2 to 4.

[0031] The process for producing this benzylamine is never limited. For example, the benzylamine can be obtained by causing a polyolefin such as propylene oligomer, polybutene, or ethylene/ α -olefin copolymer to react with phenol to prepare an alkylphenol, and then causing this to react with formaldehyde and a polyamine such as diethylenetriamine, triethylenetetramine, tetraethylenepentamine or pentaethylenehexamine by Mannich reaction.

[0032] More specific examples of the (B-3) polyamine include compounds represented by the following formula (4):



wherein R⁵ represents an alkyl or alkenyl group having a number-average molecular weight of 900 or more, and d represents an integer of 1 to 5, preferably 2 to 4.

[0033] The process for producing this polyamine is never limited. For example, the polyamine can be obtained by chlorinating a polyolefin such as propylene oligomer, polybutene, or ethylene/ α -olefin copolymer and then causing this to react with ammonia or a polyamine such as ethylenediamine, diethylenetriamine, triethylenetetramine, tetraethylenepentamine or pentaethylenehexamine.

[0034] Specific examples of a derivative of a nitrogen-containing compound such as a succinimide, benzylamine or polyamine as described above include the so-called acid-modified compounds obtained by causing a monocarboxylic acid (such as aliphatic acid) having 2 to 30 carbon atoms, oxalic acid, a polycarboxylic acid having 2 to 30 carbon atoms such as phthalic acid, trimellitic acid or pyromellitic acid, or an anhydride or ester compound thereof, an alkylene oxide having 2 to 6 carbon atoms, a hydroxy(poly)oxyalkylene carbonate, or some other compound to act on a nitrogen-containing compound as described above, and then neutralizing or amidating a part or the whole of remaining amino groups and/or imino groups; the so-called boron-modified compounds obtained by causing a boron compound such as boric acid, a boric acid salt or a boric acid ester to act on a nitrogen-containing compound as described above, and then neutralizing or amidating a part or the whole of remaining amino groups and/or imino groups; sulfur-modified compounds obtained by causing a sulfur compound to act on a nitrogen-containing compound as described above; and modified compounds wherein a nitrogen-containing compound as described above is combined with two or more kinds of modifications selected from acid modification, boron modification and sulfur modification.

[0035] The alkyl or alkenyl group having a number-average molecular weight of 900 or more in the (B) component of the invention may be in a linear chain form or a branched chain form. Specific and preferable examples thereof include branched alkyl and branched alkenyl groups derived from oligomer of an olefin such as propylene, 1-butene or isobutylene, and from cooligomer of ethylene and propylene. Preferable is a poly(iso)butenyl group. The number-average molecular weight thereof is preferably 1200 or more, more preferably 1700 or more, even more preferably 2000 or more, and particularly preferably 2300 or more. About the number-average molecular weight, the upper limit thereof is not particularly limited, and is preferably 5000 or less, more preferably 3500 or less, and particularly preferably 3000 or less. When the number-average molecular weight of the alkyl or alkenyl group is set to 900 or more, the composition easily maintains an anti-wear characteristic after the long-term use thereof. When the number-average molecular weight is set to 5000 or less, a composition good in low-temperature fluidity can be obtained.

[0036] The nitrogen content in the nitrogen-containing compound of the (B) component is arbitrary. Usually, the component having a nitrogen content by percentage of 0.01 to 10% by mass, preferably 0.1 to 10% by mass, is desirably used from the viewpoints of friction resistance, oxidation stability, frictional property and others.

[0037] An alkyl or alkenylsuccinimide having a number-average molecular weight as described above can be more preferably used as the (B) component in the lubricating oil composition of the invention since the anti-wear characteristic is easily maintained. It is particularly preferable to incorporate, as an essential component, a boron-modified compound of an alkyl or alkenylsuccinimide having a number-average molecular weight as described above, in particular, a number-average molecular weight of 2000 or more since the composition is best in the anti-wear characteristic at the initial use thereof and after the long-term use thereof.

[0038] The ratio by mass of boron to nitrogen (B/N ratio) in the boron-modified compound of the nitrogen-containing compound is not particularly limited. The ratio is usually from 0.01 to 3, preferably 0.05 or more, more preferably 0.1 or more, and even more preferably 0.2 or more, and is preferably 1 or less, more preferably 0.8 or less, and even more preferably 0.6 or less. When the B/N ratio of the boron-modified compound is selected in the above-mentioned range, a composition good in the anti-wear characteristic at the initial use thereof and after the long-term use thereof can be obtained. When the above-mentioned nitrogen-containing compound is used together with the boron-modified compound, for example, when a bis type succinimide having a poly(iso)butenyl group having a number-average molecular weight of 900 or more and less than 2000 is used together with a bis type succinimide having a poly(iso)butenyl group

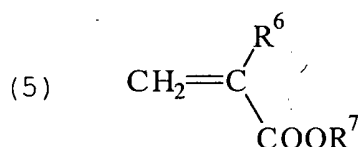
having a number-average molecular weight of 2000 or more, the anti-wear characteristic at the initial use and after the long-term use can be synergistically improved. Even in this case, it is preferable to set the ratio by mass of boron resulting from the boron-modified compound to the whole of nitrogen resulting from the (B) component into the above-mentioned range.

[0039] The lower limit of the content of the (B) component in the lubricating oil composition of the invention is 0.01% by mass, preferably 0.02% by mass of the total of the lubricating oil composition, this limit being a limit in terms of the content of nitrogen. On the other hand, the upper limit of the content thereof in terms of the content of nitrogen is 0.2% by mass, preferably 0.18% by mass of the total of the lubricating oil composition. If the content of the (B) component in terms of the nitrogen content is less than 0.01% by mass of the total of the lubricating oil composition, the anti-wear characteristic after the long-term use, on the basis of the incorporation of the (B) component, is not easily maintained. If the content is more than 0.2% by mass, the low-temperature fluidity of the lubricating oil composition deteriorates. Thus, these cases are each not preferred. In the case that the (B) component is a nitrogen-containing compound having two alkyl or alkenyl groups having a number-average molecular weight of 1200 or more, the upper limit of the content thereof can be set to 0.07% or less by mass also. Furthermore, in the case that the (B) component is a nitrogen-containing compound having two alkyl or alkenyl groups having a number-average molecular weight of 2000 or more, a sufficient anti-wear characteristic can be exhibited even if the upper limit is set to 0.04% or less by mass.

[0040] In the case that a boron-modified compound of the nitrogen-containing compound, as the (B) component, is incorporated as an essential component, the lower limit of the content thereof in terms of the amount of boron is preferably 0.002% or more by mass, more preferably 0.004% or more by mass, and particularly preferably 0.008% or more. The upper limit of the content thereof in terms of the boron amount is preferably 0.05% or less by mass, more preferably 0.02% or less by mass, and particularly preferably 0.015% or less by mass. When the amount of the boron-modified compound as the (B) component in terms of the boron amount is set into the above-mentioned preferred range, the composition can maintain a better anti-wear characteristic at the initial use and after the long-term use.

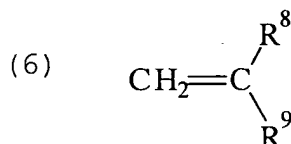
[0041] The (C) component in the invention is a viscosity index improver having a weight-average molecular weight of 40,000 or less, and may be specifically a non-dispersion type viscosity index improver and/or dispersion type viscosity index improver having a weight-average molecular weight of 40,000 or less.

[0042] Specific examples of the non-dispersion type viscosity index improver include copolymers of one or more monomers (C-1) selected from compounds represented by formulae (5), (6) and (7) illustrated below, or hydrogenated products thereof. Specific examples of the dispersion type viscosity index improver include copolymers of two or more monomers selected from compounds represented by general formulae (8) and (9), or compounds wherein oxygen-containing groups are introduced into hydrogenated products of the copolymers; and copolymers of one or more monomers (C-1) selected from compounds represented by the general formulae (5) to (7) and one or more monomers (C-2) selected from compounds represented by the general formulae (8) and (9), or hydrogenated products thereof.



wherein R^6 represents hydrogen or a methyl group, and R^7 represents an alkyl group having 1 to 18 carbon atoms.

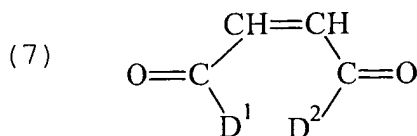
[0043] Specific examples of the alkyl group represented by R^7 , which has 1 to 18 carbon atoms, include methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, and octadecyl groups, (which may be in a linear chain form or a branched chain form).



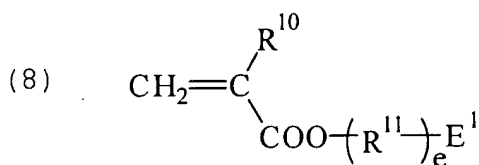
wherein R^8 represents hydrogen or a methyl group, and R^9 represents a hydrocarbon group having 1 to 12 carbon atoms.

[0044] Specific examples of the hydrocarbon group represented by R^9 , which has 1 to 12 carbon atoms, include alkyl groups, such as methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, octyl, nonyl, decyl, undecyl, and dodecyl groups, (which may be in a linear chain form or a branched chain form); alkenyl groups, such as butenyl, pentenyl, hexenyl,

heptenyl, octenyl, nonenyl, decenyl, undecenyl, and dodecenyl groups, (which may be in a linear chain form or a branched chain form and may have a double bond at any position); cycloalkyl groups having 5 to 7 carbon atoms, such as cyclopentyl, cyclohexyl and cycloheptyl groups; alkylcycloalkyl groups having 6 to 11 carbon atoms, such as methylcyclopentyl, dimethylcyclopentyl, methylethylcyclopentyl, diethylcyclopentyl, methylcyclohexyl, dimethylcyclohexyl, methylethylcyclohexyl, diethylcyclohexyl, methylcycloheptyl, dimethylcycloheptyl, methylethylcycloheptyl, and diethylcycloheptyl, (these alkyl groups being allowable to be substituted onto any position of the respective cycloalkyl groups); aryl groups, such as phenyl and naphthyl groups; alkylaryl groups having 7 to 12 carbon atoms, such as tolyl, xylyl, ethylphenyl, propylphenyl, butylphenyl, pentylphenyl, and hexylphenyl (these alkyl groups being allowable to be in a linear chain form or a branched chain form and be substituted onto any position of the respective aryl groups); phenylalkyl groups having 7 to 12 carbon atoms, such as benzyl, phenylethyl, phenylpropyl, phenylbutyl, phenylpentyl, and phenylhexyl groups, (which may be in a linear chain form or a branched chain form).



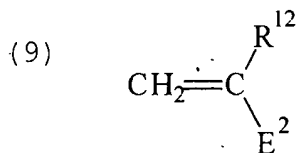
wherein D¹ and D² each independently represent a hydrogen atom, a residue of an alkylalcohol having 1 to 18 carbon atoms (-OR wherein R is an alkyl group having 1 to 18 carbon atoms), or a residue of a monoalkylamine having 1 to 18 carbon atoms (-NHR wherein R is an alkyl group having 1 to 18 carbon atoms).



wherein R¹⁰ represents a hydrogen atom or a methyl group, R¹¹ represents an alkylene group having 1 to 18 carbon atoms, E¹ represents an amine residue or heterocyclic residue containing 1 or 2 nitrogen atoms or 0 to 2 oxygen atoms, and e is an integer of 0 or 1.

[0045] Specific examples of the alkylene group represented by R¹¹, which has 1 to 18 carbon atoms, include ethylene, propylene, butylene, pentylene, hexylene, heptylene, octylene, nonylene, decylene, undecylene, dodecylene, tridecylene, tetradecylene, pentadecylene, hexadecylene, heptadecylene, and octadecylene group, (which may be in a linear chain form or a branched chain form).

[0046] Specific examples of the group which represents E¹ include dimethylamino, diethylamino, dipropylamino, dibutylamino, anilino, toluidino, xylidino, acetylamino, benzoylamino, morpholino, pyrrolyl, pyrrolino, pyridyl, methylpyridyl, pyrrolidinyl, piperidinyl, quinolyl, pyrrolidonyl, pyrrolidono, imidazolyno, and pyrazino groups.



wherein R¹² represents a hydrogen atom or a methyl group, and E² represents an amine residue or heterocyclic residue containing 1 or 2 nitrogen atoms and 0 to 2 oxygen atoms.

[0047] Specific examples of the group which represents E² include dimethylamino, diethylamino, dipropylamino, dibutylamino, anilino, toluidino, xylidino, acetylamino, benzoylamino, morpholino, pyrrolyl, pyrrolino, pyridyl, methylpyridyl, pyrrolidinyl, piperidinyl, quinolyl, pyrrolidonyl, pyrrolidono, imidazolyno, and pyrazino groups.

[0048] Specific and preferable examples of the monomer of the (C-1) component include alkyl acrylates having 1 to 18 carbon atoms, alkyl methacrylates having 1 to 18 carbon atoms, olefins having 2 to 20 carbon atoms, styrene, methylstyrene, anhydrous maleic acid esters, and anhydrous maleic amide, and mixtures thereof.

[0049] Specific and preferable examples of the monomer of the (C-2) component include dimethylaminomethyl meth-

acrylate, diethylaminomethyl methacrylate, dimethylaminoethyl methacrylate, diethylaminoethyl methacrylate, 2-methyl-5-vinylpyridine, morpholinomethyl methacrylate, morpholinoethyl methacrylate, N-vinylpyrrolidone and mixtures thereof.

[0050] When one or more monomers selected from the (C-1) compounds are copolymerized with one or more monomers selected from the (C-2) compounds, the mole ratio of the (C-1) component(s) to the (C-2) component(s) is arbitrary, and is generally from about 80/20 to about 95/5. The reaction process for the copolymerization is arbitrary. Usually, a copolymer is easily obtained by subjecting the (C-1) component(s) and the (C-2) component(s) to radical solvent polymerization in the presence of a polymerization initiator such as benzoylperoxide.

[0051] Specific examples of the (C) component include non-dispersion type and dispersion type polymethacrylates, non-dispersion type and dispersion type ethylene/ α -olefin copolymers and hydrogenated products thereof, polyisobutylene and hydrogenated products thereof, styrene/diene hydrogenated copolymers, styrene/anhydrous maleic acid ester copolymers, and polyalkylstyrenes.

[0052] About the molecular weight of the (C) component in the invention, the weight-average molecular weight is desirably 40,000 or less, more preferably 35,000 or less. If the weight-average molecular weight is more than 40,000, the anti-wear characteristic after the long-term use is unfavorably poor. The weight-average molecular weight of the (C) component is not particularly limited, and a substance having a weight-average molecular weight of 900 or more can be used. The weight-average molecular weight is preferably 10,000 or more, more preferably 20,000 or more. It is advisable to set the weight-average molecular weight of the (C) component to 10,000 or more for the following reason: a viscosity index improver of the (C) component having a weight-average molecular weight of less than 10,000, for example, polyisobutylene having a weight-average molecular weight of 900 or more and less than 10,000 is small in viscosity-index-improving effect; thus, the improver needs to be incorporated in a large amount, and the anti-wear characteristic after the long-term use is not easily maintained.

[0053] It is particularly preferable that the (C) component in the invention is a non-dispersion type polymethacrylate having a weight-average molecular weight of 10,000 to 40,000 since the composition easily maintains an anti-wear characteristic after the long-term use.

[0054] The content of the (C) component in the lubricating oil composition of the invention is desirably set so as to fix the viscosity index of the composition into 160 or more, preferably 165 or more. The content of the (C) component is desirably set so as to fix the upper limit of the viscosity index of the composition preferably into less than 210, more preferably into less than 190. It is necessary to incorporate the (C) component in such an amount that the kinematic viscosity of the composition becomes 20 to 30 mm²/s at 40°C. Preferably, the (C) component is incorporated in such an amount that the viscosity becomes 22 to 28 mm²/s. One or more compounds selected at will from viscosity index improvers as described above can be incorporated in any amount into the present invention so as to set the viscosity index of the composition and the kinematic viscosity thereof at 40°C into the above-defined ranges. For example, the content thereof by percentage is 1% or more by mass, preferably 5% or more by mass, and particularly preferably 6% or more by mass, and is 20% or less by mass, preferably 15% or less by mass, and particularly preferably 12% or less by mass. If the viscosity index of the composition is less than 160, the anti-wear characteristic after the long-term use unfavorably deteriorates. If the kinematic viscosity at 40°C is less than the above-mentioned range, the anti-wear characteristic at the initial use and after the long-term use unfavorably deteriorates. If the kinematic viscosity at 40°C is more than the above-mentioned range, the composition does not favorably gain fuel consumption saving performance, based on a decrease in the stirring resistance, with ease.

[0055] In the invention, the specified amount of the (B) component is incorporated into the (A) component as a base oil, and further the (C) component is incorporated therein so as to set the viscosity index of the composition to 160 or more and set the kinematic viscosity of the composition into the range of 20 to 30 mm²/s at 40°C, thereby yielding a lubricating oil composition having a low viscosity. Only in this way, the resultant lubricating oil composition is good in the anti-wear characteristic at the initial use and after the long-term use thereof. In order to make the performance high, other kinds of additives may be added thereto if necessary. Examples of such additives include an anti-wear agent or extreme-pressure agent, a metal detergent, a friction modifier, a rust inhibitor, a corrosion inhibitor, a pour point depressant, a rubber swelling agent, an antifoamer, and a colorant. These compounds may be used alone or in combination of two or more thereof.

[0056] The anti-wear agent or extreme-pressure agent that can be used together in the lubricating oil composition of the invention may be any compound that is usually used as an anti-wear agent or extreme-pressure agent for lubricating oil, such as a sulfur-based additive, phosphorus-based additive, and sulfur- and phosphorus-based additive. For example, the following can be used: sulfur-based compounds such as disulfides, olefin sulfides and oil and fat sulfides; phosphorus-based compounds such as phosphoric acid monoesters, phosphoric acid diesters, phosphoric acid triesters, phosphorous acid monoesters, phosphorous acid diesters, phosphorous acid triesters, and salts of these esters and amines or alkanolamines; sulfur- and phosphorus-based compounds such as zinc dithiophosphate and thiophosphoric acid esters. The content of these anti-wear agents or extreme-pressure agents is not particularly limited, and is usually from 0.01 to 5.0% by mass of the total of the lubricating oil composition.

[0057] The metal detergent that can be used together in the lubricating oil composition of the invention may be any compound that is usually used as a metal detergent for lubricating oil. For example, the following can be used alone or in combination of two or more thereof in the composition of the invention: sulfonates, phenates, salicylates, and naphthenates of alkali metal or alkaline earth metal. Examples of the alkali metal include sodium, and potassium, and examples of the alkaline earth metal include calcium, and magnesium. Specifically, a sulfonate, phenate or salicylate of calcium or magnesium is preferably used as the metal detergent. The total base number of these metal detergents and the content thereof can be selected at will in accordance with required lubricating oil performance. Usually, the total base number is from 0 to 500 mgKOH, which is according to the perchloric acid process, and the content is from 0.01 to 10% by mass.

[0058] The friction modifier that can be used together in the lubricating oil composition of the invention may be any compound that is usually used as a friction modifier for lubricating oil. Examples thereof include amine compounds, aliphatic acid esters, aliphatic acid amides, and aliphatic acid metal salts each of which has in the molecule thereof an alkyl or alkenyl group having 6 to 30 carbon atoms, in particular, a linear alkyl group or linear alkenyl group having 6 to 30 carbon atoms.

[0059] Example of the amine compounds are linear or branched, preferably linear, aliphatic monoamines having 6 to 30 carbon atoms; linear or branched, preferably linear, aliphatic polyamines; or alkyleneoxide adducts of these aliphatic amines. Examples of the aliphatic acid esters are esters of linear or branched, preferably linear, aliphatic acid having 7 to 31 carbon atoms and aliphatic monohydric alcohol or aliphatic polyhydric alcohol. Examples of the aliphatic acid amides are amides of linear or branched, preferably linear, aliphatic acid having 7 to 31 carbon atoms and aliphatic monoamine or aliphatic polyamine. Examples of the aliphatic acid metal salts are alkaline earth metal salts (such as magnesium salt and calcium salts) or zinc salts of linear or branched, preferably linear, aliphatic acid having 7 to 31 carbon atoms.

[0060] One or more compounds selected at will from these friction modifiers can be incorporated in any amount into the invention. Usually, it is desired that the content thereof is from 0.01 to 5.0% by mass, preferably from 0.03 to 3.0% by mass of the total of the lubricating oil composition.

[0061] The antioxidant that can be used together in the lubricating oil composition of the invention may be any material that is generally used in lubricating oil, such as a phenol type compound or amine type compound.

[0062] Specific examples thereof include alkylphenols such as 2,6-di-tert-butyl-4-methylphenol; bisphenols such as methylene-4,4-bisphenol(2,6-di-tert-butyl-4-methylphenol); naphthylamines such as phenyl- α -naphthylamine; dialkyldiphenylamines; zinc dialkyldithiophosphates such as zinc di-2-ethylhexyldithiophosphate; and esters of (3,5-di-tert-butyl-4-hydroxyphenyl) or (3-methyl-5-tert-butyl-4-hydroxyphenyl)aliphatic acid (such as propionic acid) and a monohydric or polyhydric alcohol, such as methanol, octanol, octadecanol, 1,6 hexadiol, neopentylglycol, thiodiethylene glycol, triethylene glycol, and pentaerythritol.

[0063] One or more compounds selected at will therefrom can be incorporated in any amount. Usually, it is desired that the content thereof is from 0.01 to 5.0% by mass of the total of the lubricating oil composition.

[0064] The corrosion inhibitor that can be used together in the lubricating oil composition of the invention may be any compound that is usually used as a corrosion inhibitor for lubricating oil. Examples thereof include benzotriazole type, tolyltriazole type, thiadiazole type, and imidazole type compounds. One or more compounds selected at will therefrom can be incorporated in any amount. Usually, it is desired that the content thereof is from 0.01 to 3.0% by mass of the total of the lubricating oil composition.

[0065] The antifoamer that can be used together in the lubricating oil composition of the invention may be any compound that is usually used as an antifoamer for lubricating oil. Examples thereof include silicones such as dimethylsilicone and fluorosilicone. One or more compounds selected at will therefrom can be incorporated in any amount. Usually, it is desired that the content thereof is from 0.001 to 0.05% by mass of the total of the lubricating oil composition.

[0066] The lubricating oil composition of the invention is good in the anti-wear characteristic at the initial use and after the long-term use thereof, and makes it possible to decrease stirring resistance resulting from the lubricating oil; therefore, when the composition is used as a lubricating oil for automobiles, for example, for internal combustion engines or transmissions, in particular, automatic transmissions or continuously variable transmissions, the composition can contribute to an improvement in the fuel efficiency of the automobiles.

Examples

[0067] The details of the present invention are more specifically described by way of the following examples and comparative examples. However, the invention is not limited at all thereby.

(Examples 1 to 5 and Comparative Examples 1 to 3)

[0068] Lubricating oil compositions according to the invention (Examples 1 to 5 in Table 1) and lubricating oil com-

positions for comparison (Comparative Examples 1 to 3 in Table 2) were prepared by blending various kinds of lubricant base oils and additives shown in Table 1 or 2. The added amount of each of the additives is an amount on the basis of the total amount of the composition.

[0069] About each of the resultant compositions, the anti-wear characteristics at the initial use and after the long-term use were evaluated in an abrasion test in item (1) described below. For the evaluation of the initial anti-wear characteristic, the oil which was in a new state was used, and for the evaluation of the anti-wear characteristic after the long-term use, the oil deteriorated beforehand by carrying out an ultrasonic shearing test described in item (2) was used. Results of the performance evaluations also are each described in Tables 1 and 2.

(1) Abrasion test

[0070] An LFW-1 abrasion test was made under conditions described below in accordance with testing conditions prescribed in ASTM D2714, and the abrasion width of each block testing piece was measured after the test. As the abrasion width is smaller, the anti-wear characteristic is better.

(Testing conditions)

[0071]

Ring: Falex S-10 test ring
(SAE 4620 steel)
Block: Falex H-30 test block
(SAE 01 steel)
Test Oil temperature: 100°C
Test Load: 50 lb
Slip velocity: 100 cm/s

(2) Ultrasonic shearing test

[0072] A shearing test was carried out for 8 hours in accordance with an automatic transmission oil shearing stability test method prescribed in JASO M347-95. The present test is a test for evaluating the degree of a fall in the kinematic viscosity of automatic transmission oil in the step of the use thereof. It appears that the shearing test for 8 hours corresponds to an actual running of 100000 km and oil after the present test corresponds to oil after a running of 100000 km.

Table 1

	Example 1	Example 2	Example 3	Example 4	Example 5
(A) Base oil 1 ¹⁾	35.5	36.5	35.6	35.9	36.4
(A) Base oil 2 ²⁾	43.4	44.6	43.6	43.8	44.4
Kinematic viscosity of base oil (100°C)	3	3	3	3	3
Viscosity index of base oil	95	95	95	95	95
(B) Nitrogen-containing compound 1 ³⁾	4	4		2	
(B) Nitrogen-containing compound 2 ⁴⁾			4		
(B) Nitrogen-containing compound 3 ⁵⁾				2	4
Nitrogen content resulting from (B) component	0.08	0.08	0.06	0.05	0.02
Boron content resulting from (B) component	0	0	0	0.005	0.01
(C) Viscosity index improver 1 ⁶⁾	9.1		8.8	8.3	7.2
(C) Viscosity index improver 2 ⁷⁾		6.9			
Viscosity index improver 3 ⁸⁾					
Dispersing-agent-free package additives for ATF ⁹⁾	8	8	8	8	8
Kinematic viscosity of composition (40°C)	25	25	25	25	25
Viscosity index of composition	167	176	175	173	175
LFW-1 abrasion test					
abrasion loss using new oil	1.50	1.48	1.44	1.41	1.40
LFW-1 abrasion test					
abrasion loss using deteriorated oil	1.54	1.52	1.45	1.42	1.41

1) Refined mineral oil (kinematic viscosity at 100°C: 2.0 mm²/s, and viscosity index: 95)

2) Refined mineral oil (kinematic viscosity at 100°C: 4.4 mm²/s, and viscosity index: 95)

3) Polybutenylsuccinimide (bis type, number-average molecular weight of polybutenyl groups: 1,000, and nitrogen content: 2.0% by mass)

4) Polybutenylsuccinimide (bis type, number-average molecular weight of polybutenyl groups: 1,300, and nitrogen content: 1.6% by mass)

5) Boron-containing polybutenylsuccinimide (bis type, number-average molecular weight of polybutenyl groups: 2,500, nitrogen content: 0.5% by mass, and boron content: 0.25% by mass)

6) Non-dispersion type polymethacrylate (weight-average molecular weight: 20,000)

7) Non-dispersion type polymethacrylate (weight-average molecular weight: 30,000)

8) Non-dispersion type polymethacrylate (weight-average molecular weight: 50,000)

9) Package additives containing an anti-wear agent, an extreme-pressure agent, a metal detergent, a corrosion inhibitor, an antioxidant, a friction modifier, a rubber swelling agent, an antifoamer, and others.

Table 2

	Comparative example 1	Comparative example 2	Comparative example 3
(A) Base oil 1 ¹⁾	34.8	12.9	37.4
(A) Base oil 2 ²⁾	42.6	73.1	45.7
Kinematic viscosity of base oil (100°C)			
Viscosity index of base oil	3	3.8	3
	95	95	95
(B) Nitrogen-containing compound 1 ³⁾	0.3	4	4
(B) Nitrogen-containing compound 2 ⁴⁾			
(B) Nitrogen-containing compound 3 ⁵⁾			
Nitrogen content resulting from (B) component	0.006	0.08	0.08
Boron content resulting from (B) component	0	0	0
(C) Viscosity index improver 1 ⁶⁾	14.3	2	
(C) Viscosity index improver 2 ⁷⁾			
Viscosity index improver 3 ⁸⁾			5
Dispersing-agent-free package additives for ATF ⁹⁾	8	8	8
Kinematic viscosity of composition (40°C)	25	25	25
Viscosity index of composition	188	152	195
LFW-1 abrasion test			
abrasion loss using new oil	1.61	1.59	1.51
LFW-1 abrasion test			
abrasion loss using deteriorated oil	1.85	1.64	1.79

1) Refined mineral oil (kinematic viscosity at 100°C: 2.0 mm²/s, and viscosity index: 95)

2) Refined mineral oil (kinematic viscosity at 100°C: 4.4 mm²/s, and viscosity index: 95)

3) Polybutenylsuccinimide (bis type, number-average molecular weight of polybutenyl groups: 1,000, and nitrogen content: 2.0% by mass)

4) Polybutenylsuccinimide (bis type, number-average molecular weight of polybutenyl groups: 1,300, and nitrogen content: 1.6% by mass)

5) Boron-containing polybutenylsuccinimide (bis type, number-average molecular weight of polybutenyl groups: 2,500, nitrogen content: 0.5% by mass, and boron content: 0.25% by mass)

6) Non-dispersion type polymethacrylate (weight-average molecular weight: 20,000)

7) Non-dispersion type polymethacrylate (weight-average molecular weight: 30,000)

8) Non-dispersion type polymethacrylate (weight-average molecular weight: 50,000)

9) Package additives containing an anti-wear agent, an extreme-pressure agent, a metal detergent, a corrosion inhibitor, an antioxidant, a friction modifier, a rubber swelling agent, an antifoamer, and others.

[0073] As is evident from the results shown in Table 1, about all the lubricating oil compositions of Examples 1 to 5 according to the present invention, the anti-wear characteristic of the deteriorated oils thereof hardly became lower than that of the initial oils thereof. In the case that a non-dispersion type polymethacrylate having a weight-average molecular weight of 30000 was used as the (C) component (Example 2), in the case that a bis type succinimide having a polybutenyl group having a number-average molecular weight of 1,300 was used as the (B) component, and in the case that a boron-modified compound of a bis type succinimide having a polybutenyl group having a number-average molecular weight of 2,500 was used as the essential (B) component (Examples 4 and 5), better results in the anti-wear characteristic at the initial use and after the long-term use were exhibited than in the case of the composition of Example 1. Furthermore, in the case that the succinimide used in Example 1 was used together with the boron-modified compound of the succinimide used in Example 5 (Example 4), the anti-wear characteristic was synergistically improved.

[0074] On the other hand, in the case that the content of the (B) component did not satisfy the range defined in the invention (Comparative Example 1), in the case that the viscosity index of the composition was less than 160 (Comparative Example 2) and in the case that the weight-average molecular weight of the (C) component was over 40,000 (Comparative Example 3), the anti-wear characteristic of the new oils thereof was poor. The anti-wear characteristic of the deteriorated oils thereof got worse. In the case that the kinematic viscosity of a composition was less than 20 at 40°C, the anti-wear characteristic thereof was poorer than that of Comparative Examples.

Industrial Applicability

[0075] The lubricating oil composition of the present invention has a low viscosity and further has a good anti-wear characteristic at the initial use and after the long-term use, so as to make it possible that stirring resistance resulting from the lubricating oil is decreased. Therefore, when the composition is used as a lubricating oil for automobiles, for example, for internal combustion engines and transmissions, in particular, automatic transmissions or continuously variable transmission, the composition can contribute to an improvement in the fuel efficiency of the automobiles. In lubricating oils for machinery/apparatus other than automobiles, the energy consumption of the machinery/apparatus can be restrained while the anti-wear characteristic thereof is maintained. Accordingly, the composition is useful for various purposes, for example, for wet brakes, hydraulic apparatus, compressors, turbines, gears and shaft bearings.

Claims

1. A lubricating oil composition which is obtained by incorporating, into (A) a lubricant base oil comprising a mineral oil, a synthetic oil or a mixture thereof,

(B) a nitrogen-containing compound having at least one alkyl group or alkenyl group having a number-average molecular weight of 900 or more and/or a derivative thereof in an amount of 0.01 to 0.20% by mass in terms of the content of nitrogen of the total of the composition, and

(C) a viscosity index improver having a weight-average molecular weight of 40,000 or less, so as to set the viscosity index of the composition to 160 or more and set the kinematic viscosity of the composition into the range of 20 to 30 mm²/s at 40°C.

2. The lubricating oil composition according to claim 1, wherein the (B) component is a nitrogen-containing compound having two alkyl groups or alkenyl groups having a number-average molecular weight of 1200 or more and/or a derivative thereof.

3. The lubricating oil composition according to claim 1 or 2, which essentially comprises, as the (B) component, a boron-modified compound of a nitrogen-containing compound having at least one alkyl group or alkenyl group having a number-average molecular weight of 900 or more in an amount of 0.002% or more by mass in terms of the content of boron of the total of the composition.

4. The lubricating oil composition according to claim 3, wherein the boron-modified compound is a boron-modified compound of a nitrogen-containing compound having at least one alkyl group or alkenyl group having a number-average molecular weight of 1200 or more.

5. The lubricating oil composition according to claim 3, wherein the boron-modified compound is a boron-modified compound of a nitrogen-containing compound having two alkyl groups or alkenyl groups having a number-average molecular weight of 1200 or more.

6. The lubricating oil composition according to claim 3, wherein the boron-modified compound is a boron-modified compound of a nitrogen-containing compound having two alkyl groups or alkenyl groups having a number-average molecular weight of 1700 or more.
7. The lubricating oil composition according to any one of claims 3 to 6, wherein the ratio by mass of boron to nitrogen (B/N ratio) in the boron-modified compound is from 0.01 to 3.
8. The lubricating oil composition according to claim 1, wherein the (B) component comprises a boron-modified compound of a bis type succinimide having a poly (iso) butenyl group having a number-average molecular weight of 2000 or more.
9. The lubricating oil composition according to claim 1, wherein the (B) component comprises both of a bis type succinimide having a poly(iso)butenyl group having a number-average molecular weight of 900 or more and less than 2000 and boron-modified compounds of a bis type succinimide having a poly (iso) butenyl group having a number-average molecular weight of 2000 or more.
10. The lubricating oil composition according to any one of claims 1 to 9, which is used in an automatic transmission or a continuously variable transmission.
11. Use of the lubricating oil composition according to any one of claims 1 to 9 in an automatic transmission or a continuously variable transmission.
12. A method for maintaining the anti-wear characteristic of an automatic transmission or a continuously variable transmission by use of the lubricating oil composition according to any one of claims 1 to 9.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP03/14032

A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl.⁷ C10M161/00// (C10M161/00, 133:00, 143:00, 145:10, 149:02),
C10N20:02, 20:04, 30:06, 40:04

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)

Int.Cl.⁷ C10M161/00, 133/00-133/58, 143/00-143/18, 145/10-145/16,
149/02-149/10, C10N20:02-20:04, 30:06, 40:04

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

Jitsuyo Shinan Koho 1922-1996 Toroku Jitsuyo Shinan Koho 1994-2004
Kokai Jitsuyo Shinan Koho 1971-2004 Jitsuyo Shinan Toroku Koho 1996-2003

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	EP 949319 A2 (NIPPON MITSUBISHI OIL CORP.), 13 October, 1999 (13.10.99), & JP 11-293265 A & JP 11-349968 A & JP 11-349969 A & JP 11-349971 A & JP 11-349972 A & US 6242393 B1	1-12
Y	EP 1118654 A1 (NIPPON MITSUBISHI OIL CORP.), 25 July, 2001 (25.07.01), & JP 2000-303085 A & JP 2001-40381 A & JP 2001-55591 A & US 2001/0010293 A1 & WO 00/63323 A	1-12
Y	EP 699738 A1 (HONDA GIKEN KOGYO KABUSHIKI KAISHA), 06 March, 1996 (06.03.96), & DE 69513588 T2 & JP 8-127789 A	1-12

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

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"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

"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

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search
22 January, 2004 (22.01.04)

Date of mailing of the international search report
10 February, 2004 (10.02.04)

Name and mailing address of the ISA/
Japanese Patent Office

Authorized officer

Facsimile No.

Telephone No.