DESCRIPTION

Title of Invention: MIXTURE OF ALKYL-SUBSTITUTED DIPHENYL ETHER COMPOUNDS AND LUBRICANT

Technical Field

[0001]

The present invention relates to a mixture of alkyl-substituted diphenyl ether compounds and a lubricant. This application claims priority to Japanese Patent Application No. 2024-027640, filed on February 27, 2024, the entire contents of which are incorporated herein by reference.

Background Art

[0002]

Lubricants that generate less dust and have a low vapor pressure even in vacuum are desired as lubricants to be used in semiconductor production. As such a technique, for example, Patent Literature 1 discloses a low dust-generating lubricating oil having a vapor pressure (25°C) of 1×10^{-5} Pa or less, in which a lubricating oil with a surface tension (25°C) of 25 mN/m or more is used. Patent Literature 1 (JP 2001-152175 A) discloses alkyl-substituted diphenyl ether as one of the specific examples of low dust-generating lubricating oils.

[0003]

Patent Literature 2 (JP 5659422 B) discloses a lubricant containing an alkyl-substituted diphenyl ether composition, as a lubricating oil which has an excellent heat resistance, and a low evaporation loss in a high temperature environment. The alkyl-substituted diphenyl ether composition contained in the lubricating oil disclosed in Patent Literature 2 has a hydrocarbon group having 12 or 14 carbon atoms, wherein the number of alkyl groups attached is from 3.6 to 4.0.

Citation List

Patent Literature

[0004]

Patent Literature 1: JP 2001-152175 A

Patent Literature 2: JP 5659422 B

Summary of Invention

Technical Problem

[0005]

In semiconductor production, lubricants having a low vapor pressure are desired in order to maintain a clean operating environment. Conventionally, fluorine lubricants have been widely used as such low-vapor pressure lubricants. In light of social demands such as PFAS regulations, however, lubricants to replace fluorine lubricants are needed. As disclosed in Patent Literatures 1 and 2, lubricating oils composed of alkyl-substituted diphenyl ethers are known as fluorine-free lubricating oils, and such lubricating oils have been used as lubricating oils for semiconductor production, because of their high heat resistance and low vapor pressure. With advancements in high vacuum technology in semiconductor production in recent years, however, there is a demand for lubricants that release even fewer substances into a clean environment.

[0006]

An object of the present invention is to provide a lubricant containing alkyl-substituted diphenyl ethers with a low vapor pressure and low outgassing, and to provide an alkyl-substituted diphenyl ether mixture included in such a lubricant.

Solution to Problem

[0007]

In the mixture of alkyl-substituted diphenyl ether compounds according to the

present disclosure,

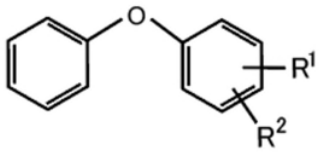
the average number of alkyl groups attached is from 2 to 3.6;

the proportion of alkyl-substituted diphenyl ether compounds to which zero or one alkyl group is attached is 1 mol% or less;

the proportion of alkyl-substituted diphenyl ether compounds to which two alkyl groups are attached is 25 mol% or more; and

the proportion of alkyl-substituted diphenyl ether compounds represented by the following formula (1) is 25 mol% or less.

[Chem. 1]



(1)

(In the formula (1), R^1 and R^2 are alkyl groups which may be the same as, or different from, each other.)

Advantageous Effects of Invention

[0008]

The present invention provides a lubricant containing alkyl-substituted diphenyl ethers with a low vapor pressure and low outgassing. The present invention also provides an alkyl-substituted diphenyl ether mixture included in such a lubricant.

Brief Description of Drawings

[0009]

FIG. 1 shows the HPLC chromatogram of the alkyl-substituted diphenyl ether mixture of Example 2.

FIG. 2 shows the ^{1}H-NMR spectrum of the alkyl-substituted diphenyl ether mixture of Example 2.

FIG. 3 shows the ^{13}C-NMR spectrum of the alkyl-substituted diphenyl ether

mixture of Example 2.

FIG. 4 shows the GC-MS chromatograms of the alkyl-substituted diphenyl ether mixtures of Example 6 and Comparative Example 1.

Description of Embodiments

[0010]

[Summary of Embodiments]

First, embodiments of the mixture of alkyl-substituted diphenyl ether compounds according to the present disclosure will be listed and described. In the present specification, the expression "(from) A to B" used to indicate a numerical range means "A or more and B or less" unless otherwise specified.

[0011]

In the mixture of alkyl-substituted diphenyl ether compounds according to present disclosure,

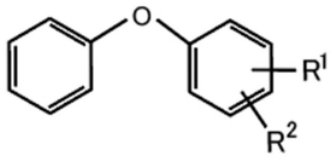
the average number of alkyl groups attached is from 2 to 3.6;

the proportion of alkyl-substituted diphenyl ether compounds to which zero or one alkyl group is attached is 1 mol% or less;

the proportion of alkyl-substituted diphenyl ether compounds to which two alkyl groups are attached is 25 mol% or more; and

the proportion of alkyl-substituted diphenyl ether compounds represented by the following formula (1) is 25 mol% or less.

[Chem. 2]



(1)

(In the formula (1), R^1 and R^2 are alkyl groups which may be the same as, or different from, each other.)

[0012]

Alkyl-substituted diphenyl ether used as a lubricant or a base oil thereof is usually a mixture of a plurality of kinds of diphenyl ether compounds varying in the number and/or position of alkyl groups attached, and is not composed of a single kind of compound. Conventional studies (such as Patent Literature 2) have proposed to control the number of alkyl groups attached to a diphenyl ether skeleton and the distribution thereof within specific ranges.

[0013]

The present inventors focused on the proportions and the structures of the respective components (namely, the compounds constituting the alkyl-substituted diphenyl ether mixture) in the alkyl-substituted diphenyl ether mixture obtained by an alkyl substitution reaction, in order to achieve an even lower vapor pressure and low outgassing in a lubricant composed of alkyl-substituted diphenyl ethers. As a result of further studies, the present inventors have discovered that a mixture of alkyl-substituted diphenyl ether compounds, in which the average number of alkyl groups attached is within a specific range, in which compounds to which zero or one alkyl group is attached and compounds to which two alkyl groups are attached are respectively present at specific proportions, and in which the proportion of compounds having two alkyl groups on one of the benzene rings of a diphenyl ether skeleton is equal to or less than a certain amount, has a low vapor pressure and low outgassing, thereby completing the present invention.

[0014]

In the mixture of alkyl-substituted diphenyl ether compounds described above, the mixture may have a number average molecular weight of from 330 to 1,800.

[0015]

In the mixture of alkyl-substituted diphenyl ether compounds described above, the average number of carbon atoms in the alkyl group may be from 6 to 28.

[0016]

In the mixture of alkyl-substituted diphenyl ether compounds described above, the proportion of alkyl-substituted diphenyl ether compounds having two alkyl groups attached thereto or alkyl-substituted diphenyl ether compounds having three alkyl groups attached thereto may be the highest.

[0017]

The mixture of alkyl-substituted diphenyl ether compounds described above may have a kinematic viscosity at 40°C of 139 mm^2/s or less.

[0018]

The mixture of alkyl-substituted diphenyl ether compounds described above may have a vapor pressure at 25°C of 1.0×10^{-6} Pa or less.

[0019]

The lubricant according to the present disclosure may be a lubricant containing the mixture of alkyl-substituted diphenyl ether compounds described above.

[0020]

[Specific Examples of Embodiments]

The mixture of alkyl-substituted diphenyl ether compounds according to the present disclosure will be described more specifically. In the following description, the “mixture of alkyl-substituted diphenyl ether compounds” is sometimes referred to as “alkyl-substituted diphenyl ether mixture”, or simply referred to as “mixture”.

[0021]

[Compounds Constituting Alkyl-substituted Diphenyl Ether Mixture]

The mixture according to the present disclosure is a mixture of alkyl-substituted diphenyl ether compounds. The “alkyl-substituted diphenyl ether mixture” as used herein is a mixture composed of alkyl-substituted diphenyl ethers and unsubstituted diphenyl ether. In the case of a composition containing an additive such as an antioxidant, in addition to alkyl-substituted diphenyl ethers, the

portion consisting of alkyl-substituted diphenyl ethers corresponds to the alkyl-substituted diphenyl ether mixture. Hereinafter, alkyl-substituted diphenyl ether is sometimes referred to as “ADE”.

[0022]

The alkyl-substituted diphenyl ether mixture according to the present disclosure includes a plurality of kinds of alkyl-substituted diphenyl ether compounds varying in the number and/or position of alkyl groups attached. The number of alkyl groups attached to each alkyl-substituted diphenyl ether compound is from 0 to 10, but not particularly limited thereto. The alkyl-substituted diphenyl ether mixture may include one or more compounds selected from the group consisting of: the compound having zero alkyl groups attached thereto (hereinafter, sometimes referred to as “non-adduct”, “diphenyl ether” or “diphenyl oxide (DPO)”), compounds having one alkyl group attached thereto (hereinafter, sometimes referred to as “monoalkyl adducts”), compounds having two alkyl groups attached thereto (hereinafter, sometimes referred to as “dialkyl adducts”), compounds having three alkyl groups attached thereto (hereinafter, sometimes referred to as “trialkyl adducts”), compounds having four alkyl groups attached thereto (hereinafter, sometimes referred to as “tetraalkyl adducts”), and compounds having five or more alkyl groups attached thereto. The number of alkyl groups attached to each alkyl-substituted diphenyl ether compound is preferably from 2 to 6, and more preferably from 2 to 5. When the number of alkyl groups attached is within the range described above, the resulting mixture tends to have a low viscosity while maintaining a low vapor pressure.

[0023]

The number of carbon atoms in each alkyl group contained in the alkyl-substituted diphenyl ether compounds constituting the mixture may be from 6 to 28, and is preferably from 6 to 24, and more preferably from 12 to 20, but not particularly limited thereto. The alkyl group may be a linear alkyl group or a branched alkyl

group, and both a linear alkyl group and a branched alkyl group may be contained in a single compound. The alkyl group is preferably a linear alkyl group from the viewpoint of heat resistance.

[0024]

Specific examples of alkyl groups include: linear alkyl groups such as hexyl group, heptyl group, octyl group, nonyl group, decyl group, undecyl group, dodecyl group, tridecyl group, tetradecyl group, pentadecyl group, hexadecyl group, heptadecyl group, octadecyl group, icosyl group, docosyl group and tetracosyl group; and branched alkyl groups such as 1-methylundecyl group, 1-ethyldecyl group, 1-methyltridecyl group, 1-ethyldodecyl group, 1-methylpentadecyl group, 1-ethyltetradodecyl group, 1-methylheptadecyl group, 1-ethyloctadecyl group, 1-methylnonadecyl group, 1-ethyloctadecyl group, 2-ethylhexyl group, 2-octyldodecyl group, 2-decyltetradecyl group, 2-dodecyl hexadecyl group, 1-butyl-1-methylpentyl group, 1-butyl-1-methylheptyl group, 1-methyl-1-pentyloctyl group, 1-hexyl-1-methylnonyl group, 1-heptyl-1-methyldecyl group, 1-methyl-1-octylundecyl group and 1-decyl-1-methyltridecyl group.

[0025]

[Alkyl-Substituted Diphenyl Ether Mixture]

The composition or the characteristics of the physical properties of the mixture as a whole will be described below, for the mixture of alkyl-substituted diphenyl ether compounds.

[0026]

As described above, the alkyl-substituted diphenyl ether mixture according to the present disclosure is a mixture of alkyl-substituted diphenyl ether compounds varying in the number of alkyl groups attached. The average number of alkyl groups contained in each of the compounds constituting the mixture is referred to as the “average number of alkyl groups attached” in the mixture. The average number of

alkyl groups attached in the alkyl-substituted diphenyl ether mixture according to the present disclosure is from 2 to 3.6, and preferably from 2.1 to 3.3. When the average number of alkyl groups attached is within the range described above, the alkyl-substituted diphenyl ether mixture is liquid at normal temperature (20°C) and is capable of exhibiting physical properties suitable for use as a base oil for a lubricant. The method of calculating the average number of alkyl groups attached will be described in detail in the Examples.

[0027]

The proportion of alkyl-substituted diphenyl ether compounds having zero or one alkyl group is 1 mol% or less, preferably 0.6 mol% or less, more preferably 0.3 mol% or less, and particularly preferably 0 mol%, with respect to the total amount of the mixture. When the proportion of the non-adduct and the monoalkyl adducts is 1 mol% or less, the amount of outgassing can be reduced. In the present specification, the proportions of alkyl-substituted diphenyl ether compounds are values determined by HPLC under the conditions shown in the Examples to be described later.

[0028]

The proportion of alkyl-substituted diphenyl ether compounds having two alkyl groups is 25 mol% or more, preferably 30 mol% or more, and more preferably 35 mol% or more, with respect to the total amount of the mixture. When the proportion of alkyl-substituted diphenyl ether compounds having two alkyl groups is 25 mol% or more, it is thought that the viscosity of the mixture can be maintained low, leading to a low torque when used as a lubricant.

[0029]

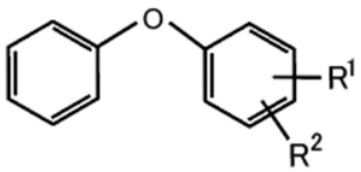
In the alkyl-substituted diphenyl ether mixture according to the present disclosure, the proportion of alkyl-substituted diphenyl ether compounds having zero or one alkyl group attached thereto is 1 mol% or less, and the proportion of alkyl-substituted diphenyl ether compounds having two alkyl groups attached thereto is 25

mol% or more.

[0030]

The proportion of alkyl-substituted diphenyl ether compounds represented by the formula (1) is 25 mol% or less, preferably 20 mol% or less, and more preferably 15 mol% or less, with respect to the total amount of the mixture. The alkyl-substituted diphenyl ether compounds represented by the formula (1) are dialkyl adducts, and compounds in which two alkyl groups are attached to one of the two aromatic rings of a diphenyl ether skeleton. The dialkyl adducts are composed of compounds in which one alkyl group is substituted on each of the two aromatic rings of diphenyl ether, and compounds in which two alkyl groups are substituted on one of the two aromatic rings. When the proportion of the compounds in which two alkyl groups are substituted on one of the two aromatic rings, of these two kinds of compounds, is 25 mol% or less, the vapor pressure can be reduced. Conventionally, it has been considered that the vapor pressure of alkyl-substituted diphenyl ether is correlated with the molecular weight of alkyl-substituted diphenyl ether compounds, and the substitution positions of the alkyl groups do not affect the vapor pressure. However, the present inventors have surprisingly discovered that the vapor pressure decreases when the amount of diphenyl ether compounds having a structure represented by the formula (1) in the mixture is small, even if the number average molecular weight of the mixture is the same.

[Chem. 3]



(1)

(In the formula (1), R^1 and R^2 are alkyl groups which may be the same as, or different from, each other.)

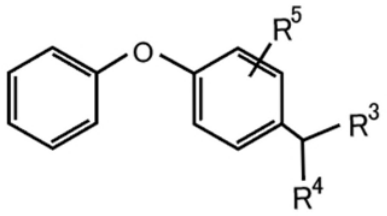
[0031]

R^1 and R^2 in the formula (1) shown above may be the alkyl groups as described above, and are preferably alkyl groups having from 6 to 24 carbon atoms.

[0032]

Further, the proportion of compounds represented by the following formula (2), among dialkyl adducts, is preferably 15 mol% or less. The alkyl-substituted diphenyl ether compounds represented by the formula (2) are dialkyl adducts in which: two alkyl groups are attached to one of the two aromatic rings of a diphenyl ether skeleton; the alkyl group attached to the para position is a branched alkyl group, and the alkyl group corresponding to the shorter branch of the branched chain has a length longer than that of butyl group or a longer alkyl group. It is thought that vapor pressure can be reduced when the proportion of such compounds is 15 mol% or less.

[Chem. 4]



(2)

(In the formula (2), R^3 and R^4 are alkyl groups, and the shorter alkyl chain of R^3 and R^4 has four or more carbon atoms; and R^5 is an alkyl group.)

[0033]

R^3, R^4 and R^5 in the formula (2) above may be the alkyl groups as described above, R^5 is preferably an alkyl group having from 6 to 24 carbon atoms, and the sum of the carbon atoms in R^3 and R^4 is preferably from 5 to 23. Typically, R^3 may be a methyl group, an ethyl group or a propyl group, and R^4 may be a nonyl group, a decyl group or an undecyl group.

[0034]

In the mixture, it is preferred that the proportion of alkyl-substituted diphenyl ether compounds to which two alkyl groups are attached (dialkyl adducts) or alkyl-

substituted diphenyl ether compounds to which three alkyl groups are attached (trialkyl adducts) is the highest. When the proportion of such compounds is the highest, the mixture can have a low viscosity while maintaining a low vapor pressure. When the proportion of the non-adduct and the monoalkyl adducts is 1 mol% or less and the proportion of the dialkyl adducts or the trialkyl adducts is the highest, in the mixture, it is possible to obtain an alkyl-substituted diphenyl ether mixture which has a low vapor pressure and low outgassing while having an appropriate kinematic viscosity as a lubricant.

[0035]

In the alkyl-substituted diphenyl ether mixture according to the present disclosure, the average number of carbon atoms in the alkyl group (namely, the average number of carbon atoms contained in one alkyl group) is preferably from 6 to 28. The average number of carbon atoms in the alkyl group is a real number that represents the average number of carbon atoms per alkyl group contained in the alkyl-substituted diphenyl ether compounds included in the alkyl-substituted diphenyl ether mixture. The average number of carbon atoms in the alkyl group can be determined by the method shown in the Examples to be described later.

[0036]

Further, the lower limit of the average total number of carbon atoms in the alkyl groups in the alkyl-substituted diphenyl ether mixture according to the present invention is 12 or more, preferably 24 or more, and more preferably 28 or more. The upper limit of the average total number of carbon atoms in the alkyl groups in the mixture is 100 or less, preferably 80 or less, more preferably 70 or less, and still more preferably 60 or less. The term “total number of carbon atoms in the alkyl groups” refers to the sum of the number of carbon atoms in the alkyl group or groups contained in one molecule of the alkyl-substituted diphenyl ether compounds included in the alkyl-substituted diphenyl ether mixture. The term “average total number of carbon

atoms in the alkyl groups" refers to the average total number of carbon atoms in the alkyl group or groups in each alkyl-substituted diphenyl ether compound included in the alkyl-substituted diphenyl ether mixture. The average total number of carbon atoms in the alkyl groups in the alkyl-substituted diphenyl ether mixture is a real number calculated as the product of the average number of carbon atoms in the alkyl group and the average number of alkyl groups attached.

[0037]

The alkyl-substituted diphenyl ether mixture according to the present disclosure preferably has a number average molecular weight of from about 330 to 1,800, more preferably from about 500 to 1,200, and still more preferably from about 500 to 1,000. A higher number average molecular weight tends to result in an excellent heat resistance but an excessively high kinematic viscosity. A lower number average molecular weight leads to a lower kinematic viscosity but tends to result in poor heat resistance. When the number average molecular weight is within the range described above, it is possible to obtain an alkyl-substituted diphenyl ether mixture having a low viscosity and an excellent heat resistance. In the present specification, the number average molecular weight of the alkyl-substituted diphenyl ether mixture is a value determined by ^{1}H-NMR, as will be shown in the Examples to be described later. Hereinafter, the number average molecular weight is sometimes simply referred to as "average molecular weight".

[0038]

[Physical Properties of Alkyl-Substituted Diphenyl Ether Mixture]

The viscosity of the alkyl-substituted diphenyl ether mixture according to the present disclosure is not limited as long as the effects of the present disclosure can be obtained. However, the kinematic viscosity at 40°C of the mixture may be from 25 to 139 mm^2/s, and is preferably from 50 to 130 mm^2/s. Further, the kinematic viscosity at 100°C of the mixture may be from 3 to 16 mm^2/s, and is preferably from

8 to 16 mm^2/s. The viscosity index (VI) is not particularly limited as well, and may be from 100 to 135, and is preferably from 110 to 135. When the kinematic viscosities and VI are within the ranges described above, it is possible to obtain a lubricant which has a low volatility and low outgassing while having an appropriate viscosity in a wide temperature range as a lubricant. The method of measuring the kinematic viscosities will be described in detail in the Examples.

[0039]

The alkyl-substituted diphenyl ether mixture according to the present disclosure preferably has a vapor pressure (Pa) at 25°C of 1.0×10^{-6} Pa or less, and more preferably 1.0×10^{-7} Pa or less. Since the alkyl-substituted diphenyl ether mixture according to the present disclosure has an extremely low vapor pressure, the dispersion of droplets is reduced even in a high-vacuum environment, making it possible to provide a lubricant capable of maintaining a clean environment. The method of measuring the vapor pressure at 25°C will be described in detail in the Examples.

[0040]

The alkyl-substituted diphenyl ether mixture according to the present disclosure preferably has low outgassing. For example, in the case of measuring outgassing by the method to be described in detail in the Examples, the peak area in the corresponding region of the resulting chromatogram is preferably 5.0×10^8 or less, and more preferably 1.0×10^8 or less. The alkyl-substituted diphenyl ether mixture according to the present disclosure can provide a lubricant which has low outgassing, in which the dispersion of droplets is reduced even in a high-vacuum environment and which is capable of maintaining a clean environment, in addition to having an extremely low vapor pressure.

[0041]

[Production Method]

The alkyl-substituted diphenyl ether mixture described above can be produced, for example, by the following method, but not particularly limited thereto.

[0042]

A mixture including alkyl-substituted diphenyl ether compounds can be obtained by the Friedel-Crafts reaction of diphenyl ether and an olefin or an alkyl halide, using aluminum chloride as a catalyst. Subsequently, the resulting mixture is purified. Low boiling components such as unreacted raw materials are removed by adsorption or distillation. Further, distillation is repeated to obtain the mixture of alkyl-substituted diphenyl ether compounds according to the present disclosure.

[0043]

More specifically, for example, aluminum chloride as a catalyst is added to diphenyl ether, and the resulting mixture is heated to a temperature of from 80 to 130°C to homogeneously dissolve the catalyst in diphenyl ether. Thereafter, an α-olefin(s) in an equivalent of from 1 to 4 with respect to one equivalent of diphenyl ether is/are added dropwise to the resulting solution over 1 to 5 hours, while maintaining the temperature within the range of from 80 to 125°C. After the completion of the dropwise addition, the resulting mixture is aged at a temperature of from 80 to 110°C for 5 to 90 minutes. The resulting reaction mixture is allowed to cool naturally until the temperature thereof decreases to normal temperature. An alkali neutralizer is added thereto, the mixture is stirred at 80 to 90°C for about 30 minutes, and then activated white clay is further added, followed by stirring at 80 to 90°C for 0.5 to 3 hours. Subsequently, the resulting mixture is allowed to cool naturally to 30°C, and then filtered under reduced pressure, followed by distillation under reduced pressure. Further, distillation is carried out once or a plurality of times, under the conditions of a temperature of from about 150 to 220°C and a pressure of from about 1.0×10^{-2} to 1.0×10^{-4} Pa. Any ordinary distillation method such as reduced-pressure (topping) distillation or molecular distillation can be used to carry out the distillation described

above, but molecular distillation is suitable. The distillation is preferably carried out at a pressure (from about 1.0×10^{-2} to 1.0×10^{-4} Pa) lower than that in conditions conventionally used. Carrying out the distillation at a pressure lower than usual makes it easier to obtain the mixture of alkyl-substituted diphenyl ether compounds according to the present disclosure.

[0044]

[Lubricant]

The alkyl-substituted diphenyl ether mixture according to the present disclosure can be suitably used as a lubricant (such as a lubricating oil or grease). Further, the alkyl-substituted diphenyl ether mixture according to the present disclosure is used singly as a lubricant, or alternatively, combined with another compound or composition to be used as a base oil for a lubricant. For example, in order to further improve the performance of the resulting lubricant, or to give an additional performance(s) as necessary, the lubricant may contain, in addition to the alkyl-substituted diphenyl ether mixture according to the present disclosure, a mineral oil, or a synthetic oil such as an α-olefin oligomer, a polyol ester, a diester, a polyalkylene glycol, a silicone oil, a modified silicone oil, an alkyl-substituted benzene oil, a multiply-alkylated cyclopentane oil or a silahydrocarbon oil, as long as the effects according to the present disclosure are not impaired. If necessary, the lubricant may further contain any of various types of additives, such as, for example, an antioxidant, an extreme pressure agent, a friction modifier, a metal inactivator, a defoaming agent, a viscosity increasing agent, a colorant and a thickener, singly or in combination of a plurality of kinds thereof.

[0045]

Any antioxidant commonly used in a lubricant can be used as an additive, without particular limitation. Examples of the antioxidant include phenolic compounds, amine compounds, phosphorus compounds and sulfur compounds.

[0046]

Examples of the extreme pressure agent include phosphorus compounds and sulfur compounds.

Examples of the friction modifier include: molybdenum compounds such as molybdenum dithiocarbamate; and fatty acid derivatives such as glycerol monostearate.

Examples of the metal inactivating agent include benzotriazole compounds, tolyl triazole compounds, thiadiazole compounds and imidazole compounds.

Examples of the defoaming agent include polyacrylates and styrene ester polymers.

Examples of the thickener include inorganic thickeners such as metal soaps (such as lithium soaps), silica, graphite, clays (such as hectorite and bentonite); and organic thickeners such as polyureas, polyether ether ketone (PEEK), polyphenylsulfide (PPS), melamine cyanurate (MC), PTFE, polyesters and polyacrylic acid.

[0047]

In the case of incorporating the alkyl-substituted diphenyl ether mixture according to the present disclosure into a lubricant, the content ratio of the mixture is preferably from about 50 to 100% by mass with respect to the total amount (total mass) of the lubricant, but not particularly limited thereto. In such a case, the content of the additive(s) and the like in the lubricant is preferably from about 0 to 50% by mass. When the content ratio and the content thereof are within the ranges described above, a lubricant with a low volatility and low outgassing can be obtained.

[0048]

Further, it is also possible to use the alkyl-substituted diphenyl ether mixture according to the present disclosure as an additive for a lubricant. In such a case, the content of the alkyl-substituted diphenyl ether mixture is preferably from about 1 to

49% by mass with respect to the total amount (total mass) of the lubricant. In addition, the alkyl-substituted diphenyl ether mixture according to the present disclosure can also be used as a component of a lubricant or a grease to be used in a high-vacuum environment.

[0049]

The lubricant containing the alkyl-substituted diphenyl ether mixture can be suitably used as a lubricating oil for bearings, a grease base oil, a refrigerating machine oil, a sealing oil or the like. In particular, the lubricant can be suitably used as any of various types of lubricating oils to be used in a high-vacuum environment, such as, for example, bearing oils (fluid bearing oils, oils for oil-impregnated bearings), grease base oils, oils for oil-impregnated plastics, gear oils, jet engine oils, adiabatic engine oils, gas turbine oils, automatic transmission oils, vacuum pump oils, hydraulic fluids, and oils for drive members (such as guide rails, bearings and ball screws) in semiconductor production equipment and measuring apparatuses.

Examples

[0050]

The present invention will be described below more specifically. However, the present invention is in no way limited to these Examples.

[0051]

[Preparation of Alkyl-Substituted Diphenyl Ether Mixtures (Crude Mixtures)]

Diphenyl ether (manufactured by The Dow Chemical Japan Limited) and aluminum chloride were introduced into a flask, and the mixture was stirred at 85°C. Subsequently, an olefin(s) was/were added dropwise to the flask. After the completion of dropwise addition, the resulting mixture was stirred at 95°C for one hour. Each mixture was allowed to cool to normal temperature, and sequentially subjected to an adsorption treatment with activated white clay and Kyoward

(registered trademark). Thereafter, low boiling components such as unreacted skeletal compounds were removed by topping distillation (at a pressure of from 40 to 200 Pa), to obtain each of ADEs 1 to 6. The amounts (g) of diphenyl ether, olefins and aluminum chloride introduced and the types thereof are shown in Table 1.

[0052]

[Table 1]

	ADE-1	ADE-2	ADE-3	ADE-4	ADE-5	ADE-6
Diphenyl ether (DPO) (g)	80	200	180	160	95	160
Olefin 1	1-dodecene	1-dodecene	1-octadecene	1-dodecene	1-dodecene	1-dodecene
Equivalent Relative to DPO Skeleton	1.9	0.46	0.7	2.0	1.1	2.0
Amount Introduced (g)	150.0	91.8	180.0	321.2	107.5	321.2
Olefin 2	-	1-tetradecene	-	1-tetradecene	1-tetradecene	1-tetradecene
Equivalent Relative to DPO Skeleton		0.40		1.7	1.0	1.7
Amount Introduced (g)		91.8		321.2	107.5	321.2
Aluminum chloride						
Equivalent Relative to Olefin	0.0084	0.0074	0.0082	0.0267	0.0063	0.0095
Amount Introduced (g)	1.0	1.0	1.0	12.6	1.0	4.5
Appearance	light-yellow liquid	light-yellow liquid	light-yellow liquid	light-yellow liquid	light-yellow liquid	light-yellow liquid
Amount Obtained (g)	170	115	126	683	230	683
Yield (mol%)	73.9%	30.0%	35.0%	85.1%	74.2%	85.1%

[0053]

[Purification of Alkyl-Substituted Diphenyl Ether Mixtures (Crude Mixtures)]

ADEs 1 to 6 were subjected to distillation under the following conditions to obtain alkyl-substituted diphenyl ether mixtures varying in the proportions of alkyl-substituted products. In the distillation conditions used in the following procedures, the pressure is set lower than that in distillation conditions conventionally used.

[0054]

(1) Distillation of ADE-1

ADE-1 was subjected to molecular distillation at 170°C and at 1.0×10^{-3} Pa to obtain a colorless transparent liquid from which high molecular weight components had been removed, as a distillate. The distillate was subjected to molecular distillation at 150°C and at 1.0×10^{-3} Pa to obtain a colorless transparent liquid from which low molecular weight components had been removed, as a distillation residue. The distillation residue was subjected to molecular distillation at 160°C and at 1.0×10^{-3} Pa to obtain a colorless transparent liquid (Comparative Example 3) as a distillate, and a colorless transparent liquid (Example 1) as a distillation residue.

[0055]

(2) Distillation of ADE-2 and ADE-3

Fifty percent by weight each of ADE-2 and ADE-3 were mixed, and the resulting mixture was subjected to molecular distillation at 1×10^{-2} Pa or less and at 180°C to remove low molecular weight components, thereby obtaining a distillation residue (Example 2). The mixture before the molecular distillation was used as Comparative Example 4.

[0056]

(3) Distillation of ADE-4 (Material A)

ADE-4 was subjected to molecular distillation at 1×10^{-2} Pa or less and at

215°C to obtain a light-yellow transparent liquid (Material A) from which low molecular weight components had been removed.

[0057]

(4) Distillation of ADE-5, Part 1 (Material B)

ADE-5 was subjected to topping distillation at 320°C and at 60 Pa to obtain a yellow transparent liquid from which low molecular weight components had been removed, as a distillation residue. The distillation residue was further subjected to molecular distillation at 1.0×10^{-2} Pa or less and at 220°C to obtain a light-yellow transparent liquid (Material B) from which high molecular weight components had been removed, as a distillate.

[0058]

(5) Distillation of ADE-5, Part 2

ADE-5 was subjected to molecular distillation at 1.0×10^{-2} Pa or less and at 150°C to remove low molecular weight components, thereby obtaining a light-yellow liquid (Example 6).

[0059]

[Preparation of Evaluation Samples]

Example 1: The colorless transparent liquid obtained as the distillation residue in the section (1) described above was used as Example 1.

Example 2: The distillation residue obtained in the section (2) described above was used as Example 2.

Example 3: A liquid obtained by mixing Example 1 and Material B at a weight ratio of 50:50 was used as Example 3.

Example 4: A liquid obtained by mixing Example 1 and Material B at a weight ratio of 20:80 was used as Example 4.

Example 5: A liquid obtained by mixing Example 1 and Material A at a weight ratio of 35:65 was used as Example 5.

Example 6: The light-yellow liquid obtained as the distillation residue in the section (5) described above was used as Example 6.

Comparative Example 1: ADE-5 was used as Comparative Example 1.

Comparative Example 2: ADE-6 was used as Comparative Example 2.

Comparative Example 3: The colorless transparent liquid obtained as the distillate in the section (1) described above was used as Comparative Example 3.

Comparative Example 4: The mixture which had been obtained by mixing 50% by weight each of ADE-2 and ADE-3 and which is before the molecular distillation, was used as Comparative Example 4.

[0060]

[Composition Analysis of Alkyl-Substituted Diphenyl Ether Mixtures]

The composition analysis was carried out by HPLC and NMR, for each of the alkyl-substituted diphenyl ether mixtures of Examples 1 to 6 and Comparative Examples 1 to 4. The results are shown in Table 2.

[0061]

[Table 2]

		Example 1	Comparative Example 3	Example 3	Example 4	Example 5	Example 6	Comparative Example 1	Example 2	Comparative Example 4	Comparative Example 2
Composition	Number of Carbon Atoms in Alkyl Group	C12	C12	C12, C14	C12, C14	C12, C14	C12, C14	C12, C14	C12, C14, C18	C12, C14, C18	C12, C14
	Average Number of Alkyl Groups Attached	2.1	2.0	2.5	2.7	3.3	2.9	2.9	2.3	2.3	3.7
	Average Number of Carbon Atoms in Alkyl Group	26	24	31	35	41	38	38	36	33	48
	Molecular Weight	528	508	624	664	767	702	698	686	625	843
	Non-Adduct + Monoalkyl Adducts (mol%)	0.0	0.0	0.0	0.0	0.0	0.0	1.1	0.6	1.3	0.4
	Dialkyl Adducts (di-adducts) (mol%)	86.6	99.0	52.5	32.0	31.5	25.3	30.3	74.0	80.1	5.3
	Trialkyl Adducts (tri-adducts) (mol%)	13.4	1.0	42.8	60.4	21.7	45.5	43.9	23.7	10.6	22.4
	Tetraalkyl Adducts (tetra-adducts) (mol%)	0.0	0.0	4.8	7.7	33.2	24.9	24.7	1.7	8.0	44.5
	Adducts with Five or More Alkyl Groups (mol%)	0.0	0.0	0.0	0.0	13.6	4.3	0.0	0.0	0.0	27.4
	Components Having Highest Content	di-adducts	di-adducts	di-adducts	tri-adducts	tetra-adducts	tri-adducts	tri-adducts	di-adducts	di-adducts	tetra-adducts
	Compounds Represented by Formula (1) (%)	21.5	46.8	≤ 19.9	≤ 18.9	≤ 8.8	≤ 10.0	10.0	23.5	-	≤ 5.3
Physical Properties	Kinematic Viscosity at 40°C (mm^2/s)	58.6	51.0	75.7	86.3	105.3	110.7	101.6	96.0	81.6	139.9
	Kinematic Viscosity at 100°C (mm^2/s)	8.3	7.1	9.9	11.1	13.2	13.4	12	12.0	10.9	16.5
	Viscosity Index VI	111	96	111	116	122	118	108	121	120	126
	Vapor Pressure at 25°C (Pa)	9.5×10^{-7}	1.4×10^{-4}	6.6×10^{-7}	6.3×10^{-8}	8.0×10^{-8}	9.3×10^{-8}	5.6×10^{-8}	1.3×10^{-8}	6.4×10^{-8}	1.8×10^{-9}
	Amount of Outgassing	1.2×10^{8}	1.0×10^{8}	6.7×10^{7}	7.0×10^{7}	6.5×10^{7}	4.5×10^{7}	5.1×10^{8}	1.5×10^{8}	1.2×10^{9}	7.4×10^{8}

[0062]

The composition of each mixture shown in Table 2 was determined by the measurement and calculation in accordance with the following methods.

For each of the alkyl-substituted diphenyl ether mixtures of Examples 1 to 6 and Comparative Examples 1 to 4, the proportions (mol%) of the diphenyl ether compounds varying in the number of alkyl groups attached, and the average number of alkyl groups attached were calculated, and the structural analysis of dialkyl adducts were carried out.

[0063]

[Measurement and Calculation of Diphenyl Ether Compounds Varying in the Number of Alkyl Groups Attached]

1. HPLC Analysis

Diphenyl ether compounds varying in molecular weight were separated using HPLC. From the resulting peak areas, the proportions (mol%) of the diphenyl ether compounds varying in the number of alkyl substituents were calculated.

· HPLC apparatus: 1220 Infinity LC, manufactured by Agilent Technologies Inc.

· Developing solvent: $CHCl_3$/acetonitrile = 50:50

· Column: TSKgel ODS-100V, 5 μm, 4.6 mm × 15 cm, manufactured by Tosoh Corporation

· Flow velocity: 1.00 mL/min

· Detector: UV 254 nm

[0064]

FIG. 1 shows the results of the HPLC analysis of the mixture of Example 2. Referring to FIG. 1, it can be seen that the peaks of the dialkyl adducts were observed at retention times from 3.0 to 4.3 min., the peaks of trialkyl adducts were observed at retention times from 4.3 to 6.6 min, and the peaks of tetraalkyl adducts

were observed at retention times from 6.6 to 10.0 min.

[0065]

2. NMR Analysis

Each synthesized mixture was identified by NMR. The NMR measurements were carried out using the following apparatus and under the following measurement conditions.

· NMR apparatus: FT NMR apparatus, JNM-ECX series, 400 MHz, manufactured by JEOL Ltd.

· Solvent: None

· Temperature: 70°C

· Measurement conditions:

^{1}H- NMR: the number of scans: 8

^{13}C- NMR: the number of scans: 1,024

[0066]

(a) Determination of Number of Alkyl Groups Attached

A description will be given below with reference to Example 2 as an example.

FIG. 2 shows the entire ^{1}H-NMR spectrum of the mixture of Example 2. The respective integration ratios of the peaks corresponding to: the protons bound to the aromatic rings; the protons bound to the benzylic carbons of the alkyl groups; and the protons bound to the carbons excluding the benzylic carbons of the alkyl groups; were calculated from the ^{1}H-NMR spectrum. The letters a to c in the ^{1}H-NMR spectrum represent the following:

· The protons bound to the aromatic rings: the integration ratio of the peaks within the range of from 6.5 to 7.5 ppm is defined as a (a = 1 as a reference).

· The protons bound to the benzylic carbons of the alkyl groups: the integration ratio of the peaks within the range of from 2.2 to 3.4 ppm is defined as b.

· The protons bound to the carbons excluding the benzylic carbons of the alkyl groups: the integration ratio of the peaks within the range of from 0.7 to 1.7 ppm is defined as c.

Further, the number of protons in the attached alkyl chains is defined as d. For example, in cases where 1-dodecene and 1-tetradecene are attached, the average composition of the attached alkyl chains corresponds to $C_{13}H_{27}$, and therefore, d = 27.

The average number of alkyl groups attached was calculated by the following equation.

Average number of alkyl groups attached = $10 \times (b + c) / (b + c + a \times d)$

[0067]

(b) Structural Analysis of Dialkyl Adducts

Dialkyl adducts in the mixture were separated by the HPLC described above. The resulting dialkyl adduct components were determined by ^{13}C-NMR measurement.

FIG. 3 shows the ^{13}C-NMR spectrum of the dialkyl adduct components separated from the mixture of Example 2. The respective integration ratios of the peaks corresponding to “the carbon atoms adjacent to the carbon atoms to which the oxygen atom of ADEs in which two alkyl groups are attached to one of the benzene rings is bound”, and “the carbon atoms adjacent to the carbon atoms to which the oxygen atom of ADEs in which one alkyl group is attached to each of the two benzene rings is bound” were calculated from the ^{13}C-NMR spectrum.

· The carbon atoms adjacent to the carbon atoms to which the oxygen atom of ADEs in which two alkyl groups are attached to one of the benzene rings is bound: A1 (within the range of from 158 to 160 ppm) + A3 (within the range of from 152 to 154 ppm)

· The carbon atoms adjacent to the carbon atoms to which the oxygen atom of ADEs in which one alkyl group is attached to each of the two benzene rings is bound: A2

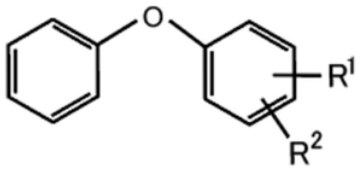
(within the range of from 154 to 157 ppm)

· The proportion of the dialkyl adducts in which alkyl groups are attached to one of the aromatic rings:

(S1) = (A1 + A3) / (A1 + A2 + A3)

· The proportion of the compounds represented by the following general formula (1) (namely, alkyl-substituted diphenyl ether compounds in which alkyl groups are attached to one of the aromatic rings) in the mixture = the proportion of dialkyl adducts × (S1)

[Chem. 5]



(1)

(In the formula (1), R^1 and R^2 are alkyl groups which may be the same as, or different from, each other.)

[0068]

[Analysis of Physical Properties of Alkyl-Substituted Diphenyl Ether Mixtures]

The vapor pressure at 25°C, the kinematic viscosities (at 40°C and 100°C), the viscosity index and outgassing were measured, for each of the alkyl-substituted diphenyl ether mixtures of Examples 1 to 6 and Comparative Examples 1 to 4. The results are summarized in Table 2. Each measurement was carried out by the following procedure.

[0069]

(a) Vapor Pressure at 25°C

Using a simultaneous thermal analyzer (STA7200RV, manufactured by Hitachi High-Tech Science Corporation), the amount of evaporation was measured under the following conditions, and the vapor pressure at 25°C of each measurement sample was calculated using the following Equations 1 to 3.

· Test container: Deep aluminum pan

· Amount of Sample: 5 mg

· Gas flow rate: N_2, 100 mL/min

· Measurement temperatures: Isothermal measurements at four different temperatures (the temperatures vary depending on the measurement sample)

· Measurement time: 4 hours

The lowest measurement temperature T1 was set to a temperature at which the weight loss (amount of evaporation) at the end of the measurement was from 3 to 20%. The other three measurement temperatures were set in increments of 10°C from T1. In other words, T_2 was set to T_1 + 10°C, T_3 was set to T_1 + 20°C, and T_4 was set to T_1 + 30°C.

The evaporation rate per unit area was calculated using Equation 1 from the slope of the curve within the 2 to 3% range of the amount of evaporation. The thus calculated evaporation rate A was substituted into Equation 2 to calculate the vapor pressure p at each of the temperatures T_1 to T_4. The same calculation was carried out for each measurement sample.

[0070]

[Math 1]

$$A = -\frac{1}{U} \times \frac{dm}{dt} \qquad \text{(Equation 1)}$$

[Math 2]

$$p = \frac{A}{\alpha}\sqrt{\frac{2\pi RT}{M}} = \frac{A}{4.38\times10^3} \times \frac{1}{\sqrt{M/T}} \qquad \text{(Equation 2)}$$

p: vapor pressure [Pa], U: cross-sectional area of sample pan [m^2], m: weight [kg], t:

time [s], A: evaporation rate [$kg \cdot s^{-1} \cdot m^{-2}$]

α: condensation coefficient (set to 3.9×10^{-5}), T: temperature [K], R: gas constant [$J \cdot mol^{-1} \cdot K^{-1}$], M: molecular weight [$g \cdot mol^{-1}$]

The condensation coefficient is 1 in an absolute vacuum. However, in the present measurement, α was determined by measuring a substance whose vapor pressure is known by this method to calculate the value of $p \times \alpha$, and dividing the calculated value by the known vapor pressure.

[0071]

A regression curve was generated from the vapor pressures at the temperatures T_1 to T_4, by plotting the common logarithm of the vapor pressure against the reciprocal of the absolute temperature, and the vapor pressure at 25°C was then extrapolated using Equation 3.

[Math 3]

$$\mathrm{Log}P = \frac{B}{T} + C \qquad \text{(Equation 3)}$$

P: vapor pressure [Pa], T: temperature [K], B: slope of regression line, C: intercept of regression line

[0072]

(b) Kinematic Viscosities and Viscosity Index

The kinematic viscosities and the viscosity index (VI) of each of the resulting mixtures and compositions were measured in accordance with JIS K 2283.

[0073]

(c) Measurement of Amount of Outgassing

The amount of outgassing of each measurement sample was calculated from the peak area (A + B) obtained under the following conditions, using a gas chromatography mass spectrometer (GC-MS).

· Apparatus: Gas chromatography mass spectrometer (5977B MS, 7890B GC) manufactured by Agilent Technologies Inc.

· Sampling conditions

· Sampling method: Dynamic headspace method

· Sampling tube / Purge gas: Tenax TA (manufactured by GERSTEL GmbH & Co. KG) / Inert gas

· Heating temperature / Heating time / Sampling time / Amount of purge gas: 150°C / 60 minutes / 10 minutes / 1,000 mL

· GC Conditions

· Injection mode: Splitless

· Column: DB-1 (60 m× 0.25 mm × 0.25 μm), manufactured by Agilent Technologies Inc.

· Temperature range: 40°C (5 minutes) to 300°C

· Heating rate: 10°C/min

A (retention time: 20 to 22 minutes) indicates the non-adduct outgas component. The peak area in this region of the chromatogram was evaluated as the amount of outgassing.

B (retention time: 30 to 36 minutes) indicates monoalkyl adduct outgas components. The peak area in this region of the chromatogram was evaluated as the amount of outgassing.

[0074]

FIG. 4 shows the chromatograms of the mixtures of Comparative Example 1 and Example 6 obtained using a gas chromatography mass spectrometer. The amount of outgassing was evaluated in the same manner for each of the mixtures of Examples 1 to 6 and Comparative Examples 1 to 4.

[0075]

As shown in Table 2, all of Examples 1 to 6, which are the mixtures of alkyl-

substituted diphenyl ether compounds according to the present disclosure, had a vapor pressure at 25°C of 1.0×10^{-6} or less and an amount of outgassing of 5.0×10^{8} or less. Further, all of the above mixtures had a kinematic viscosity at 40°C of 139 mm^2/s or less.

[0076]

In contrast, the mixtures of Comparative Example 1 and Comparative Example 4 in which the proportion of alkyl-substituted diphenyl ether compounds having zero or one alkyl group is more than 1 mol%, had a larger amount of outgassing. The mixture of Comparative Example 2 in which the average number of alkyl groups attached is more than 3.6 and the proportion of alkyl-substituted diphenyl ether compounds having two alkyl groups is less than 25 mol%, had a larger amount of outgassing, and a higher kinematic viscosity at 40°C. Further, the mixture of Comparative Example 3 in which the proportion of the compounds represented by the formula (1) is more than 25 mol% had a higher vapor pressure.

[0077]

The comparison of Example 1 and Comparative Example 3 confirmed that the proportion of the compounds represented by the formula (1) affects the vapor pressure. Further, the comparison of Examples 3 to 6 and Comparative Example 1 as well as the comparison of Example 2 and Comparative Example 4 suggest that the proportion of the non-adduct and the monoalkyl adducts affects the amount of outgassing. In addition, the comparison of Example 2 and Comparative Example 2 suggests that, when the average number of alkyl groups attached is 3.6 or less and the proportion of alkyl-substituted diphenyl ether compounds is 25 mol% or more, the resulting mixture has low kinematic viscosities at 40°C and 100°C, and thus has a good lubricity. These results have confirmed that, in addition to the average number of alkyl groups attached, the proportions of the non-adduct, the monoalkyl adducts and the dialkyl adducts as well as the proportion of the compounds represented by the formula (1)

need to be within predetermined ranges, in order to obtain a lubricant containing alkyl-substituted diphenyl ethers that has good lubricity, a low vapor pressure and low outgassing.

[0078]

[Preparation of Lubricants (Greases)]

The alkyl-substituted diphenyl ether mixture of Example 2 and a thickener were mixed and stirred using a planetary centrifugal mixer (Awatori Neritaro AR-100, manufactured by THINKY Corporation), to obtain each of Greases 1 to 6 having a grayish white color. The resulting greases were evaluated for the following physical properties. The mixing ratio of the respective materials and the measurement results are shown in Table 3. In Table 3, empty cells each indicate that the corresponding component is not added, and the symbols "-" each indicate that no measurement is performed.

(Thickener)

· PTFE 1: Dyneon TF-9207Z, manufactured by 3M Company

· PTFE 2: TLP-10F-1, manufactured by The Chemours Company

· PEEK: KETASPIRE KT-820 UFP, manufactured by Solvay Specialty Polymers Japan K.K.

· MC 1: MC-4000, manufactured by Nissan Chemical Industries, Ltd.

· MC 2: MC-6000, manufactured by Nissan Chemical Industries, Ltd.

· PPS: Ryton M1100 UFP, manufactured by Solvay Specialty Polymers Japan K.K.

[Evaluation of Lubricants (Greases)]

The cone penetration of each grease was measured using a penetrometer (RPM-201, manufactured by Rigosha & Co., Ltd.) in accordance with JIS K 2220.

The oil separation was measured using a wire mesh cone filter (manufactured by Rigosha & Co., Ltd.) in accordance with Method B defined in JIS K 2220.

The dropping point was measured using an automatic dropping point and

softening point system, DP70, manufactured by Mettler-Toledo International Inc., in accordance with JIS K 2220.

The friction coefficient is the average value within the range of from 3,000 to 3,600 seconds, measured under the following conditions using an SRV tester manufactured by Optimol Instruments Prüftechnik GmbH. Each sample was prepared by applying 0.1 g of each grease on a SUJ2 disk.

· Stroke: 4 mm

· Load / Frequency / Time: (Running-in) 1 N / 1 Hz / 60 seconds; (Main run) 10 N / 1 Hz / 3,600 seconds

· Temperature: 25°C / 100 °C

· Material and size: An SUJ2 ball with 10 mm diameter, a disk with 24 mm diameter × 7.9 mm thickness

The vapor pressure at 25°C and the amount of outgassing were measured in the same manner as in the section of “Analysis of Physical Properties of Alkyl-Substituted Diphenyl Ether Mixtures” described above.

[0079]

[Table 3]

		Grease 1	Grease 2	Grease 3	Grease 4	Grease 5	Grease 6
Base oil (mass%)	Example 2	56	58	46	52	55	47
Thickener (mass%)	PTFE 1	44					
	PTFE 2		42				
	PEEK			54			
	MC 1				48		
	MC 2					45	
	PPS						53
Evaluation	Cone Penetration / NLGI Grade	2	1	2	2	1	2
	Oil Separation / %	2.1	-	< 0.1	0.6	-	< 0.1
	Dropping Point / °C	-	> 260	> 260	> 260	> 260	> 260
	Friction Coefficient (25°C)	-	-	0.10	0.14	-	0.21
	Friction Coefficient (100°C)	-	-	0.28	0.12	-	0.23
	Vapor Pressure at 25°C / Pa	-	3.2×10^{-7}	9.4×10^{-12}	3.0×10^{-10}	-	1.3×10^{-7}
	Amount of outgassing	-	2.1×10^{6}	1.5×10^{8}	3.5×10^{5}	5.5×10^{6}	1.6×10^{8}

[0080]

As shown in Table 3, it has been confirmed that the lubricants each containing the alkyl-substituted diphenyl ether mixture according to the present disclosure have a good cone penetration and good evaluation results for the respective properties. Because of their low vapor pressure of the base oil, the lubricants whose vapor pressure and amount of outgassing are not measured are also assumed to have a low volatility and low outgassing as with other lubricants. These results have confirmed that the lubricants according to the present disclosure have a cone penetration, an oil separation, a dropping point and a friction coefficient equivalent to those of conventional lubricants, and also have a lower volatility and lower outgassing than those of conventional lubricants.

[0081]

The embodiments disclosed herein are to be considered in all respects as illustrative and not restrictive. The scope of the invention is defined by the appended claims rather than by the foregoing description, and all changes that come within the meaning and range of equivalency of the claims are intended to be embraced therein.

[0082]

Embodiments of the invention will now be further described with reference to the following numbered clauses.

[1] A mixture of alkyl-substituted diphenyl ether compounds,

wherein:

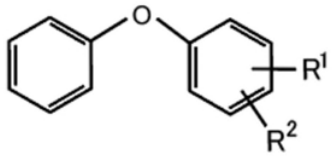
the average number of alkyl groups attached is from 2 to 3.6;

the proportion of alkyl-substituted diphenyl ether compounds having zero or one alkyl group attached thereto is 1 mol% or less;

the proportion of alkyl-substituted diphenyl ether compounds having two alkyl groups attached thereto is 25 mol% or more; and

the proportion of alkyl-substituted diphenyl ether compounds represented by the following formula (1) is 25 mol% or less:

[Chem. 6]



(1)

(wherein in the formula (1), R^1 and R^2 are alkyl groups which may be the same as, or different from, each other).

[2] The mixture of alkyl-substituted diphenyl ether compounds according to [1], wherein the mixture has a number average molecular weight of from 330 to 1,800.

[3] The mixture of alkyl-substituted diphenyl ether compounds according to [1] or [2], wherein the average number of carbon atoms in the alkyl group is from 6 to 28.

[4] The mixture of alkyl-substituted diphenyl ether compounds according to any one of [1] to [3],

wherein the proportion of alkyl-substituted diphenyl ether compounds having two alkyl groups attached thereto or alkyl-substituted diphenyl ether compounds having three alkyl groups attached thereto is the highest.

[5] The mixture of alkyl-substituted diphenyl ether compounds according to any one of [1] to [4], wherein the mixture has a kinematic viscosity at 40°C of 139 mm^2/s or less.

[6] The mixture of alkyl-substituted diphenyl ether compounds according to any one of [1] to [5] wherein the mixture has a vapor pressure at 25°C of 1.0×10^{-6} Pa or less.

[7] A lubricant containing the mixture of alkyl-substituted diphenyl ether compounds according to any one of [1] to [6].

CLAIMS

[Claim 1] A mixture of alkyl-substituted diphenyl ether compounds, wherein:

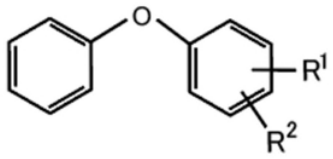
average number of alkyl groups attached is from 2 to 3.6;

proportion of alkyl-substituted diphenyl ether compounds having zero or one alkyl group attached thereto is 1 mol% or less;

proportion of alkyl-substituted diphenyl ether compounds having two alkyl groups attached thereto is 25 mol% or more; and

proportion of alkyl-substituted diphenyl ether compounds represented by the following formula (1) is 25 mol% or less:

[Chem. 1]



(1)

(wherein in the formula (1), R^1 and R^2 are alkyl groups which may be the same as, or different from, each other).

[Claim 2] The mixture of alkyl-substituted diphenyl ether compounds according to claim 1, wherein the mixture has a number average molecular weight of from 330 to 1,800.

[Claim 3] The mixture of alkyl-substituted diphenyl ether compounds according to claim 1 or claim 2, wherein average number of carbon atoms in the alkyl group is from 6 to 28.

[Claim 4] The mixture of alkyl-substituted diphenyl ether compounds according

to claim 1 or claim 2,

wherein the proportion of alkyl-substituted diphenyl ether compounds having two alkyl groups attached thereto or alkyl-substituted diphenyl ether compounds having three alkyl groups attached thereto is the highest.

[Claim 5] The mixture of alkyl-substituted diphenyl ether compounds according to claim 1 or claim 2, wherein the mixture has a kinematic viscosity at 40°C of 139 mm^2/s or less.

[Claim 6] The mixture of alkyl-substituted diphenyl ether compounds according to claim 1 or claim 2, wherein the mixture has a vapor pressure at 25°C of 1.0×10^{-6} Pa or less.

[Claim 7] A lubricant comprising the mixture of alkyl-substituted diphenyl ether compounds according to claim 1 or claim 2.