(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) **EP 2 535 397 A1**

(12) **EUROPEAN PATENT APPLICATION**
published in accordance with Art. 153(4) EPC

(43) Date of publication:
19.12.2012 Bulletin 2012/51

(21) Application number: **10845787.0**

(22) Date of filing: **08.10.2010**

(51) Int Cl.:
C10M 169/00 *(2006.01)* ***C10M 137/06*** *(2006.01)*
C10M 137/10 *(2006.01)* ***C10M 143/00*** *(2006.01)*
C10M 159/04 *(2006.01)* ***C10N 10/04*** *(2006.01)*
C10N 20/00 *(2006.01)* ***C10N 20/02*** *(2006.01)*
C10N 30/00 *(2006.01)*

(86) International application number:
PCT/JP2010/067774

(87) International publication number:
WO 2011/099195 (18.08.2011 Gazette 2011/33)

(84) Designated Contracting States:
AL AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HR HU IE IS IT LI LT LU LV MC MK MT NL NO PL PT RO RS SE SI SK SM TR

(30) Priority: **12.02.2010 JP 2010029340**

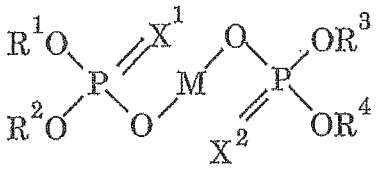
(71) Applicant: **JX Nippon Oil & Energy Corporation**
Chiyoda-ku
Tokyo 100-8162 (JP)

(72) Inventor: **YAGISHITA, Kazuhiro**
Chiyoda-ku
Tokyo (JP)

(74) Representative: **Piotrowicz, Pawel Jan Andrzej et al**
Venner Shipley LLP
Byron House
Cambridge Business Park
Cowley Road
Cambridge CB4 0WZ (GB)

(54) LUBRICANT ADDITIVE COMPOSITION

(57) The present invention provides an additive composition for a lubricating oil that comprises a metal salt of a phosphorus-containing compound, is easy to produce, does not have adverse effects such as causing rubber seals to swell, and is excellent in storage stability. The additive composition comprises a metal salt of a phosphorus-containing compound represented by formula (1) in an amount of 10 to 90 percent by mass on the basis of the composition, contained in a hydrocarbon solvent selected from a mineral oil, a hydrocarbon synthetic oil and a mixture thereof, the hydrocarbon solvent having a 100°C kinematic viscosity of 0.5 to 4.5 mm^2/s, a $\%C_A$ of 3 or less, and a sulfur content of 0.05 percent by mass or less:



(1)

wherein X^1 and X^2 are each independently oxygen or sulfur, R^1, R^2, R^3, and R^4 are each independently a straight-chain alkyl group having 3 to 12 carbon atoms, the average of the carbon number is 5 or greater, and M is a divalent metal atom.

EP 2 535 397 A1

Description

Technical Field

[0001] The present invention relates to additive compositions containing a metal salt of a phosphorus-containing compound for lubricating oils.

Background Art

[0002] Metal salts of phosphorus-containing compounds such as dialkylthiophosphoric acid esters, dialkylphosphoric acid esters or the like, which are solids or viscous liquids at ambient temperatures have problems that they are poor in handleability during the production and transportation thereof, cannot be dissolved in a base oil kept at ambient temperature, or are poor in storage stability after being dissolved. A technology has been, therefore, proposed wherein such metal salts are dissolved in an amine compound or the like beforehand to be liquefied (see Patent Literature 1 below). However, since addition of a metal salt of a phosphorus-containing compound having been dissolved in such an amine compound to a lubricating oil would lead to adverse effects such as causing rubber seals to swell, the use of the metal salt are restricted to a large extent.

Citation List

Patent Literature

[0003] Patent Literature 1: WO2004/003118

Summary of Intention

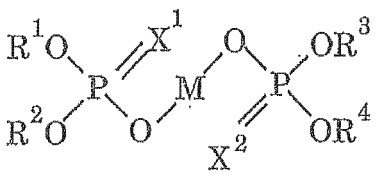
Technical Problem

[0004] The present invention provides an additive composition for a lubricating oil that does not give adverse effects such as causing rubber seals to swell, is excellent in storage stability and contains a metal salt of a dialkylthiophosphoric acid ester or a dialkylphosphoric acid ester that and is solid or viscous liquid at ambient temperatures, in a dissolved state and also a process for producing an additive composition for a lubricating oil comprising dissolving the above-mentioned compound in a base oil at a relatively low temperature of 60° or lower for a short period of time.

Solution to Problem

[0005] As the results of the extensive studies and research conducted by the inventors, the present invention has been accomplished on the basis of the finding that the use of a specific hydrocarbon solvent enables a metal salt of a phosphorus-containing compound to dissolve at a high concentration and a relatively low temperature of 60°C or lower and the dissolved metal salt is excellent in storage stability.

[0006] That is, the present invention relates to an additive composition for a lubricating oil, comprising a metal salt of a phosphorus-containing compound represented by formula (1) in an amount of 10 to 90 percent by mass on the basis of the composition, contained in a hydrocarbon solvent selected from a mineral oil, a hydrocarbon synthetic oil and a mixture thereof, the hydrocarbon solvent having a 100°C kinematic viscosity of 0.5 to 4.5 mm^2/s, a $\%C_A$ of 3 or less, and a sulfur content of 0.05 percent by mass or less:

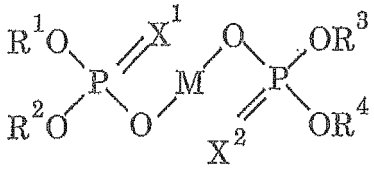


(1)

wherein X^1 and X^2 are each independently oxygen or sulfur, R^1, R^2, R^3, and R^4 are each independently a straight-chain alkyl group having 3 to 12 carbon atoms, the average of the carbon number is 5 or greater, and M is a divalent metal atom.

[0007] The present invention also relates to a process for producing an additive composition for a lubricating oil, comprising dissolving a metal salt of a phosphorus-containing compound represented by formula (1) in an amount of 10 to 90 percent by mass on the basis of the composition in a hydrocarbon solvent selected from a mineral oil, a

hydrocarbon synthetic oil and a mixture thereof, the hydrocarbon solvent having a 100°C kinematic viscosity of 0.5 to 4.5 mm^2/s, a $\%C_A$ of 3 or less, and a sulfur content of 0.05 percent by mass or less:



(1)

wherein X^1 and X^2 are each independently oxygen or sulfur, R^1, R^2, R^3, and R^4 are each a straight-chain alkyl group having 3 to 12 carbon atoms, the average of the carbon number is 5 or greater, and M is a divalent metal atom.

[0008] The present invention also relates to a lubricating oil composition comprising the foregoing additive composition.

Advantageous Effects of Invention

[0009] The additive composition for a lubricating oil of the present invention can be produced at a relative low temperature that is 60°C or lower for a short period of time, does not have adverse effects such as causing rubber seals to swell, and is excellent in storage stability.

Best Mode for Carrying out the Invention

[0010] The present invention will be described in more details below.
The hydrocarbon solvent used in the lubricating oil additive composition of the present invention is a solvent selected from a mineral oil, a hydrocarbon synthetic oil, and a mixture thereof, having a 100°C kinematic viscosity of 0.5 to 4.5 mm^2/s, a $\%C_A$ of 3 or less, and a sulfur content of 0.05 percent by mass or less.

[0011] Specific examples of the mineral base oil include those which can be produced by subjecting a lubricating oil fraction produced by vacuum-distilling an atmospheric distillation bottom oil resulting from atmospheric distillation of a crude oil, to any one or more treatments selected from solvent deasphalting, solvent extraction, hydrocracking, solvent dewaxing, and hydrorefining; wax-isomerized mineral oils; and those produced by isomerizing GTL WAX (Gas to Liquid Wax) produced through Fischer-Tropsch process.

[0012] Specific examples of the synthetic base oil include polybutenes and hydrogenated compounds thereof; poly-α-olefins such as 1-octene oligomer and 1-decene oligomer, and hydrogenated compounds thereof; aromatic synthetic oils such as alkylnaphthalenes and alkylbenzenes; and mixtures of two or more of these oils.

[0013] The hydrocarbon solvent used in the present invention may be a mineral oil, a hydrocarbon synthetic oil or any mixture of two more types selected from these oils. For example, the solvent may be a mixture of more than one mineral oils, a mixture of more than one hydrocarbon synthetic oils, or a mixture of one or more mineral oil and one or more hydrocarbon synthetic oil.

[0014] The hydrocarbon solvent used in the present invention has a 100°C kinematic viscosity of necessarily 0.5 to 4.5mm^2/s, preferably 1.0 to 4.3 mm^2/s, more preferably 1.5 to 4.2 mm^2/s, most preferably 2.0 to 4.1 mm^2/s. A hydrocarbon solvent having a 100°C kinematic viscosity of less than 0.5 mm^2/s is not preferable with the objective of evaporation loss. A hydrocarbon solvent having a 100°C kinematic viscosity of greater than 4.5 mm^2/s is not also preferable because of its poor dissolubility of a metal salt of a phosphorus-containing compound.

[0015] The hydrocarbon solvent used in the present invention has a $\%C_A$ of necessarily 3 or less, preferably 2.5 or less, more preferably 2.0 or less, more preferably 1.5 or less. A hydrocarbon solvent having a $\%C_A$ of greater than 3 is not preferable because it tends to be reduced in dissolubility of a metal salt of a phosphorus-containing compound. The hydrocarbon solvent may have a $\%C_A$ of 0.
The $\%C_A$ used herein denote the percentages of the aromatic carbon number in the total carbon number, determined by a method (n-d-M ring analysis) in accordance with ASTM D 3238-85.

[0016] The hydrocarbon solvent used in the present invention has a sulfur content of necessarily 0.05 percent by mass or less, preferably 0.04 percent by mass or less, more preferably 0.03 percent by mass or less, particularly preferably 0.02 percent by mass or less. A hydrocarbon solvent having a sulfur content of more than 0.05 percent by mass is not preferable because it tends to be reduced in dissolubility of a metal salt of a phosphorus-containing compound.

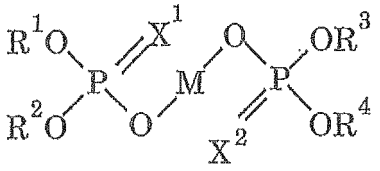
[0017] Generally, the sulfur content of a hydrocarbon solvent varies on the sulfur content of its raw material. For example, in the case of using a raw material containing substantially no sulfur such as a synthetic wax produced through a Fischer-Tropsch reaction, a hydrocarbon solvent containing substantially no sulfur can be produced. In the case of using a raw material containing sulfur such as a slack wax produced through the refining process of a hydrocarbon solvent or a micro wax produced through wax refining, the sulfur content of the resulting hydrocarbon solvent is usually

0.01 percent by mass or more. In the present invention, the sulfur content is necessarily 0.05 percent by mass or less. The sulfur content referred herein is measured in accordance with the method described in JIS K2541-1996.

[0018] No particular restriction is imposed on the viscosity index of the hydrocarbon solvent used in the present invention. However, the viscosity index is preferably 80 or greater, 100 or greater, most preferably 120 or greater such that excellent viscosity characteristics can be obtained at from low temperatures to high temperatures. No particular restriction is imposed on the upper limit of the viscosity index. Those with a viscosity index of 135 to 180 such as normal paraffins, slack waxes, GTL waxes or isoparaffin mineral oil produced by isomerizing them may be used. A hydrocarbon solvent with a viscosity index of less than 80 would tend to be reduced in dissolubility of a metal salt of a phosphorus-containing compound.

[0019] The additive composition for a lubricating oil of the present invention comprises the above-described hydrocarbon solvent and one or more metal salt of a phosphorus-containing compound represented by formula (1) below (hereinafter simply referred to as "a metal salt of a phosphorus-containing compound) in an amount of 10 to 90 percent by mass on the basis of the total mass of the composition.

[0020]



(1)

[0021] In formula (1) , X^1 and X^2 are each oxygen or sulfur. R^1, R^2, R^3 and R^4 are each independently a straight-chain alkyl group having 3 to 12, preferably 4 to 11, more preferably 4 to 10, most preferably 4 to 8 carbon atoms. An alkyl group having fewer than 3 carbon atoms would cause the resulting metal salt to be poor in dissolubility while an alkyl group having more than 12 carbon atoms would makes it difficult to produce the metal salt. The average carbon number of R^1, R^2, R^3 and R^4 is necessarily 5 or greater. M is a divalent metal atom, specific examples of which include zinc, copper, nickel, cobalt, calcium and magnesium. Preferred is zinc.

[0022] Examples of the straight-chain alkyl group includen-propyl, n-butyl, n-pentyl, n-hexyl, n-heptyl, n-octyl, n-nonyl, n-decyl, n-undecyl, and n-dodecyl groups.

[0023] Specific examples of the phosphorus-containing compound include di-n-butylphosphate, di-n-hexylphosphate, di-n-octylphosphate, di-n-decylphosphate, di-n-dodecylphosphate, n-butyl-n-hexylphosphate, n-butyl-n-octylphosphate, di-n-butylthiophosphate, di-n-hexylthiophosphate, di-n-octylthiophospahte, n-butyl-n-hexylthiophosphate, n-butyl-n-octylthiophosphate, and mixtures thereof.

Specific examples of the compounds represented by formula (1) include metal salts, such as zinc, copper, nickel, cobalt, calcium, and magnesium salts of the above-described phosphorus-containing compounds.

[0024] The mix ratio of the metal salt of the phosphorus-containing compound to the hydrocarbon solvent is from 10 to 90 percent by mass, preferably from 30 to 85 percent by mass, more preferably from 35 to 80 percent by mass, most preferably from 40 to 75 percent by mass on the basis of the total mass of the composition. When the mix ratio is less than 10 percent by mass, the amount of the additive to be added during the process of production of a lubricating oil would be increased, possibly causing an increase in the evaporation loss of the lubricating oil. When the mix ratio is more than 90 percent by mass, the additive would separate from the lubricating oil during the storage thereof.

[0025] The additive composition for a lubricating oil of the present invention has a 100°C kinematic viscosity of preferably 0.5 to 4.5 mm^2/s, more preferably 1.0 to 4.3 mm^2/s, more preferably 1.5 to 4.2 mm^2/s.

[0026] The additive composition of the present invention can be easily produced by, for example, adding a metal salt of a phosphorus-containing compound having been finely crushed, to a hydrocarbon solvent heated to a temperature of 30 to 60 °C and stirring the mixture at a revolution number of about 300 rpm or alternatively dissolving a metal salt of a phosphorous-containing compound in an organic solvent such as hexane and then mixing the metal salt with a hydrocarbon solvent, followed by removal of the organic solvent.

[0027] The resulting additive composition for a lubricating oil of the present invention is used as an additive for various lubricating oils.

Examples

[0028] The present invention will be described more specifically with reference to the following Examples and Comparative Examples but not limited thereto.

[0029] [Examples 1 to 8, Comparative Examples 1 to 16]

The following metal salts of phosphorous-containing compounds were added to the following hydrocarbon solvents to

prepare additive compositions for lubricating oils (Examples 1 to 8, Comparative Examples 1 to16) having formulations set forth in Tables 1 and 2 below. The hydrocarbon solvents and metal salts of phosphorous-containing compounds in the tables are as follows.

[0030] (1) Hydrocarbon solvents

Solvent 1: Paraffinic solvent refined mineral oil (100°C kinematic viscosity: 2.1 mm^2/s, $\%C_A$: 5, sulfur content: 800 ppm by mass)

Solvent 2: Hydrogenated refined mineral oil (100°C kinematic viscosity: 2.7 mm^2/s, $\%C_A$: 0, sulfur content: 0 ppm by mass)

Solvent 3: Poly-α-olefin (PAO) (100°C kinematic viscosity: 4.0 mm^2/s, $\%C_A$: 0, sulfur content: 0 ppm by mass)

[0031] (2) Metal salts of phosphorus-containing compounds

Phosphorus compound A: zinc n-butyl-n-hexylphosphate (average carbon number: 5, phosphorus content: 11.5 percent by mass, zinc content: 11.9 percent by mass)

Phosphorus compound B: zinc di-n-hexylphosphate (average carbon number: 6, phosphorus content: 10.4 percent by mass, zinc content: 10.7 percent by mass)

Phosphorus compound C: zinc n-butyl-n-octylphosphate (average carbon number: 6, phosphorus content: 10.4 percent by mass, zinc content: 10.7 percent by mass)

Phosphorus compound D: zinc n-butyl-n-octylthiophosphate (average carbon number: 6, phosphorus content: 9.8 percent by mass, zinc content: 10.1 percent by mass, sulfur content: 10.1 percent by mass)

Phosphorus compound E: zinc di-n-butylphosphate (average carbon number: 4, phosphorus content: 12.8 percent by mass, zinc content: 13.1 percent by mass)

Phosphorus compound F: zinc di-n-butylthiophosphate (average carbon number: 4, phosphorus content: 11.9 percent by mass, zinc content: 12.3 percent by mass, sulfur content: 12.3 percent by mass)

Phosphorus compound G: zinc n-butyl-n-hexylphosphate (average carbon number: 4.5, phosphorus content: 12.2 percent by mass, zinc content: 12.6 percent by mass)

Phosphorus compound H: zinc di-2-ethylhexylphosphate (phosphorus content: 8.8 percent by mass, zinc content: 9.1 percent by mass)

Phosphorus compound I: zinc di-2-ethylhexyldithiophosphate (phosphorus content: 8.4 percent by mass, zinc content: 8.7 percent by mass, sulfur content: 8.7 percent by mass)

[0032] The additive compositions for a lubricating oil of Examples 1 to 8 and Comparative Examples 1 50 16 were subjected to a storage test at normal temperature to observe whether they were liquefied at a temperature of 60°C and whether insoluble precipitated after 1 week, 2 weeks, 3 weeks and 4 weeks. The results are set forth in Tables 1 and 2. A stable additive composition for a lubricating oil cannot be produced in the case of using solvent 1 not having the properties defined by the present invention and phosphorus-containing compounds having a branched alkyl group.

[0033] [Table 1]

Test Oil No.		1 Example 1	2 Example 2	3 Example 3	4 Example 4	5 Example 5	6 Example 6	7 Example 7	8 Example 8	9 Comparative Example 1	10 Comparative Example 2	11 Comparative Example 3	12 Comparative Example 4
Solvent I	mass%	-	-	-	-	-	-	-	-	balance	balance	balance	balance
Solvent 2	mass%	balance	-	balance	-	balance	-	balance	-	-	-	-	-
Solvent 3	mass%	-	balance	-	balance	-	balance	-	balance	-	-	-	-
Phosphorus Compound A	mass%	50	50	-	-	-	-	-	-	50	-	-	-
Phosphorus Compound B	mass%	-	-	50	50	-	-	-	-	-	50	-	-
Phosphorus Compound C	mass%	-	-	-	-	50	50	-	-	-	-	50	-
Phosphorus Compound D	mass%	-	-	-	-	-	-	50	50	-	-	-	50
Phosphorus Compound E	mass%	-	-	-	-	-	-	-	-	-	-	-	-
Phosphorus Compound F	mass%	-	-	-	-	-	-	-	-	-	-	-	-

(continued)

Test Oil No.		1 Example 1	2 Example 2	3 Example 3	4 Example 4	5 Example 5	6 Example 6	7 Example 7	8 Example 8	9 Comparative Example 1	10 Comparative Example 2	11 Comparative Example 3	12 Comparative Example 4
Phosphorus Compound G	mass%	-	-	-	-	-	-	-	-	-	-	-	-
Phosphorus Compound H	mass%	-	-	-	-	-	-	-	-	-	-	-	-
Phosphorus Compound I	mass%	-	-	-	-	-	-	-	-	-	-	-	-
Average Carbon Number of Phosphorus Compound		5	5	6	6	6	6	6	6	5	6	6	6
Liquefaction at 60°C		Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Stability at room temperature (Insoluble)	one week	No	No	No	No	No	No	No	No	No	No	No	No
	two weeks	No	No	No	No	No	No	No	No	Yes	No	Yes	Yes

(continued)

Test Oil No.		1 Example 1	2 Example 2	3 Example 3	4 Example 4	5 Example 5	6 Example 6	7 Example 7	8 Example 8	9 Comparative Example 1	10 Comparative Example 2	11 Comparative Example 3	12 Comparative Example 4
	three weeks	No	No	No	No	No	No	No	No	Yes	Yes	Yes	Yes
	four weeks	No	No	No	No	No	No	No	No	Yes	Yes	Yes	Yes

[Table 2]

Test Oil No.		13 Comparative Example 5	14 Comparative Example6	15 Comparative Example 7	16 Comparative Example8	17 Comparative Example 9	18 Comparative Example 10	19 Comparative Example 11	20 Comparative Example 12	21 Comparative Example 13	22 Comparative Example 14	23 Comparative Example 15	24 Comparative Example 16
Solvent 1	mass %	-	-	-	-	-	-	-	-	-	-	balance	balance
Solvent 2	mass %	balance	-	balance	-	balance	-	balance	-	balance	-	-	-
Solvent 3	mass %	-	balance	-	balance	-	balance	-	balance	-	balance	-	-
Phosphorus Compound A	mass %	-	-	-	-	-	-	-	-	-	-	-	-
Phosphorus Compound B	mass %	-	-	-	-	-	-	-	-	-	-	-	-
Phosphorus Compound C	mass %	-	-	-	-	-	-	-	-	-	-	-	-
Phosphorus Compound D	mass %	-	-	-	-	-	-	-	-	-	-	-	-

(continued)

Test Oil No.		13 Comparative Example 5	14 Comparative Example6	15 Comparative Example 7	16 Comparative Example8	17 Comparative Example 9	18 Comparative Example 10	19 Comparative Example 11	20 Comparative Example 12	21 Comparative Example 13	22 Comparative Example 14	23 Comparative Example 15	24 Comparative Example 16
Phosphorus Compound E	mass %	50	50	-	-	-	-	-	-	-	-	50	-
Phosphorus Compound F	mass %	-	-	50	50	-	-	-	-	-	-	-	-
Phosphorus Compound G	mass %	-	-	-	-	50	50	-	-	-	-	-	-
Phosphorus Compound H	mass %	-	-	-	-	-	-	50	50	-	-	-	50
Phosphorus Compound I	mass %	-	-	-	-	-	-	-	-	50	50	-	-
Average Carbon Number of Phosphorus Compound		4	4	4	4	4.5	4.5	8	8	8	8	4	8

(continued)

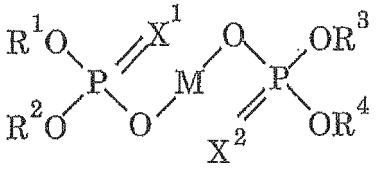
Test Oil No.		13 Comparative Example 5	14 Comparative Example6	15 Comparative Example 7	16 Comparative Example8	17 Comparative Example 9	18 Comparative Example 10	19 Comparative Example 11	20 Comparative Example 12	21 Comparative Example 13	22 Comparative Example 14	23 Comparative Example 15	24 Comparative Example 16
Liquefaction at 60°C		Yes	Yes	No	No	Yes	Yes	No	No	No	No	Yes	No
Stability at room temperature (Insoluble)	one week	Yes	Yes			Yes	Yes					Yes	No
	two weeks												
	three weeks												
	four weeks												

Industrial Applicability

[0034] The additive composition for a lubricating oil of the present invention is significantly large in industrial value because it can be produced at a relatively low temperature, which is 60°C or lower for a short period of time, does not have any adverse effect such as causing rubber seals to swell, and is excellent in storage stability.

Claims

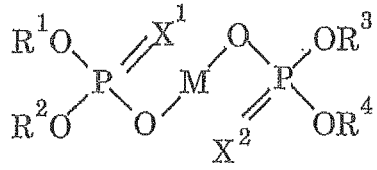
1. An additive composition for a lubricating oil, comprising a metal salt of a phosphorus-containing compound represented by formula (1) in an amount of 10 to 90 percent by mass on the basis of the composition, contained in a hydrocarbon solvent selected from a mineral oil, a hydrocarbon synthetic oil and a mixture thereof, the hydrocarbon solvent having a 100°C kinematic viscosity of 0.5 to 4.5 mm^2/s, a $\%C_A$ of 3 or less, and a sulfur content of 0.05 percent by mass or less:



(1)

wherein X^1 and X^2 are each independently oxygen or sulfur, R^1, R^2, R^3, and R^4 are each independently a straight-chain alkyl group having 3 to 12 carbon atoms, the average of the carbon number is 5 or greater, and M is a divalent metal atom.

2. A process for producing an additive composition for a lubricating oil, comprising dissolving a metal salt of a phosphorus-containing compound represented by formula (1) in an amount of 10 to 90 percent by mass on the basis of the composition in a hydrocarbon solvent selected from a mineral oil, a hydrocarbon synthetic oil and a mixture thereof, the hydrocarbon solvent having a 100°C kinematic viscosity of 0.5 to 4.5 mm^2/s, a $\%C_A$ of 3 or less, and a sulfur content of 0.05 percent by mass or less:



(1)

wherein X^1 and X^2 are each independently oxygen or sulfur, R^1, R^2, R^3, and R^4 are each a straight-chain alkyl group having 3 to 12 carbon atoms, the average of the carbon number is 5 or greater, and M is a divalent metal atom.

3. A lubricating oil composition comprising the additive composition for a lubricating oil according to claim 1.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2010/067774

A. CLASSIFICATION OF SUBJECT MATTER
C10M169/00(2006.01)i, *C10M137/06*(2006.01)n, *C10M137/10*(2006.01)n, *C10M143/00*(2006.01)n, *C10M159/04*(2006.01)n, *C10N10/04*(2006.01)n, *C10N20/00*(2006.01)n, *C10N20/02*(2006.01)n, *C10N30/00*(2006.01)n
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C10M169/00, C10M137/06, C10M137/10, C10M143/00, C10M159/04, C10N10/04, C10N20/00, C10N20/02, C10N30/00

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 1996-2010
Kokai Jitsuyo Shinan Koho 1971-2010 Toroku Jitsuyo Shinan Koho 1994-2010

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 63-99082 A (Osaka Koryo Kabushiki Kaisha), 30 April 1988 (30.04.1988), claims; page 2, lower left column, lines 4 to 9; table 1 (Family: none)	1-3
Y	WO 2009/116225 A1 (Nippon Oil Corp.), 24 September 2009 (24.09.2009), claims; paragraphs [0002] to [0004], [0019] to [0021] & JP 2009-227769 A	1-3
A	WO 2008/123227 A1 (Nippon Oil Corp.), 16 October 2008 (16.10.2008), claims & CN 101646758 A & JP 2008-239762 A	1-3

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

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
14 December, 2010 (14.12.10)	28 December, 2010 (28.12.10)
Name and mailing address of the ISA/ Japanese Patent Office Facsimile No.	Authorized officer Telephone No.

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

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2010/067774

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 2008/123226 A1 (Nippon Oil Corp.), 16 October 2008 (16.10.2008), claims & JP 2008-239776 A	1-3
A	JP 2006-299279 A (Nippon Oil Corp.), 02 November 2006 (02.11.2006), claims & EP 1227145 A1 & JP 2002-294271 A & US 2002/0142922 A1	1-3

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

REFERENCES CITED IN THE DESCRIPTION

This list of references cited by the applicant is for the reader's convenience only. It does not form part of the European patent document. Even though great care has been taken in compiling the references, errors or omissions cannot be excluded and the EPO disclaims all liability in this regard.

Patent documents cited in the description

- WO 2004003118 A **[0003]**