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- (54) LUBRICATING OIL COMPOSITION, MECHANICAL DEVICE EQUIPPED WITH LUBRICATING OIL COMPOSITION, AND METHOD FOR PRODUCING LUBRICATING OIL COMPOSITION
- (57) A lubricating oil composition containing a lubricating base oil (A), a neutral phosphorus-based compound (B), an acidic phosphorus-based compound (C), a sulfur-based compound (D) and an organic molybdenum compound (E).

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### Description

Technical Field

[0001] The present invention relates to a lubricating oil composition, a mechanical device including the lubricating oil composition, and a method for producing the lubricating oil composition.

**Background Art** 

10 [0002] In recent years, reduction of carbon dioxide has been strongly required from the viewpoint of protection of the global environment, and thus efforts have been put into development of a fuel saving technology in the field of automobile. Examples of a fuel saving automobile include a hybrid automobile and an electric automobile, and it is expected that these automobiles will rapidly become widespread in the future. A hybrid automobile and an electric automobile are provided with an electric motor, a generator, an inverter, a storage battery etc., and travel by utilizing power generated 15 by an electric motor.

[0003] For cooling of an electric motor and a generator in such a hybrid automobile and an electric automobile, existing automatic transmission fluid (hereinafter, referred to as ATF) and continuously variable transmission fluid (hereinafter, referred to as CVTF) are primarily used. Since some hybrid automobiles and electric automobiles are in the form of those having gear reducers, a lubricating oil composition is required to have both cooling ability and lubricating ability.

[0004] Therefore, a lubricating oil composition has been proposed which is obtained by blending a base oil, a neutral phosphorus-based compound, at least one acidic phosphorus-based compound selected from a group consisting of an amine salt of an acidic phosphoric acid ester having a specified structure and an acidic phosphorous acid ester having a specified structure, and a sulfur-based compound (Patent Literature 1: International publication No. WO11/080970).

25 Citation List

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Patent Literature

[0005] Patent Literature 1 International Publication No. WO11/080970

Summary of Invention

**Technical Problem** 

[0006] However, although volume resistivity, abrasion resistance between metals and solubility were improved in the lubricating oil composition described in Patent Literature 1, a lubricating oil composition satisfying all of abrasion resistance, seizure resistance and low friction at a higher level is required. Furthermore, a lubricating oil composition having even higher cooling ability is also required.

Solution to Problem

[0007] Therefore, the present inventors have solved the problem to be solved by the present invention by further adding an organic molybdenum compound to a lubricating oil composition comprising a base oil, a neutral phosphorusbased compound, an acidic phosphorus-based compound and a sulfur-based compound.

[0008] The present invention comprises the inventions of the following aspects.

- [1] A lubricating oil composition, comprising a lubricating base oil (A), a neutral phosphorus-based compound (B), an acidic phosphorus-based compound (C), a sulfur-based compound (D) and an organic molybdenum compound (E).
- [2] A mechanical device, comprising the lubricating oil composition according to [1].
- [3] A method for producing a lubricating oil composition, comprising a step of mixing a lubricating base oil (A), a neutral phosphorus-based compound (B), an acidic phosphorus-based compound (C), a sulfur-based compound (D) and an organic molybdenum compound (E).

Advantageous Effects of Invention

[0009] The lubricating oil composition according to an aspect of the present invention exhibits excellent properties

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with respect to all of abrasion resistance, seizure resistance, and low friction. The lubricating oil composition according to an aspect of the present invention further has an excellent cooling ability.

### Description of Embodiments

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**[0010]** Hereinafter, the embodiments of the present invention will be described in detail. The present invention is not limited to the following embodiments, and optionally changes may be made in the range without departing from the gist of the invention to practice the present invention. All documents and publications cited in the present specification are incorporated herein by reference in their entirety regardless of the purpose.

**[0011]** The lubricating oil composition of the present invention comprises a lubricating base oil (A), a neutral phosphorus-based compound (B), an acidic phosphorus-based compound (C), a sulfur-based compound (D) and an organic molybdenum compound (E).

In the lubricating oil composition of the present invention, the total content of a lubricating base oil (A), a neutral phosphorus-based compound (B), an acidic phosphorus-based compound (C), a sulfur-based compound (D) and an organic molybdenum compound (E) is preferably 70 mass% or more, more preferably 80 mass% or more, still more preferably 85 mass% or more, even more preferably 90 mass% or more based on the total amount of the composition.

[0012] Hereinafter, each component contained in the lubricating oil composition will be described in detail.

#### [Lubricating base oil (A)]

[0013] The lubricating base oil (A) (hereinafter, also simply referred to as "base oil") contained in the lubricating oil composition is not particularly limited as long as it is an oil having lubricity, and may be a mineral oil or a synthetic oil. The types of these base oils are not particularly limited, and any type of oil may be appropriately selected from mineral oils and synthetic oils which are conventionally used as a base oil of a lubricating oil for a transmission for an automobile, and may be used.

**[0014]** Examples of mineral oils include, for example, a refined mineral oil or a wax obtained by refining a lubricating oil fraction obtained by reduced-pressure distillation of an atmospheric residue obtained by atmospheric pressure distillation of crude oil, using one or more treatment process of solvent deasphalting, solvent extraction, hydrocracking, solvent dewaxing, contact dewaxing, hydrorefining etc.; and a mineral oil (GTL) produced by isomerizing GTL WAX (gas-to-liquid wax) produced from natural gas by Fischer Tropsh process. Among these, a refined mineral oil obtained by treatment process of hydrorefining and a mineral oil (GTL) produced by isomerizing GTL WAX are preferable from the viewpoint of %C<sub>P</sub> and viscosity index described below.

[0015] Examples of synthetic oils include, for example, polybutene; poly- $\alpha$ -olefins such as an  $\alpha$ -olefin homopolymer and an  $\alpha$ -olefin copolymer (for example, ethylene- $\alpha$ -olefin copolymer); various esters such as a polyol ester, a dibasic acid ester and a phosphoric acid ester; various ethers such as polyphenyl ether; polyglycol; alkylbenzene; and alkylnaphthalene. Among these synthetic oils, poly- $\alpha$ -olefins and esters are preferable. These synthetic oils may be used alone or in combination of two or more.

**[0016]** The above base oil may contain one mineral oil or may contain two or more mineral oils. For the base oil, one synthetic oil may be used, or two or more synthetic oils may be used in combination. The above base oil may contain one or more mineral oils and one or more synthetic oils.

**[0017]** The base oil is a main component of the lubricating oil composition, and the content of the base oil is usually preferably 65-98 mass%, more preferably 70-97 mass%, still more preferably 75-96 mass% based on the total amount of the composition.

[0018] Although the flash point of the lubricating base oil (A) is not limited, it is preferable to use a base oil having a high flash point since the obtained lubricating oil composition tends to also have a higher flash point. Specifically, the flash point of the lubricating base oil (A) is preferably 172°C or higher, still more preferably the flash point is 174°C or higher, particularly preferably the flash point is 176°C or higher. When the lubricating base oil (A) contains more than one mineral oils or synthetic oils etc., all of these mineral oils or synthetic oils are not required to have a flash point of 172°C or higher, but it is sufficient that the lubricating base oil (A) obtained by mixing these oils has a flash point of 172°C or higher.

**[0019]** The flash point as used herein means the value obtained by measurement using COC method according to JIS-K-2265-4.

**[0020]** The viscosity of the base oil is not particularly limited, and the kinetic viscosity at a temperature of 100°C is preferably 21-30 mm²/s, more preferably 21.5-15 mm²/s, still more preferably 2-10 mm²/s though it varies depending on the intended use of the lubricating oil composition. When the kinetic viscosity at 100°C is 2 mm²/s or more, vaporization loss is lower. When the kinetic viscosity is 30 mm²/s or less, power loss due to viscous resistance is lower, which results in the effect of improvement of fuel efficiency.

[0021] The kinetic viscosity of the base oil at 40°C is not particularly limited, and preferably 5-65 mm<sup>2</sup>/s, more preferably

8-40 mm²/s, still more preferably 10-25 mm²/s. When the kinetic viscosity at 40°C is 5 mm²/s or more, vaporization loss is lower. When the kinetic viscosity is 65 mm²/s or less, power loss due to viscous resistance is lower, which results in the effect of improvement of fuel efficiency.

[0022] Herein, "kinetic viscosity at 100°C" and "kinetic viscosity at 40°C" can be measured by the method according to JIS-K-2283:2000. When the lubricating base oil (A) contains two or more oils, "kinetic viscosity at 100°C" and "kinetic viscosity at 40°C" mean the kinetic viscosity of the whole mixed base oil.

**[0023]** The viscosity index of the base oil is not particularly limited, and preferably 70 or more, more preferably 80 or more, still more preferably 90 or more. In the base oil having the above viscosity index of 70 or more, viscosity change due to temperature change is smaller. When the viscosity index of the base oil is within the above range, better viscosity properties of the lubricating oil composition can be obtained, and the effect of improvement of fuel efficiency can be obtained. Herein, "viscosity index" can be calculated by the method according to JIS-K-2283:2000.

**[0024]** The content of aromatic moieties ( ${}^{\circ}C_A$ ) of the base oil obtained by ring analysis and the sulfur content are not particularly limited, and the base oil having  ${}^{\circ}C_A$  of 3.0 or less and the sulfur content of 10 mass ppm or less is preferably used. In this context,  ${}^{\circ}C_A$  obtained by ring analysis refers to a ratio of aromatic moieties (percentage) calculated by the ring analysis n-d-M method in which measurement is conducted according to ASTM D 3238. The base oil having the above  ${}^{\circ}C_A$  of 3.0 or less and the sulfur content of 10 mass ppm or less can provide a lubricating oil composition having good oxidation stability and capable of suppressing increase in acid value and production of sludge. More preferably  ${}^{\circ}C_A$  is 1.0 or less, still more preferably  ${}^{\circ}C_A$  is 0.5 or less. More preferably the sulfur content is 7 mass ppm or less, still more preferably the sulfur content is 5 mass ppm or less.

**[0025]** The paraffin content ( ${}^{\circ}C_P$ ) of the base oil obtained by ring analysis is not particularly limited, and preferably 70 or more, more preferably 75 or more, still more preferably 79 or more. When the above  ${}^{\circ}C_P$  is 70 or more, the oxidation stability of the base oil is better. The upper limit of  ${}^{\circ}C_P$  is not particularly limited, and is for example 98 or less. In this context,  ${}^{\circ}C_P$  obtained by ring analysis refers to a ratio of paraffin component (percentage) calculated by the ring analysis n-d-M method in which measurement is conducted according to ASTM D 3238.

**[0026]** NOACK evaporation amount of the base oil is not particularly limited, and preferably 15.0 mass% or less, more preferably 14.0 mass% or less, more preferably 13.0 mass% or less. NOACK evaporation amount can be measured according to ASTM D 5800 (250°C, 1 hour).

[Neutral phosphorus-based compound (B)]

**[0027]** The neutral phosphorus-based compound (B) is added for the purpose of improving the abrasion resistance between metals. The abrasion resistance between metals cannot be enhanced without using the neutral phosphorus-based compound (B).

**[0028]** The neutral phosphorus-based compound (B) is not particularly limited as long as it is a compound which is neutral and contains a phosphorus atom, but a compound represented by the following general formula (2) or (3) is preferably used.

## [Formula 2]

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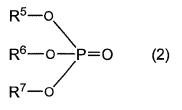
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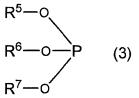
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**[0029]** In the above general formulae (2) and (3), hydrocarbon groups R<sup>5</sup>, R<sup>6</sup> and R<sup>7</sup> each independently represent an aryl group having 6 to 30 carbon atoms, an alkyl group having 1 to 30 carbon atoms or an alkenyl group having 2 to 30 carbon atoms, preferably an aryl group having 8 to 28 carbon atoms, an alkyl group having 2 to 28 carbon atoms or an alkenyl group having 4 to 28 carbon atoms, still more preferably represent an aryl group having 10 to 26 carbon

atoms, an alkyl group having 4 to 26 carbon atoms or an alkenyl group having 6 to 26 carbon atoms, and particularly preferably an aryl group having 12 to 24 carbon atoms, an alkyl group having 6 to 24 carbon atoms or an alkenyl group having 6 to 24 carbon atoms.  $R^5$ ,  $R^6$  and  $R^7$  may be the same or may be different.

**[0030]** Examples of the neutral phosphorus-based compound (B) include, for example, neutral aromatic phosphoric acid esters such as tricresyl phosphate, triphenyl phosphate, trixylenyl phosphate, tricresyl thiophosphate and triphenyl thiophosphate; neutral aliphatic phosphoric esters such as tributyl phosphate, tri(2-ethyl-hexyl) phosphate, tributoxy phosphate and tributyl thiophosphate; neutral aromatic phosphite esters such as triphenyl phosphite, tricresyl phosphite, trisnonylphenyl phosphite, diphenyl mono(2-ethylhexyl) phosphite, diphenyl monotridecyl phosphite, tricresyl thiophosphite and triphenyl thiophosphite; and neutral aliphatic phosphorous esters such as tributyl phosphite, trioctyl phosphite, trisdecyl phosphite, tris(trydecyl) phosphite, trioleyl phosphite, tributyl thiophosphite and trioctyl thiophosphite. Among these neutral phosphorus-based compounds, neutral aromatic phosphoric acid esters, neutral aliphatic phosphoric acid esters etc. are preferably used from the viewpoint of abrasion resistance between metals. Furthermore, these neutral phosphorus-based compounds may be used alone or may be used in combination of two or more.

[0031] The content of the neutral phosphorus-based compound (B) in the lubricating oil composition is preferably 2.5 mass% or less, more preferably 0.12 mass% or more and 2.5 mass% or less, particularly preferably 0.25 mass% or more and 1.3 mass% or less based on the total amount of the composition. When the content of the phosphorus-based compound (B) is 0.12 mass% or more based on the total amount of the composition, abrasion resistance between metals provided by the lubricating oil composition can be further enhanced. On the other hand, when the content of the neutral phosphorus-based compound (B) is 2.5 mass% or less based on the total amount of the composition, solubility of the neutral phosphorus-based compound (B) to the base oil can be enhanced.

[0032] The content of the neutral phosphorus-based compound (B) in terms of phosphorus atom is preferably 2000 mass ppm or less, more preferably 100 mass ppm or more and 2000 mass ppm or less, particularly preferably 200 mass ppm or more and 1000 mass ppm or less based on the total amount of the composition. When the content of the neutral phosphorus-based compound (B) in terms of phosphorus atom is 2000 mass ppm or less based on the total amount of the composition, solubility of the neutral phosphorus-based compound (B) to the base oil can be enhanced. When the content of the neutral phosphorus-based compound (B) in terms of phosphorus atom is 100 mass ppm or more based on the total amount of the composition, abrasion resistance between metals provided by the lubricating oil composition can be further enhanced.

[0033] Herein, the content of phosphorus atom means the value obtained by measurement according to JPI-5S-38-92.

[Acidic phosphorus-based compound (C)]

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**[0034]** The acidic phosphorus-based compound (C) is added for the purpose of enhancement of seizure resistance. When the acidic phosphorus-based compound (C) is not used, seizure resistance cannot be enhanced.

**[0035]** The acidic phosphorus-based compound (C) is not particularly limited as long as it is a compound which is acidic and contains a phosphorus atom, but is preferably at least one acidic phosphorus-based compound selected from the group consisting of acidic phosphoric acid esters represented by the following general formula (4) and the group consisting of acidic phosphorous acid esters represented by the following general formula (5).

[Formula 3]

$$R^8 - O$$
 P—OH (5)

[0036] In the above general formula (4) and the above general formula (5),  $R^B$  and  $R^9$  represent a hydrogen atom or a hydrocarbon group having 8 to 30 carbon atoms.  $R^8$  and  $R^9$  may be the same or may be different. At least one of  $R^8$  and  $R^9$  is a hydrocarbon group having 8 to 30 carbon atoms, preferably both of them are hydrocarbon groups having 8

to 30 carbon atoms, still more preferably 10 to 28 carbon atoms, particularly preferably 12 to 26 carbon atoms. When the number of carbon atoms in the above hydrocarbon group is 8 or more, oxidation stability of the lubricating oil composition enhances, and on the other hand, when the number or carbon atoms in the above hydrocarbon group is 30 or less, seizure resistance between metals becomes satisfactory. Examples of hydrocarbon groups of R<sup>8</sup> and R<sup>9</sup> include, for example, an alkyl group, an alkenyl group, an aryl group, an alkylaryl group and arylalkyl group.

**[0037]** Examples of acidic phosphoric acid esters represented by the above general formula (4) and amine salts thereof include, for example, acidic aliphatic phosphoric acid esters such as di-2-ethylhexyl acid phosphate, dilauryl acid phosphate and dioleyl acid phosphate; acidic aromatic phosphoric acid esters such as diphenyl acid phosphate and dicresyl acid phosphate; and sulfur-containing acidic phosphoric acid esters such as S-octyl thioethyl acid phosphate and S-dodecyl thioethyl acid phosphate. These acidic phosphoric acid esters and amine salts thereof may be used alone or may be used in combination of two or more.

**[0038]** Examples of acidic phosphorous acid esters represented by the above general formula (5) and amine salts thereof include, for example, acidic aliphatic phosphorous acid esters such as dibutyl hydrogen phosphite, di-2-ethylhexyl hydrogen phosphite, dilauryl hydrogen phosphite and dioleyl hydrogen phosphite; acidic aromatic phosphite esters such as diphenyl hydrogen phosphite and dicresyl hydrogen phosphite; and sulfur-containing acidic phosphorous acid esters such as S-octyl thioethyl hydrogen phosphite and S-dodecyl thioethyl hydrogen phosphite. The lubricating oil composition may contain these acidic phosphorous acid esters as amine salts thereof. These acidic phosphorous acid esters and amine salts thereof may be used alone or may be used in combination of two or more.

**[0039]** In the lubricating oil composition, the content of the acidic phosphorus-based compound (C) is preferably 0.8 mass% or less, more preferably 0.1 mass% or more and 0.8 mass% or less, particularly preferably 0.1 mass% or more and 0.5 mass% or less based on the total amount of the composition. When the content of the acidic phosphorus-based compound (C) is 0.8 mass% or less based on the total amount of the composition, volume resistivity of the lubricating oil composition may become satisfactory. When the content of the acidic phosphorus-based compound (C) is 0.1 mass% or more based on the total amount of the composition, seizure resistance between metals provided by the lubricating oil composition can be further enhanced.

**[0040]** The content of the acidic phosphorus-based compound (C) in terms of phosphorus atom is preferably 400 mass ppm or less, more preferably 50 mass ppm or more and 400 mass ppm or less, particularly preferably 50 mass ppm or more and 250 mass ppm or less based on the total amount of the composition. When the content of the acidic phosphorus-based compound (C) in terms of phosphorus atom is 400 mass ppm or less based on the total amount of the composition, volume resistivity of the lubricating oil composition may become satisfactory. When the content of the acidic phosphorus-based compound (C) in terms of phosphorus atom is 50 mass ppm or more based on the total amount of the composition, abrasion resistance between metals provided by the lubricating oil composition may be further enhanced.

[Sulfur-based compound (D)]

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**[0041]** The sulfur-based compound (D) is added for the purpose of enhancement of seizure resistance. When the sulfur-based compound (D) is not used, seizure resistance may not be enhanced.

**[0042]** The sulfur-based compound (D) is not particularly limited as long as it is a compound which contains a sulfur atom. As the sulfur-based compound (D), a known compound can be used, but specifically, a thiadiazole-based compound, a polysulfide-based compound, a thiocarbamate-based compound, a sulfurized fat and oilbased compound, a sulfurized olefin-based compound etc. can be exemplified. Among these sulfur-based compounds, a thiadiazole-based compound and a polysulfide-based compound are preferable from the viewpoint of seizure resistance between metals and abrasion resistance between metals. These sulfur-based compounds may be used alone or may be used in combination of two or more.

**[0043]** As the above thiadiazole-based compound, a known compound can be used as necessary, but for example, a compound represented by the following general formula (6) can be exemplified.

$$R^{10}$$
- $(S)_{X1}$ - $C$ - $C$ - $C$ - $C$ - $(S)_{X2}$ - $R^{11}$  (6)

**[0044]** In the above general formula (6), R<sup>10</sup> and R<sup>11</sup> each independently represent an alkyl group having 1 to 30 carbon atoms, but are preferably alkyl groups having 6 to 20 carbon atoms, still more preferably alkyl groups having 8 to 18 carbon atoms. The alkyl group may be linear or branched. R<sup>10</sup> and R<sup>11</sup> may be the same or may be different. X1

and X2 each independently represent an integer of 1 to 3, and refer to the number of sulfur atoms, but a compound having the number of sulfur atoms of 2 is preferably used.

[0045] As a thiadiazole-based compound represented by the above general formula (6), 2,5-bis(n-hexyldithio)-1,3,4-thiadiazole, 2,5-bis(n-octyldithio)-1,3,4-thiadiazole, 2,5-bis(n-octyldithio)-1,3,4-thiadiazole, 2,5-bis(n-octyldithio)-1,3,4-thiadiazole, 3,5-bis(n-hexyldithio)-1,2,4-thiadiazole, 3,6-bis(n-octyldithio)-1,2,4-thiadiazole, 3,5-bis(n-octyldithio)-1,2,4-thiadiazole, 3,5-bis(n-octyldithio)-1,2,3-thiadiazole, 4,5-bis(n-octyldithio)-1,2,3-thiadiazole and 4,5-bis(1,1,3,3-tetramethylbutyldithio)-1,2,3-thiadiazole are preferable, 2,5-bis(n-hexyldithio)-1,3,4-thiadiazole, 2,5-bis(n-nonyldithio)-1,3,4-thiadiazole are more preferable, and 2,5-bis(1,1,3,3-tetramethylbutyldithio)-1,3,4-thiadiazole is particularly preferable.

**[0046]** As the above polysulfide-based compound, a known compound can be used as necessary, but for example, a compound represented by the following general formula (7) can be exemplified.

$$R^{12}$$
- $(S)_{v}$ - $R^{13}$  (7)

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**[0047]** In the above general formula (7),  $R^{12}$  and  $R^{13}$  each independently represent an alkyl group having 1 to 24 carbon atoms, an aryl group having 6 to 20 carbon atoms or an alkylaryl group having 7 to 20 carbon atoms. The number of carbon atoms in the alkyl group is preferably 3 or more and 20 or less, still more preferably 6 or more and 16 or less. The number of carbon atoms in the aryl group is preferably 6 or more and 20 or less, still more preferably 6 or more and 16 or less. The number of carbon atoms in the alkylaryl group is preferably 8 or more and 20 or less, still more preferably 9 or more and 18 or less.  $R^{12}$  and  $R^{13}$  may be the same or may be different.

**[0048]** Y represents the number of sulfur atoms, and is preferably an integer of 2 or more and 8 or less taking into consideration of abrasion resistance, fatigue life, availability and corrosion, etc., more preferably an integer of 2 or more and 7 or less, still more preferably an integer of 2 or more and 6 or less.

**[0049]** Examples of a group represented by R<sup>12</sup> and R<sup>13</sup> include aryl groups such as a phenyl group, a naphthyl group, a benzyl group, a tolyl group and a xyl group; and alkyl groups such as a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, a hexyl group, an octyl group, a nonyl group, a decyl group, a dodecyl group, a cyclohexyl group and a cyclooctyl group. These groups may be linear or branched. These groups may be used alone or may be used in combination of two or more.

**[0050]** Among the polysulfide-based compounds represented by the above general formula (7), dibenzyl polysulfide, di-tert-nonyl polysulfide, didodecyl polysulfide, di-tert-butyl polysulfide, dioctyl polysulfide, diphenyl polysulfide and dicyclohexyl polysulfide are more preferred, and disulfides of these compounds are particularly preferred.

**[0051]** In the lubricating oil composition, the content of the sulfur-based compound (D) is preferably 0.3 mass% or less, more preferably 0.03 mass% or more and 0.3 mass% or less, particularly preferably 0.03 mass% or more and 0.15 mass% or less based on the total amount of the composition. When the content of the sulfur-based compound (D) is 0.3 mass% or less based on the total amount of the composition, volume resistivity of the lubricating oil composition can be expected to be maintained. When the content of the sulfur-based compound (D) is 0.03 mass% or more based on the total amount of the composition, seizure resistance between metals provided by the lubricating oil composition can be further enhanced.

[0052] The content of the sulfur-based compound (D) in terms of sulfur atom preferably 1000 mass ppm or less based on the total amount of the composition, more preferably 125 mass ppm or more and 1000 mass ppm or less and furthermore, particularly preferably 125 mass ppm or more and 500 mass ppm or less from the viewpoint of achievement of both volume resistivity and seizure resistance of the lubricating oil composition. When the content of the sulfur-based compound (D) in terms of sulfur atom is 1000 mass ppm or less based on the total amount of the composition, volume resistivity of the lubricating oil composition can be expected to allow to be maintained. When the content of the sulfur-based compound (D) in terms of sulfur atom is 125 mass ppm or more based on the total amount of the composition, seizure resistance between metals provided by the lubricating oil composition can be further enhanced.

[0053] As used herein, the content of sulfur refers to the value obtained by measurement according to JIS K2541-6.

[Organic molybdenum compound (E)]

**[0054]** The lubricating oil composition is characterized in that it further contains the organic molybdenum compound (E) in addition to the lubricating base oil (A), the neutral phosphorus-based compound (B), the acidic phosphorus-based compound (C) and the sulfur-based compound (D). By adding the organic molybdenum compound (E), the lubricating oil composition can provide low friction in addition to seizure resistance and abrasion resistance. When the organic molybdenum compound (E) is not used, low friction may not be provided.

**[0055]** As the molybdenum-based friction modifier (E), any organic compound having a molybdenum atom can be used, but the organic compound represented by formula (1) is preferable.

[Formula 5]

**[0056]** In formula (1), R<sup>1</sup> to R<sup>4</sup> are each independently a hydrocarbon group having 4 to 18 carbon atoms, preferably an alkyl group having 4 to 18 carbon atoms, an alkenyl group having 4 to 18 carbon atoms, a cycloalkyl group having 4 to 18 carbon atoms, an aryl group having 6 to 18 carbon atoms, an alkylaryl group having 7 to 18 carbon atoms or an arylalkyl group having 7 to 18 carbon atoms.

**[0057]** Examples of hydrocarbon group of R¹ to R⁴ include, for example, alkyl groups having 5 to 18 carbon atoms such as pentyl group, hexyl group, heptyl group, octyl group, nonyl group, decyl group, undecyl group, dodecyl group, tridecyl group, tetradecyl group, pentadecyl group, hexadecyl group, heptadecyl group and octadecyl group; alkenyl groups having 5 to 18 carbon atoms such as octenyl group, nonenyl group, decenyl group, undecenyl group, dodecenyl group, tridecenyl group, tetradecenyl group and pentadecenyl group; cycloalkyl groups having 5 to 18 carbon atoms such as a cyclohexyl group, a dimethylcyclohexyl group, an ethylcyclohexyl group, a methylcyclohexyl group; aryl groups having 6 to 18 carbon atoms such as a phenyl group, a naphthyl group, an anthracenyl group, a biphenyl group and a terphenyl group; alkylaryl groups such as a tolyl group, a dimethylphenyl group, a butylphenyl group, a nonylphenyl group, a methylbenzyl group and a dimethylnaphthyl group; and arylalkyl groups having 7 to 18 carbon atoms such as a phenylmethyl group, a phenylethyl group and a diphenylmethyl group.

**[0058]** In formula (1),  $X^1$  to  $X^4$  are each independently an oxygen atom or a sulfur atom. The molar ratio of sulfur atoms to oxygen atoms [sulfur atoms/oxygen atoms] in  $X^1$  to  $X^4$  is preferably 1/3-3/1, more preferably 1.5/2.5-3/1 from the viewpoint of enhancement of solubility to the lubricating base oil (A).

**[0059]** In the lubricating oil composition, the content of the organic molybdenum compound (E) is preferably 0.01 mass% or more and 1.0 mass% or less, still more preferably 0.1 mass% or more and 0.9 mass% or less, particularly preferably 0.2 mass% or more and 0.8 mass% or less based on the total amount of the lubricating oil composition from the viewpoint of achievement of low friction of the lubricating oil composition.

**[0060]** The ratio of the content (mass%) of the sulfur-based compound (D) to the content of the organic molybdenum compound (E) (sulfur-based compound (D)/organic molybdenum compound (E)) is preferably within the range of 1-10, since low friction of the lubricating oil composition is enhanced.

**[0061]** The ratio of the total content(mass%) of the acidic phosphorus-based compound (C) and the sulfur-based compound (D) to the content (mass%) of the organic molybdenum compound (E) ((acidic phosphorus-based compound (C)+sulfur-based compound (D))/organic molybdenum compound (E)) is preferably within the range of 0.3-1, since abrasion resistance of the lubricating oil composition is enhanced.

[Additives]

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**[0062]** A viscosity index enhancer, a detergent dispersant, an antioxidant, a metal deactivator, an anti-rust agent, a surfactant/demulsifying agent, an anti-foaming agent, a corrosion inhibitor, an oiliness improver, an acid scavenger, etc. can be appropriately added to the lubricating oil composition within the range not inhibiting the effect of the present invention.

**[0063]** Examples of a viscosity index enhancer include, for example, a non-dispersion type polymethacrylate, a dispersion type polymethacrylate, an olefin-based copolymer, a dispersion type olefin-based copolymer and a styrene-based copolymer. The mass average molecular weight of these viscosity index enhancers is preferably 5000 or more and 300000 or less for a dispersion type and a non-dispersion type polymethacrylate, for example. For an olefin-based copolymer, the mass average molecular weight is preferably 800 or more and 100000 or less. These viscosity index enhancers may be used alone or may be used in combination of two or more. The amount of the viscosity index enhancer to be added is not particularly limited, and preferably 0.5 mass% or more and 15 mass% or less, more preferably 1 mass% or more and 10 mass% or less based on the total amount of the composition.

[0064] As a detergent dispersant, an ashless dispersant and a metal-based detergent dispersant can be used.

**[0065]** Examples of an ashless dispersant include, for example, a succinimide compound, a boron-based imide compound, a Mannich dispersant and an acid amide-based compound. These may be used alone or may be used in combination of two or more. The amount of an ashless dispersant to be added is not particularly limited, and is preferably 0.1 mass% or more and 20 mass% or less based on the total amount of the composition.

[0066] Examples of a metal-based detergent dispersant include, for example, an alkali metal sulfonate, an alkali metal

phenate, an alkali metal salicylate, an alkali metal naphthenate, an alkali earth metal sulfonate, an alkali earth metal phenate, an alkali earth metal salicylate and an alkali earth metal naphthenate. These may be used alone or may be used in combination of two or more. The amount of a metal-based detergent dispersant to be added is not particularly limited, and is preferably 0.1 mass% or more and 10 mass% or less based on the total amount of the composition.

**[0067]** Examples of an antioxidant include, for example, an amine-based antioxidant, phenol-based antioxidant and sulfur-based antioxidant. These may be used alone or may be used in combination of two or more. The amount of an antioxidant to be added is not particularly limited, and preferably 0.05 mass% or more and 7 mass% or less based on the total amount of the composition.

**[0068]** Examples of a pour point depressant include a polymethacrylate, an ethylene-vinyl acetate copolymer, a condensate of chlorinated paraffin and naphthalene, a condensate of chlorinated paraffin and phenol, a polyalkylstyrene and a poly(meth)acrylate. The mass average molecular weight (Mw) of the pour point depressant is preferably 20,000-100,000, more preferably 30,000-80,000, still more preferably 40,000-60,000. The molecular weight distribution (Mw/Mn) is preferably 5 or less, more preferably 3 or less, still more preferably 2 or less. The content of a pour point depressant may be appropriately determined depending on the desired MRV viscosity etc., and is preferably 0.01 mass% or more and 5 mass% or less, more preferably 0.02 mass% or more and 2 mass% or less based on the total amount of the composition.

**[0069]** Examples of the metal deactivator include, for example, a benzotriazole-based metal deactivator, a tolyltriazole-based metal deactivator, a thiadiazole-based metal deactivator and an imidazole-based metal deactivator. These may be used alone or may be used in combination of two or more. The amount of the metal deactivator to be added is not particularly limited, and is preferably 0.01 mass% or more and 3 mass% or less, more preferably 0.01 mass% or more and 1 mass% or less based on the total amount of the composition.

**[0070]** Examples of the anti-rust agent include, for example, a petroleum sulfonate, an alkylbenzene sulfonate, a dinonylnaphthalene sulfonate, an alkenylsuccinate ester and a polyhydric alcohol ester. These may be used alone or may be used in combination of two or more. The amount of the anti-rust agent to be added is not particularly limited, and preferably 0.01 mass% or more and 1 mass% or less, more preferably 0.05 mass% or more and 0.5 mass% or less based on the total amount of the composition.

**[0071]** Examples of a surfactant/demulsifying agent include, for example, a polyalkylene glycol-based nonionic surfactant. Specifically, polyoxyethylene alkyl ether, polyoxyethylene alkylphenyl ether and polyoxyethylene alkylnaphthyl ether can be exemplified. These may be used alone or may be used in combination of two or more. The amount of the surfactant to be added is not particularly limited, and is preferably 0.01 mass% or more and 3 mass% or less, more preferably 0.01 mass% or more and 1 mass% or less based on the total amount of the composition.

**[0072]** Examples of an anti-foaming agent include, for example, a fluorosilicone oil and a fluoroalkyl ether. These may be used alone or may be used in combination of two or more. The amount of the anti-foaming agent to be added is not particularly limited, and is preferably 0.005 mass% or more and 0.5 mass% or less, more preferably 0.01 mass% or more and 0.2 mass% or less based on the total amount of the composition.

**[0073]** Examples of the corrosion inhibitor include, for example, a benzotriazole-based corrosion inhibitor, a benzimidazole-based corrosion inhibitor, a benzothiazole-based corrosion inhibitor and a thiadiazole-based corrosion inhibitor. These may be used alone or may be used in combination of two or more. The amount of the corrosion inhibitor to be added is not particularly limited, and is preferably within the range of 0.01 mass% or more and 1 mass% or less based on the total amount of the composition.

**[0074]** Examples of the oiliness improver include, for example, an aliphatic monocarboxylic acid, a polymerized fatty acid, a hydroxyl fatty acid, an aliphatic monoalcohol, an aliphatic monoamine, an aliphatic monocarboxylic acid amide, a partial ester of a polyhydric alcohol and an aliphatic monocarboxylic acid. These may be used alone or may be used in combination of two or more. The amount of the oiliness improver to be added is not particularly limited, and is preferably within the range of 0.01 mass% or more and 10 mass% or less based on the total amount of the composition.

[0075] As the acid scavenger, an epoxy compound can be used. Specifically, phenyl glycidyl ether, an alkyl glycidyl ether, an alkylene glycol glycidyl ether, cyclohexene oxide, an  $\alpha$ -olefin oxide and an epoxydized soybean oil can be exemplified. These may be used alone or may be used in combination of two or more. The amount of the acid capturing agent to be added is not particularly limited, and is preferably within the range of 0.005 mass% or more and 5 mass% or less based on the total amount of the composition.

[Properties of lubricating oil composition and the like]

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**[0076]** The kinetic viscosity of the lubricating oil composition can be measured by the method according to JIS-K-2283:2000.

**[0077]** The kinetic viscosity of the lubricating oil composition at  $100^{\circ}$ C is preferably  $14.0 \text{ mm}^2$ /s or less, more preferably  $12.5 \text{ mm}^2$ /s or less, still more preferably  $10.0 \text{ mm}^2$ /s or less, and preferably  $2.0 \text{ mm}^2$ /s or more, more preferably  $2.2 \text{ mm}^2$ /s or more, still more preferably  $2.5 \text{ mm}^2$ /s or more from the viewpoint of enhancement of lubricity, viscosity property

and fuel-saving performance.

**[0078]** The kinetic viscosity of the lubricating oil composition at  $40^{\circ}$ C is preferably  $80.0 \text{ mm}^2$ /s or less, more preferably  $70.0 \text{ mm}^2$ /s or less, still more preferably  $65.0 \text{ mm}^2$ /s or less, and preferably  $5.0 \text{ mm}^2$ /s or more, more preferably  $7.0 \text{ mm}^2$ /s or more, still more preferably  $10.0 \text{ mm}^2$ /s or more from the viewpoint of enhancement of lubricity, viscosity property and fuel-saving performance.

**[0079]** The viscosity index of the lubricating oil composition can be calculated by the method according to JIS-K-2283:2000. The viscosity index of the lubricating oil composition is preferably 90 or more, more preferably 100 or more, still more preferably 103 or more so that viscosity change due to temperature change is suppressed and fuel saving performance is enhanced.

[Flash point]

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**[0080]** The flash point means the value according to JIS-K-2265-4 and obtained by measurement using COC method. The flash point of the lubricating oil composition is preferably 172°C or higher, still more preferably 174°C or higher, particularly preferably 176°C or higher.

**[0081]** When the flash point of the lubricating oil composition is 172°C or higher, the ability of cooling the mechanical device in which the lubricating oil composition is used can be better. Increase in the flash point of the lubricating oil composition can be achieved by, for example, using an oil having high flash point as each oil constituting the lubricating base oil (A).

[Application of lubricating oil composition]

**[0082]** The above described lubricating oil composition of the present invention has a flash point within the specified range, and can exhibit lubricity (abrasion resistance, seizure resistance, low friction). Therefore, the lubricating oil composition of the present invention can be preferably applied to a device in which a motor and a speed reducer are integrated, for example mechanical devices such as a hydraulic system, a stationary transmission device, an automobile transmission device and a cooling device of a motor/battery.

[Method for producing lubricating oil composition]

**[0083]** A method for producing the lubricating oil composition of the present invention is not particularly limited, and it preferably comprises a step of mixing the lubricating base oil (A), the neutral phosphorus-based compound (B), the acidic phosphorus-based compound (C), the sulfur-based compound (D) and the organic molybdenum compound (E).

35 [Mechanical device]

**[0084]** The lubricating oil composition enhances lubricity of the mechanical device, and can be used for mechanical devices which are a hydraulic system, a stationary transmission device, an automobile transmission device and a cooling device of a motor/battery. For example, the lubricating oil composition can be used for a motor mounted in a hybrid automobile, electric automobile etc., an engine for a diesel engine or a gasoline engine, a transmission of an automobile etc. and the like. In particular, it is preferably used for a transmission mounted in a hybrid automobile, electric automobile and the like.

[Examples]

**[0085]** Hereinafter, the present invention will be described in more detail with reference to Examples, but the present invention is not limited to these Examples.

[0086] The properties and abilities in Examples and Comparative Examples were measured as follows.

50 (1) Kinetic viscosity

[0087] The kinetic viscosity at 40°C and the kinetic viscosity at 100°C were measured according to JIS-K-2283:2000 using a glass capillary type viscometer.

55 (2) Viscosity index

[0088] The viscosity index was calculated by the method according to JIS-K-2283:2000.

(3) Flash point

[0089] The flash point was measured by COC method according to JIS-K-2265-4.

5 (4) Abrasion resistance

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**[0090]** The abrasion resistance was evaluated by Four-ball wear test. Specifically, the abrasion resistance between metals was evaluated by measuring a diameter of an abrasion mark (unit: mm) under the test condition of revolution number of 1800 rpm, test temperature of 80°C, load of 392 N, and test time of 30 minutes, according to the method described in ASTM D4172. When the diameter of the abrasion mark is smaller, the abrasion resistance between metals is more excellent.

- (5) Seizure resistance
- [0091] The fusion load WL (N) was measured by conducting a test according to ASTM D2783-03(2014) under the condition of revolution number of 1800 rpm at room temperature (25°C). When this value is higher, the seizure resistance is more excellent.
  - (6) Friction property

**[0092]** The inter-metal friction coefficient was measured by LFW-1 test according to JASO method (high load method) M358:2005. When this value is lower, the seizure resistance is more excellent.

[Examples 1-3, Comparative Examples 1-4]

**[0093]** The lubricating oil composition was prepared according to the composition shown in Table 1 using the lubricating base oil (A), the neutral phosphorus-based compound (B), the acidic phosphorus-based compound (C), the sulfur-based compound (D), the organic molybdenum compound (E) etc. described below. Each component constituting the lubricating oil composition and described in Table 1 is as follows.

[Lubricating base oil (A)]

[0094] Mineral oil-1: The mineral oil having a kinetic viscosity at 100°C of 2.4 mm<sup>2</sup>/s, a viscosity index of 110 and a flash point of 186°C.

[0095] Mineral oil-2: The mineral oil having a kinetic viscosity at 100°C of 2.4 mm<sup>2</sup>/s, a viscosity index of 105 and a flash point of 176°C.

**[0096]** Synthetic oil-1: The synthetic oil having a kinetic viscosity at 100°C of 2.4 mm<sup>2</sup>/s, a viscosity index of 110 and a flash point of 186°C.

40 [Neutral phosphorus-based compound (B)]

**[0097]** Tricresyl phosphate (TCP) (the compound of the above general formula (2) wherein R<sup>5</sup>-R<sup>7</sup> are methylphenyl groups)

45 [Acidic phosphorus-based compound (C)]

**[0098]** Dioleyl acid phosphate (acidic phosphoric acid ester of the above general formula (4) wherein R<sup>8</sup> and R<sup>9</sup> are oleyl groups)

50 [Sulfur-based compound (D)]

**[0099]** 2,5-bis(1,1,3,3-tetramethylbutyldithio)-1,3,4-thiadiazole (the thiadiazole-based compound of the above general formula (6) wherein X1 and X2 are 2, R<sup>10</sup> and R<sup>11</sup> are 1,1,3,3-tetramethylbutyl groups)

<sup>55</sup> [Organic molybdenum compound (E)]

**[0100]** Molybdenum dithiocarbamate (MoDTC) (The compound of the formula (1) wherein  $R^1$  to  $R^4$  are each independently a hydrocarbon group having 8 or 13 carbon atoms,  $X^1$  to  $X^4$  are oxygen atoms. The content of molybdenum

atoms = 10.0 mass%, the content of sulfur atoms = 11.5 mass%)

**[0101]** Other additives (balance) contained in the composition of Examples and Comparative Examples consist of a viscosity index enhancer, an antioxidant, a detergent dispersant, a pour point depressant, an anti-foaming agent and the like.

	Г				П				T									
5		Comparative Example 4	95.10			08'0	099	0.20	120	0.10	300		Balance	100	10.74	2.82	108	186
10		Comparative Example 3	95.10			0.80	099	0.20	120		0	0.40	Balance	100	10.71	2.82	108	186
15	•	Comparative Example 2	95.20			08.0	099		0	0.10	300	0.40	Balance	100	10.70	2.82	109	188
20	•	Comparative Example 1	95.80				0	0.20	120	0.10	300	0.40	Balance	100	10.60	2.81	110	186
25		Example 3			95.00	08.0	099	0.20	120	0.10	300	0.40	Balance	100	10.79	2.85	112	188
30 :	[Table 1]	Example 2		00'56		08'0	099	0.20	120	0.10	300	0.40	Balance	100	10.81	2.83	107	178
35		Example 1	95.00			08.0	099	0.20	120	0.10	300	0.40	Balance	100	10.81	2.86	111	186
40			Mineral oil-1	Mineral oil-2	Synthetic oil-1	punodwoo	mass	(C) punodu	mass	(D) pu	mass ppm	pound (E)			s/zww	s/zww		ပ္
<i>45 50</i>		Lubricating base oil (A)				Neutral phosphorus-based compound (B)	Content of component (B) in terms of phosphorus atom	Acidic phosphorus-based compound (C)	Content of component (C) in terms of phosphorus atom	Sulfur-based compound (D)	Content of component (D) in terms of sulfur atom	Organic molybdenum compound (E)	Other additives	Total	Kinetic viscosity at 40°C	Kinetic viscosity at 100°C	Viscosity index	Flash point
55			Composition of components (mass%)							Property								

5		Comparative Example 4	0.48	618	0.110
10		Comparative Example 3	0.48	490	0.046
15		Comparative Example 2	0.48	490	0.044
20		Comparative Example 1	0.64	618	0.044
25	(p		0.43	618	0.047
30	(continued)	Example Example 1 2 3	0.46	618	0.046
35		Example 1	0.45	618	0.046
40			шш	z	
<b>45</b>		Lubricating base oil (A)	Abrasion resistance	Seizure resistance	Friction property
55				Ability	

**[0102]** As shown in Table 1, when Examples 1-3 and Comparative Examples 1-4 were compared, it was found that the lubricating oil compositions containing all of the lubricating base oil (A), the neutral phosphorus-based compound (B), the acidic phosphorus-based compound (C), the sulfur-based compound (D) and the organic molybdenum compound (E) had excellent abilities with respect to all of abrasion resistance, seizure resistance and friction property.

**[0103]** When Examples 1-3 and Comparative Example 4 were compared, it was found that the friction property of the obtained lubricating oil composition was enhanced by using the organic molybdenum compound (E).

**[0104]** In Examples 1-3, the flash point of the lubricating oil composition increased when the base oil having a high flash point was used as the lubricating base oil (A). In particular, since the lubricating base oil (A) had a flash point of 176°C or higher in Examples 1 and 3, the flash point of the obtained lubricating oil composition was also higher.

#### Claims

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- 1. A lubricating oil composition, comprising a lubricating base oil (A), a neutral phosphorus-based compound (B), an acidic phosphorus-based compound (C), a sulfur-based compound (D) and an organic molybdenum compound (E).
- 2. The lubricating oil composition according to claim 1, wherein the organic molybdenum compound (E) is a compound represented by a following formula (1)

wherein  $R^1$  to  $R^4$  are each independently a hydrocarbon group having 4 to 18 carbon atoms, and  $X^1$  to  $X^4$  are each independently an oxygen atom or a sulfur atom.

- **3.** The lubricating oil composition according to claim 2, wherein R<sup>1</sup> to R<sup>4</sup> are each independently an alkyl group having 4 to 18 carbon atoms, an alkenyl group having 4 to 18 carbon atoms, a cycloalkyl group having 4 to 18 carbon atoms, an aryl group having 6 to 18 carbon atoms, an alkylaryl group having 7 to 18 carbon atoms or an arylalkyl group having 7 to 18 carbon atoms.
- 4. The lubricating oil composition according to any one of claims 1 to 3, wherein a content of the organic molybdenum compound (E) is 0.01 mass% or more and 1.0 mass% or less based on a total amount of the lubricating oil composition.
- 5. The lubricating oil composition according to any one of claims 1 to 4, wherein a flash point of the lubricating base oil (A) is 172°C or higher.
  - **6.** The lubricating oil composition according to any one of claims 1 to 5, wherein the lubricating oil composition is used for a mechanical device.
- **7.** The lubricating oil composition according to claim 6, wherein the mechanical device is a hydraulic device, a stationary transmission device, an automobile transmission device or a cooling device of a motor/battery.
  - 8. A mechanical device, comprising the lubricating oil composition according to any one of claims 1 to 5.
- 50 **9.** The mechanical device according to claim 8, wherein the mechanical device is a hydraulic device, a stationary transmission device, an automobile transmission device or a cooling device of a motor/battery.
  - **10.** A method for producing a lubricating oil composition, comprising a step of mixing a lubricating base oil (A), a neutral phosphorus-based compound (B), an acidic phosphorus-based compound (C), a sulfur-based compound (D) and an organic molybdenum compound (E).

#### INTERNATIONAL SEARCH REPORT International application No. PCT/JP2019/040538 CLASSIFICATION OF SUBJECT MATTER 5 Int. Cl. C10M169/04(2006.01)i, C10M135/00(2006.01)n, C10M135/18(2006.01)n, C10M137/00(2006.01)n, C10N10/12(2006.01)n, C10N20/00(2006.01)n, C10N30/00(2006.01)n, C10N30/06(2006.01)n, C10N40/04(2006.01)n, C10N40/08(2006.01)n According to International Patent Classification (IPC) or to both national classification and IPC B. FIELDS SEARCHED 10 Minimum documentation searched (classification system followed by classification symbols) Int. Cl. C10M101/00-177/00, C10N10/12, C10N20/00, C10N30/00, C10N30/06, C10N40/04, C10N40/08 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched 15 Published examined utility model applications of Japan Published unexamined utility model applications of Japan Registered utility model specifications of Japan Published registered utility model applications of Japan 1922-1996 1971-2019 Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) 20 C. DOCUMENTS CONSIDERED TO BE RELEVANT Category\* Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. Χ JP 2003-336089 A (CHEVRON TEXACO JAPAN LTD.) 28 1-6, 8, 10November 2003, claim 1, paragraphs [0001], [0051]-25 [0069], examples 1-5 (Family: none) Χ JP 2011-6635 A (IDEMITSU KOSAN CO., LTD.) 13 1-4, 6-10 January 2011, comparative example 13, paragraphs 30 [0065]-[0076] (Family: none) 35 40 Further documents are listed in the continuation of Box C. See patent family annex. Special categories of cited documents: 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 document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international document of particular relevance; the claimed invention cannot be filing date 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 being obvious to a person skilled in the art document referring to an oral disclosure, use, exhibition or other means document published prior to the international filing date but later than "&" document member of the same patent family the priority date claimed Date of the actual completion of the international search Date of mailing of the international search report 50 26.11.2019 14.11.2019 Name and mailing address of the ISA/ Authorized officer Japan Patent Office 3-4-3, Kasumigaseki, Chiyoda-ku, 55 Tokyo 100-8915, Japan Telephone No.

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# INTERNATIONAL SEARCH REPORT

International application No. PCT/JP2019/040538

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### REFERENCES CITED IN THE DESCRIPTION

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