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Shimizu(10) **Pub. No.: US 2006/0148663 A1**(43) **Pub. Date: Jul. 6, 2006**(54) **ADDITIVES FOR LUBRICATING OILS AND
FUEL OILS, LUBRICATING OIL
COMPOSITIONS, AND FUEL OIL
COMPOSITIONS**(75) Inventor: **Nobuaki Shimizu**, Chiba (JP)

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Feb. 5, 2003 (JP) 2003-28642**Publication Classification**(51) **Int. Cl.****C10M 135/26** (2006.01)**C10L 1/24** (2006.01)(52) **U.S. Cl.** **508/465; 44/390**(57) **ABSTRACT**

Additives for lubricating oils and for fuel oils which comprise as the main component a disulfide represented by the following general formula (I) or (IV):

(R¹ and R² each independently represent a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, A¹ and A² each independently represent a group represented by CR³R⁴, R³ and R⁴ each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and x represents 2.)(R¹ and R² are as defined above, A³ and A⁴ each independently represent a group represented by CR⁵R⁶—CR⁷R⁸, R⁵ to R⁸ each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and x represents 2.) The additives exhibit excellent functions as the extreme pressure additive and the antiwear agent and the suppressed corrosive property to nonferrous metals and are advantageously used as the sulfur-based extreme pressure additives for lubricating oils and fuel oils.

ADDITIVES FOR LUBRICATING OILS AND FUEL OILS, LUBRICATING OIL COMPOSITIONS, AND FUEL OIL COMPOSITIONS

TECHNICAL FIELD

[0001] The present invention relates to additives for lubricating oils and fuel oils, lubricating oil compositions and fuel oil compositions. More particularly, the present invention relates to additives for lubricating oils and fuel oils which comprise a disulfide compound having a specific structure as the main component and exhibit excellent functions as the friction modifier, in particular, as the extreme pressure additive and the antiwear agent, and lubricating oil compositions and fuel compositions each comprising the additive.

BACKGROUND ART

[0002] Heretofore, lubricating oils are used for apparatuses and instruments used for driving and apparatuses such as internal combustion engines, automatic transmissions, shock absorbers and power steering to achieve smooth working of the apparatuses and instruments. It is well known that the lubricated surface is worn by friction under conditions of great output powers and great loads since the lubricating property becomes insufficient, and seizure takes place under extreme conditions. Therefore, lubricating oils containing extreme pressure additives and antiwear agents are used. However, conventional extreme pressure additives are not always satisfactory in that the sufficient effect of preventing the seizure is not exhibited due to interactions with other additives, metals are corroded, and wear resistance is insufficient.

[0003] It has been attempted that metal working oils used for metal working such as cutting, grinding and plastic working is prepared by adding oiliness agents and extreme pressure additives such as alcohols, esters of fatty acids and fatty acids to mineral oils and synthetic hydrocarbon oils to improve the working property.

[0004] However, a novel metal working oil which can provide a further improved working property is desired from the standpoint of improvement in the productivity and energy saving. On the other hand, the use of chlorine-based extreme pressure additives which have been widely used as the extreme pressure additive tends to be suppressed due to adverse effects on the working environment such as formation of rash on the human body and formation of rust on metals for the use.

[0005] As the metal working oil expected to overcome the above problems, an oil agent prepared by adding a sulfurized olefin containing active sulfur and an overbased sulfonate to a base oil is commercially available.

[0006] The above commercial metal working oil exhibits excellent resistance to welding and has the property of preventing abnormal wear of tools such as fracture and tearing of the surface of working. However, in working in which friction takes place repeatedly under a relatively small load, the efficiency of the production is frequently adversely affected since corrosive wear of tools with the active sulfur proceeds and the period of time before exchange of the tool with a new tool or grinding of the tool to refresh its surface decreases. On the contrary, the efficiency of production is frequently decreased when the problem of the abnormal wear is absent in the metal working.

[0007] Hydraulic oil is the fluid transmitting power which is used for transmitting power, controlling force and buffering in hydraulic systems such as hydraulic apparatuses and instruments and also exhibits the function of lubrication of sliding portions.

[0008] The excellent property of preventing seizure under loads and the excellent wear resistance are indispensable fundamental properties required for the hydraulic oil. These properties are provided by adding an extreme pressure additive and an antiwear agent to a base oil such as a mineral oil and a synthetic oil. However, conventional extreme pressure additives are not always satisfactory due to insufficient wear resistance or occurrence of corrosive wear although a sufficient property of preventing the seizure under loads may be exhibited.

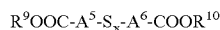
[0009] As for gear oil, in particular, gear oil for automobiles, the improvement in the wear resistance and the stability under oxidation is urgently required since the driving condition is becoming severer due to the increase in the amount of carried loads and the increase in the long distance transportation brought about by the improved highway network, and also the interval before the oil is renewed is increasing.

[0010] Heretofore, it is conducted that an extreme pressure additives or an antiwear additive such as sulfurized oil and fat, a sulfurized olefin, a phosphoric acid-based compound, a thiophosphoric acid-based compound and zinc dithiophosphate is added to the base oil for a lubricating oil as the main agent for the improvement. Further improvements in the wear resistance and the stability under oxidation and a further decrease in the ratio of the friction coefficients (at low speeds/at high speeds) have been required.

[0011] On the other hand, as for fuel oil, it is known that the lubricating property becomes insufficient as the fuel oil is hydrogenated to a greater degree. It is pointed out that the wear of a fuel pump takes place more quickly when a highly refined fuel is used. An excellent lubricating property is required for recent high performance fuel oils for turbines. Therefore, an excellent additive for fuel oils which is adsorbed on the surface of a metal in apparatuses and instruments of the fuel system to form an extreme pressure film so that the lubricating property is improved and the wear is decreased, is desired.

[0012] Conventionally, sulfur-based extreme pressure additives are frequently used as the extreme pressure additive for lubricating oils. The sulfur-based extreme pressure additive has sulfur atom in the molecule and is dissolved or uniformly dispersed in the base oil to exhibit the extreme pressure effect. Examples of the sulfur-based extreme pressure additive include sulfurized oils and fats, sulfurized fatty acids, sulfides of esters, polysulfides, sulfurized olefins, thiocarbamates, sulfurized terpenes and dialkyl thiodipropionates. However, the above sulfur-based extreme pressure additives are not always satisfactory since these additives have problems in that metals are corroded, the effect of preventing seizure is not sufficiently exhibited due to interactions with other additives, and the wear resistance is insufficient.

[0013] Recently, as the sulfur-based extreme pressure additive, a compound represented by general formula (VII):



(VII)

(in the above general formula, R^9 and R^{10} each represent a hydrocarbon group having 1 to 20 carbon atoms, A^5 and A^6 each represent a hydrocarbon group having 0 to 20 carbon atoms, and x represents an integer of 1 to 6) is disclosed (for example, Japanese Unexamined Patent Application Laid-Open No. 2001-288490).

[0014] However, in accordance with Japanese Unexamined Patent Application Laid-Open No. 2001-288490, the compound represented by general formula (VII) is prepared by the reaction of a chlorinated ester such as an ester of monochloroacetic acid and sodium polysulfide, and it is inevitable that the obtained product of the reaction is a mixture of monosulfides, disulfides and polysulfides which are the trisulfide or greater. The number of carbon atom alone is described on the divalent hydrocarbon group represented by A^5 and A^6 , and the preferable structure of the group is obscure since no descriptions can be found on the structure of the group.

DISCLOSURE OF THE INVENTION

[0015] Under the above circumstances, the present invention has an object of providing a sulfur-based extreme pressure additive which exhibits more excellent load carrying capacity and wear resistance than those exhibited by conventional sulfur-based additives and the suppressed corrosive property to nonferrous metals and can be used for lubricating oils and fuel oils, and a lubricating oil composition and a fuel oil composition comprising the additive.

[0016] As the result of intensive studies by the present inventor to achieve the above object, it was found that the object could be achieved by an additive for lubricating oils and fuel oils which comprised a disulfide compound having a specific structure as the main component. The present invention has been completed based on the knowledge.

[0017] The present invention provides:

(1) An additive for lubricating oils which comprises as a main component a disulfide compound represented by general formula (I):



wherein R^1 and R^2 each independently represent a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, A^1 and A^2 each independently represent a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and x represents 2;

(2) An additive for lubricating oils which comprises as a main component a disulfide compound which is obtained by oxidative coupling of an ester of a mercaptoalkanecarboxylic acid represented by at least one of general formula (II):



wherein R^1 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^1 represents a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and

general formula (III):



wherein R^2 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^2 represent a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms;

(3) An additive for lubricating oils which comprises as a main component a disulfide compound represented by general formula (IV):

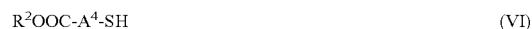


wherein R^1 and R^2 each independently represent a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, A^3 and A^4 each independently represent a group represented by CR^5R^6 — CR^7R^8 , R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and x represents 2;

(4) An additive for lubricating oils which comprises as a main component a disulfide compound which is obtained by oxidative coupling of an ester of a mercaptoalkanecarboxylic acid represented by at least one of general formula (V):



wherein R^1 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^3 represents a group represented by CR^5R^6 — CR^7R^8 , R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and general formula (VI):



wherein R^2 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^4 represents a group represented by CR^5R^6 — CR^7R^8 , R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms;

(5) An additive for lubricating oils described in the above item (1), wherein a content of polysulfide compounds represented by general formula (I) in which x represents a number of 3 or greater is 30% by mass or smaller based on a total of an amount of the disulfide compound and amounts of the polysulfide compounds;

(6) An additive for lubricating oils described in the above item (3), wherein a content of polysulfide compounds represented by general formula (IV) in which x represents a number of 3 or greater is 30% by mass or smaller based on a total of an amount of the disulfide compound and amounts of the polysulfide compounds;

(7) An additive for fuel oils which comprises as a main component a disulfide compound represented by general formula (I):



wherein R^1 and R^2 each independently represent a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, A^1 and A^2 each independently represent a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and x represents 2;

(8) An additive for fuel oils which comprises as a main component a disulfide compound which is obtained by oxidative coupling of an ester of a mercaptoalkanecarboxylic acid represented by at least one of general formula (II):

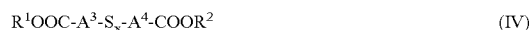


wherein R^1 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^1 represents a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and general formula (III):



wherein R^2 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^2 represent a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms;

(9) An additive for fuel oils which comprises as a main component a disulfide compound represented by general formula (IV):



wherein R^1 and R^2 each independently represent a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, A^3 and A^4 each independently represent a group represented by CR^5R^6 — CR^7R^8 , R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and x represents 2;

(10) An additive for fuel oils which comprises as a main component a disulfide compound which is obtained by oxidative coupling of an ester of a mercaptoalkanecarboxylic acid represented by at least one of general formula (V):



wherein R^1 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^3 represents a group represented by CR^5R^6 — CR^7R^8 , R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and general formula (VI):



wherein R^2 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^4 represents a group represented by CR^5R^6 — CR^7R^8 , R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms;

(11) An additive for fuel oils described in the above item (7), wherein a content of polysulfide compounds represented by general formula (I) in which x represents a number of 3 or greater is 30% by mass or smaller based on a total of an amount of the disulfide compound and amounts of the polysulfide compounds;

(12) An additive for fuel oils described in the above item (9), wherein a content of polysulfide compounds represented by general formula (IV) in which x represents a number of 3 or greater is 30% by mass or smaller based on a total of an amount of the disulfide compound and amounts of the polysulfide compounds;

(13) A lubricating oil composition which comprises a component (A): a base oil for a lubricating oil and a component (B): an additive for lubricating oils described in any one of the above items (1) to (6);

(14) A lubricating oil composition described in the above item (13), wherein a content of the component (B) is in a range of 0.01 to 50% by mass;

(15) A fuel oil composition which comprises a component (X): a fuel oil and a component (Y): an additive for fuel oils described in any one of the above items (7) to (12); and

(16) A fuel oil composition described in the above item (15), wherein a content of the component (Y) is in a range of 0.01 to 1,000 ppm by mass.

THE MOST PREFERRED EMBODIMENT TO CARRY OUT THE INVENTION

[0018] The compound of the present invention represented by general formula (I) which is used as the additive for lubricating oils and fuel oils is a disulfide compound having the following structure:



[0019] In the above general formula (I), R^1 and R^2 each independently represent a hydrocarbyl group having 1 to 30 carbon atoms, preferably 1 to 20 carbon atoms, more preferably 2 to 18 carbon atoms and most preferably 2 to 18 carbon atoms. The hydrocarbyl group may be linear, branched or cyclic and may have oxygen atom, sulfur atom or nitrogen atom. R^1 and R^2 may represent the same group or different groups. From the standpoint of the preparation, it is preferable that R^1 and R^2 represent the same group.

[0020] A^1 and A^2 each independently represent a group represented by CR^3R^4 . R^3 and R^4 each independently represent hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms. As the hydrocarbyl group, hydrocarbyl groups having 1 to 12 carbon atoms are preferable, and hydrocarbyl groups having 1 to 8 carbon atoms are more preferable. Examples of the group represented by A^1 and A^2 include methylene group, ethylidene group and propylidene group. Among these groups, methylene group is preferable. The atoms or the groups represented by A^1 and A^2 may be the same with or different from each other. From the standpoint of the preparation, it is preferable that the atoms and the groups represented by R^1 and R^2 are the same with each other. x represents 2.

[0021] In the additives for lubricating oils and fuel oils of the present invention, it is preferable that the content of polysulfide compounds represented by general formula (IV) in which x represents a number of 3 or greater is 30% by mass or smaller based on the total of the amount of the compounds represented by general formula (IV) in which x represents 2 and the amounts of the above polysulfide compounds. The corrosive property to nonferrous metals can be sufficiently suppressed when the above content is 30% or smaller. It is more preferable that the content of the polysulfide compounds represented by general formula (IV) in which x represents a number of 3 or greater is 10% by mass or smaller and still more preferably 5% or smaller.

[0022] Therefore, it is important that, in the production of the disulfide compound represented by general formula (I), a process providing the polysulfide compounds represented

by general formula (IV) in which x represents a number of 3 or greater as the byproducts in an amount within the above range be used. In the present invention, it is preferable that the disulfide compound is produced, for example, in accordance with the process shown in the following.

[0023] The oxidative coupling is conducted using as the raw material an ester of a mercaptoalkancarboxylic acid represented by general formula (II) and/or general formula (III):



wherein R^1 , R^2 , A^1 and A^2 are as defined above. The formation of polysulfide compounds, i.e., trisulfides and greater, as byproducts is substantially absent when the above process is conducted.

[0024] Specifically, compounds represented by $R^1OOC-A_1-S_2-A^2-COOR^2$, $R^1OOC-A_1-S_2-A^1-COOR^1$ and $R^1OOC-A^2-S_2-A^2-COOR^2$ are produced.

[0025] Examples of the oxidizing agent include oxygen, hydrogen peroxide, halogens such as iodine and bromine, hypohalous acids, hypohalites, sulfoxides such as dimethyl sulfoxide and diisopropyl sulfoxide and manganese (IV) oxide. Among these oxidizing agents, oxygen, hydrogen peroxide and dimethyl sulfoxide are preferable since the price is low and the production of the disulfide is facilitated.

[0026] Examples of the disulfide compound represented by general formula (I) include bis(methoxycarbonylmethyl) disulfide, bis(ethoxycarbonylmethyl) disulfide, bis(n-propoxycarbonylmethyl) disulfide, bis(isopropoxycarbonylmethyl) disulfide, bis(cyclopropoxycarbonylmethyl) disulfide, 1,1-bis(1-methoxycarbonylethyl) disulfide, 1,1-bis(1-methoxycarbonyl-n-propyl) disulfide, 1,1-bis(1-methoxycarbonyl-n-butyl) disulfide, 1,1-bis(1-methoxycarbonyl-n-hexyl) disulfide, 1,1-bis(1-methoxycarbonyl-n-octyl) disulfide, 2,2-bis(2-methoxycarbonyl-n-propyl) disulfide and α,α -bis(α -methoxycarbonylbenzyl) disulfide.

[0027] The compound represented by general formula (IV) of the present invention which is used for lubricating oils and fuel oils is a disulfide compound having the following structure:



[0028] In the above general formula (IV), R^1 and R^2 are as defined above for R^1 and R^2 in general formula (I). R^1 and R^2 may represent the same group or different groups. From the standpoint of the preparation, it is preferable that R^1 and R^2 represent the same group.

[0029] A^3 and A^4 each independently represent a group represented by $CR^5R^6-CR^7R^8$. R^5 to R^8 each independently represent hydrogen atom or a hydrocarbonyl group having 1 to 20 carbon atoms. As the hydrocarbonyl group, hydrocarbonyl groups having 1 to 12 are preferable, and hydrocarbonyl groups having 1 to 8 carbon atoms are more preferable. From the standpoint of the preparation, it is preferable that the atoms and the groups represented by R^1 and R^2 are the same with each other. x represents 2.

[0030] It is preferable that, in the additives for lubricating oils and fuel oils of the present invention, the content of polysulfide compounds represented by general formula (IV) in which x represents a number of 3 or greater is 30% by

mass or smaller based on the total of the amount of the compounds represented by general formula (IV) in which x represents 2 and the amounts of the above polysulfide compounds. The corrosive property to nonferrous metals can be sufficiently suppressed when the above content is 30% or smaller. It is more preferable that the content of the polysulfide compounds represented by general formula (IV) in which x represents a number of 3 or greater is 10% by mass or smaller and still more preferably 5% or smaller.

[0031] Therefore, it is important that, in the production of the disulfide compound represented by general formula (IV), a process providing the polysulfide compounds represented by general formula (IV) in which x represents a number of 3 or greater in an amount within the above range be used. In the present invention, it is preferable that the disulfide compound is produced, for example, in accordance with the process shown in the following.

[0032] The oxidative coupling is conducted using as the raw material an ester of a mercaptoalkancarboxylic acid represented by general formula (V) and/or general formula (VI):



wherein R^1 , R^2 , A^3 and A^4 are as defined above. The formation of polysulfide compounds, i.e., trisulfides or a greater compound, as byproducts is substantially absent in accordance with the above process.

[0033] Specifically, compounds represented by $R^1OOC-A^3-S_2-A^4-COOR^2$, $R^1OOC-A^3-S_2-A^3-COOR^1$ and $R^1OOC-A^4-S_2-A^4-COOR^2$ are produced.

[0034] As the oxidizing agent used for oxidation of an ester of an α -mercaptocarboxylic acid to produce the corresponding disulfide, oxidizing agent used in the production of a disulfide from a mercaptan can be used. Examples of the oxidizing agent include oxygen, hydrogen peroxide, halogens such as iodine and bromine, hypohalous acids, hypohalites, sulfoxides such as dimethyl sulfoxide and diisopropyl sulfoxide and manganese (IV) oxide. Among these oxidizing agents, oxygen, hydrogen peroxide and dimethyl sulfoxide are preferable since the price is low and the production of the disulfide is facilitated.

[0035] Examples of the disulfide compound represented by the above general formula (IV) include 1,1-bis(2-methoxycarbonylethyl) disulfide, 1,1-bis(2-ethoxycarbonylethyl) disulfide, 1,1-bis(2-n-propoxycarbonylethyl) disulfide, 1,1-bis(2-isopropoxycarbonylethyl) disulfide, 1,1-bis(2-cyclopropoxycarbonylethyl) disulfide, 1,1-bis(2-methoxycarbonyl-n-propyl) disulfide, 1,1-bis(2-methoxycarbonyl-n-butyl) disulfide, 1,1-bis(2-methoxycarbonyl-n-hexyl) disulfide, 1,1-bis(2-methoxycarbonyl-n-propyl) disulfide, 2,2-bis(3-methoxycarbonyl-n-pentyl) disulfide and 1,1-bis(2-methoxy-carbonyl-1-phenylethyl) disulfide.

[0036] These disulfide compounds exhibit excellent load carrying capacity and wear resistance when they are used as the sulfur-based extreme pressure additive and are used as the additives for lubricating oils and fuel oils.

[0037] The additives for lubricating oils and fuel oils of the present invention may comprise the above disulfide compound represented by general formula (I) and/or the

above disulfide compound represented by general formula (IV) singly or as a combination of two or more.

[0038] The lubricating oil composition of the present invention comprises a component (A): a base oil for lubricating oils and a component (B): an additive for lubricating oils comprising the disulfide compound described above. The lubricating oil composition in the present invention includes lubricating oils for automobiles used for driving apparatuses and instruments and gears such as internal engines, automatic transmissions, shock absorbers and power steering, metal working oils used for metal working such as cutting, grinding and deformation processing, and hydraulic oils which are also fluids for transmitting power used for transmission of power, control of power and shock absorbing in hydraulic systems such as hydraulic apparatuses and instruments.

[0039] In the lubricating oil composition of the present invention, the base oil for lubricating oils used as the component (A) is not particularly limited and suitably selected from mineral oils and synthetic oils in accordance with the object and the conditions of the use of the composition. Examples of the mineral oil include distillates obtained by atmospheric distillation of paraffinic crude oils, intermediate crude oils and naphthenic crude oils, distillates obtained by vacuum distillation of the residue of the atmospheric distillation and refined oils obtained by refining these oils in accordance with a conventional process such as oils refined with solvents, oils refined by hydrogenation, oils treated by dewaxing and oils treated with clay.

[0040] Examples of the synthetic oil include low molecular weight polybutenes, low molecular weight polypropylenes, oligomers of α -olefins having 8 to 14 carbon atoms, hydrogenation products of these oligomers, ester-based compounds such as polyol esters (such as esters of fatty acids with trimethylolpropane and esters of fatty acids with pentaerythritol), esters of dibasic acids, esters of aromatic polycarboxylic acids and esters of phosphoric acid, alkylaromatic compounds such as alkylbenzenes and alkyl-naphthalenes, polyglycol oils such as polyalkylene glycol, and silicone oils.

[0041] The base oil may be used singly or in combination of two or more.

[0042] The content of the additive for lubricating oils of the component (B) in the lubricating oil composition of the present invention is suitably selected in accordance with the object and the condition of the use of the composition. In general, the content is in the range of 0.01 to 50% by mass. In the case of the lubricating oil for automobiles and the hydraulic oil, the content is selected, in general, in the range of 0.01 to 30% by mass and preferably in the range of 0.01 to 10% by mass. In the case of the metal working oil, the content is selected, in general, in the range of 0.1 to 60% by mass and preferably in the range of 0.1 to 50% by mass although the additive can be used singly without mixing with other components.

[0043] The lubricating oil composition of the present invention may suitably comprise various other additives such as other friction modifiers (oiliness agents and other extreme pressure additives), antiwear agents, ashless dispersants, metal-based detergents, viscosity index improvers, pour point depressants, rust preventive, corrosion inhibitors

for metals, defoaming agents, surfactants and antioxidants in accordance with the object of the use.

[0044] Examples of the other friction modifier and the antiwear agent include sulfur-based compounds such as sulfurized olefins, dialkyl polysulfides, diarylalkyl polysulfides and diaryl polysulfides; phosphorus-based compounds such as esters of phosphoric acid, esters of thiophosphoric acid, esters of phosphorous acid, alkyl hydrogenphosphites, amine salts of esters of phosphoric acid and amine salts of esters of phosphorous acid; chlorine-based compounds such as chlorinated esters of fatty acids and chlorinated fatty acids; ester-based compounds such as esters of alkylmaleic acids and alkenyl maleic acids and esters of alkylsuccinic acids and alkenylsuccinic acids; organic acid-based compounds such as alkylmaleic acids, alkenylmaleic acids, alkylsuccinic acids and alkenylsuccinic acids; and organo-metallic compounds such as salts of naphthenic acid, zinc dithiophosphate (ZnDTP), zinc dithiocarbamate (ZnDTC), molybdenum dithiophosphate (MoDTP) and molybdenum dithiocarbamate (MoDTC).

[0045] Examples of the ashless dispersant include succinimides, succinimides containing boron, benzylamines, benzylamines containing boron, esters of succinic acid and amides of monovalent and divalent carboxylic acids typical examples of which include fatty acids and succinic acid. Examples of the metal-based detergent include neutral metal sulfonates, neutral metal phenates, neutral metal salicylates, neutral metal phosphonates, basic sulfonates, basic phenates, basic salicylates, basic phosphonates, overbased sulfonates, overbased phenates, overbased salicylates and overbased phosphonates.

[0046] Examples of the viscosity index improver include polymethacrylates, polymethacrylates of the dispersion type and olefin-based copolymers such as ethylene-propylene copolymers. Examples of the pour point depressant include polymethacrylates.

[0047] Examples of the rust preventive include alkenylsuccinic acid and partial esters thereof. Examples of the corrosion inhibitor for metals include benzotriazole-based agents, benzimidazole-based agents, benzothiazole-based agents and thiadiazole-based agents. Examples of the defoaming agent include dimethylpolysiloxane and polyacrylates. Examples of the surfactant include polyoxyethylene alkylphenyl ethers.

[0048] Examples of the antioxidant include amine-based antioxidants such as alkylated diphenylamines, phenyl- α -naphthylamine and alkylated naphthylamines; and phenol-based antioxidants such as 2,6-di-*t*-butyl-*p*-cresol and 4,4'-methylenebis(2,6-di-*t*-butylphenol).

[0049] The lubricating oil composition of the present invention is used as the lubricating oil for automobiles used for driving apparatuses and instruments and gears such as internal engines, automatic transmissions, shock absorbers and power steering, the metal working oil used for metal working such as cutting, grinding and deformation processing, and the hydraulic oil which are also fluids for transmitting power used for transmission of power, control of power and shock absorbing in hydraulic systems such as hydraulic apparatuses and instruments.

[0050] The fuel oil composition of the present invention comprises a component (X): a fuel oil and a component (Y): an additive for fuel oils comprising the disulfide compound described above.

[0051] In the fuel oil composition of the present invention, fuel oils highly refined by hydrogenation such as high performance fuel oils for turbines are preferable as the fuel oil of the component (X).

[0052] The content of the additive for fuel oils of the component (Y) in the fuel oil composition of the present invention is, in general, in the range of 0.01 to 1,000 ppm by mass and preferably in the range of 0.01 to 100 ppm by mass.

[0053] The fuel oil composition of the present invention may further comprise various types of additives, where necessary. Examples of the additive include conventional additives for fuel oils, examples which include antioxidants such as phenylenediamine-based antioxidants, diphenylamine-based antioxidants, alkylphenol-based antioxidants and aminophenol-based antioxidants; detergents such as polyether amines and polyalkylamines; metal inactivators such as compounds of the Schiff type and compounds of the thioamide type; agents for preventing surface ignition such as organic phosphorus-based compounds; agents for preventing freezing such as polyhydric alcohols; combustion improvers such as sulfuric acid esters of higher alcohols; antistatic agents such as anionic surfactants, cationic surfactants and amphoteric surfactants; rust preventives such as esters of alkenylsuccinic acids; identifiers such as quinizarin and coumarine; scenting agents such as natural spirits and synthetic perfumes; and coloring agents such as azo dyes.

EXAMPLES

[0054] The present invention will be described more specifically with reference to examples in the following. However, the present invention is not limited to the examples.

[0055] The friction coefficient, the diameter of wear marks and the corrosive property were evaluated in accordance with the following methods.

(1) Friction Coefficient and Diameter of Wear Marks

[0056] The four-ball test of the Soda type was conducted under the following conditions.

[0057] The test was conducted at a speed of rotation of 500 rpm and a temperature of the oil of 80° C. while the load was increased stepwise. The loads in the steps were 0.5, 0.7, 0.9, 1.1, 1.3 and 1.5 kgf/cm² ($\times 0.09807$ MPa). The load was kept at each step for 180 seconds, and the entire time of the test was 1,080 seconds. The friction coefficient was obtained at each step, and the width of wear marks was measured at the end of the test.

(2) Corrosive Property

[0058] The test of the corrosive property was conducted in accordance with the method of Japanese Industrial Standard K-2513 "Test method of corrosion of a copper plate with petroleum products" at a test temperature of 100° C. for a test time of 3 hours by the test tube method. The change in the color of a copper plate was observed in accordance with the standard for corrosion of a copper plate", and the corrosive property was evaluated in accordance with the

classification of 1a to 4c. The smaller the number in the classification, the smaller the corrosive property. The corrosive property increases in the order of the alphabet.

Preparation Example 1

Preparation of bis(ethoxycarbonylmethyl) disulfide

[0059] Ethyl mercaptoacetate was oxidized with dimethyl sulfoxide in accordance with the following process, and bis(ethoxycarbonylmethyl) disulfide was prepared. No polysulfides of trisulfides and greater were present in the obtained compound.

[0060] Into a 100 ml recovery flask, 20.9 g of methyl mercaptoacetate and 30.8 g of dimethyl sulfoxide were placed, and the resultant mixture was heated in an oil bath at 120° C. for 8 hours. After being cooled, the obtained reaction mixture was dissolved into 100 ml of toluene and washed ten times with water, and the unreacted dimethyl sulfoxide was removed. Toluene was removed by distillation under a reduced pressure, and 16.0 g of bis(ethoxycarbonylmethyl) disulfide was obtained.

Preparation Example 2

Preparation of bis(n-butoxycarbonylmethyl) disulfide

[0061] The oxidation was conducted in accordance with the same procedures as those conducted in Preparation Example 1 except that n-butyl mercaptoacetate was used, and bis(n-butoxycarbonylmethyl) disulfide was prepared. No polysulfides of trisulfides and greater were present in the obtained compound.

Preparation Example 3

Preparation of bis(n-octoxycarbonylmethyl) disulfide

[0062] The oxidation was conducted in accordance with the same procedures as those conducted in Preparation Example 1 except that n-octyl mercaptoacetate was used, and bis(n-octoxycarbonylmethyl) disulfide was prepared. No polysulfides of trisulfides and greater were present in the obtained compound.

Preparation Example 4

Preparation of bis(2-ethylhexoxycarbonylmethyl) disulfide

[0063] The oxidation was conducted in accordance with the same procedures as those conducted in Preparation Example 1 except that 2-ethylhexyl mercaptoacetate was used, and bis(2-ethylhexoxycarbonyl-methyl) disulfide was prepared. No polysulfides of trisulfides and greater were present in the obtained compound.

Preparation Example 5

Preparation of bis(isooctoxycarbonylmethyl) disulfide

[0064] The oxidation was conducted in accordance with the same procedures as those conducted in Preparation Example 1 except that isooctyl mercaptoacetate was used,

and bis(isooctoxycarbonylmethyl) disulfide was prepared. No polysulfides of trisulfides and greater were present in the obtained compound.

Preparation Example 6

Preparation of bis(n-stearoxycarbonylmethyl) disulfide

[0065] The oxidation was conducted in accordance with the same procedures as those conducted in Preparation Example 1 except that stearyl mercaptoacetate was used, and bis(n-stearoxycarbonylmethyl) disulfide was prepared. No polysulfides of trisulfides and greater were present in the obtained compound.

Preparation Example 7

Preparation of 1,1-bis(1-ethoxycarbonylethyl) disulfide

[0066] The oxidation was conducted in accordance with the same procedures as those conducted in Preparation Example 1 except that ethyl α -mercaptopropionate was used, and 1,1-bis(1-ethoxycarbonylethyl) disulfide was prepared. No polysulfides of trisulfides and greater were present in the obtained compound.

Comparative Preparation Example 1

Preparation of bis(n-butoxy-carbonylmethyl) polysulfide

[0067] Sodium polysulfide was prepared from sodium sulfide and sulfur, and bis(n-butoxycarbonylmethyl) polysulfide was prepared by the reaction of the obtained sodium polysulfide and n-butyl chloroacetate. In the preparation, the ratio of the amounts by mole of sodium sulfide to sulfur was adjusted so that sodium polysulfide (Na_2S_x) having an average number of sulfur (x) of 2 could be obtained for comparison with the disulfide of the present invention. Sodium polysulfide was used in an excess amount by 5% so that n-butyl chloroacetate was not left remaining. Specifically, bis(n-butoxycarbonylmethyl) polysulfide was prepared by the reaction in accordance with the following process. Into a 500 ml four-necked glass flask equipped with a stirrer and a reflux condenser, 26.4 g of sodium sulfide nonahydrate, 3.52 g of sulfur and 150 ml of a 95% ethanol were placed, and the resultant mixture was heated under stirring in an oil bath at 80° C. for 5 hours. After the reaction mixture was cooled to the room temperature, 30.12 g of n-butyl chloroacetate was added in small portions, and the obtained mixture was stirred at the room temperature for 2 hours. The reaction fluid was transferred to a separation funnel, dissolved into 500 ml of toluene and washed ten times with water. Toluene was removed by distillation under a reduced pressure, and 26.5 g of bis(n-butoxycarbonylmethyl) polysulfide was obtained.

[0068] The obtained compound was analyzed in accordance with the high performance liquid chromatography [the column: ODS; the solvent: acetonitrile; the detector: a refractive index (RI) detector]. The result was as follows: the

monosulfide: 21%; the disulfide: 40%; the trisulfide: 20%; the tetrasulfide: 12%; and the pentasulfide: 4%. These values are expressed as % by mass.

Comparative Preparation Example 2

Preparation of bis(2-ethylhexyloxy-carbonylmethyl) sulfide

[0069] Into a 500 ml egg plant shaped glass flask equipped with a Dean Stark dehydrator, 45.1 g of 2,2'-thiodiglycolic acid, 101.6 g of 2-ethylhexanol and 2.0 g of p-toluene-sulfonic acid monohydrate were placed, and the resultant mixture was heated under the refluxing condition for 5 hours. After being cooled, the reaction fluid was transferred to a separation funnel and washed twice with an aqueous solution of sodium hydrogencarbonate and five times with water. Toluene was removed by distillation under a reduced pressure, and 120.0 g of bis(2-ethylhexyloxy-carbonylmethyl) sulfide was obtained.

Example 1

[0070] To a mineral oil P500N of a 500 neutral fraction (the kinematic viscosity at 100° C.: 10.9 mm²/s; % CAO: 0.1% or smaller), bis(ethoxy-carbonylmethyl) disulfide obtained in Preparation Example 1 was added in an amount such that the content in the entire composition was 1% by mass, and a lubricating oil composition was prepared. The properties of the obtained lubricating oil composition were evaluated. The results are shown in Table 1.

Examples 2 to 7

[0071] To a mineral oil P500N of a 500 neutral fraction, the additives obtained in Preparation Examples 2 to 7 were added in amounts such that the content in the entire composition was 1% by mass as shown in Table 1, and lubricating oil compositions were prepared. The properties of the obtained lubricating oil compositions were evaluated. The results are shown in Table 1.

Comparative Example 1

[0072] The same procedures as those conducted in Example 1 were conducted except that bis(2-ethylhexyloxy-carbonylmethyl) sulfide obtained in Comparative Preparation Example 1 was used. The results are shown in Table 1.

Comparative Example 2

[0073] The same procedures as those conducted in Example 1 were conducted except that bis(n-butoxycarbonylmethylene) polysulfide obtained in Comparative Preparation Example 2 was used. The results are shown in Table 1.

Comparative Example 3

[0074] A mineral oil P500N of a 500 neutral fraction was evaluated without adding an additive in accordance with the same procedures as those conducted in Example 1. The results are shown in Table 1.

TABLE 1

	Example										
	1	2	3	4	5	6	7	1	2	3	
	Preparation of additive						Preparation Example				
	1	2	3	4	5	6	7	1	2	—	
Friction coefficient load (kgf/cm ²)	0.5	0.044	0.032	0.040	0.061	0.041	0.034	0.044	0.047	0.048	0.044
	0.7	0.051	0.041	0.050	0.066	0.052	0.045	0.052	0.054	0.061	0.055
	0.9	0.056	0.048	0.055	0.066	0.057	0.053	0.058	0.056	0.072	0.064
	1.1	0.059	0.052	0.058	0.066	0.058	0.057	0.059	0.057	0.080	0.070
	1.3	0.060	0.055	0.058	0.066	0.059	0.056	0.061	0.059	0.082	0.072
	1.5	0.061	0.054	0.056	0.066	0.058	0.057	0.062	0.06	0.085	0.076
Width of wear marks (mm)		0.35	0.40	0.34	0.30	0.36	0.42	0.33	0.46	0.47	0.52
Corrosion of copper plate		1B	1B	1B	1B	1B	1B	1B	4B	1B	1B

Preparation Example 8

Preparation of 1,1-bis(2-ethoxycarbonyl-ethyl) disulfide

[0075] Ethyl α -mercaptopropionate was oxidized with dimethyl sulfoxide in accordance with the following process, and 1,1-bis(2-ethoxycarbonyl-ethyl) disulfide was prepared. No polysulfides of trisulfides and greater were present in the obtained compound.

[0076] Into a 100 ml recovery flask, 20.9 g of ethyl α -mercaptopropionate and 30.8 g of dimethyl sulfoxide were placed, and the resultant mixture was heated in an oil bath at 120° C. for 8 hours. After being cooled, the obtained reaction mixture was dissolved into 100 ml of toluene and washed ten times with water, and the unreacted dimethyl sulfoxide was removed. Toluene was removed by distillation under a reduced pressure, and 16.0 g of 1,1-bis(2-ethoxycarbonyl-ethyl) disulfide was obtained.

Preparation Example 9

Preparation of 1,1-bis(2,2-ethylhexoxycarbonyl-ethyl) disulfide

[0077] 2-Ethylhexyl mercaptoacetate was oxidized in accordance with the same procedures as those conducted in Preparation Example 1, and 1,1-bis(2,2-ethylhexoxycarbonyl-ethyl) disulfide was prepared. No polysulfides of trisulfides and greater were present in the obtained compound.

Example 8

[0078] To a mineral oil P500N of a 500 neutral fraction (the kinematic viscosity at 100° C.: 10.9 mm²/s; % CAO: 0.1% or smaller), 1,1-bis(2-ethoxycarbonyl-ethyl) disulfide obtained in Preparation Example 8 was added in an amount such that the content in the entire composition was 1% by mass, and a lubricating oil composition was prepared. The properties of the obtained lubricating oil composition were evaluated. The results are shown in Table 2.

Example 9

[0079] The same procedures as those conducted in Examples 8 were conducted except that 1,1-bis(2,2-ethyl-

hexoxycarbonyl-ethyl) disulfide obtained in Preparation Example 9 was used. The results are shown in Table 2.

TABLE 2

		Example	
		8	9
Preparation Example		8	9
Preparation of additive			
Friction coefficient	0.5	0.056	0.037
load (kgf/cm ²)	0.7	0.062	0.047
	0.9	0.066	0.054
	1.1	0.066	0.059
	1.3	0.067	0.061
	1.5	0.068	0.066
Diameter of wear marks (mm)		0.37	0.36
Corrosion of copper plate		1B	1B

[0080] As clearly shown by the results in Examples and Comparative Examples, the lubricating oil composition using the additive of the present invention gave wear marks having a small diameter and exhibited very excellent load carrying capacity and wear resistance. As shown in Comparative Example 1, when the lubricating oil composition having a great content of the polysulfide compounds represented by general formula (I) or (IV) in which x was 3 or greater was used, the corrosive property to the nonferrous metal was exhibited. In contrast, the lubricating oil composition of the present invention exhibited the very excellent properties with the suppressed corrosive property.

INDUSTRIAL APPLICABILITY

[0081] In accordance with the present invention, the sulfur-based extreme pressure additive which exhibits more excellent load carrying capacity and wear resistance than those exhibited by conventional sulfur-based additives and the suppressed corrosive property to nonferrous metals and can be used for lubricating oils and fuel oils, and the lubricating oil composition and the fuel oil composition comprising the additive, can be provided.

1. An additive for lubricating oils which comprises as a main component a disulfide compound represented by general formula (I):

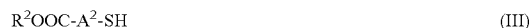


wherein R^1 and R^2 each independently represent a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, A^1 and A^2 each independently represent a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and x represents 2.

2. An additive for lubricating oils which comprises as a main component a disulfide compound which is obtained by oxidative coupling of an ester of a mercaptoalkanecarboxylic acid represented by at least one of general formula (II):



wherein R^1 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^1 represents a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and general formula (III):



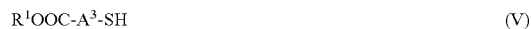
wherein R^2 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^2 represent a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms.

3. An additive for lubricating oils which comprises as a main component a disulfide compound represented by general formula (IV):

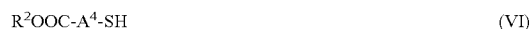


wherein R^1 and R^2 each independently represent a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, A^3 and A^4 each independently represent a group represented by $CR^5R^6-CR^7R^8$, R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and x represents 2.

4. An additive for lubricating oils which comprises as a main component a disulfide compound which is obtained by oxidative coupling of an ester of a mercaptoalkanecarboxylic acid represented by at least one of general formula (V):



wherein R^1 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^3 represents a group represented by $CR^5R^6-CR^7R^8$, R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and general formula (VI):



wherein R^2 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^4 represents a group represented by $CR^5R^6-CR^7R^8$, R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms.

5. An additive for lubricating oils according to claim 1, wherein a content of polysulfide compounds represented by general formula (I) in which x represents a number of 3 or greater is 30% by mass or smaller based on a total of an amount of the disulfide compound and amounts of the polysulfide compounds.

6. An additive for lubricating oils according to claim 3, wherein a content of polysulfide compounds represented by general formula (IV) in which x represents a number of 3 or greater is 30% by mass or smaller based on a total of an amount of the disulfide compound and amounts of the polysulfide compounds.

7. An additive for fuel oils which comprises as a main component a disulfide compound represented by general formula (I):



wherein R^1 and R^2 each independently represent a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, A^1 and A^2 each independently represent a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and x represents 2.

8. An additive for fuel oils which comprises as a main component a disulfide compound which is obtained by oxidative coupling of an ester of a mercaptoalkanecarboxylic acid represented by at least one of general formula (II):



wherein R^1 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^1 represents a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and general formula (III):



wherein R^2 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^2 represent a group represented by CR^3R^4 , R^3 and R^4 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms.

9. An additive for fuel oils which comprises as a main component a disulfide compound represented by general formula (IV):



wherein R^1 and R^2 each independently represent a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, A^3 and A^4 each independently represent a group represented by $CR^5R^6-CR^7R^8$, R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and x represents 2.

10. An additive for fuel oils which comprises as a main component a disulfide compound which is obtained by oxidative coupling of an ester of a mercaptoalkanecarboxylic acid represented by at least one of general formula (V):



wherein R^1 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^3 represents a group represented by $CR^5R^6-CR^7R^8$, R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms, and general formula (VI):



wherein R^2 represents a hydrocarbyl group which has 1 to 30 carbon atoms and may have oxygen atom, sulfur atom or nitrogen atom, and A^4 represents a group represented by $CR^5R^6-CR^7R^8$, R^5 to R^8 each independently representing hydrogen atom or a hydrocarbyl group having 1 to 20 carbon atoms.

11. An additive for fuel oils according to claim 7, wherein a content of polysulfide compounds represented by general formula (I) in which x represents a number of 3 or greater is 30% by mass or smaller based on a total of an amount of the disulfide compound and amounts of the polysulfide compounds.

12. An additive for fuel oils according to claim 9, wherein a content of polysulfide compounds represented by general formula (IV) in which x represents a number of 3 or greater is 30% by mass or smaller based on a total of an amount of

the disulfide compound and amounts of the polysulfide compounds.

13. A lubricating oil composition which comprises a component (A): a base oil for a lubricating oil and a component (B): an additive for lubricating oils described in any one of claims 1 to 6.

14. A lubricating oil composition according to claim 13, wherein a content of the component (B) is in a range of 0.01 to 50% by mass.

15. A fuel oil composition which comprises a component (X): a fuel oil and a component (Y): an additive for fuel oils described in any one of claims 7 to 12.

16. A fuel oil composition according to claim 15, wherein a content of the component (Y) is in a range of 0.01 to 1,000 ppm by mass.

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