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(54) **LUBRICANT COMPOSITION**

(57) A lubricating oil composition is used in an internal combustion engine that uses a fuel containing at least one fat and oil selected from a group consisting of natural fat and oil, hydrotreated natural fat and oil, transesterified natural fat and oil and hydrotreated transesterified natural fat and oil. A polyol ester having a kinematic viscosity in the range of 3 to 8 mm²/s at 100 degrees C and/or a

polybasic acid ester having a kinematic viscosity in the range of 3 to 8 mm²/s at 100 degrees C is mixed in an amount in the range of 5 to 30 mass% of a total amount of the lubricating oil composition as a base oil of lubricating oil.

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Description**Technical Field**

5 **[0001]** The present invention relates to a lubricating oil composition to be used in an internal combustion engine that uses a fuel originating from natural fat and oil.

Background Art

10 **[0002]** These days, environmental regulations are being increasingly tightened on a global scale, among which fuel efficiency regulations and exhaust emission regulations for automobiles are especially being further tightened. Demands for tightening of the regulations are derived from environmental issues such as global warming and resource conservation due to a concern for depletion of petroleum resources.

15 Meanwhile, plants living on the earth absorb carbon dioxide in the air, water and sunlight to photosynthetically generate carbohydrate and oxygen. So-called biofuel, which is manufactured from plant-based plant oil, has been gathering remarkable attentions because of its effects on reduction of carbon dioxide (a main cause of global warming) and reduction of atmospheric contaminants emitted from automobiles. In line with an idea of carbon neutral advocating that carbon dioxide generated due to combustion of plant biomass is not counted as a contributor to an increase of the global warming gas, ratio at which the biofuel is mixed in hydrocarbon fuel is expected to be increased in the future (cf. Non-Patent Document 1

20 **[0003]**

Non-Patent Document 1: Koji YAMANE, BIODIESEL - From Deep Fryer to Fuel Thank, (Tokyo-Tosho-Shuppankai, May of 2006)

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Disclosure of the Invention**Problems to Be Solved by the Invention**

30 **[0004]** An important problem in an internal combustion engine, especially in a diesel engine, has been how to reduce environment pollution caused by such emission gas components as particulate matters (PM) such as soot and NO_x. An effective solution is to mount such an exhaust purifying device as a diesel particulate filter (DPF) or an exhaust purifying catalyst (oxidization or reduction catalyst) on an automobile. For example, soot generated in the diesel engine adheres to the DPF to be removed by oxidization and combustion.

35 When the DPF is mounted on the diesel engine, post-injection of fuel is generally conducted so as to combust the soot accumulated on the filter. Engine oil is diluted by the fuel due to the post-injection, so that performance of the engine oil is expected to be deteriorated. Particularly, since biofuel can be easily accumulated in the engine oil due to its property and generates polar compounds when degraded and decomposed, the biofuel may adversely affect detergency of engine parts such as a piston. For example, the filter is likely to be clogged by a metal oxide, hydrosulfate, carboxylate or the like generated in the combustion. Also, a portion of the used engine oil is combusted and exhausted as exhaust gas. Accordingly, it is preferable that metal content or sulfur content in a lubricating oil is as low as possible. Furthermore, it is preferable to decrease phosphorus content or sulfur content in the lubricating oil in order to prevent degrading of exhaust gas purifying catalyst.

40 However, lubricating oil sufficiently adapted for use with biofuel has not been provided yet. Here, if content in the lubricant oil, such as metal content, phosphorus content or sulfur content is simply decreased, lubricity thereof may be damaged despite the intentions.

45 An object of the invention is to provide a lubricating oil composition that is excellent in lubricity and engine-parts detergency even when biofuel or biofuel-mixed fuel is employed in an internal combustion engine such as a diesel engine.

Means for Solving the Problems

[0005] In order to solve the above-mentioned problems, according to an aspect of the invention, lubricating oil compositions as follows are provided:

55 (1) a lubricating oil composition used in an internal combustion engine, the internal combustion engine using a fuel that contains at least one fat and oil selected from a group consisting of natural fat and oil, hydrotreated natural fat and oil, transesterified natural fat and oil and hydrotreated transesterified natural fat and oil, in which a polyol ester having a kinematic viscosity in the range of 3 to 8 mm²/s at 100 degrees C and/or a polybasic acid ester having a

kinematic viscosity in the range of 3 to 8 mm²/s at 100 degrees C is mixed in an amount in the range of 5 to 30 mass% of a total amount of the lubricating oil composition as a base oil of lubricating oil;

(2) the above-described lubricating oil composition, in which a boron derivative of polybutenyl succinimide compound including a polybutenyl group that has a number average molecular weight of 500 to 3500 and possessing a mass ratio (B/N) between boron (B) and nitrogen (N) of 0.5 or more, and an alkaline earth metal detergent are mixed;

(3) the above-described lubricating oil composition, in which a sulfated ash content is 1.1 mass% or less;

(4) the above-described lubricant oil composition, in which a phenol-based antioxidant and/or an amine-based antioxidant are contained by 0.3 mass% or more of the total amount of the composition;

(5) the above-described lubricating oil composition, in which the boron derivative of the polybutenyl succinimide compound is mixed in a boron amount of 0.01 mass% or more; and

(6) the above-described lubrication oil composition, in which a sulfur content is 0.5 mass% or less of the total amount of the composition.

[0006] The lubricating oil composition according to the aspect of the invention exhibits excellent detergency for engine parts such as a piston in the internal combustion engine using so-called biofuel made of natural fat and oil and the like even when the biofuel is mixed into the engine oil. Especially, the lubricating oil is excellent in high-temperature detergency when the engine reaches a high temperature. In addition, in the lubricating oil composition of the invention, even when used in a diesel engine with a DPF, the lubricating oil composition can reduce residual ash content on the DPF, thereby preventing performance of the DPF from being deteriorated.

Natural fat and oil used in the invention is not limited to plant-derived fat and oil but may include animal-derived fat and oil.

Best Mode for Carrying Out the Invention

[0007] Embodiments of the invention will be described in detail below.

A lubricating oil composition according to the invention is a lubricating oil composition used in an internal combustion engine, the internal combustion engine using a fuel that contains at least one fat and oil selected from a group consisting of natural fat and oil, hydrotreated natural fat and oil, transesterified natural fat and oil and hydrotreated transesterified natural fat and oil.

[0008] Although the natural fat and oil may be a variety of animal-derived or plant-derived fat and oil that is generally available in nature, the natural fat and oil is preferably plant oil that contains ester of fatty acid and glycerin as a major ingredient, examples of which are safflower oil, soybean oil, canola oil, palm oil, palm kernel oil, cotton oil, cocoanut oil, rice bran oil, benne oil, castor oil, linseed oil, olive oil, wood oil, camellia oil, earthnut oil, kapok oil, cacao oil, haze wax, sunflower seed oil, corn oil and the like.

The hydrotreated natural fat and oil is formed by hydrogenating the above fat and oil under the presence of a suitable hydrogenating catalyst.

The hydrogenating catalyst is exemplified by a nickel-based catalyst, a platinum family (Pt, Pd, Rh, Ru) catalyst, a cobalt-based catalyst, a chrome-oxide based catalyst, a copper-based catalyst, an osmium-based catalyst, an iridium-based catalyst, a molybdenum-based catalyst and the like. A combination of two or more of the catalysts may also be preferably used as the hydrogenating catalyst.

[0009] The transesterified natural fat and oil is ester formed by transesterifying triglyceride contained in the natural fat and oil under the presence of a suitable ester-synthesis catalyst. For instance, by transesterifying lower alcohol and the fat and oil under the presence of the ester-synthesis catalyst, fatty acid ester usable as biofuel is manufactured. The lower alcohol, which is used as an esterifying agent, is exemplified by alcohol having 5 or less carbon atoms such as methanol, ethanol, propanol, butanol, pentanol and the like. In view of reactivity and cost, methanol is preferable. The lower alcohol is generally used in an amount equivalent to the fat and oil or more.

The hydrotreated transesterified natural fat and oil is formed by hydrogenating the above transesterified fat and oil under the presence of a suitable hydrogenating catalyst.

The natural fat and oil, the hydrotreated natural fat and oil, the transesterified natural fat and oil, and the hydrotreated transesterified natural fat and oil can be preferably used as mixed fuel by adding the above to fuel formed of hydrocarbon such as light oil.

[0010] A base oil of lubricating oil employed for the lubricating oil composition according to the invention at least includes polyol ester and/or polybasic acid ester.

Examples of polyol ester include an ester of aliphatic polyhydric alcohol and linear or branched fatty acid. Examples of the aliphatic polyhydric alcohol that form this polyol ester include neopentyl glycol, trimethylolpropane, ditrimethylolpropane, trimethylolethane, ditrimethylolethane, pentaerythritol, dipentaerythritol, and tripentaerythritol. Fatty acid having 8 to 12 carbon atoms may be employed, and examples of preferable fatty acid include pelargonic acid, capric acid, undecylic acid, lauric acid, and tridecyl acid. Partial ester of the above-noted aliphatic polyhydric alcohol and linear or branched fatty acid may also be employed. This partial ester can be obtained by reaction of aliphatic hydric alcohol and

fatty acid accompanied by suitable adjustment of a reaction mol number.

[0011] In the polyol ester of the invention, kinematic viscosity at 100 degrees C is in the range of 3 to 8 mm²/s, preferably is in the range of 4 to 7 mm²/s, and further preferably in the range of 5 to 6 mm²/s. When the kinematic viscosity at 100 degrees C is 3 mm²/s or more, evaporation loss is small. When the kinematic viscosity at 100 degrees

[0012] In polybasic acid ester of the invention, a carboxylic acid content preferably is linear or branched aliphatic dibasic acid having 6 to 10 carbon atoms. Specific examples include adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, and others that have the same property as these. An alcohol content preferably is aliphatic alcohol having 6 to 15 carbon atoms. Specific examples include hexyl alcohol, heptyl alcohol, octyl alcohol, nonyl alcohol, decyl alcohol, undecyl alcohol, dodecyl alcohol, tridecyl alcohol, tetradecyl alcohol, pentadecyl alcohol, and isomers thereof

In the polybasic acid ester of the invention, kinematic viscosity at 100 degrees C is in the range of 3 to 8 mm²/s, preferably is in the range of 4 to 7 mm²/s, and further preferably in the range of 5 to 6 mm²/s. When the kinematic viscosity at 100 degrees C is 3 mm²/s or more, evaporation loss is small. When the kinematic viscosity at 100 degrees C is 8 mm²/s or less, power loss due to viscosity resistance is restricted, thereby improving fuel efficiency.

[0013] The above-noted polyol ester and polybasic acid ester may respectively be used as a base oil alone or be used in a mixture, for example, in complex ester. Complex ester is ester synthesized from polybasic acid and polyhydric alcohol, usually including monobasic acid. In the invention, complex ester favorably used may be formed from: aliphatic polyhydric alcohol; and linear or branched aliphatic monocarboxylic acid having 8 to 12 carbon atoms, linear or branched aliphatic dibasic acid, or aromatic dibasic acid, tribasic or tetrabasic acid.

[0014] Examples of aliphatic polyhydric alcohol used to form complex ester include trimethylolpropane, trimethylol-ethane, pentaerythritol, and dipentaerythritol. The aliphatic monocarboxylic acid may be aliphatic carboxylic acid having 8 to 12 carbon atoms, examples of which include heptadecylic acid, stearic acid, nonadecanoic acid, arachic acid, behenic acid, and lignoceric acid. Examples of aliphatic dibasic acid include succinic acid, adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, undecanedioic acid, dodecanedioic acid, tridecanedioic acid, carboxylic octadecane acid, carboxymethyl octadecane acid, and docosanedioic acid. Examples of aromatic dibasic acid include phthalic acid, isophthalic acid. Examples of aromatic tribasic acid include trimellitic acid. Examples of aromatic tetrabasic acid include pyromellitic acid.

When complex ester is employed as base oil of lubricating oil used in a lubricating oil composition of the invention, preferable viscosity range is the same as those of polyol ester and polybasic acid ester as described above.

[0015] Examples of esterification reaction for producing the above-described esters include: a reaction between alcohol (e.g., polyhydric alcohol) and carboxylic acid (e.g., aliphatic polybasic ester or aromatic polybasic acid) in a predetermined ratio; partial esterification of the same and a subsequent reaction between the partially esterified material and carboxylic acid; a reaction of the acids in a reverse order; and a similar esterification reaction with mixed acids.

[0016] Contained amount of the above-described polyol ester and/or polybasic acid ester is in the range of 5 to 30 mass% relative to a total amount of the lubricating oil composition, preferably is in the range of 10 to 25 mass%, and further preferably is in the range of 10 to 20 mass%. When the contained amount of polyol ester and/or polybasic acid ester is 5 mass% or less, sufficient detergency in an engine at a high temperature cannot be obtained. On the other hand, when the contained amount of polyol ester and/or polybasic acid ester is more than 30 mass%, an influence on rubber is unfavorably increased.

[0017] The basic oil may be suitably selected from a mineral oil and a synthetic oil used as a basic oil for a lubricating oil for an internal combustion engine to be mixed with the above-described polyol ester or polybasic acid ester.

Examples of the mineral oil include: a mineral oil refined by processing lubricating oil fractions by at least one of solvent-deasphalting, solvent-extracting, hydrocracking, solvent-dewaxing, catalytic-dewaxing and hydrotreating (the lubricating oil fractions are obtained by vacuum-distilling atmospheric residual oil obtained by atmospherically distilling crude oil); and a mineral oil manufactured by isomerizing wax and GTL (gas-to-liquid) WAX.

On the other hand, examples of the synthetic oil include polybutene, polyolefin (e.g., α -olefin homopolymer or copolymer), various ethers (such as polyphenylether), polyglycol, alkylbenzene, and alkyl naphthalene. Among the above, polyolefin is particularly preferable in view of viscosity characteristic and vaporizability.

[0018] In the invention, one of the above mineral oils may be singularly used or a combination of two or more thereof may be used as the base oil to be mixed with polyol ester and polybasic acid ester. In addition, one of the above synthetic oil may be singularly used or a combination of two or more thereof may be used. Further, a combination of at least one of the above mineral oil and at least one of the above synthetic oil may be used. When these base oils are mixed and used, kinematic viscosity at 100 degrees C of the base oils as a whole preferably is in the range of 3 to 8 mm²/s. When the kinematic viscosity at 100 degrees C is 3 mm²/s or more, evaporation loss is small. When the kinematic viscosity at 100 degrees C is 8 mm²/s or less, power loss due to viscosity resistance is restricted, thereby improving fuel efficiency.

[0019] As the base oil mixed with polyol ester and polybasic acid ester, oil whose %CA measured by a ring analysis is 3 or less and whose sulfur content is 50 ppm by mass or less can be preferably used. The %CA measured by the ring analysis means a proportion (percentage) of aromatic content calculated by the n-d-M method (a ring analysis). The

sulfur content is measured based on Japanese Industrial Standard (hereinafter called, JIS) K 2541.

The base oil whose %CA is 3 or less and whose sulfur content is 50 ppm by mass or less exhibits a favorable oxidation stability. Such base oil can restrict an increase of acid number and a generation of sludge, thereby providing a lubricating oil composition that is less corrosive to metal. The sulfur content is more preferably 30 ppm by mass or less. The %CA is more preferably 1 or less, further more preferably 0.5 or less.

[0020] In addition, viscosity index of polyol ester, polybasic acid ester, and the base oil mixed with these and used is preferably 70 or more, more preferably 100 or more, further more preferably 120 or more. In the base oil whose viscosity index is 70 or more, a viscosity change due to a temperature change is small.

[0021] A lubricating oil composition of the invention preferably includes: a boron derivative of polybutenyl succinimide compound whose polybutenyl group has a number average molecular weight of 500 to 3500 and whose mass ratio (B/N) between boron (B) and nitrogen (N) is 0.5 or more; and an alkaline earth metal detergent.

Such a boron derivative of the polybutenyl succinimide compound can be obtained by, for example, reacting (a) a succinic acid substituted by a polybutenyl group having the number average molecular weight of 500 to 3500 or an anhydride of the succinic acid, (b) polyalkylene polyamine and (c) a boron compound.

[0022] As the material (a), the succinic acid substituted by the polybutenyl group or an anhydride of the succinic acid is used. The number average molecular weight (hereinafter may be abbreviated as molecular weight or Mn) of the polybutenyl group is typically 500 to 3500, preferably 1000 to 3000. When the molecular weight of the polybutenyl group is less than 500, the eventually-obtained boron derivative of the succinimide compound may not be sufficiently dissolved in the base oil of the lubricating oil. When the molecular weight is more than 3500, the succinimide compound may become so highly viscous as to impair the usability.

The polybutenyl substituted succinic acid or an anhydride of the succinic acid as the material (a) may be obtained by reacting polybutene having the molecular weight equivalent to that of the polybutenyl group with maleic anhydride by a conventional method.

[0023] Although polyalkylene polyamine is used for the material (b), 5 mol% or more of the total material is preferably formed from polyalkylene polyamine having a terminal ring structure. The entirety of the material (b) may be formed from polyalkylene polyamine having a terminal ring structure, or may be a mixture of polyalkylene polyamine having a terminal ring structure and polyalkylene polyamine having no terminal ring structure. When polyalkylene polyamine having a terminal ring structure is contained by 5 mol% or more, engine-parts detergency is further improved, which is an object of the invention. When the content of the polyalkylene polyamine is 10 mol% or more, further 20 mol% or more, the detergency is further improved, especially detergency at a high temperature is enhanced.

[0024] As the material (c), a boron compound is used. Examples of the boron compound are boracic acid, boric anhydride, borate ester, boric oxide and boron halogenide. Among the above, boracic acid is particularly preferable.

The boron derivative of polybutenyl succinimide according to the invention can be obtained by reacting the materials (a), (b) and (c). Without special limitations, any known methods of reacting can be used. For instance, by reacting the materials by the following manner, the target substance can be obtained. The materials (a) and (b) are initially reacted with each other, then its reaction product is reacted with the material (c). A mixing ratio of the materials (a) to (b) in the reaction of the material (a) and (b) is preferably 0.1-to-10 to 1 (mole ratio), more preferably 0.5-to-2 to 1 (mole ratio). A reaction temperature of the materials (a) and (b) is preferably in a range of approximately 80 to 250 degrees C, more preferably in a range of approximately 100 to 200 degrees C. At the time of reacting, depending on the materials, or in order to adjust the reaction, solvents such as an organic solvent exemplified by hydrocarbon oil may be used as necessary.

[0025] Subsequently, the thus-obtained reaction product of the materials (a) and (b) is reacted with the material (c). A mixing ratio of polyalkylene polyamine to the boron compound as the reaction material (c) is typically 1 to 0.05-to-10, preferably 1 to 0.5-to-5 (mole ratio). A reaction temperature therefor is typically approximately 50 to 250 degrees C, preferably 100 to 200 degrees C. At the time of reacting, as in the reaction of the materials (a) and (b), depending on the materials or in order to adjust the reaction, solvents such as an organic solvent exemplified by hydrocarbon oil may be used as necessary.

As a product of the above reaction, a boron derivative of a succinimide compound substituted by a polybutenyl group having a number average molecular weight of 200 to 3500 is obtained. In the invention, one of the boron derivative may be singularly used or a combination of two or more thereof may be used.

[0026] The content of boron derivative of polybutenyl succinimide compound in the lubricating composition of the invention preferably is 0.01 mass% or more in terms of boron (atoms) relative to the total amount of the composition. The content more preferably is 0.01 to 0.2 mass%, further more preferably is 0.01 to 0.15 mass%, and the most preferably is 0.01 to 0.1 mass%.

Since a predetermined amount or more of boron is contained in the boron derivative, even when biofuel is mixed into the lubricating oil composition, favorable piston detergency can be obtained in an internal combustion engine at a high temperature. When the content of boron derivative is less than 0.01 mass%, sufficient high-temperature detergency cannot be obtained. When the content of boron exceeds 0.2 mass%, no further improvement is made on the high-temperature detergency, which is of little practical use.

A mass ratio (B/N) of boron (B) contained in the boron derivative and nitrogen (N) is preferably 0.5 or more, more preferably 0.6 or more, further more preferably 0.8 or more. When B/N is 0.5 or more, high-temperature detergency for engine parts is greatly enhanced.

Although a boronated succinimide-based compound can be obtained by initially reacting the materials (a) and (b) and subsequently reacting the reaction product thereof with the material (c), the reaction order may be changed such that the materials (a) and (c) are initially reacted and the reaction product thereof is subsequently reacted with the material (b). With this reaction order, the target boronated succinimide compound may be likewise obtained.

[0027] An alkaline earth metal detergent as well as the polybutenyl succinide compound preferably is mixed to the lubricating oil composition of the invention.

Examples of the alkaline earth metal detergent include one selected from a group consisting of alkaline earth metal sulfonate, alkaline earth metal phenate and alkaline earth metal salicylate and a mixture of two or more selected from the group.

An example of alkaline earth metal sulfonate is alkaline earth metal salt of alkyl aromatic sulfonic acid obtained by sulfonating an alkyl aromatic compound having a molecular weight of 300 to 1500 (preferably 400 to 700). The alkaline earth metal salt is exemplified by magnesium salt and/or calcium salt and the like, among which calcium salt is preferably used.

An example of alkaline earth metal phenate is alkaline earth metal salt of alkylphenol, alkylphenol sulfide and a Mannich reaction product of alkylphenol. The alkaline earth metal salt is exemplified by magnesium salt and/or calcium salt and the like, among which calcium salt is preferably used.

An example of alkaline earth metal salicylate is alkaline earth metal salt of alkyl salicylic acid. The alkaline earth metal salt is exemplified by magnesium salt and/or calcium salt and the like, among which calcium salt is preferably used. An alkyl group forging the alkaline earth metal-based detergent preferably has 4 to 30 carbon atoms. The alkyl group is more preferably a linear or branched alkyl group having 6 to 18 carbon atoms, in which 6 to 18 carbon atoms may be in a linear chain or in a branched chain. The alkyl group may be a primary alkyl group, a secondary alkyl group or a tertiary alkyl group.

[0028] In addition, alkaline earth metal sulfonate, alkaline earth metal phenate and alkaline earth metal salicylate may be neutral alkaline earth metal sulfonate, neutral alkaline earth metal phenate and neutral alkaline earth metal salicylate obtained by: directly reacting the above-described alkyl aromatic sulfonic acid, alkylphenol, alkylphenol sulfide, a Mannich reaction product of alkylphenol, alkyl salicylic acid or the like with alkaline earth metal base exemplified by an oxide or a hydroxide of alkaline earth metal such as magnesium and/or calcium; or converting the above-described substance into alkali metal salt such as sodium salt or potassium salt and subsequently substituting the alkali metal salt with alkaline earth metal salt. Alternatively, alkaline earth metal sulfonate, alkaline earth metal phenate and alkaline earth metal salicylate may be: basic alkaline earth metal sulfonate, basic alkaline earth metal phenate and basic alkaline earth metal salicylate obtained by heating neutral alkaline earth metal sulfonate, neutral alkaline earth metal phenate and neutral alkaline earth metal salicylate with excess alkaline earth metal salt or alkaline earth metal base under the presence of water; or overbased alkaline earth metal sulfonate, overbased alkaline earth metal phenate and overbased alkaline earth metal salicylate obtained by reacting neutral alkaline earth metal sulfonate, neutral alkaline earth metal phenate and neutral alkaline earth metal salicylate with carbonate or borate of alkaline earth metal under the presence of carbon dioxide gas.

[0029] In the invention, the content of the alkaline earth metal-based detergent is preferably 0.35 mass% or less in terms of alkaline earth metal, more preferably 0.01 to 0.35 mass%, further more preferably 0.1 to 0.35 mass%. When the content of the alkaline earth metal-based detergent is 0.01 mass% or more, the lubricating oil composition exhibits more excellent oxidation stability, base-number retention and high-temperature detergency. On the other hand, when the content of the alkaline earth metal-based detergent exceeds 0.35 mass%, performance of catalyst for purifying exhaust gas may be deteriorated. In addition, when such is applied to a diesel engine with a DPF, an amount of ash content adhering to the DPF may be increased, thereby shortening the life of the DPF.

[0030] The lubricating oil composition according to the invention preferably contains a phenol-based antioxidant and/or an amine-based antioxidant as the antioxidant.

Examples of the phenol-based antioxidant are: octadecyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate;

4,4'-methylenebis(2,6-di-t-butylphenol); 4,4'-bis(2,6-di-t-butylphenol);

4,4'-bis(2-methyl-6-t-butylphenol); 2,2'-methylenebis(4-ethyl-6-t-butylphenol);

2,2'-methylenebis(4-methyl-6-t-butylphenol);

4,4'-butylidenebis(3-methyl-6-t-butylphenol);

4,4'-isopropylidenebis(2,6-di-t-butylphenol);

2,2'-methylenebis(4-methyl-6-nonylphenol); 2,2'-isobutylidenebis(4,6-dimethylphenol);

2,2'-methylenebis(4-methyl-6-cyclohexylphenol); 2,6-di-t-butyl-4-methylphenol;

2,6-di-t-butyl-4-ethylphenol; 2,4-dimethyl-6-t-butylphenol; 2,6-di-t-amyl-p-cresol;

2,6-di-t-butyl-4-(N,N'-dimethylaminomethylphenol);

4,4'-thiobis(2-methyl-6-t-butylphenol); 4,4'-thiobis(3-methyl-6-t-butylphenol);
 2,2'-thiobis(4-methyl-6-t-butylphenol); bis(3-methyl-4-hydroxy-5-t-butylbenzyl)sulfide;
 bis(3,5-di-t-butyl-4-hydroxybenzyl)sulfide;
 n-octyl-3-(4-hydroxy-3,5-di-t-butylphenyl)propionate;

n-octadecyl-3-(4-hydroxy-3,5-di-t-butylphenyl)propionate;

2,2'-thio[diethyl-bis-3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate] and the like. Among the above, bisphenol-based antioxidant and ester group-containing phenol-based antioxidant are preferable.

[0031] Examples of the amine-based antioxidant are: an antioxidant based on monoalkyldiphenylamine such as mono-octyldiphenylamine and monononyldiphenylamine; an antioxidant based on dialkyl diphenylamine such as 4,4'-dibutyldiphenylamine, 4,4'-dipentyldiphenylamine, 4,4'-dihexyldiphenylamine, 4,4'-diheptyldiphenylamine, 4,4'-dioctyldiphenylamine and 4,4'-dinonyldiphenylamine; an antioxidant based on polyalkyldiphenylamine such as tetrabutyl-diphenylamine, tetrahexyldiphenylamine, tetraoctyldiphenylamine and tetranonyldiphenylamine; and an antioxidant based on naphthylamine, specifically alkyl-substituted phenyl- α -naphthylamine such as α -naphthylamine, phenyl- α -naphthylamine, butylphenyl- α -naphthylamine, pentylphenyl- α -naphthylamine, hexylphenyl- α -naphthylamine, heptylphenyl- α -naphthylamine, octylphenyl- α -naphthylamine and nonylphenyl- α -naphthylamine. Among the above, a dialkyl diphenylamine-based antioxidant and a naphthylamine-based antioxidant are preferable.

[0032] Incidentally, as another antioxidant, a molybdenum-amine complex-based antioxidant may be used. As the molybdenum-amine complex-based antioxidant, a hexahydric molybdenum compound, an example of which is a reaction product obtained by reacting molybdenum trioxide and/or molybdenum acid with an amine compound, may be used.

The reaction product may be, for example, a compound obtained by the manufacturing method disclosed in JP-A-2003-252887. The amine compound to be reacted with the hexahydric molybdenum compound subjects to no particular limitation, and examples thereof are monoamine, diamine, polyamine and alkanolamine. Specific examples of the amine compound are: alkyl amine having an alkyl group of 1 to 30 carbon atoms (the alkyl group may contain a linear chain or a branched chain), exemplified by methylamine, ethylamine, dimethylamine, diethylamine, methylethylamine, methylpropylamine and the like; alkenyl amine having an alkenyl group of 2 to 30 carbon atoms (the alkenyl group may contain a linear chain or a branched chain), exemplified by ethenylamine, propenylamine, butenylamine, octenylamine and oleylamine; alkanol amine having an alkanol group of 1 to 30 carbon atoms (the alkanol group may contain a linear chain or a branched chain), exemplified by methanolamine, ethanolamine, methanolethanolamine and methanolpropanolamine; alkylenediamine having an alkylene group of 1 to 30 carbon atoms, exemplified by methylenediamine, ethylenediamine, propylenediamine and butylenediamine; polyamine such as diethylenetriamine, triethylenetetramine, tetraethylenepentamine and pentaethylenhexamine; a heterocyclic compound obtained by reacting monoamine, diamine and polyamine with a compound having an alkyl or alkenyl group of 8 to 20 carbon atoms or imidazoline, monoamine, diamine and polyamine being exemplified by undecyldiethylamine, undecyldiethanolamine, dodecyldipropylamine, oleyldiethanolamine, oleylpropylenediamine and stearyl-tetraethylenepentamine; an alkylene-oxide adduct of the compounds; and a mixture thereof. In addition, sulfur-containing molybdenum complexes of succinimide as disclosed in JP-B-03-22438 and JP-A-2004-2866 may be used.

A mixing content of the antioxidant is preferably 0.3 mass% or more of the total amount of the composition, more preferably 0.5 mass% or more. On the other hand, when the content exceeds 2 mass%, the antioxidant may not be dissolved in the base oil of the lubricating oil. Accordingly, the content of the antioxidant is preferably in a range from 0.3 to 2 mass% of the total amount of the composition.

[0033] The lubricating oil composition according to the invention may be added as necessary with other additives such as a viscosity index improver, a pour point depressant, antiwear agent, an ashless-type friction modifier, a rust inhibitor, a metal deactivator, a surfactant and antifoaming agent as long as effects of the invention are not hampered.

[0034] Examples of the viscosity index improver are polymethacrylate, dispersed polymethacrylate, an olefin-based copolymer (such as an ethylene-propylene copolymer), a dispersed olefin-based copolymer, a styrene-based copolymer (such as a styrene-diene copolymer and a styrene-isoprene copolymer) and the like. In view of blending effects, a content of the viscosity index improver is 0.5 to 15 mass% of the total amount of the composition, preferably 1 to 10 mass%.

[0035] An example of the pour point depressant is polymethacrylate having a weight-average molecular weight of 5000 to 50000.

Examples of the antiwear agent are: sulfur-containing compounds such as zinc dithiophosphate, zinc dithiocarbamate, zinc phosphate, disulfides, sulfurized olefins, sulfurized fats and oils, sulfurized esters, thiocarbonates, thiocarbamates (such as Mo-DTC) and the like; phosphorus-containing compounds such as phosphite esters, phosphate esters, phosphonate esters and amine salts or metal salts thereof; and a sulfur and phosphorus-containing antiwear agent such as thiophosphite esters, thiophosphate esters (such as Mo-DTP), thiophosphonate esters and amine salts or metal salts thereof.

[0036] As the ashless-type friction modifier, any compounds generally used as the ashless-type friction modifier for lubricating oil may be used, examples of which are fatty acid, aliphatic alcohol, aliphatic ether, aliphatic ester, aliphatic amine and aliphatic amide that have at least one alkyl or alkenyl group of 6 to 30 carbon atoms in the molecule.

[0037] Examples of the rust inhibitor are petroleum sulfonate, alkylbenzene sulfonate, dinonylnaphthalene sulfonate, alkenyl succinic ester, multivalent alcohol ester and the like. In view of blending effects, a content of the rust inhibitor is generally 0.01 to 1 mass% of the total amount of the composition, preferably 0.05 to 0.5 mass%.

[0038] Examples of the metal deactivator (copper corrosion inhibitor) are benzotriazole-based compounds, tolyltriazole-based compounds, thiadiazole-based compounds, imidazole-based compounds, and pyrimidine-based compounds. Among the above, the benzotriazole-based compounds are preferable. By adding the metal deactivator, the engine parts can be prevented from being metallically corroded and degraded due to oxidation. In view of blending effects, a content of the metal deactivator is preferably 0.01 to 0.1 mass% of the total amount of the composition, more preferably 0.03 to 0.05 mass%.

[0039] Examples of the surfactant are nonionic surfactants based on polyalkylene glycol such as polyoxyethylene-alkylether, polyoxyethylenealkylphenylether and polyoxyethylenealkylnaphthylether.

[0040] Examples of the antifoaming agent are silicone oil, fluorosilicone oil, fluoroalkylether and the like. In view of a balance between antifoaming effects and economics, a content of the antifoaming agent is preferably approximately 0.005 to 0.1 mass% of the total amount of the compound.

[0041] Sulfur content of the lubricating oil composition according to the invention is preferably 0.5 mass% or less of the total amount of the composition, more preferably 0.3 mass% or less, further more preferably 0.2 mass% or less. When the sulfur content is 0.5 mass% or less, deterioration of the catalyst performance for purifying exhaust gas can be effectively prevented.

Phosphorus content of the lubricating oil composition according to the invention is preferably 0.12 mass% or less of the total amount of the composition, more preferably 0.1 mass% or less. When the phosphorus content is 0.12 mass% or less, deterioration of the catalyst performance for purifying exhaust gas can be effectively prevented.

[0042] Sulfated ash content of the lubricating oil composition according to the invention is preferably 1.1 mass% or less, more preferably 1 mass% or less. When the sulfated ash content is 1.1 mass% or less, deterioration of the catalyst performance for purifying exhaust gas can be effectively prevented. In addition, in a case of a diesel engine, the ash content accumulated on the filter of the DPF can be reduced, thereby preventing the filter blockage due to the ash content and contributing to a long life of the DPF. The sulfated ash content means ash content obtained by adding sulfuric acid to carbonized residue caused by combustion of samples for heating so that the residue has a constant mass. The sulfated ash is generally used to know a rough amount of metal-based additives contained in the lubricating oil composition. Specifically, the sulfated ash is measured by a method prescribed in "5. Experiment Method of Sulfated Ash" of JIS K 2272.

Incidentally, when vaporizability of lubricating oil in an internal combustion engine is increased, more lubricating oil is consumed, which leads to a shortened lifetime of lubricating oil. In addition, because more lubricating oil is dispersed within the exhaust gas catalyst, catalyst performance is lowered and catalyst lifetime is shortened. In view of the above, in the lubricating oil composition of the invention, NOACK evaporation measured according to JPI (abbreviation for Japan Petroleum Institute, the same applies hereinafter)-5S-41-93 is preferably 15 mass% or less, more preferably 13 mass% or less, and further preferably 10 mass% or less.

Examples

[0043] Next, the invention will be further described in detail based on Examples, which by no means limit the invention.

Examples 1 to 7 and Comparatives 1 to 4

[0044] Lubricating oil compositions containing components shown in Table 1 respectively were prepared, which were then subjected to such a hot tube test as follows. The components used for preparing the lubricating oil compositions are as follows.

(1) Base Oil A: hydrorefining base oil; kinematic viscosity at 40 degrees C of 21 mm²/s; kinematic viscosity at 100 degrees C of 4.5 mm²/s; viscosity index of 127; %CA of 0.0; sulfur content of less than 20 ppm by mass; and NOACK evaporation of 13.3 mass%.

(2) Base Oil B: PAO; kinematic viscosity at 40 degrees C of 17.5 mm²/s; kinematic viscosity at 100 degrees C of 3.9 mm²/s; viscosity index of 120; and NOACK evaporation of 15 mass%.

(3) Base Oil C: ester oil (polyol fatty acid ester; polyol content being trimethylolpropane, and fatty acid content being refined coconut oil (of C8 to C12)); kinematic viscosity at 40 degrees C of 19.8 mm²/s; kinematic viscosity at 100 degrees C of 4.3 mm²/s; viscosity index of 139; NOACK evaporation of 3 mass%; and saponification number of 0.1 mg KOH/g.

(4) Base Oil D: tridecyl adipate; kinematic viscosity at 40 degrees C of 27 mm²/s; kinematic viscosity at 100 degrees C of 5.3 mm²/s; viscosity index of 132; and NOACK evaporation of 4 mass%.

[0045]

(5) Viscosity Index Improver: polymethacrylate; mass average molecular weight of 420000; and resin content of 39 mass%.

(6) Pour Point Depressant: polyalkylmethacrylate; and mass average molecular weight of 6000.

(7) Metal-Based Detergent: overbased calcium salicylate; base number of 225 mg KOH/g (perchloric acid method); calcium content of 7.8 mass%; and sulfur content of 0.3 mass%.

(8) Polybutenyl Succinic Monoimide A: number average molecular weight of the polybutenyl group being 1000; nitrogen content of 1.76 mass%; boron content of 2.0 mass%; and B/N of 1.1.

(9) Polybutenyl Succinic Bisimide B: number average molecular weight of the polybutenyl group being 2000; nitrogen content of 0.99 mass%; and B/N of 0.

(10) Phenol-Based Antioxidant: octadecyl-3-(3, 5-tert-butyl-4-hydroxyphenyl)propionate.

(11) Amine-Based Antioxidant: dialkyl diphenylamine; nitrogen content of 4.62 mass%.

(12) Zinc Dialkyl Dithio Phosphate: Zn content of 9.0 mass%; phosphorus content of 8.2 mass%; sulfur content of 17.1 mass%; and the alkyl group being a mixture of a secondary butyl group and a secondary hexyl group.

(13) Copper Corrosion Inhibitor: 1-[N,N-bis(2-ethylhexyl) aminomethyl] methyl benzotriazole.

(14) Biodiesel Fuel: canola oil methyl ester.

(15) Other Additives: a rust inhibitor, a surfactant and an antifoaming agent.

[0046] Measurement of properties of the lubricating oil compositions and the hot tube test were conducted in the following manner.

(Calcium Content)

[0047] Measurement was conducted based on JIS-5S-38-92.

(Boron Content)

[0048] Measurement was conducted based on JIS-5S-38-92.

(Nitrogen Content)

[0049] Measurement was conducted based on JIS K2609.

(Phosphorus Content)

[0050] Measurement was conducted based on JPI-5S-38-92.

(Sulfur Content)

[0051] Measurement was conducted based on JIS K2541.

(Sulfated Ash Content)

[0052] Measurement was conducted based on JIS K2272.

(Noack)

[0053] Measurement was conducted based on JPI-5S-41-93.

(Hot Tube Test)

[0054] As the lubricating oil composition to be tested, mixed oil in which biofuel (fuel obtained by transesterifying canola oil with methyl alcohol) was mixed by 5 mass% of each of the lubricating oil compositions (new oil) was used, assuming a mixing ratio of the fuel and the lubricating oil in an internal combustion engine. The measurement was conducted with the test temperature being set at 280 degrees C and 320 degrees C, and other conditions being based on JPI-5S-55-99. In addition, since the hot tube test may be affected by the amount of the viscosity index improver, the content of the viscosity index improver was made constant among Examples and Comparatives. The smaller an amount

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of fouling on the glass tube after the test was, the more favorable the detergency is.

The properties of the lubricating oil compositions and the results of the hot tube test are shown in Table 1.

[0055]

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(Table 1)

		EXAMPLES							COMPARATIVES			
		1	2	3	4	5	6	7	1	2	3	4
COMPOSITION (mass%)	BASE OIL A	62.61	62.61	62.61	-	61.61	61.61	-	82.61	62.61	79.61	79.61
	BASE OIL B	-	-	-	62.61	-	-	61.61	-	20.00	-	-
	BASE OIL C	20.00	-	10.00	20.00	20.00	-	20.00	-	-	3.00	-
	BASE OIL D	-	20.00	1000	-	-	20.00	-	-	-	-	3.00
	VISCOSITY INDEX IMPROVER	6.50	6.50	6.50	650	6.50	6.50	6.50	6.50	6.50	6.50	6.50
	POUR POINT DEPRESSANT	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30
	METAL-BASED DETERGENT	2.82	282	2.82	2.82	2.82	2.82	2.82	282	2.82	2.82	2.82
	POLYBUTENYL SUCCINIC MONOIMIDEA -		-	-	-	1.00	1.00	1.00	-	-	-	-
	POLYBUTENYL SUCCINIC BISMIDE B	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00
	PHENOL-BASED ANTIOXIDANT	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50
	AMINE-BASED ANTIOXIDANT	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50
	ZINC DIALKYL DITHIO PHOSPHATE	1.22	1.22	1.22	1.22	1.22	1.22	1.22	1.22	1.22	1.22	1.22
	COPPER CORROSION INHIBITOR	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
	OTHERS	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50
	TOTAL	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00

(continued)												
		EXAMPLES							COMPARATIVES			
		1	2	3	4	5	6	7	1	2	3	4
CHARACTERISTICS OF COMPOSITION (mass%)	CALCIUM CONTENT	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22
	BORON CONTENT	0.00	0.00	0.00	0.00	0.02	0.02	0.02	0.00	0.00	0.02	0.02
	NITROGEN CONTENT	0.07	0.07	0.07	0.07	0.09	0.09	0.09	0.07	0.07	0.09	0.09
	SULFUR CONTENT	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
	PHOSPHORUS CONTENT	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10
	SULFATE ASH CONTENT	0.96	0.96	0.96	0.96	0.97	0.97	0.97	0.96	0.96	0.97	0.97
	NOACK	8.7	9.0	8.8	9.5	8.8	9.0	9.8	11.2	10.5	11.0	11.0
HOT TUBE TEST/FOULING (mg)												
NEW OIL 95% + BIODIESEL FUEL 5% @280°C		0.8	0.8	0.8	0.8	0.1	0.1	0.1	10.3	9.7	9.9	10.1
NEW OIL 95% + BIODIESEL FUEL 5% @320° C		25.2	28.0	26.2	26.6	3.9	4.4	4.1	157.8	83.3	155.5	153.5

(Evaluation Result)

[0056] As is understood from the results of the hot tube test shown in Table 1, in Examples 1 to 7 where the lubricating oil composition according to the invention was used, irrespective of the addition of the biofuel to the new oil, the amount of fouling in the hot tube was not notably large. Such results indicate excellent detergency for the engine of the lubricating oil composition. On the other hand, in Comparatives 1 and 2, since neither polyol ester nor polybasic acid ester was used as a base oil, a very large amount of fouling was observed. In addition, even in the case where polyol ester or polybasic acid ester was used as a base oil as in Comparatives 3 and 4, when the content thereof was small, a fouling prevention effect was hardly observed. Such a result indicates that detergency for an engine should not be expected.

Industrial Applicability

[0057] The lubricating oil composition according to the present invention is favorably applied to an internal combustion engine in which biofuel or biofuel-fixed fuel is employed.

Claims

1. A lubricating oil composition used in an internal combustion engine, the internal combustion engine using a fuel that contains at least one fat and oil selected from a group consisting of natural fat and oil, hydrotreated natural fat and oil, transesterified natural fat and oil and hydrotreated transesterified natural fat and oil, wherein a polyol ester having a kinematic viscosity in the range of 3 to 8 mm²/s at 100 degrees C and/or a polybasic acid ester having a kinematic viscosity in the range of 3 to 8 mm²/s at 100 degrees C is mixed in an amount in the range of 5 to 30 mass% of a total amount of the lubricating oil composition as a base oil of lubricating oil.
2. The lubricant oil composition according to claim 1, wherein a boron derivative of polybutenyl succinimide compound including a polybutenyl group that has a number average molecular weight of 500 to 3500 and possessing a mass ratio (B/N) between boron (B) and nitrogen (N) of 0.5 or more, and an alkaline earth metal detergent are mixed.
3. The lubricant oil composition according to claim 1 or 2, wherein a sulfated ash content is 1.1 mass% or less.
4. The lubricant oil composition according to any one of claims 1 to 3, wherein a phenol-based antioxidant and/or an amine-based antioxidant are contained by 0.3 mass% or more of the total amount of the composition.
5. The lubricant oil composition according to any one of claims 1 to 4, wherein the boron derivative of the polybutenyl succinimide compound is mixed in a boron amount of 0.01 mass% or more.
6. The lubricant oil composition according to any one of claims 1 to 5, wherein a sulfur content is 0.5 mass% or less of the total amount of the composition.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2008/068860

A. CLASSIFICATION OF SUBJECT MATTER

See extra sheet.

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)

C10M105/32, C10M169/04, C10M105/36, C10M105/38, C10M129/10, C10M129/70, C10M133/04, C10M135/10, C10M139/00, C10M159/22, C10M159/24, C10N10/04, C10N20/00, C10N20/02, C10N20/04, C10N30/04, C10N30/06, C10N40/25

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

Jitsuyo Shinan Koho	1922-1996	Jitsuyo Shinan Toroku Koho	1996-2008
Kokai Jitsuyo Shinan Koho	1971-2008	Toroku Jitsuyo Shinan Koho	1994-2008

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	JP 2002-206096 A (Nippon Mitsubishi Oil Corp.), 26 July, 2002 (26.07.02), Claims; Par. Nos. [0001], [0006], [0009] to [0010], [0013] to [0017], [0040]; example 3 (Family: none)	1, 3, 4, 6 2, 5
X A	JP 2002-275488 A (Nippon Oil Corp.), 25 September, 2002 (25.09.02), Claims; Par. Nos. [0005] to [0007], [0019] to [0021], [0039], [0046]; examples 1 to 6 (Family: none)	1, 3, 4, 6 2, 5

☒ 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
"A" document defining the general state of the art which is not considered to be of particular relevance	"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
"E" earlier application or patent but published on or after the international filing date	"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
"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)	"&" document member of the same patent family
"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
07 November, 2008 (07.11.08)Date of mailing of the international search report
18 November, 2008 (18.11.08)Name and mailing address of the ISA/
Japanese Patent Office

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INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2008/068860

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	JP 2001-348591 A (Nippon Mitsubishi Oil Corp.), 18 December, 2001 (18.12.01), Claims; Par. Nos. [0007] to [0013], [0046], [0049] to [0051], [0054]; example 1 (Family: none)	1, 3, 4, 6 2, 5
A	JP 2005-264066 A (Nippon Oil Corp.), 29 September, 2005 (29.09.05), Claims; Par. No. [0029] & WO 2005/090532 A1 & EP 1736529 A1 & US 2007/0179070 A1	1-6
A	JP 2004-269707 A (Idemitsu Kosan Co., Ltd.), 30 September, 2004 (30.09.04), Claims; Par. Nos. [0010] to [0015] (Family: none)	1-6

Form PCT/ISA/210 (continuation of second sheet) (April 2007)

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2008/068860

Continuation of A. CLASSIFICATION OF SUBJECT MATTER

(International Patent Classification (IPC))

C10M105/32(2006.01)i, C10M169/04(2006.01)i, C10M105/36(2006.01)n,
C10M105/38(2006.01)n, C10M129/10(2006.01)n, C10M129/70(2006.01)n,
C10M133/04(2006.01)n, C10M135/10(2006.01)n, C10M139/00(2006.01)n,
C10M159/22(2006.01)n, C10M159/24(2006.01)n, C10N10/04(2006.01)n,
C10N20/00(2006.01)n, C10N20/02(2006.01)n, C10N20/04(2006.01)n,
C10N30/04(2006.01)n, C10N30/06(2006.01)n, C10N40/25(2006.01)n

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

REFERENCES CITED IN THE DESCRIPTION

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