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(54) LUBRICATING OIL COMPOSITION

(57) A lubricating oil composition of the invention contains: a base oil; at least one of acid phosphate represented by formulae (1) and (2) below; and an antioxidant.

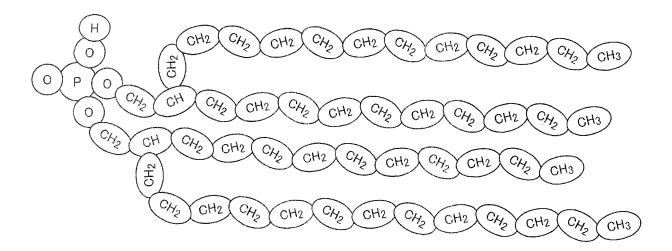
$$\begin{array}{c}
O \\
\parallel \\
P - OH \\
OR^2
\end{array}$$
(1)

$$\begin{array}{ccc}
O \\
\parallel \\
R^{3}O - P - (OH)_{2}
\end{array} (2)$$

In the above formulae: R^1 , R^2 and R^3 are alkyl groups; and at least one of R^1 and R^2 has 20 to 30 carbon atoms while R^3 has 20 to 30 carbon atoms.

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FIG.1



Description

TECHNICAL FIELD

⁵ **[0001]** The present invention relates to a lubricating oil composition, for instance, to a lubricating oil composition to be used for a shock absorber in an automobile and industrial machinery/device.

BACKGROUND ART

[0002] A shock absorber is provided at a connection portion between tires and a vehicle body in an automobile and is configured to damp vibration of the vehicle body caused by a bumpy road surface and acceleration/deceleration of the automobile. Accordingly, riding comfort on the automobile is considerably affected by a performance of the shock absorber.

[0003] Friction of the shock absorber is mainly generated on an oil seal (a rubber material) and a rod (chrome-plated) portion. Accordingly, an improvement in friction characteristics generated between the rubber material and the chrome-plated portion is a very crucial problem. Moreover, in order that the rod of the shock absorber smoothly moves in extension and contraction, an absolute value (a friction coefficient between the rubber material and metal) of rubber friction is required to be small.

[0004] The friction characteristics have been so far evaluated in terms of the absolute value of the rubber friction. Accordingly, a shock absorber oil having a small value with respect to the above parameter has been developed (see non-Patent Literatures 1 and 2).

CITATION LIST

NON-PATENT LITERATURE(S)

[0005]

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Non-Patent Literature 1: Proceedings of Petroleum Product Symposium of Petroleum Institute, page 96 (December in 2009)

Non-Patent Literature 2: Tribologists, page 567, Vol. 56, No. 9 (in 2011)

SUMMARY OF THE INVENTION

PROBLEMS TO BE SOLVED BY THE INVENTION

[0006] The absolute values of the rubber friction as described in non-Patent Literatures 1 and 2 are not enough to sufficiently evaluate friction characteristics, so that development of the lubricating oil having excellent friction characteristics has not been easy. Moreover, it has been found that a large workload of the friction (i.e., friction energy) is also crucial for damping vibration of the vehicle body. Further, since the shock absorber is not supposed to be replaced, the shock absorber is generally used continuously for 5 to 10 years. Accordingly, the lubricating oil used for the shock absorber is required to also have a high oxidation stability.

[0007] An object of the invention is to provide a lubricating oil composition having a small friction coefficient between a rubber material and metal, a large friction energy and an excellent oxidation stability.

MEANS FOR SOLVING THE PROBLEM(S)

[0008] In order to solve the above problems, a lubricating oil composition according to an aspect of the invention is provided as follows:

a lubricating oil composition provided by blending to a base oil at least one of acid phosphate represented by formulae (1) and (2) below; and an antioxidant.

55 [Formula 1]

$$\begin{array}{c}
O \\
R^{1}O - P - OH \\
OR^{2}
\end{array} (1)$$

$$\begin{array}{ccc}
O \\
R^3O - P - (OH)_2
\end{array} (2)$$

In the formulae, R^1 , R^2 and R^3 are alkyl groups. At least one of R^1 and R^2 has 20 to 30 carbon atoms while R^3 has 20 to 30 carbon atoms.

[0009] According to the above aspect of the invention, a lubricating oil composition having a small friction coefficient between a rubber material and metal, a large friction energy and an excellent oxidation stability can be provided.

BRIEF DESCRIPTION OF DRAWING(S)

20 [0010]

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Fig. 1 schematically shows an example of ditetracosyl acid phosphate.

Fig. 2 schematically shows an example of dioctacosyl acid phosphate.

Fig. 3 schematically shows an example of distearyl acid phosphate.

Fig. 4 shows a friction tester used in Examples.

Fig. 5 shows an example of a Lissajous waveform obtained by the friction tester.

DESCRIPTION OF EMBODIMENT(S)

[0011] A lubricating oil composition in an exemplary embodiment of the invention (hereinafter also referred to as "the present composition") is provided by blending a predetermined acid phosphate and an antioxidant to a base oil. The present composition will be described in detail below.

Base Oil

Dago C

[0012] The base oil used in the present composition is not particularly limited. At least one of a mineral oil and a synthetic oil may be used alone or in combination of two or more. Alternatively, a combination of the mineral oil and the synthetic oil may be used.

[0013] When the present composition is used for a shock absorber, it is preferable to use a base oil having a kinematic viscosity at 40 degrees C in a range approximately from 5 mm²/s to 40 mm²/s in order to keep suitable friction characteristics.

[0014] Moreover, a pour point of the base oil, which is an index for low-temperature fluidity, is preferably -10 degrees C or less, particularly preferably -15 degrees C or less.

[0015] Examples of the mineral oil include a naphthenic mineral oil, a paraffinic mineral oil, and GTL WAX. Specifically, the examples of the mineral oil include a light neutral oil, intermediate neutral oil, heavy neutral oil and bright stock which are obtained by solvent purification or hydrogenation purification.

[0016] On the other hand, examples of the synthetic oil include polybutene, a hydride thereof, poly-alpha-olefin (e.g., 1-octane oligomer, 1-decene oligomer), alkylbenzene, polyolester, diacid ester, polyoxyalkyleneglycol, polyoxyalkyleneglycolester, hindered ester and silicone oil.

Acid Phosphate

[0017] Acid phosphate contained in the present composition is represented by formulae (I) and (2) below.

[Formula 2]

$$\begin{array}{c}
O\\
R^3O - P - (OH)_2
\end{array} (2)$$

[0018] In the formulae, R¹, R² and R³ are alkyl groups. At least one of R¹ and R² has 20 to 30 carbon atoms while R³ has 20 to 30 carbon atoms.

[0019] Examples of the alkyl group having 20 to 30 carbon atoms include an eicosyl group, heneicosyl group, docosyl group, tricosyl group, tetracosyl group, pentacosyl group, hexacosyl group, heptacosyl group, octacosyl group, nonacosyl group and triacontyl group.

[0020] Examples of the acid phosphate represented by the formulae (1) and (2) include tetracosyl acid phosphate, ditetracosyl acid phosphate, and dioctacosyl acid phosphate when R to R³ are a tetracosyl group or an octacosyl group. For reference, an example of ditetracosyl acid phosphate and an example of dioctacosyl acid phosphate are schematically shown in Figs 1 and 2, respectively.

[0021] Since the acid phosphate has an alkyl group having 20 or more carbon atoms, a friction coefficient and a friction energy are effectively improvable. Moreover, the oxidation stability is also improvable in the presence of the above-described acid phosphate.

[0022] Further, the number of the carbon atoms of the above-described alkyl group is preferably 30 or less since solubility of the acid phosphate in the base oil can be secured. The number of the carbon atoms of the above-described alkyl group is preferably in a range from 20 to 26, more preferably in a range from 20 to 24.

[0023] On the other hand, when the number of the carbon atoms of the alkyl group falls below the lower limit described above, the friction coefficient becomes large, so that the friction energy also cannot be expected to be increased. For instance, in Fig. 3 schematically showing an example of distearyl acid phosphate, since the number of the carbon atoms of the alkyl group is small, the friction coefficient becomes large and the friction energy becomes small.

[0024] Although the alkyl group of the formulae (1) and (2) may be linear, it is desirable that the alkyl group has a side chain in order to decrease the friction coefficient and increase the friction energy. Further, since the side chain serves as steric hindrance, the oxidation stability is also improvable. Specifically, at least one of R¹ and R² preferably has a side chain in the formula (1). Moreover, R³ preferably has a side chain in the formula (2). Further, the number of the carbon atoms of the above-described side chain is preferably in a range from 6 to 18. The side chain having 6 or more carbon atoms is preferable in terms of a decrease in the friction coefficient and an increase in the friction energy. Moreover, the side chain having 18 or less carbon atoms is preferable since solubility of the acid phosphate in the base oil becomes favorable.

[0025] The acid phosphate of the formula (1) is preferable to the acid phosphate of the formula (2) in terms of the decrease in the friction coefficient, the increase in the friction energy and the oxidation stability.

[0026] In the present composition, a content of the above-described acid phosphate blended with the base oil is preferably in a range of 0.01 mass% to 3 mass% of the total amount of the composition, more preferably in a range of 0.1 mass% to 2 mass%, further preferably in a range of 0.2 mass% to 1 mass%. The content of the acid phosphate of 0.01 mass% or more is preferable not only in terms of the decrease in the friction coefficient and the increase in the friction energy but also the oxidation stability. Moreover, the content of the acid phosphate of 3 mass% or less is preferable since solubility of the acid phosphate in the base oil can be secured.

Antioxidant

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[0027] The present composition further contains an antioxidant. Although the predetermined acid phosphate described above has an effect for improving the oxidation stability, use of the antioxidant in combination with the acid phosphate exhibits an outstanding oxidation stability.

[0028] As the antioxidant, at least one of an amine antioxidant, a phenolic antioxidant and a sulfur antioxidant is preferably usable. One of the antioxidant may be used alone or a combination of two or more thereof may be used.

[0029] Examples of the amine antioxidant includes: monoalkyldiphenylamine compounds such as monooctyldiphenylamine and monononyldiphenylamine; dialkyldiphenylamine compounds such as 4,4'-dibutyldiphenylamine, 4,4'-dipentyldiphenylamine, 4,4'-dihenylamine, 4,4'-dihenylamine, 4,4'-diphenylamine, and 4,4'-dinonyldiphenylamine; polyalkyldiphenylamine compounds such as tetrabutyldiphenylamine, tetrahexyldiphenylamine,

tetraoctyldipihenylamine and tetranonyldiphenylamine; and naphthylamine compounds such as α -naphthylamine, phenyl- α -naphthylamine, butylphenyl- α -naphthylamine, pentylphenyl- α -naphthylamine, heptylphenyl- α -naphthylamine, octylphenyl- α -naphthylamine and nonylphenyl- α -naphthylamine.

[0030] Examples of the phenolic antioxidant include: monophenol compounds such as 2,6-di-tert-butyl-4-methylphenol and 2,6-di-tert-butyl-4-ethylphenol; and diphenol compounds such as 4,4'-methylenebis(2,6-di-tert-butylphenol) and 2,2'-methylenebis(4-ethyl-5-tert-butylphenol).

[0031] Examples of the sulfur antioxidant include: thioterpene compounds such as 2,6-di-tert-butyl-4-(4,6-bis(octylthio)-1,3,5-triazine-2-yl amino)phenol and a reactant of phosphorus pentasulfide and pinene; and dialkylthio dipropionates such as dilauryl thiodipropionate and distearyl thiodipropionate.

[0032] A content of the antioxidant is approximately from 0.01 mass% to 10 mass% based on the total amount of the composition, preferably approximately from 0.03 mass% to 5 mass%.

[0033] In the invention, a "lubricating oil composition provided by blending a predetermined acid phosphate and an antioxidant to a base oil" encompasses not only a "lubricating oil composition containing a base oil, a predetermined acid phosphate and an antioxidant" but also a composition containing a modified substance of at least one of the "base oil," "predetermined acid phosphate" and "antioxidant" in place of the at least one of the "base oil," "predetermined acid phosphate" and a composition containing a reaction product obtained by reacting the "base oil," "predetermined acid phosphate" and "antioxidant."

Other Components

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[0034] The present composition may be added as needed with other additives such as a viscosity index improver, a pour point depressant, an antiwear agent, a friction modifier, a metal detergent, an ashless dispersant, a rust inhibitor, a metal deactivator and an antifoaming agent as long as advantages of the invention are not hampered. Moreover, the present composition containing the above additives encompasses a composition containing a modified substance of the additives and a composition containing a reaction product obtained by reacting the additives.

[0035] Examples of the viscosity index improver include polymethacrylate, dispersed polymethacrylate, olefin copolymer (e.g. ethylene-propylene copolymer), dispersed olefin copolymer and styrene copolymer (e.g. styrene-diene copolymer and styrene-isoprene copolymer). A content of the viscosity index improver is approximately in a range from 0.5 mass% to 15 mass% of the total amount of the present composition in view of the blending effect thereof.

[0036] An example of the pour point depressant is polymethacrylate having a mass average molecular weight approximately in a range from 10000 to 150000. A preferable content of the pour point depressant is approximately in a range from 0.01 mass% to 10 mass% of the total amount of the present composition.

[0037] Examples of the antiwear agent include: a sulfur antiwear agent such as a thiophosphoric acid metal salt (e.g., Zn, Pb and Sb) and a thiocarbamic acid metal salt (e.g., Zn); and a phosphorus antiwear agent such as a phosphate (tricresyl phosphate). A preferable content of the antiwear agent is approximately in a range from 0.05 mass% to 5 mass% of the total amount of the present composition.

[0038] Examples of the friction modifier include a polyhydric alcohol partial ester such as neopentyl glycol monolaurate, trimethyrol propanemonolaurate, and glycerin monooleate (oleic acid monoglyceride). A preferable content of the friction modifier is approximately in a range from 0.05 mass% to 4 mass% of the total amount of the present composition.

[0039] The metal detergent is preferably at least one of metal salicylate, metal phenate and metal sulfonate. The metal is preferably alkaline earth metal, more preferably Ca. Ca salicylate is particularly preferable in order to retain the detergency. A base value of the metal detergent according to a hydrochloric acid method is preferably in a range from 100 mgKOH/g to 250 mgKOH/g in order to retain the detergency. A content of the metal detergent is preferably in a range from 60 mass ppm to 6000 mass ppm based on the total amount of the composition and in terms of the metal amount.

[0040] Examples of the ashless dispersant include succinimides, boron-containing succinimides, benzylamines, boron-containing benzylamines, succinic acid esters, and mono- or di-carboxylic acid amides respectively represented by a fatty acid or succinic acid. A preferable content of the ashless dispersant is approximately in a range from 0.1 mass% to 20 mass% of the total amount of the present composition.

[0041] Examples of the rust inhibitor include a fatty acid, alkenylsuccinic acid half ester, fatty acid soap, alkyl sulfonate, fatty acid ester of polyhydric alcohol, fatty acid amide, oxidized paraffin and alkyl polyoxyethylene ether. A preferable content of the rust inhibitor is approximately in a range from 0.01 mass% to 3 mass% of the total amount of the present composition.

[0042] One of the metal deactivators such as benzotriazole and thiadiazole may be used alone, or a combination of two or more thereof may be used. A preferable content of the metal deactivator is approximately in a range from 0.01 mass% to 5 mass% of the total amount of the present composition.

[0043] One of the antifoaming agents such as a silicone compound and an ester compound may be used alone, or a combination of two or more thereof may be used. A preferable content of the antiwear agent is approximately in a range from 0.05 mass% to 5 mass% of the total amount of the present composition.

[0044] Since the present composition contains the acid phosphate having a predetermined structure and the antioxidant, the friction coefficient between the rubber material and the metal is small, the friction energy is large, and the oxidation stability is excellent. Accordingly, the present composition is suitably usable for the shock absorber, particularly suitably usable for the shock absorber of a four-wheel automobile (e.g., a passenger car, bus and truck) in which riding comfort is of great interest.

[0045] Moreover, the present composition is also suitably usable for a shock absorber of a two-wheel vehicle. Further, the present composition is suitably usable as hydraulic fluid. Examples

[0046] The invention will be further described in detail below by reference to Examples and Comparatives, which by no means limit the invention. Properties and performance of the lubricating oil composition (sample oil) in each of Examples and Comparatives were obtained in accordance with the following methods.

(1) Kinematic Viscosity at 40 Degrees C

[0047] A kinematic viscosity at 40 degrees C of the present composition was measured in accordance with JIS K 2283.

(2) increase Ratio of Kinematic Viscosity at 40 Degrees C

[0048] After the present composition was subjected to an ISOT test (in accordance with JIS K 2514: 130°C, 24 hours), a kinematic viscosity at 40 degrees C of the present composition was measured. An increase ratio in the kinematic viscosity at 40 degrees C of the present composition after the ISOT test relative to the present composition before the ISOT test was obtained.

- (3) Friction Coefficient and Friction Energy
- 25 [0049] The friction coefficient (kinematic friction coefficient) between the rubber and the metal and the friction energy were obtained using a tester shown in Fig. 4. Specifically, as shown in Fig. 4, rubber and a Cr (chrome)-plated steel plate were reciprocatingly slid while being in pressure contact with each other at a predetermined load through a sample oil, and a Lissajous waveform was recorded. Fig. 5 shows an example of the Lissajous waveform. A friction coefficient (μ) was obtained from the maximum value of a friction force. An area (which is a product of an amplitude and the friction force and corresponds to workload) of the Lissajous waveform was obtained as the friction energy.

Test conditions are shown below. Temperature: 30 degrees C Amplitude: ±0.4 mm (sine wave)

Load: 3 kgf (29.4N)

Upper test piece: NBR (nitrile rubber)
Lower test piece: hard Cr-plated steel plate

Excitation Frequency: 5Hz Sample oil amount: 100 mL

Examples 1 to 2, Comparatives 1 to 12

[0050] Lubricating oil compositions were prepared according to blending compositions shown in Tables 1 and 2. Properties and performance of each of the sample oils were evaluated in accordance with the above-described methods. The results are also shown in Tables 1 and 2.

Table 1

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Table 2

			Comparative 6	Comparative 7	Comparative 8	Comparative 9	Comparative 10	Comparative 11	Comparative 12
Ba	Base oils	paraffinic mineral oil ¹⁾	99.00	00'66	99.00	99.00	00.66	00'66	99.00
		antioxdant ²⁾	0.50	0.50	0.50	0.50	0.50	0:20	0.50
		ditetracosyl acid phosphate	ı	1	ı	ı	ı	ı	ı
and the same of th		dioctacosyl acid phosphate	ı	ı	ı	ı	ı	ı	1
		N-oleylsarcosine	1	1	ı	ı	ı	ı	1
noi		monooleyl amine	ı	ı	ŀ	i ana	-	man.	1
tisoo		oleyl alcohol	ı	1	1		-	_	1
% s		oleic acid	I	ı	ı	ļ	l	_	1
	Additives	monoglyceride oleate	0.50	1	ı	ı	ı	ı	1
nibn		dioleyl acid phosphate	ı	0.50	1	I	I	ı	1
Ble		dioleyl hydrogen phosphite	1	1	0.50		ı	1	
		acid phosphate amine salt ³⁾	ı	ı	1	0.50	ı	_	I
		ZnDTP ⁴⁾	ı	ı	ľ	ı	0.50	_	1
		Z _n DTP ⁵⁾	1	1	,	ı	ı	0.50	1
www.com		distearyl acid phosphate		1	1	ı	I	*	0.50
		total	100.00	100.00	100.00	100.00	100.00	100.00	100.00
		friction coefficient	0.265	0.226	0.250	0.358	0.275	0.345	0.321
Evaluation Results	Results	friction energy (kgf·mm)	0.468	0.549	0.372	0.592	0.601	0.539	0.401
****		increase ratio in 40°C KV (%)	9.0+	+2.7	9'0-	+2.0	+1.2	+0.1	-0.2

[0051]

¹⁾ Base oil: paraffinic mineral oil (60N, a kinematic viscosity at 40 degrees C of 7.8 mm²/s)

- 2) Antioxidant: DBPC (2,6-di-tert-butyl-p-cresol)
- 3) Acid phosphate amine salt: An alkyl group of phosphate is mainly exemplified by a monoethyl group and a monomethyl group.
- 4) ZnDTP: in a type of a primary alkyl group having 12 carbon atoms
- 5) ZnDTP: in a type of a primary alkyl group having 6 carbon atoms (having a small amount of an isopropyl group and isobutyl group)

Evaluation Results

[0052] As understood from Examples 1 and 2, in the sample oils containing a predetermined acid phosphate and the antioxidant, the friction coefficient between the rubber material and the metal is small, the friction energy is large, and the oxidation stability is excellent. According to the invention, it is understandable that a shock absorber oil providing an excellent riding comfort and usable for a long period of time can be provided.

[0053] In contrast, the sample oils of Comparatives, in which various agents such as an oiliness agent and a phosphorus extreme pressure agent are blended, cannot simultaneously satisfy all of the friction coefficient, the friction energy and the oxidation stability. For instance, the sample oils in Comparatives 2, 3, 5 and 10 exhibit poor oxidation stability in spite of containing the same amount of the same antioxidant as in Examples 1 and 2. In Comparative 12, the sample oil contains acid phosphate diester having an alkyl group with a relatively large number of carbon atoms. Since the number of the carbon atoms of the alkyl group is 18, which is smaller than the lower limit of the number of the carbon atoms of the alkyl group in the invention, the friction coefficient is large and an increase in the friction energy is hardly recognized as compared with the sample oil with only the base oil (i.e., Comparative 1 only containing the antioxidant).

Claims

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1. A lubricating oil composition comprising:

a base oil;

at least one of acid phosphate represented by formulae (1) and (2) below; and an antioxidant.

[Formula 1]

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$$\begin{array}{c}
O \\
R^{1}O - P - OH \\
OR^{2}
\end{array}$$
(1)

40

$$\begin{array}{ccc}
O \\
R^3O - P - (OH)_2
\end{array}$$
(2)

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where: R¹, R² and R³ are alkyl groups; and at least one of R¹ and R² has 20 to 30 carbon atoms while R³ has 20 to 30 carbon atoms.

- 2. The lubricating oil composition according to claim 1, wherein at least one of R¹ and R² has a side chain.
- The lubricating oil composition according to claim 1, wherein R³ has a side chain.
- The lubricating oil composition according to claim 2 or 3, wherein the side chain has 6 to 18 carbon atoms.
 - 5. The lubricating oil composition according to any one of claims 1 to 4, wherein

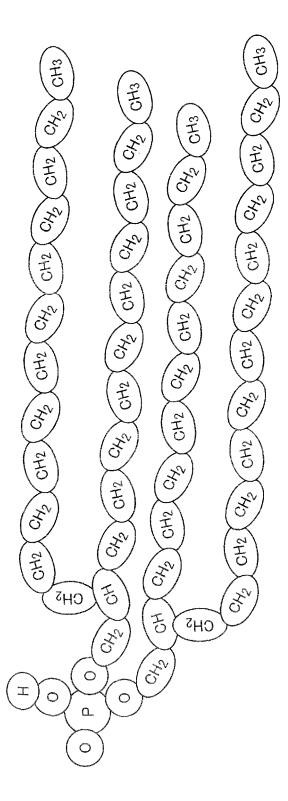
the antioxidant is at least one of a phenolic antioxidant, an amine antioxidant and a sulfur antioxidant.

6.	The lubricating oil composition according to any one of claims 1 to 5, wherein
	the acid phosphate is contained in a range from 0.01 mass% to 3 mass% of a total amount of the composition.

7. The lubricating oil composition according to any one of claims 1 to 6. wherein the lubricating oil composition is suitable for use in a shock absorber.

8.	The lubricating oil composition according to clair	7, wherein the shock absorber is suitable for use in a four-whee
	automobile.	

<u>Б</u>



EF3

CH3 CF3 CH3 CF3 1 cho, CF3 10401 CH3 , Ch2 > CH2 , C/45 , CH2, CH2, , CH2 1 1 640 CHZ , Ch2, CH2, CH2, 1 ch2 1 CH2 1 CH2 Y , CH2 1 CH2, CHZ , CH2 1 , CH2 1 CH2, , Ch2, O.H.Z. CH2 , C/42 , CHZ CHO , CH2 , CHZ CH2 . CHZ CH2 1 CHZ 5 CH^{5} CH^{5} \circ I 0 0 ۵. 0

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FIG.3

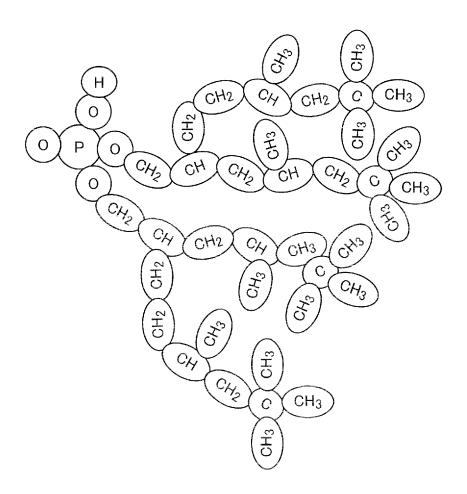


FIG.4

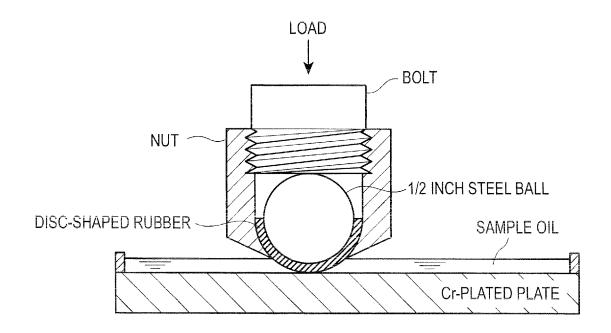
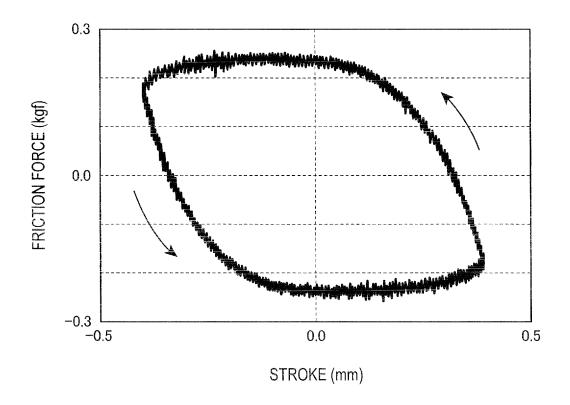


FIG.5



INTERNATIONAL SEARCH REPORT International application No. PCT/JP2015/064722 A. CLASSIFICATION OF SUBJECT MATTER C10M169/04(2006.01)i, C10M101/02(2006.01)i, C10M129/10(2006.01)i, 5 C10M133/12(2006.01)i, C10M137/04(2006.01)i, C10N30/06(2006.01)n, C10N30/10(2006.01)n, C10N40/06(2006.01)n According to International Patent Classification (IPC) or to both national classification and IPC FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) 10 C10M169/04, C10M101/02, C10M129/10, C10M133/12, C10M137/04, C10N30/06, C10N30/10, C10N40/06 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 1996-2015 15 Kokai Jitsuyo Shinan Koho 1971-2015 Toroku Jitsuyo Shinan Koho 1994-2015 Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) CAplus/REGISTRY(STN) DOCUMENTS CONSIDERED TO BE RELEVANT 20 Category* Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. Α JP 2011-68801 A (JX Nippon Oil & Energy Corp.), 1 - 807 April 2011 (07.04.2011), claims 1 to 4; paragraph [0021]; examples 1 to 25 & WO 2011/036919 A1 JP 2009-235268 A (Nippon Oil Corp.), Α 1 - 815 October 2009 (15.10.2009), claims 1 to 4; paragraphs [0096] to [0100]; 30 examples 1 to 7 & US 2011/0021394 A1 & WO 2009/119332 A1 & KR 10-2010-0125289 A & CN 101981170 A 35 |X|Further documents are listed in the continuation of Box C. See patent family annex. 40 Special categories of cited documents: later document published after the international filing date or priority "A" document defining the general state of the art which is not considered to be of particular relevance date and not in conflict with the application but cited to understand the principle or theory underlying the invention "E" earlier application or patent but published on or after the international filing 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 "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) 45 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 "O" document referring to an oral disclosure, use, exhibition or other means being obvious to a person skilled in the art document published prior to the international filing date but later than the document member of the same patent family priority date claimed Date of the actual completion of the international search Date of mailing of the international search report 50 06 August 2015 (06.08.15) 18 August 2015 (18.08.15) Name and mailing address of the ISA/ Authorized officer Japan Patent Office 3-4-3, Kasumigaseki, Chiyoda-ku, Tokyo 100-8915, Japan 55 Telephone No.

Form PCT/ISA/210 (second sheet) (July 2009)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2015/064722

	C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT				
5	Category*	Citation of document, with indication, where appropriate, of the relevan	nt passages	Relevant to claim No.	
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REFERENCES CITED IN THE DESCRIPTION

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- Proceedings of Petroleum Product Symposium of Petroleum Institute, December 2009, 96 [0005]
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