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(54) **LUBRICATING OIL COMPOSITION**

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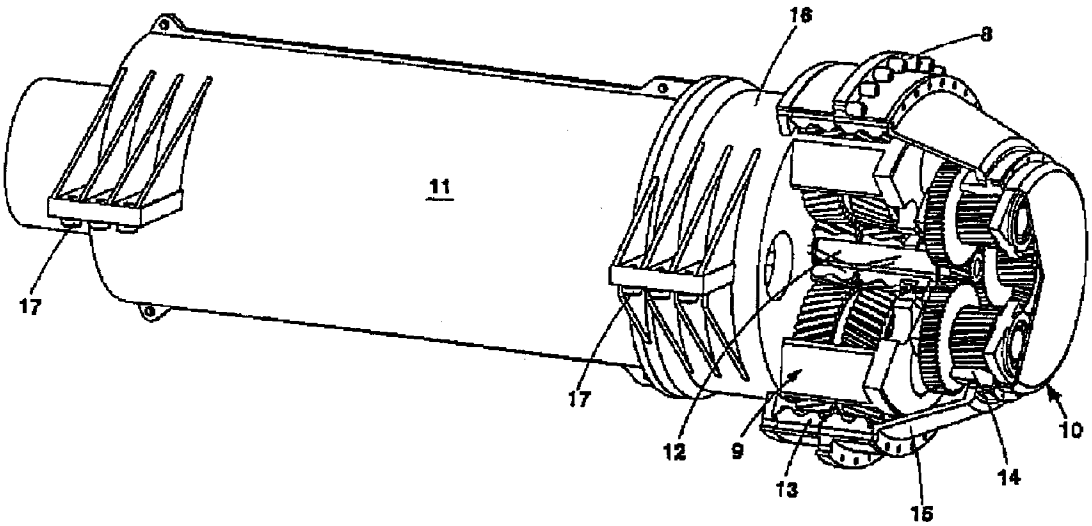
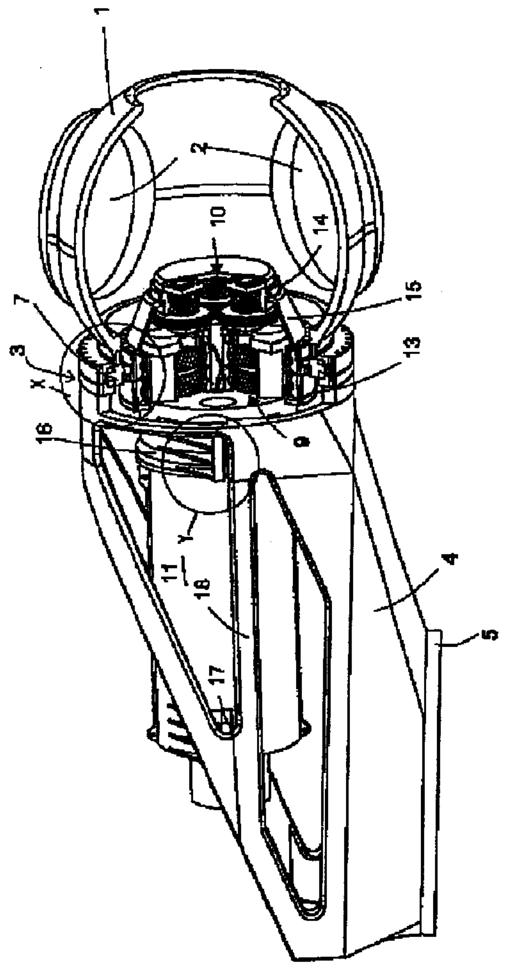
(57) **ABSTRACT**

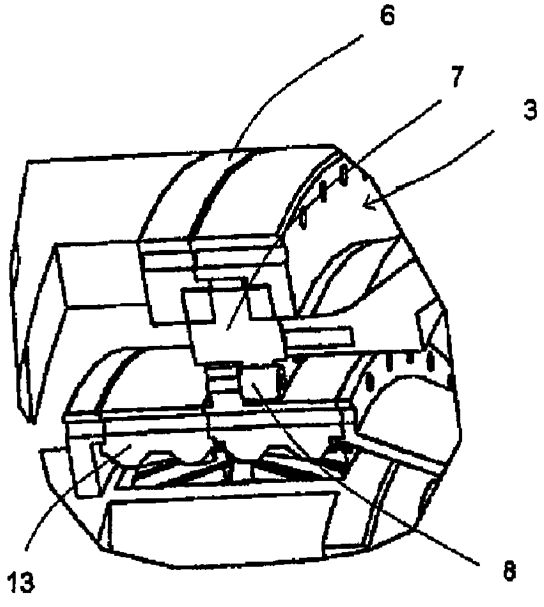
A lubricating oil composition comprising: boron atoms from about 5-75 ppm; and 100% by mass being (i) 90 to 10% of a (co)polymer of specified kinematic viscosity and acid values, primarily derived from 1-decene, (ii) 5 to 85% of a (co) polymer having specified kinematic viscosity and molecular weight distribution, mostly derived from 1-octene, and (iii) 5 to 15% of a fatty acid ester.



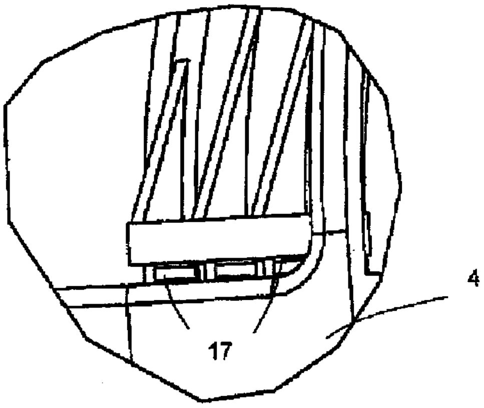
[Figure 1]



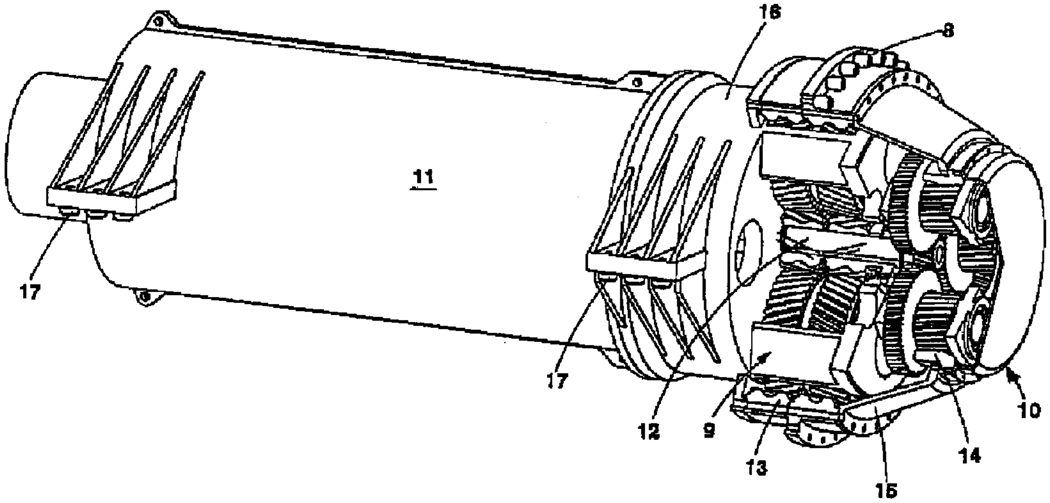
[Figure 2]



[Figure 3]



[Figure 4]



LUBRICATING OIL COMPOSITION

TECHNICAL FIELD

[0001] The present invention relates to a lubricating oil composition and a gear for a wind power generator, which is filled with the lubricating oil composition.

BACKGROUND ART

[0002] For grease lubricating oils, gear oils, hydraulic oils, etc., various properties are required, and in recent years, as the internal combustion engines and industrial machines are made to have higher performance and higher output power or as the operation conditions become severer, higher properties of engine parts, such as higher abrasion resistance, heat resistance, sludge resistance, lubricating oil consumption properties and fuel saving, have been required. The environment where the lubricating oils are used is becoming severer, while life lengthening of lubricating oils tends to be required in consideration of environmental problems, and on that account, decrease in lowering of viscosity attributable to shear stress given by a gear during the operation of engine, that is, improvement in shear stability of lubricating oils has been desired. On the other hand, in order to improve energy conversion efficiency of engine or ensure favorable lubricating properties of engine in an extremely low temperature environment, temperature viscosity properties such that oil films of lubricating oils are maintained at high temperatures and fluidity of lubricating oils is rather maintained at low temperatures are regarded as important.

[0003] Accordingly, as grease lubricating oils, gear oils, hydraulic oils, etc., those having long life, namely, low-evaporation properties, and having favorable temperature viscosity properties are desired.

[0004] Particularly in wind power generators which have been paid attention as generators of renewable energy in recent years, gear boxes to convert wind force into electric power are installed at elevated spots in order to receive more stable wind force. In addition, from the viewpoint of stable supply of wind force and direction of wind, they are often installed not only in the inland areas but also on the sea. On this account, maintenance of gears, that is, exchange of lubricating oils is very difficult, and as a result, higher shear stability is required for the lubricating oils. Moreover, from the viewpoint of efficiency of energy conversion from wind force into electric power, favorable lubricating properties at extremely low temperatures such as −40° C., that is, high fluidity of lubricating oils in a low temperature environment is desired.

[0005] Furthermore, high anti-micropitting performance is desired for gear oils for wind power generators. Micropitting is a fatigue process of gear brought about immediately before gear damage because of a cycle of excess stress in the rolling elasto-hydrodynamic lubrication (EHL) region under high load. Separation of small fragments from gear tooth surface due to repeated compression stress in this high-load state is referred to as "micropitting", and for lubricating oils used for gear oils for wind power generators, high anti-micropitting performance is desired.

[0006] In view of the above background, developments of various synthetic lubricating oils have been made (patent literatures 1 and 2, etc.).

PRIOR ART LITERATURE

Patent Literature

[0007] Patent literature 1: National Publication of International Patent No. 542524/2008

[0008] Patent literature 2: National Publication of International Patent No. 500489/2009

SUMMARY OF THE INVENTION

Problem to be Solved by the Invention

[0009] However, particularly in the fields where high shear stability is desired, such as a field of gear oils for wind power generators, there is room for improvement in the lubricating oil compositions described in the patent literature 1 and the patent literature 2.

[0010] The problem to be solved by the present invention relates to a lubricating oil composition and lubricating oil each of which has high oxidation stability and heat resistance and is excellent in temperature viscosity properties, and it is an object of the present invention to provide a lubricating oil composition having particularly high shear stability and excellent low-temperature viscosity properties that are compatible with each other and further having excellent anti-micropitting performance.

Means to Solve the Problem

[0011] In order to solve the above problem, earnest studies have been made, and as a result, it has been found that by blending specific lubricating base oil, a low-molecular weight polymer having a specific molecular weight distribution which is obtained by homopolymerizing an α-olefin of a specific number of carbon atoms or copolymerizing it with another α-olefin, and a specific ester in a specific blending ratio and introducing boron atoms in a specific amount, the resulting lubricating oil composition exhibits extremely excellent shear stability, temperature viscosity properties, low-temperature viscosity properties and anti-micropitting performance, and the present invention has been accomplished.

[0012] That is to say, the lubricating oil composition of the present invention is a lubricating oil composition comprising the following (i) to (iii), having a kinematic viscosity at 40° C. of not less than 30 mm^2/s but not more than 750 mm^2/s and containing boron atoms in an amount of not less than 5 ppm but not more than 75 ppm,

[0013] (i) 90 to 10% by mass of a (co)polymer having a kinematic viscosity at 100° C. of not more than 45 mm^2/s and having an acid value of less than 0.1 mgKOH/g, not less than 60% by mol of its constituent units being derived from 1-decene,

[0014] (ii) 5 to 85% by mass of a (co)polymer having a kinematic viscosity at 100° C. of not less than 35 mm^2/s but not more than 1,500 mm^2/s and having a molecular weight distribution, as measured by gel permeation chromatography, of not more than 1.8, not less than 90% by mol of its constituent units being derived from 1-octene, and

[0015] (iii) 5 to 15% by mass of a fatty acid ester having a kinematic viscosity at 100° C. of not more than 20 mm^2/s,

[0016] with the proviso that the total amount of (i) to (iii) is 100% by mass.

Effects of the Invention

[0017] By the present invention, a lubricating oil composition having excellent shear stability as compared with conventional lubricating oils and further having high temperature viscosity properties, excellent low-temperature viscosity properties and anti-micropitting performance is obtained. In particular, a lubricating oil composition capable of maintaining the above properties even under high-load conditions which are also referred to as "extreme pressure lubricating conditions" is obtained.

BRIEF DESCRIPTION OF THE DRAWINGS

[0018] FIG. **1** is a view to explain an example of a wind power generating apparatus of the present invention, and is a partially cutaway sectional view of the wind power generating apparatus in the longitudinal direction, and is a perspective view of the wind power generating apparatus partially visually observed from the front side of the apparatus.

[0019] FIG. **2** is an enlarged perspective view of X portion of the wind power generating apparatus shown in FIG. **1**, said X portion being visually observed.

[0020] FIG. **3** is an enlarged perspective view of Y portion of the wind power generating apparatus shown in FIG. **1**, said Y portion being visually observed.

[0021] FIG. **4** is a diagrammatical perspective view of a driving strung module used for driving a wind power generating apparatus.

MODE FOR CARRYING OUT THE INVENTION

Lubricating Oil Composition

[0022] The lubricating oil composition of the present invention has a kinematic viscosity at 40° C. of not less than 30 mm^2/s but not more than 750 mm^2/s, and more preferably has a kinematic viscosity at 40° C. of not less than 190 mm^2/s but not more than 750 mm^2/s. When the kinematic viscosity is not more than 750 mm^2/s, the lubricating oil composition is excellent in fuel saving performance and energy conversion efficiency. When the kinematic viscosity at 40° C. is not less than 30 mm^2/s, the lubricating oil composition exhibits good lubricating properties at high temperatures, and when the kinematic viscosity is not less than 190 mm^2/s, the lubricating oil composition exhibits good lubricating properties as gear oil in high-load state. When the kinematic viscosity at 40° C. is in the above range, oil films of the lubricating oils can be sufficiently maintained at high temperatures, and the lubricating oil composition exhibits more excellent shear stability.

[0023] The kinematic viscosity of the lubricating oil composition at 40° C. can be controlled by properly changing the kinematic viscosity of the below-described decene (co)polymer (i) and octene (co)polymer (ii) at 100° C. or the amounts of the decene (co)polymer (i) and the octene (co)polymer (ii) blended. For example, the kinematic viscosity of the lubricating oil composition can be controlled based on the blending ratio of the (co)polymers that is estimated by the method described in JIS K-2283 Annex 1.

[0024] The lubricating oil composition of the present invention contains (i) a (co)polymer comprising 1-decene and having a kinematic viscosity at 100° C. of not more than 45 mm^2/s and (ii) a (co)polymer comprising 1-octene and having a kinematic viscosity at 100° C. of not less than 35 mm^2/s but not more than 1,500 mm^2/s. In the present specification, the "(co)polymer" includes both of a homopolymer and a copolymer.

[0025] The total amount of the (co)polymer comprising 1-decene and the (co)polymer comprising 1-octene is preferably not less than 80% by mass in the lubricating oil composition of the present invention. When the total amount thereof is in this range, the lubricating oil composition has high oxidation stability and heat resistance.

[0026] In the present invention, the (co)polymer (i) comprising 1-decene and having a kinematic viscosity at 100° C. of not more than 45 mm^2/s is industrially known by the common name of poly-α-olefin (PAO) and is generally used as base oil for lubricating oils. Such (co)polymers comprising 1-decene are on the market with the trademarks of, for example, NEXBASE (available from NESTE OIL CORPORATION), SPECTRASYN (available from EXXONMOBIL CHEMICAL COMPANY) and DURASYN (available from INEOS), and they are readily obtainable. Although this (co)polymer is a homopolymer of 1-decene or a copolymer of 1-decene and 1-octene and/or 1-dodecene, not less than 60% by mol of its constituent units are formed from 1-decene. If the (co)polymer is a copolymer of 1-decene and 1-octene, low-temperature viscosity properties are sometimes lowered, and if it is a copolymer of 1-decene and 1-dodecene, shear stability is sometimes lowered. Therefore, a homopolymer of 1-decene is preferably used.

[0027] The acid value of the (co)polymer (i) comprising 1-decene, as measured by JIS K2501, is preferably less than 0.1 mgKOH/g, and is more preferably less than 0.05 mgKOH/g. When the acid value is in this range, the resulting lubricating oil composition is excellent in oxidation stability and heat resistance.

[0028] The kinematic viscosity of the (co)polymer (i) comprising 1-decene at 100° C. is not more than 45 mm^2/s, and is preferably not more than 11 mm^2/s. When the kinematic viscosity is not more than 11 mm^2/s, the lubricating oil composition exhibits more excellent low-temperature viscosity properties.

[0029] The (co)polymer (i) comprising 1-decene may be used singly or may be used as a mixture of two or more kinds.

[0030] The 1-decene (co)polymer can be prepared by a hitherto publicly known process. For example, by using a catalyst such as BF_3 or $AlCl_3$ and controlling polymerization temperature, polymerization time, etc., a (co)polymer having a desired molecular weight can be obtained. Through this control of molecular weight (that is, high-molecular weight gives high viscosity, and low-molecular weight gives low viscosity), the kinematic viscosity at 100° C. can be adjusted. In order to adjust the acid value to less than 0.1 mgKOH/g, the resulting (co)polymer is generally subjected to hydrogenation process in a publicly known way. The processes for preparing the 1-decene (co)polymer are disclosed in U.S. Pat. No. 3,149,178, No. 3,382,291, No. 3,742,082, No. 3,780,128, No. 4,172,855, No. 4,956,122, etc.

[0031] The (co)polymer (ii) having a kinematic viscosity at 100° C. of not less than 35 mm^2/s but not more than 1,500 mm^2/s is a (co)polymer obtained by polymerizing a monomer having 8 carbon atoms, namely, 1-octene, and the content of the constituent units of the monomer is in the range of 90 to 100% by mol, preferably 95 to 100% by mol. In other words, this (co)polymer is a homopolymer of 1-octene or a copolymer of 1-octene and another monomer. When the content is in this range, the lubricating oil composition of the present invention has properties substantially equal to those of a lubricating oil composition containing a polymer obtained by homopolymerizing 1-octene. By combining the decene (co)polymer (i) with such an octene (co)polymer (ii) in the lubricating oil composition, shear stability and low-temperature viscosity properties of the lubricating oil composition can be improved, and besides, oxidation stability, heat resistance and temperature viscosity properties can be also improved. In order to further improve the shear stability and the low-temperature viscosity properties of the lubricating oil composition, it is preferable to use a homopolymer of 1-octene together with the decene (co)polymer (i).

[0032] The monomer to mainly constitute the (co)polymer (ii) is 1-octene. If the monomer is a monomer having 6 carbon atoms, namely, 1-hexene, temperature viscosity properties and low-temperature viscosity properties are sometimes markedly lowered. If the monomer is a monomer having 10 carbon atoms, namely, 1-decene, shear stability of the lubricating oil composition is sometimes markedly lowered.

[0033] The shear stability is evaluated by a KRL shear stability test based on the method described in Deutsche Industrie Normen DIN 52350-6. The lubricating oil composition is allowed to stand at 60° C. for 20 hours under the shearing conditions (1450 rpm), and a kinematic viscosity decrease ratio, that is a ratio of a kinematic viscosity at 40° C. after the test to a kinematic viscosity at 40° C. before the test, is evaluated. As this value is decreased, the rubricating oil composition has more excellent shear stability.

[0034] 1-Octene that is a raw material can be obtained inexpensively as compared with 1-decene. In the lubricating oil composition of the present invention, therefore, the amount of 1-decene used is decreased relatively to the amount of 1-decene in such a synthetic lubricating oil composition obtained by the prior art technique as described in the patent literature 1 or the patent literature 2, so that the lubricating oil composition of the present invention has an advantage that industrial production at a low cost becomes feasible.

[0035] Constituent units other than the constituent units of 1-octene to constitute the (co)polymer (ii) are preferably those of an α-olefin (except 1-octene) of 2 to 14 carbon atoms, and the content of such constituent units is in the range of 0 to 10% by mol, preferably 0 to 5% by mol. When α-olefins are selected from the above range, and/or the α-olefin content is in the above range, oxidation stability, heat resistance and low-temperature viscosity properties of the lubricating oil composition are excellent.

[0036] The kinematic viscosity of the (co)polymer comprising 1-octene at 100° C. in the present invention is not less than 35 mm^2/s but not more than 1,500 mm^2/s, preferably not less than 150 mm^2/s but not more than 1,200 mm^2/s. When the kinematic viscosity is not less than 150 mm^2/s, the content of the (co)polymer comprising 1-octene, which has a high viscosity, is decreased relatively to the content of the (co)polymer comprising 1-decene, which is lubricating base oil in gear oil, industrial lubricating oil, gear oil for a wind power generator or lubricating oil for bearing, and therefore, the lubricating oil composition exhibits excellent low-temperature viscosity properties. When the kinematic viscosity is not more than 1,200 mm^2/s, the lubricating oil composition exhibits more excellent shear stability.

[0037] A number-average molecular weight can be measured by gel permeation chromatography (GPC) calibrated by the use of a standard substance (monodisperse polystyrene (PSt)) whose molecular weight is already known, and the number-average molecular weight (Mn) of the (co)polymer using 1-octene as a monomer in the present invention is in the range of preferably 500 to 15,000, more preferably 3,000 to 12,000. When the number-average molecular weight is in this range, the resulting lubricating oil composition is excellent in a balance between low-temperature viscosity properties and shear stability.

[0038] The molecular weight distribution (Mw/Mn) of the (co)polymer comprising 1-octene in the present invention is in the range of 1.8, preferably in the range of 1.1 to 1.8, more preferably in the range of 1.2 to 1.7. When the molecular weight distribution is in this range, shear stability of the resulting lubricating oil composition is excellent.

[0039] The (co)polymer (ii) comprising octene may be used singly or may be used as a mixture of two or more kinds.

Preparation of (Co)Polymer (Ii) Comprising Octene in the Present Invention

[0040] Such a (co)polymer comprising 1-octene as above can be prepared referring to such catalysts used in the preparation of α-olefin (co)polymers as described in Japanese Patent Laid-Open Publication No. 41303/1990, Japanese Patent Laid-Open Publication No. 41305/1990, Japanese Patent Laid-Open Publication No. 274703/1990, Japanese Patent Laid-Open Publication No. 274704/1990, Japanese Patent Laid-Open Publication No. 179005/1991, Japanese Patent Laid-Open Publication No. 179006/1991, Japanese Patent Laid-Open Publication No. 193796/1991, Japanese Patent Laid-Open Publication No. 69394/1992, Japanese Patent Laid-Open Publication No. 17589/1993, Japanese Patent Laid-Open Publication No. 122718/1994, Japanese Patent Laid-Open Publication No. 120127/1996, Japanese Patent Laid-Open Publication No. 239414/1996, Japanese Patent Laid-Open Publication No. 087716/1998, Japanese Patent Laid-Open Publication No. 212194/2000, WO 01/27124, WO 02/074855, WO 04/029062, EP 0881236 and EP 1416000.

[0041] More specifically, in the preparation of the (co)polymer using 1-octene as a monomer, a homopolymer or a copolymer can be obtained by homopolymerizing 1-octene or copolymerizing 1-octene and the above-mentioned another α-olefin in the presence of, for example, an olefin polymerization catalyst comprising:

[0042] (A) a transition metal compound (transition metal compound of a Group 4 transition metal of the periodic table, or the like), and

[0043] (B) at least one compound selected from:

[0044] (B-1) an organometallic compound,

[0045] (B-2) an organoaluminum compound,

[0046] (B-3) an organoaluminum oxy-compound, and

[0047] (B-4) a compound which reacts with the Group 4 transition metal compound (A) to form an ion pair.

[0048] The transition metal compound (A) and the compound (B) are described below in more detail.

(A) Transition Metal Compound

[0049] As the transition metal compound (A) for use in the present invention, publicly known periodic table Group IV to Group VI transition metal compounds having olefin polymerizability are employable without any restriction. For example, halogenation products of transition metals of Group IV to Group VI of the periodic table, alkylation products thereof, alkoxylation products thereof, and non-bridged or bridged metallocene compounds are employable. Of these, preferable are halogenation products of periodic table Group IV transition metals (more preferable are halogenations products of Group IVB transition metals), alkylation products thereof, alkoxylation products thereof, and non-bridged or bridged metallocene compounds. Preferred embodiments of the transition metal compound (A) are described below.

[0050] Examples of the transition metal halogenation products, the transition metal alkylation products and the transition metal alkoxylation products include titanium tetrachloride, dimethyltitanium dichloride, tetrabenzyltitanium, tetrabenzylzirconium and tetrabutoxytitanium.

[0051] The non-bridged or bridged metallocene compound is, for example, a compound represented by the following formula (1) which is a periodic table Group 4 transition metal compound having cyclopentadienyl skeleton.

$$ML_x \quad (1)$$

[0052] In the above formula, M is a transition metal atom selected from Group IV of the periodic table, preferably zirconium, titanium or hafnium of a Group IVB transition metal. x is a valence of the transition metal and is a number of L. L is a ligand or a group coordinated to the transition metal; at least one L is a ligand having cyclopentadienyl skeleton; and L other than the ligand having cyclopentadienyl skeleton is a group or an atom selected from a halogen, a hydrogen atom, a hydrocarbon group of 1 to 10 carbon atoms, a neutral, conjugated or non-conjugated diene of 10 or less carbon atoms, an anionic ligand and a neutral ligand capable of coordination with a lone pair, two or more of said L being the same or different.

[0053] Examples of the ligands having cyclopentadienyl skeleton include a cyclopentadienyl group, an alkyl substituted cyclopentadienyl group, an indenyl group, an alkyl substituted indenyl group, a 4,5,6,7-tetrahydroindenyl group, a fluorenyl group and an alkyl substituted fluorenyl group. These groups may be substituted with halogen atoms, trialkylsilyl groups or the like.

[0054] When the compound represented by the formula (1) contains two or more ligands having cyclopentadienyl skeleton, two ligands having cyclopentadienyl skeleton among them may be bonded to each other through an alkylene group, a substituted alkylene group, a silylene group, a substituted silylene group or the like. (Such a compound is sometimes referred to as a "bridged metallocene compound" hereinafter, and a compound other than such a compound is sometimes referred to as a "non-bridged metallocene compound" hereinafter.)

[0055] Of the above compounds, preferable is, for example, a compound represented by the following formula (2).

$$ML_aX_b \quad (2)$$

[0056] In the above formula, M is a transition metal atom selected from Group IV of the periodic table, preferably zirconium, titanium or hafnium of a Group IVB transition metal, L is a ligand coordinated to the transition metal, a is an integer of 1 or greater and is a number of L, X is a group or an atom bonded to the transition metal and selected from a halogen, a hydrogen atom, a hydrocarbon group of 1 to 10 carbon atoms, a neutral, conjugated or non-conjugated diene of 10 or less carbon atoms, an anionic ligand and a neutral ligand capable of coordination with a lone pair, each X being the same or different, and b is an integer of 1 or greater and is a number of X.

[0057] In the formula (2), L is a ligand coordinated to the transition metal, and at least one L is a ligand having cyclopentadienyl skeleton. Examples of the ligands having cyclopentadienyl skeleton include a cyclopentadienyl group; alkyl substituted cyclopentadienyl groups, such as methylcyclopentadienyl group, dimethylcyclopentadienyl group, trimethylcyclopentadienyl group, tetramethylcyclopentadienyl group, pentamethylcyclopentadienyl group, ethylcyclopentadienyl group, methylethylcyclopentadienyl group, propylcyclopentadienyl group, methylpropylcyclopentadienyl group, butylcyclopentadienyl group, methylbutylcyclopentadienyl group and hexylcyclopentadienyl group; an indenyl group; a 4,5,6,7-tetrahydroindenyl group; and a fluorenyl group. These groups may be substituted with halogen atoms, trialkylsilyl groups or the like.

[0058] In the formula (2), a is an integer of 1 or greater and is a number of L.

[0059] In the formula (2), M is zirconium, titanium or hafnium.

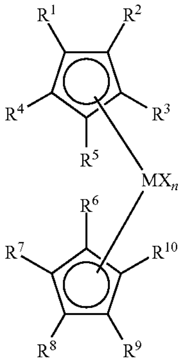
[0060] X is a group or an atom bonded to the transition metal and selected from a halogen, a hydrogen atom, a hydrocarbon group of 1 to 10 carbon atoms, a neutral, conjugated or non-conjugated diene of 10 or less carbon atoms, an anionic ligand and a neutral ligand capable of coordination with a lone pair, and each X is the same or different. Examples of the halogens are fluorine, chlorine, bromine and iodine. Examples of the hydrocarbon groups include methyl, ethyl, n-propyl, isopropyl, 2-methylpropyl, 1,1-dimethylpropyl, 2,2-dimethylpropyl, 1,1-diethylpropyl, 1-ethyl-1-methylpropyl, 1,1,2,2-tetramethylpropyl, sec-butyl, tert-butyl, 1,1-dimethylbutyl, 1,1,3-trimethylbutyl, neopentyl, cyclohexylmethyl, cyclohexyl and 1-methyl-1-cyclohexyl. Examples of the neutral, conjugated or non-conjugated dienes of 10 or less carbon atoms include s-cis- or s-trans-η^4-1,3-butadiene, s-cis- or s-trans-η^4-1,4-diphenyl-1,3-butadiene, s-cis- or s-trans-η^4-3-methyl-1,3-pentadiene, s-cis- or s-trans-η^4-1,4-dibenzyl-1,3-butadiene, s-cis- or s-trans-η^4-2,4-hexadiene, s-cis- or s-trans-η^4-1,3-pentadiene, s-cis- or s-trans-η^4-1,4-ditolyl-1,3-butadiene, and s-cis- or s-trans-η^4-1,4-bis(trimethylsilyl)-1,3-butadiene. Examples of the anionic ligands include alkoxy groups, such as methoxy, tert-butoxy and phenoxy; carboxylate groups, such as acetate and benzoate; and sulfonate groups, such as mesylate and tosylate. Examples of the neutral ligands capable of coordination with a lone pair include organic phosphorus compounds, such as trimethylphosphine, triethylphosphine, triphenylphosphine and diphenylmethylphosphine; and ethers, such as tetrahydrofuran, diethyl ether, dioxane and 1,2-dimethoxyethane. Each X may be the same or different.

[0061] In the formula (2), b is an integer of 1 or greater and is a number of X.

[0062] When the compound represented by the formula (2) contains two or more ligands having cyclopentadienyl skeleton, two ligands having cyclopentadienyl skeleton among them may be bonded to each other through an alkylene group, such as ethylene or propylene; a substituted alkylene group, such as diphenylmethylene; an alkylidene group, such as isopropylidene; a silylene group; or a substituted silylene group, such as dimethylsilylene group, diphenylsilylene group or methylphenylsilylene group. Two or more ligands having cyclopentadienyl skeleton may be the same or different.

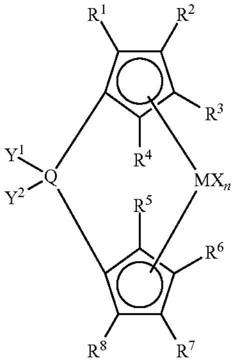
[0063] When the compound represented by the formula (2) contains two ligands having cyclopentadienyl group, this compound is more specifically represented by the following formula (3) or (4).

(3)



[0064] In the above formula, M is a transition metal atom selected from Group IV of the periodic table, preferably zirconium, titanium or hafnium of a Group IVB transition metal, R^1, R^2, R^3, R^4, R^5, R^6, R^7, R^8, R^9 and R^{10} are each selected from hydrogen, a hydrocarbon group and a silicon-containing hydrocarbon group and may be the same or different, neighboring substituents among R^1 to R^{10} may be bonded to each other to form a ring, X is a group or an atom selected from a halogen, a hydrogen atom, a hydrocarbon group of 1 to 10 carbon atoms, a neutral, conjugated or non-conjugated diene of 10 or less carbon atoms, an anionic ligand and a neutral ligand capable of coordination with a lone pair, each X being the same or different, and n is 1 or 2 and is a number of X.

(4)



[0065] In the above formula, M is a transition metal atom selected from Group IV of the periodic table, preferably zirconium, titanium or hafnium of a Group IVB transition metal, R^1, R^2, R^3, R^4, R^5, R^6, R^7 and R^8 are each selected from hydrogen, a hydrocarbon group and a silicon-containing hydrocarbon group and may be the same or different, neighboring substituents among R^1 to R^8 may be bonded to each other to form a ring, X is a group or an atom selected from a halogen, a hydrogen atom, a hydrocarbon group of 1 to 10 carbon atoms, a neutral, conjugated or non-conjugated diene of 10 or less carbon atoms, an anionic ligand and a neutral ligand capable of coordination with a lone pair, each X being the same or different, n is 1 or 2 and is a number of X, Q is selected from carbon, silicon and germanium, and Y^1 and Y^2 are each selected from hydrogen, a hydrocarbon group and a silicon-containing group, may be the same or different and may be bonded to each other to form a ring.

[0066] In the formulas (3) and (4), the hydrocarbon group is preferably an alkyl group of 1 to 20 carbon atoms, an arylalkyl group of 7 to 20 carbon atoms, an aryl group of 6 to 20 carbon atoms or an alkylaryl group of 7 to 20 carbon atoms, and may contain one or more ring structures. Examples of such groups include methyl, ethyl, n-propyl, isopropyl, 2-methylpropyl, 1,1-dimethylpropyl, 2,2-dimethylpropyl, 1,1-diethylpropyl, 1-ethyl-1-methylpropyl, 1,1,2,2-tetramethylpropyl, sec-butyl, tert-butyl, 1,1-dimethylbutyl, 1,1,3-trimethylbutyl, neopentyl, cyclohexylmethyl, cyclohexyl, 1-methyl-1-cyclohexyl, 1-adamantyl, 2-adamantyl, 2-methyl-2-adamantyl, menthyl, norbornyl, benzyl, 2-phenylethyl, 1-tetrahydronaphthyl, 1-methyl-1-tetrahydronaphthyl, phenyl, naphthyl and tolyl.

[0067] In the formulas (3) and (4), the silicon-containing hydrocarbon group is preferably an alkylsilyl or arylsilyl group having 1 to 4 silicon atoms and 3 to 20 carbon atoms. Examples of such groups include trimethylsilyl, tert-butyldimethylsilyl and triphenylsilyl.

[0068] In the present invention, R^1, R^2, R^3, R^4, R^5, R^6, R^7, R^8, R^9 and R^{10} in the formula (3) are each selected from hydrogen, a hydrocarbon group and a silicon-containing hydrocarbon group, and they may be the same or different. Preferred examples of the hydrocarbon groups and the silicon-containing hydrocarbon groups include those previously described. R^1, R^2, R^3, R^4, R^5, R^6, R^7 and R^8 in the formula (4) are each selected from hydrogen, a hydrocarbon group and a silicon-containing hydrocarbon group, and they may be the same or different. Preferred examples of the hydrocarbon groups and the silicon-containing hydrocarbon groups include those previously described.

[0069] Neighboring substituents among R^1 to R^{10} on the cyclopentadienyl rings in the formula (3) may be bonded to each other to form a ring. Examples of such substituted cyclopentadienyl groups include indenyl, 2-methylindenyl, tetrahydroindenyl, 2-methyltetrahydroindenyl, 2,4,4-trimethyltetrahydroindenyl, fluorenyl, benzofluorenyl, dibenzofluorenyl, octahydrodibenzofluorenyl and octamethyloctahydrodibenzofluorenyl. Neighboring substituents among R^1 to R^8 on the cyclopentadienyl rings in the formula (4) may be bonded to each other to form a ring. Examples of such substituted cyclopentadienyl groups include indenyl, 2-methylindenyl, tetrahydroindenyl, 2-methyltetrahydroindenyl, 2,4,4-trimethyltetrahydroindenyl, fluorenyl, benzofluorenyl, dibenzofluorenyl, octahydrodibenzofluorenyl and octamethyloctahydrodibenzofluorenyl.

[0070] In the present invention, M in the formulas (3) and (4) is a transition metal atom selected from Group IV of the periodic table, preferably zirconium, titanium or hafnium of a Group IVB transition metal.

[0071] X is a group or an atom selected from a halogen, a hydrogen atom, a hydrocarbon group of 1 to 10 carbon atoms, a neutral, conjugated or non-conjugated diene of 10 or less carbon atoms, an anionic ligand and a neutral ligand capable of coordination with a lone pair, and each X is the same or different. Examples of the halogens are fluorine, chlorine, bromine and iodine. Examples of the hydrocarbon groups include methyl, ethyl, n-propyl, isopropyl, 2-methylpropyl, 1,1-dimethylpropyl, 2,2-dimethylpropyl, 1,1-diethylpropyl, 1-ethyl-1-methylpropyl, 1,1,2,2-tetramethylpropyl, sec-butyl, tert-butyl, 1,1-dimethylbutyl, 1,1,3-trimethylbutyl, neopentyl, cyclohexylmethyl, cyclohexyl and 1-methyl-1-cyclohexyl. Examples of the neutral, conjugated or non-conjugated dienes of 10 or less carbon atoms include s-cis- or s-trans-η^4-1,3-butadiene, s-cis- or s-trans-η^4-1,4-diphenyl-1,3-butadiene, s-cis- or s-trans-η^4-3-methyl-1,3-pentadiene, s-cis- or s-trans-η^4-1,4-dibenzyl-1,3-butadiene, s-cis- or s-trans-η^4-2,4-hexadiene, s-cis- or s-trans-η^4-1,3-pentadiene, s-cis- or s-trans-η^4-1,4-ditolyl-1,3-butadiene, and s-cis- or s-trans-η^4-1,4-bis(trimethylsilyl)-1,3-butadiene. Examples of the anionic ligands include alkoxy groups, such as methoxy, tert-butoxy and phenoxy; carboxylate groups, such as acetate and benzoate; and sulfonate groups, such as mesylate and tosylate. Examples of the neutral ligands capable of coordination with a lone pair include organic phosphorus compounds, such as trimethylphosphine, triethylphosphine, triphenylphosphine and diphenylmethylphosphine; and ethers, such as tetrahydrofuran, diethyl ether, dioxane and 1,2-dimethoxyethane. Each X may be the same or different.

[0072] n is 1 or 2 and is a number of X.

[0073] Examples of the compounds represented by the formula (3) include cyclopentadienyl trichloride, cyclopentadienylzirconium trichloride, bis(cyclopentadienyl)titanium dichloride, bis(cyclopentadienyl)zirconium dichloride, bis(pentamethylcyclopentadienyl)zirconium dichloride, (cyclopentadienyl)(pentamethylcyclopentadienyl)zirconium dichloride, bis(methylcyclopentadienyl)zirconium dichloride, bis(ethylcyclopentadienyl)zirconium dichloride, bis(propylcyclopentadienyl)zirconium dichloride, bis(butylcyclopentadienyl)zirconium dichloride such as bis(n-butylcyclopentadienyl)zirconium dichloride, bis(dimethlcyclopentadienyl)zirconium dichloride such as bis(1,3-dimethlcyclopentadienyl)zirconium dichloride, bis(diethylcyclopentadienyl)zirconium dichloride such as bis(1,3-diethylcyclopentadienyl)zirconium dichloride, bis(methylethylcyclopentadienyl)zirconium dichloride, bis(methylpropylcyclopentadienyl)zirconium dichloride such as bis(1-methyl-3-n-propylcyclopentadienyl)zirconium dichloride, and bis(methylbutylcyclopentadienyl)zirconium dichloride such as bis(1-methyl-3-n-butylcyclopentadienyl)zirconium dichloride. Examples of the compounds represented by the formula (4) include

[0074] ethylenebis(indenyl)zirconium dichloride,

[0075] ethylenebis(1-indenyl)titanium dichloride,

[0076] ethylenebis(4,5,6,7-tetrahydroindenyl)zirconium dichloride,

[0077] dimethylsilylenebis(methylcyclopentadienyl)zirconium dichloride,

[0078] dimethylsilylenebis(dimethylcyclopentadienyl)zirconium dichloride,

[0079] dimethylsilylenebis(di-tert-butylcyclopentadienyl)zirconium dichloride,

[0080] dimethylsilylenebis(indenyl)zirconium dichloride,

[0081] dimethylsilylenebis(2-methylindenyl)zirconium dichloride,

[0082] dimethylsilylenebis(2-methyl-4,5-benzindenyl)zirconium dichloride,

[0083] dimethylsilylenebis(2-methyl-4-phenylindenyl)zirconium dichloride,

[0084] dimethylsilylenebis(2-methyl-4-naphthylindenyl)zirconium dichloride,

[0085] dimethylethylenebis(2-methylindenyl)zirconium dichloride,

[0086] dimethylethylenebis(2-methyl-4,5-benzindenyl)zirconium dichloride,

[0087] dimethylethylenebis(2-methyl-4-phenylindenyl)zirconium dichloride,

[0088] dimethylethylenebis(2-methyl-4-naphthylindenyl)zirconium dichloride,

[0089] isopropylidene(cyclopentadienyl)(fluorenyl)zirconium dichloride,

[0090] isopropylidene(cyclopentadienyl)(3,6-di-t-butylfluorenyl)zirconium dichloride,

[0091] isopropylidene(methylcyclopentadienyl)(fluorenyl)zirconium dichloride,

[0092] isopropylidene(tert-butylcyclopentadienyl)(fluorenyl)zirconium dichloride,

[0093] isopropylidene(methyl-tert-butylcyclopentadienyl)(fluorenyl)zirconium dichloride,

[0094] isopropylidene(methyl-tert-butylcyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0095] isopropylidene(methyl-tert-butylcyclopentadienyl)(octamethyloctahydridodibenzofluorenyl)zirconium dichloride,

[0096] diphenylmethylene(cyclopentadienyl)(fluorenyl)zirconium dichloride,

[0097] diphenylmethylene(cyclopentadienyl)(3,6-di-t-butylfluorenyl)zirconium dichloride,

[0098] diphenylmethylene(methylcyclopentadienyl)(fluorenyl)zirconium dichloride,

[0099] diphenylmethylene(tert-butylcyclopentadienyl)(fluorenyl)zirconium dichloride,

[0100] diphenylmethylene(methyl-tert-butylcyclopentadienyl)(fluorenyl)zirconium dichloride,

[0101] diphenylmethylene(methyl-tert-butylcyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0102] diphenylmethylene(methyl-tert-butylcyclopentadienyl)(octamethyloctahydridodibenzofluorenyl)zirconium dichloride,

[0103] cyclohexylene(cyclopentadienyl)(fluorenyl)zirconium dichloride,

[0104] cyclohexylene(cyclopentadienyl)(3,6-di-t-butylfluorenyl)zirconium dichloride,

[0105] cyclohexylene(methylcyclopentadienyl)(fluorenyl)zirconium dichloride,

[0106] cyclohexylene(tert-butylcyclopentadienyl)(fluorenyl)zirconium dichloride,

[0107] cyclohexylene(methyl-tert-butylcyclopentadienyl)(fluorenyl)zirconium dichloride,

[0108] cyclohexylene(methyl-tert-butylcyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0109] cyclohexylene(methyl-tert-butylcyclopentadienyl)(octamethyloctahydridodibenzofluorenyl)zirconium dichloride,

[0110] di(p-tolyl)methylene(cyclopentadienyl)(fluorenyl)zirconium dichloride,

[0111] di(p-tolyl)methylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0112] di(p-tolyl)methylene(cyclopentadienyl)(2,7-dimethylfluorenyl)zirconium dichloride,

[0113] di(p-tolyl)methylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0114] di(p-tert-butylphenyl)methylene(cyclopentadienyl)(fluorenyl)zirconium dichloride,

[0115] di(p-tert-butylphenyl)methylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0116] di(p-tert-butylphenyl)methylene(cyclopentadienyl)(2,7-dimethylfluorenyl)zirconium dichloride,

[0117] di(p-tert-butylphenyl)methylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0118] di(p-n-butylphenyl)methylene(cyclopentadienyl)(fluorenyl)zirconium dichloride,

[0119] di(p-n-butylphenyl)methylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0120] di(p-n-butylphenyl)methylene(cyclopentadienyl)(2,7-dimethylfluorenyl)zirconium dichloride,

[0121] di(p-n-butylphenyl)methylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0122] di(m-tolyl)methylene(cyclopentadienyl)(fluorenyl)zirconium dichloride,

[0123] di(m-tolyl)methylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0124] di(m-tolyl)methylene(cyclopentadienyl)(2,7-dimethylfluorenyl)zirconium dichloride,

[0125] di(m-tolyl)methylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0126] di(p-tolyl)methylene(cyclopentadienyl)(fluorenyl)zirconium dimethyl,

[0127] di(p-tolyl)methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0128] di(p-tert-butylphenyl)methylene(cyclopentadienyl)(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0129] di(p-tolyl)methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dimethyl,

[0130] di(p-tert-butylphenyl)methylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0131] di(p-tolyl)methylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dimethyl,

[0132] di(p-isopropylphenyl)methylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0133] di(p-tert-butylpheyl)methylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0134] di(4-biphenyl)methylene(cyclopentadienyl)(fluorenyl)zirconium dichloride,

[0135] di(4-biphenyl)methylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0136] di(4-biphenyl)methylene(cyclopentadienyl)(2,7-dimethylfluorenyl)zirconium dichloride,

[0137] di(4-biphenyl)methylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0138] cyclopentylidene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0139] cyclohexylidene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0140] adamantylidene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0141] monophenylmonomethylmethylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0142] dimethylmethylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0143] diphenylmethylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0144] di(p-tolyl)methylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0145] diethylmethylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0146] cyclopentylidene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0147] cyclohexylidene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0148] adamantylidene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0149] monophenylmonomethylmethylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0150] dimethylmethylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

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[0152] di(p-tolyl)methylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0153] diethylmethylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0154] cyclopentylidene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0155] cyclohexylidene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0156] adamantylidene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0157] monophenylmonomethylmethylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0158] dimethylmethylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0159] diphenylmethylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0160] di(p-tolyl)methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0161] diethylmethylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0162] cyclopentylidene(cyclopentadienyl)(octamethyltetrahydrodicyclopentafluorenyl)zirconium dichloride,

[0163] cyclohexylidene(cyclopentadienyl)(octamethyltetrahydrodicyclopentafluorenyl)zirconium dichloride,

[0164] adamantylidene(cyclopentadienyl)(octamethyltetrahydrodicyclopentafluorenyl)zirconium dichloride,

[0165] monophenylmonomethylmethylene(cyclopentadienyl)(octamethyltetrahydrodicyclopentafluorenyl)zirconium dichloride,

[0166] dimethylmethylene(cyclopentadienyl)(octamethyltetrahydrodicyclopentafluorenyl)zirconium dichloride,

[0167] diphenylmethylene(cyclopentadienyl)(octamethyltetrahydrodicyclopentafluorenyl)zirconium dichloride,

[0168] di(p-tolyl)methylene(cyclopentadienyl)(octamethyltetrahydrodicyclopentafluorenyl)zirconium dichloride,

[0169] diethylmethylene(cyclopentadienyl)(octamethyltetrahydrodicyclopentafluorenyl)zirconium dichloride,

[0170] cyclopentylidene(cyclopentadienyl)(dibenzofluorenyl)zirconium dichloride,

[0171] cyclohexylidene(cyclopentadienyl)(dibenzofluorenyl)zirconium dichloride,

[0172] adamantylidene(cyclopentadienyl)(dibenzofluorenyl)zirconium dichloride,

[0173] monophenylmonomethylmethylene(cyclopentadienyl)(dibenzofluorenyl)zirconium dichloride,

[0174] dimethylmethylene(cyclopentadienyl)(dibenzofluorenyl)zirconium dichloride,

[0175] diphenylmethylene(cyclopentadienyl)(dibenzofluorenyl)zirconium dichloride,

[0176] di(p-tolyl)methylene(cyclopentadienyl)(dibenzofluorenyl)zirconium dichloride,

[0177] diethylmethylene(cyclopentadienyl)(dibenzofluorenyl)zirconium dichloride,

[0178] ethylene(cyclopentadienyl)(fluorenyl)zirconium dichloride,

[0179] ethylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0180] ethylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0181] ethylene(cyclopentadienyl)(octamethyltetrahydrodicyclopentafluorenyl)zirconium dichloride,

[0182] ethylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0183] propylene(cyclopentadienyl)(fluorenyl)zirconium dichloride,

[0184] propylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0185] propylene(cyclopentadienyl) (3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0186] propylene(cyclopentadienyl)(octamethyltetrahydrodicyclopentafluorenyl)zirconium dichloride,

[0187] propylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0188] (methyl)(p-tolyl)methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0189] (methyl)(phenyl)methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride,

[0190] (methyl)(p-tolyl)methylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0191] (methyl)(phenyl)methylene (cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0192] (methyl)(p-tolyl)methylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0193] (methyl)(phenyl)methylene (cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0194] (methyl)(benzyl)methylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0195] (methyl)(benzyl)methylene(cyclopentadienyl)(3,6-di-tert-butylfluorenyl)zirconium dichloride,

[0196] (methyl)(benzyl)methylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

[0197] (methyl)(benzyl)methylene(cyclopentadienyl)(2,7-di-tert-butylfluorenyl)zirconium dichloride,

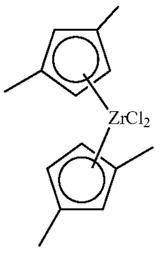
[0198] (methyl)(benzyl)methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride, and

[0199] (methyl)(benzyl)methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride.

[0200] Further, not only titanium compounds and hafnium compounds having similar stereostructure, bromides and iodides but also such transition metal compounds as described in, for example, Japanese Patent Laid-Open Publication No. 9913/1991, Japanese Patent Laid-Open Publication No. 131488/1990, Japanese Patent Laid-Open Publication No. 21607/1991, Japanese Patent Laid-Open Publication No. 106907/1991, Japanese Patent Laid-Open Publication No. 188092/1991, Japanese Patent Laid-Open Publication No. 69394/1992, Japanese Patent Laid-Open Publication No. 300887/1992 and WO 01/27124A1 can be also mentioned.

[0201] The transition metal compound (A) used in the polymerization examples of the present invention and the comparative polymerization examples is specifically a compound of the following formula (6), but in the present invention, the transition metal compound (A) is not limited to this compound. A transition metal compound (A) represented by the following formula (5), (7), (8) or (9) may be used.

(5)



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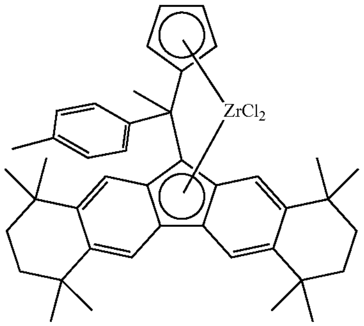

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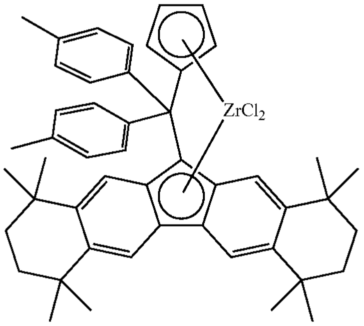

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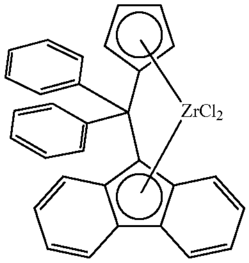

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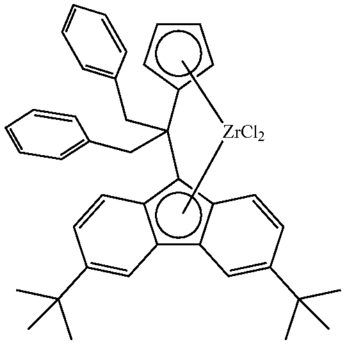


[0202] In the polymerization examples of the present invention, the compound (6) is described as

[0203] (methyl)(p-tolyl)methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride.

[0204] The structure of the transition metal compound obtained is determined by 270 MHz ^{1}H-NMR (GSH-270 of JEOL Ltd.) and FD-mass spectrometry (SX-102A of JEOL Ltd.).

[0205] The above transition metal compounds (A) may be used singly or may be used in combination of two or more kinds.

(B-1) Organometallic Compound

[0206] As the organometallic compound (B-1), an organometallic compound represented by the following formula (10) is specifically used.

[0207] A dialkyl compound of a periodic table Group 2 or Group 12 metal, which is represented by the following formula:

$$R^aR^bM^3 \tag{10}$$

[0208] wherein R^a and R^b may be the same as or different from each other and are each a hydrocarbon group of 1 to 15 carbon atoms, preferably 1 to 4 carbon atoms, and M^3 is Mg, Zn or Cd.

[0209] Such organometallic compounds (B-1) may be used singly or in combination of two or more kinds.

(B-2) Organoaluminum Compound

[0210] The organoaluminum compound (B-2) for forming the olefin polymerization catalyst is, for example, an organoaluminum compound represented by the following formula (11), or an alkyl complex compound of a Group 1 metal and aluminum, which is represented by the following formula (12).

[0211] An organoaluminum compound represented by the formula (11):

$$R^a_mAl(OR^b)_nH_pX^2_q \tag{11}$$

wherein R^a and R^b may be the same as or different from each other and are each a hydrocarbon group of 1 to 15 carbon atoms, preferably 1 to 4 carbon atoms, X^2 is a halogen atom, and m, n, p and q are numbers satisfying the conditions of $0<m\leqq 3$, $0\leqq n<3$, $0\leqq p<3$, $0\leqq q<3$ and $m+n+p+q=3$.

[0212] Examples of such compounds include trimethylaluminum, triethylaluminum, triisobutylaluminum and diisobutylaluminum hydride.

[0213] An alkyl complex compound of a periodic table Group 1 metal and aluminum, which is represented by the formula (12):

$$M^2AlR^a_4 \tag{12}$$

[0214] wherein M^2 is Li, Na or K, and R^a is a hydrocarbon group of 1 to 15 carbon atoms, preferably 1 to 4 carbon atoms.

[0215] Examples of such compounds include $LiAl\,(C_2H_5)_4$ and $LiAl\,(C_7H_{15})_4$.

[0216] The organoaluminum compound represented by the formula (11) is, for example, a compound represented by the following formula (13), (14), (15) or (16).

$$R^a_mAl(OR^b)_{3-m} \tag{13}$$

wherein R^a and R^b may be the same as or different from each other and are each a hydrocarbon group of 1 to 15 carbon atoms, preferably 1 to 4 carbon atoms, and m is preferably a number of $1.5\leqq m\leqq 3$.

$$R^a_mAlX^2_{3-m} \tag{14}$$

wherein R^a is a hydrocarbon group of 1 to 15 carbon atoms, preferably 1 to 4 carbon atoms, X^2 is a halogen atom, and m is preferably a number of $0<m<3$.

$$R^a_mAlH_{3-m} \tag{15}$$

wherein R^a is a hydrocarbon group of 1 to 15 carbon atoms, preferably 1 to 4 carbon atoms, and m is preferably a number of $2\leqq m<3$.

$$R^a_mAl(OR^b)_nX^2_q \tag{16}$$

wherein R^a and R^b may be the same as or different from each other and are each a hydrocarbon group of 1 to 15 carbon atoms, preferably 1 to 4 carbon atoms, X^2 is a halogen atom, and m, n and q are numbers satisfying the conditions of $0<m\leqq 3$, $0n\leqq n<3$, $0\leqq q<3$ and $m+n+q=3$.

[0217] Examples of the aluminum compounds represented by the formula (13), (14), (15) or (16) include tri-n-alkylaluminums, such as trimethylaluminum, triethylaluminum, tri-n-butylaluminum, tripropylaluminum, tripentylaluminum, trihexylaluminum, trioctylaluminum and tridecylaluminum; branched chain trialkylaluminums, such as triisopropylaluminum, triisobutylaluminum, tri-sec-butylaluminum, tri-tert-butylaluminum, tri-2-methylbutylaluminum, tri-3-methylbutylaluminum, tri-2-methylpentylaluminum, tri-3-methylpentylaluminum, tri-4-methylpentylaluminum, tri-2-methylhexylaluminum, tri-3-methylhexylaluminum and tri-2-ethylhexylaluminum; tricycloalkylaluminums, such as tricyclohexylaluminum and tricyclooctylaluminum; triarylaluminums, such as triphenylaluminum and tritolylaluminum; dialkylaluminum hydrides, such as diisopropylaluminum hydride and diisobutylaluminum hydride; alkenylaluminums represented by the formula $(i\text{-}C_4H_9)_xAl_y(C_5H_{10})_z$ (x, y and z are each a positive number, and $z\leqq 2x$), such as isoprenylaluminum; alkylaluminum alkoxides, such as isobutylaluminum methoxide, isobutylaluminum ethoxide and isobutylaluminum isopropoxide; dialkylaluminum alkoxides, such as dimethylaluminum methoxide, diethylaluminum ethoxide and dibutylaluminum butoxide; alkylaluminum sesquialkoxides, such as ethylaluminum sesquiethoxide and butylaluminum sesquibutoxide; partially alkoxylated alkylaluminums having average composition represented by the formula $R^a_{2.5}Al(OR^b)_{0.5}$; alkylaluminum aryloxides, such as diethylaluminum phenoxide, diethylaluminum(2,6-di-t-butyl-4-methylphenoxide), ethylaluminumbis(2,6-di-t-butyl-4-methylphenoxide), diisobutylaluminum(2,6-di-t-butyl-4-methylphenoxide) and isobutylaluminumbis(2,6-di-t-butyl-4-methylphenoxide); dialkylaluminum halides, such as dimethylaluminum chloride, diethylaluminum chloride, dibutylaluminum chloride, diethylaluminum bromide and diisobutylaluminum chloride; alkylaluminum sesquihalides, such as ethylaluminum sesquichloride, butylaluminum sesquichloride and ethylaluminum sesequibromide; partially halogenated alkylaluminums, such as alkylaluminum dihalides, specifically ethylaluminum dichloride, propylaluminum dichloride and butylaluminum dibromide; dialkylaluminum hydrides, such as diethylaluminum hydride and dibutylaluminum hydride; partially hydrogenated alkylaluminums, such as alkylaluminum dihydrides, specifically ethylaluminum dihydride and propylaluminum dihydride; and partially alkoxylated and halogenated alkylaluminums, such as ethylaluminum ethoxychloride, butylaluminum butoxychloride and ethylaluminum ethoxybromide.

[0218] Further, compounds analogous to the compound represented by the formula (11) are also employable, and for example, an organoaluminum compound wherein two or more aluminum compounds are bonded through a nitrogen atom is employable. An example of such a compound is $(C_2H_5)_2AlN(C_2H_5)Al(C_2H_5)_2$.

[0219] Examples of the compounds represented by the formula (12) include $LiAl(C_2H_5)_4$ and $LiAl(C_7H_{15})_4$.

[0220] Further, compounds by the use of which the above-mentioned organoaluminum compounds are formed in the polymerization system are also employable, and for example, a combination of aluminum halide and alkyllithium or a combination of aluminum halide and alkylmagnesium is also employable.

[0221] Of the above compounds, organoaluminum compounds are preferable.

[0222] The organoaluminum compounds represented by the formula (11) or the alkyl complex compounds of Group 1 metals and aluminum which are represented by the formula (12) are used singly or in combination of two or more kinds.

(B-3) Organoaluminum Oxy-Compound

[0223] The organoaluminum oxy-compound (B-3) may be hitherto publicly known aluminoxane, or may be such a benzene-insoluble organoaluminum oxy-compound as given as an example in Japanese Patent Laid-Open Publication No. 78687/1990.

[0224] The hitherto publicly known aluminoxane can be prepared by following processes, and is usually obtained as a solution of a hydrocarbon solvent.

[0225] (I) A process comprising adding an organoaluminum compound, such as trialkylaluminum, to a hydrocarbon medium suspension of a compound containing adsorbed water or a salt containing water of crystallization, such as magnesium chloride hydrate, copper sulfate hydrate, aluminum sulfate hydrate, nickel sulfate hydrate or cerous chloride hydrate, to allow adsorbed water or water of crystallization to react with the organoaluminum compound.

[0226] (II) A process comprising allowing water, ice or water vapor to directly act on an organoaluminum compound, such as trialkylaluminum, in a medium, such as benzene, toluene, ethyl ether or tetrahydrofuran.

[0227] (III) A process comprising allowing an organotin oxide, such as dimethyltin oxide or dibutyltin oxide, to react with an organoaluminum compound, such as trialkylaluminum, in a medium, such as decane, benzene or toluene.

[0228] The aluminoxane may contain a small amount of an organometallic component. After a solvent or an unreacted organoaluminum compound is removed from the recovered solution of aluminoxane by distillation, the residue may be redissolved in a solvent or suspended in a poor solvent for aluminoxane.

[0229] Examples of the organoaluminum compounds used for preparing aluminoxane include the same organoaluminum compounds as those given as examples of the aforesaid organoaluminum compounds (B-2).

[0230] Of those compounds, trialkylaluminums and tricycloalkylaluminums are preferable, and trimethylaluminum is particularly preferable.

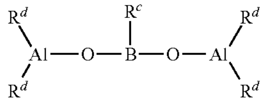
[0231] Such organoaluminum compounds are used singly or in combination of two or more kinds.

[0232] The benzene-insoluble organoaluminum oxy-compound is preferably a compound usually having a content of an Al component soluble in benzene at 60° C., of not more than 10%, preferably not more than 5%, particularly preferably not more than 2%, in terms of Al atom, that is, a compound which is insoluble or slightly soluble in benzene.

[0233] Aluminoxane prepared from trimethylaluminum is called methylaluminoxane or MAO, and is a compound particularly frequently used.

[0234] Examples of the solvents used for preparing aluminoxane include hydrocarbon solvents, e.g., aromatic hydrocarbons, such as benzene, toluene, xylene, cumene and cymene; aliphatic hydrocarbons, such as pentane, hexane, heptane, octane, decane, dodecane, hexadecane and octadecane; alicyclic hydrocarbons, such as cyclopentane, cyclohexane, cyclooctane and methylcyclopentane; petroleum fractions, such as gasoline, kerosine and gas oil; and halogenation products of the above aromatic hydrocarbons, aliphatic hydrocarbons and alicyclic hydrocarbons, particularly chlorination products and bromination products thereof. Further, ethers, such as ethyl ether and tetrahydrofuran, are also employable. Of these solvents, aromatic hydrocarbons or aliphatic hydrocarbons are particularly preferable.

[0235] As the organoaluminum oxy-compound, an organoaluminum oxy-compound containing boron, which is represented by the following formula (17), can be also mentioned.



(17)

[0236] In the above formula, R^c is a hydrocarbon group of 1 to 10 carbon atoms, and each R^d may be the same or different and is a hydrogen atom, a halogen atom or a hydrocarbon group of 1 to 10 carbon atoms.

[0237] The organoaluminum oxy-compound containing boron, which is represented by the formula (17), can be prepared by allowing an alkylboronic acid represented by the following formula (18) to react with an organoaluminum compound in an inert solvent at a temperature of −80° C. to room temperature for a period of 1 minute to 24 hours in an inert gas atmosphere.

$$R^cB(OH)_2 \qquad (18)$$

[0238] In the above formula, R^c is the same group as described above.

[0239] Examples of the alkylboronic acids represented by the formula (18) include methylboronic acid, ethylboronic acid, isopropylboronic acid, n-propylboronic acid, n-butylboronic acid, isobutylboronic acid, n-hexylboronic acid, cyclohexylboronic acid, phenylboronic acid, 3,5-difluorophenylboronic acid, pentafluorophenylboronic acid and 3,5-bis(trifluoromethyl)phenylboronic acid. Of these, preferable are methylboronic acid, n-butylboronic acid, isobutylboronic acid, 3,5-difluorophenylboronic acid and pentafluorophenylboronic acid. These alkylboronic acids are used singly or in combination of two or more kinds.

[0240] Examples of the organoaluminum compounds to be reacted with the alkylboronic acid include the same organoaluminum compounds as those given as examples of the organoaluminum compounds represented by the formula (11) or (12).

[0241] Of those compounds, preferable are trialkylaluminums and tricycloalkylaluminums, and particularly preferable are trimethylaluminum, triethylaluminum and triisobutylaluminum. These organoaluminum compounds are used singly or in combination of two or more kinds.

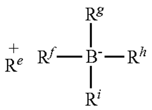
[0242] The above organoaluminum oxy-compounds (B-3) are used singly or in combination of two or more kinds.

(B-4) Compound which Reacts with the Group IV Transition Metal Compound (A) to Form Ion Pair

[0243] Examples of the compounds (B-4) which react with the Group IV transition metal compound (A) to form an ion pair include Lewis acid, an ionic compound, a borane compound and a carborane compound described in Japanese Patent Laid-Open Publication No. 501950/1989, Japanese Patent Laid-Open Publication No. 502036/1989, Japanese Patent Laid-Open Publication No. 179005/1991, Japanese Patent Laid-Open Publication No. 179006/1991, Japanese Patent Laid-Open Publication No. 207703/1991, Japanese Patent Laid-Open Publication No. 207704/1991 and U.S. Pat. No. 5,321,106.

[0244] The Lewis acid is specifically a compound represented by BR_3 (R is a phenyl group which may have a subsitituent, such as fluorine, a methyl group or a trifluoromethyl group, or fluorine), and examples thereof include trifluoroboron, triphenylboron, tris(4-fluorophenyl)boron, tris(3,5-difluorophenyl)boron, tris(4-fluoromethylphenyl)boron, tris(pentafluorophenyl)boron, tris(p-tolyl)boron, tris(o-tolyl)boron, tris(3,5-dimethylphenyl)boron, trimethylboron and triisobutylboron.

[0245] The ionic compound is, for example, a compound represented by the following formula (19).



$$\overset{+}{R^e}\quad R^f\text{—}\underset{\substack{|\\ R^i}}{\overset{\substack{R^g\\ |}}{B^-}}\text{—}R^h \qquad (19)$$

[0246] In the above formula, R^{e+} is H^+, carbenium cation, oxonium cation, ammonium cation, phosphonium cation, cycloheptyltrienyl cation, ferrocenium cation having a transition metal, or the like. R^f to R^i may be the same as or different from one another and are each an organic group, preferably an aryl group or a substituted aryl group.

[0247] Examples of the carbenium cations include tri-substituted carbenium cations, such as triphenylcarbenium cation, tris(methylphenyl)carbenium cation and tris(dimethylphenyl)carbenium cation.

[0248] Examples of the ammonium cations include trialkylammonium cations, such as trimethylammonium cation, triethylammonium cation, tri(n-propyl)ammonium cation, triisopropylammonium cation, tri(n-butyl)ammonium cation and triisobutylammonium cation; N,N-dialkylanilinium cations, such as N,N-dimethylanilinium cation, N,N-diethylanilinium cation and N,N-2,4,6-pentamethylanilinium cation; and dialkylammonium cations, such as diisopropylammonium cation and dicyclohexylammonium cation.

[0249] Examples of the phosphonium cations include triarylphosphonium cations, such as triphenylphosphonium cation, tris(methylphenyl)phosphonium cation and tris(dimethylphenyl)phosphonium cation.

[0250] Of the above cations, preferable as R^e is carbenium cation, ammonium cation or the like, and particularly preferable is triphenylcarbenium cation, N,N-dimethylanilinium cation or N,N-diethylanilinium cation.

[0251] Examples of carbenium salts include triphenylcarbenium tetraphenylborate, triphenylcarbenium tetrakis(pentafluorophenyl)borate, triphenylcarbenium tetrakis(3,5-ditrifluoromethylphenyl)borate, tris(4-methylphenyl)carbenium tetrakis(pentafluorophenyl)borate and tris(3,5-dimethylphenyl)carbenium tetrakis(pentafluorophenyl)borate.

[0252] Examples of ammonium salts include trialkyl substituted ammonium salts, N,N-dialkylanilinium salts and dialkylammonium salts.

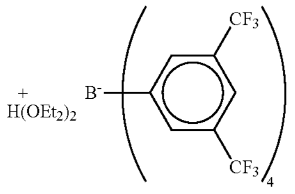
[0253] Examples of the trialkyl substituted ammonium salts include triethylammonium tetraphenylborate, tripropylammonium tetraphenylborate, tri(n-butyl)ammonium tetraphenylborate, trimethylammonium tetrakis(p-tolyl)borate, trimethylammonium tetrakis(o-tolyl)borate, tri(n-butyl)ammonium tetrakis(pentafluorophenyl)borate, triethylammonium tetrakis(pentafluorophenyl)borate, tripropylammonium tetrakis(pentafluorophenyl)borate, tripropylammonium tetrakis(2,4-dimethylphenyl)borate, tri(n-butyl)ammonium tetrakis(3,5-dimethylphenyl)borate, tri(n-butyl)ammonium tetrakis(4-trifluoromethylphenyl)borate, tri(n-butyl)ammonium tetrakis(3,5-ditrifluoromethylphenyl)borate, tri(n-butyl)ammonium tetrakis(o-tolyl)borate, dioctadecylmethylammonium tetraphenylborate, dioctadecylmethylammonium tetrakis(p-tolyl)borate, dioctadecylmethylammonium tetrakis(o-tolyl)borate, dioctadecylmethylammonium tetrakis(pentafluorophenyl)borate, dioctadecylmethylammonium tetrakis(2,4-dimethylphenyl)borate, dioctadecylmethylammonium tetrakis(3,5-dimethylphenyl)borate, dioctadecylmethylammonium tetrakis(4-trifluoromethylphenyl)borate, dioctadecylmethylammonium tetrakis(3,5-ditrifluoromethylphenyl)borate, and dioctadecylmethylammonium.

[0254] Examples of the N,N-dialkylanilinium salts include N,N-dimethylanilinium tetraphenylborate, N,N-dimethylanilinium tetrakis(pentafluorophenyl)borate, N,N-dimethylanilinium tetrakis(3,5-ditrifluoromethylphenyl)borate, N,N-diethylanilinium tetraphenylborate, N,N-diethylanilinium tetrakis(pentafluorophenyl)borate, N,N-diethylanilinium tetrakis(3,5-ditrifluoromethylphenyl)borate, N,N-2,4,6-pentamethylanilinium tetraphenylborate, and N,N-2,4,6-pentamethylanilinium tetrakis(pentafluorophenyl)borate.

[0255] Examples of the dialkylammonium salts include di(1-propyl)ammonium tetrakis(pentafluorophenyl)borate and dicyclohexylammonium tetraphenylborate.

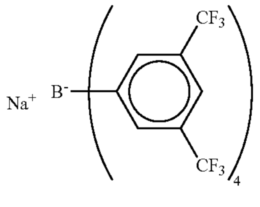
[0256] Further, ferrocenium tetrakis(pentafluorophenyl)borate, a triphenylcarbenium pentaphenylcyclopentadienyl complex, a N,N-diethylanilinium pentaphenylcyclopentadienyl complex, a borate compound represented by the following formula (20) or (21), a borate compound containing active hydrogen, which is represented by the following formula (22), a borate compound containing a silyl group, which is represented by the following formula (23), etc. can be also mentioned.

(20)



[0257] In the above formula, Et is an ethyl group.

(21)



$$[B\text{-}Qn(Gq(T\text{-}H)r)z]^{-}A^{+} \qquad (22)$$

[0258] In the formula (22), B is boron. G is a poly-linking hydrocarbon radical, and preferred poly-linking hydrocarbon is an alkylene, allylene, ethylene or alkalylene radical containing 1 to 20 carbon atoms. Preferred examples of G include phenylene, bisphenylene, naphthalene, methylene, ethylene, propylene, 1,4-butadiene and p-phenylenemethylene. The number of linkages of the poly-linking radical G is r+1, that is, one linkage is a linkage to borate anion, and other linkages r of G are linkages to (T-H) groups. A^+ is a cation.

[0259] T in the formula (22) is O, S, NR^j or PR^j, and R^j is a hydrocarbanyl radical, a trihydrocarbanylsilyl radical, a trihydrocarbanylgermanium radical or a hydride. q is an integer of 1 or greater, preferably 1. The T-H group is —OH, —SH, —NRH or —PR^{j}H, and R^j is a hydrocarbinyl radical of 1 to 18 carbon atoms, preferably 1 to 10 carbon atoms, or hydrogen. A preferred R^j group is alkyl, cycloalkyl, allyl, allylalkyl or alkylallyl having 1 to 18 carbon atoms. —OH, —SH, —NR^{j}H or —PR^{j}H may be, for example, any of —C(O)—OH, —C(S)—SH—C(O)—NR^{j}H and C(O)—PR^{j}H. A most preferred group having active hydrogen is —OH group. Q is hydride or dihydrocarbylamide, preferably dialkylamide, halide, hydrocarbyloxide, alkoxide, allyloxide, hydrocarbyl, substituted hydrocarbyl radical, or the like. n+z is 4.

[0260] [B-Qn(Gq(T-H)r)z] in the formula (22) is, for example, triphenyl(hydroxyphenyl)borate, diphenyl-di(hydroxyphenyl)borate, triphenyl(2,4-dihydroxyphenyl)borate, tri(p-tolyl)(hydroxyphenyl)borate, tris(pentafluorophenyl)(hydroxyphenyl)borate, tris(2,4-dimethylphenyl)(hydroxyphenyl)borate, tris(3,5-dimethylphenyl)(hydroxyphenyl)borate, tris[3,5-di(trifluoromethyl)phenyl](hydroxyphenyl)borate, tris(pentafluorophenyl)(2-hydroxyethyl)borate, tris(pentafluorophenyl)(4-hydroxylbutyl)borate, tris(pentafluorophenyl)(4-hydroxylcyclohexyl)borate, tris(pentafluorophenyl)[4-(4-hydroxyphenyl)phenyl]borate, or tris(pentafluorophenyl)(6-hydroxy-2-naphthyl)borate. Most preferred is tris(pentafluorophenyl)(4-hydroxyphenyl)borate. Compounds wherein —OH group in the above borate compounds is replaced with —NHR^j (R^j is methyl, ethyl or t-butyl) are also preferable.

[0261] A^+ that is a counter cation of the borate compound is carbonium cation, tropylium cation, ammonium cation, oxonium cation, sulfonium cation, phosphonium cation or the like. A cation of a metal or a cation of an organic metal which itself is apt to be reduced can be also mentioned. Examples of such cations include triphenylcarbonium ion, diphenylcarbonium ion, cycloheptatrinium, indenium, triethylammonium, tripropylammonium, tributylammonium, dimethylammonium, dipropylammonium, dicyclohexylammonium, trioctylammonium, N,N-dimethylammonium, diethylammonium, 2,4,6-pentamethylammonium, N,N-dimethlphenylammonium, di(i-propyl)ammonium, dicyclohexylammonium, triphenylphosphonium, triphosphonium, tridimethylphenylphosphonium, tri(methylphenyl)phosphonium, triphenylphosphonium ion, triphenyloxonium ion, triethyloxonium ion, pyrinium, silver ion, gold ion, platinum ion, copper ion, palladium ion, mercury ion and ferrocenium ion. Of these, ammonium ion is particularly preferable.

$$[\text{B-Q}n(\text{G}q(\text{SiR}^k\text{R}^l\text{R}^m)r)z]^-\text{A}^+ \qquad (23)$$

[0262] In the formula (23), B is boron. G is a poly-linking hydrocarbon radical, and preferred poly-linking hydrocarbon is an alkylene, allylene, ethylene or alkalylene radical containing 1 to 20 carbon atoms. Preferred examples of G include phenylene, bisphenylene, naphthalene, methylene, ethylene, propylene, 1,4-butadiene and p-phenylenemethylene. The number of linkages of the poly-linking radical G is r+1, that is, one linkage is a linkage to borate anion, and other linkages r of G are linkages to (SiR^{k}R^{l}R^m) groups. A^+ is a cation. Q is an integer of 1 or greater, preferably 1.

[0263] R^k, R^l and R^m in the formula (24) are each a hydrocarbanyl radical, a trihydrocarbanylsilyl radical, a trihydrocarbanylgermanium radical, a hydrogen radical, an alkoxy radical, a hydroxyl radical or a halogen compound radical. R^k, R^l and R^m may be the same or different. Q is hydride or dihydrocarbylamide, preferably dialkylamide, halide, hydrocarbyloxide, alkoxide, allyloxide, hydrocarbyl, substituted hydrocarbyl radical, or the like, more preferably pentafluorobenzyl radical. n+z is 4.

[0264] [B-Qn(Gq(SiR^{k}R^{l}R^m)r)z]$^-$ in the formula (23) is, for example, triphenyl(4-dimethylchlorosilylphenyl)borate, diphenyl-di(4-dimethylchlorosilylphenyl)borate, triphenyl(4-dimethylmethoxysilylphenyl)borate, tri(p-tolyl)(4-triethoxysilylphenyl)borate, tris(pentafluorophenyl)(4-dimethylchlorosilylphenyl)borate, tris(pentafluorophenyl)(4-dimethylmethoxysilylphenyl)borate, tris(pentafluorophenyl)(4-trimethoxysilylphenyl)borate, or tris(pentafluorophenyl)(6-dimethylchlorosilyl-2-naphthyl)borate.

[0265] As A^+ that is a counter cation of the borate compound, the same cation as that of A^+ in the formula (22) can be mentioned.

[0266] Examples of the borane compounds include salts of anions, such as decaborane, bis[tri(n-butyl)ammonium]nonaborate, bis[tri(n-butyl)ammonium]decaborate, bis[tri(n-butyl)ammonium]undecaborate, bis[tri(n-butyl)ammonium]dodecaborate, bis[tri(n-butyl)ammonium]decachlorodecaborate and bis[tri(n-butyl)ammonium]dodecachlorododecaborate, and salts of metallic borane anions, such as tri(n-butyl)ammoniumbis(dodecahydridododecaborate)cobaltate(III) and bis[tri(n-butyl)ammonium]bis(dodecahydridododecaborate)nickelate (III)

[0267] Examples of the carborane compounds include salts of anions, such as 4-carbanonaborane, 1,3-dicarbanonaborane, 6,9-dicarbadecaborane, dodecahydrido-1-phenyl-1,3-dicarbanonaborane, dodecahydrido-1-methyl-1,3-dicarbanonaborane, undecahydrido-1,3-dimethyl-1,3-dicarbononaborane, 7,8-dicarboundecaborane, 2,7-dicarbaundecaborane, undecahydrido-7,8-dimethyl-7,8-dicarbaundecaborane, dodecahydrido-11-methyl-2,7-dicarboundecaborane, tri(n-butyl)ammonium 1-carbadecaborate, tri(n-butyl)ammonium 1-carbaundecaborate, tri(n-butyl)ammonium 1-carbadodecaborate, tri(n-butyl)ammonium 1-trimethylsilyl-1-carbadecaborate, tri(n-butyl)ammonium bromo-1-carbadodecaborate, tri(n-butyl)ammonium 6-carbadecaborate, tri(n-butyl)ammonium 6-carbadecaborate, tri(n-butyl)ammonium 7-carbaundecaborate, tri(n-butyl)ammonium 7,8-dicarbaundecaborate, tri(n-butyl)ammonium 2,9-dicarbaundecaborate, tri(n-butyl)ammonium dodecahydrido-8-methyl-7,9-dicarbaundecaborate, tri(n-butyl)ammonium dodecahydrido-8-ethyl-7,9-dicarbaundecaborate, tri(n-butyl)ammonium undecahydrido-8-butyl-7,9-dicarbaundecaborate, tri(n-butyl)ammonium undecahydrido-8-allyl-7,9-dicarbaundecaborate, tri(n-butyl)ammonium undecahydrido-9-trimethylsilyl-7,8-dicarbaundecaborate and tri(n-butyl)ammonium undecahydrido-4,6-dibromo-7-carbaundacaborate; and salts of metallic carborane anions, such as tri(n-butyl)ammonium bis(nonahydrido-1,3-dicarbanonaborate)cobaltate(III), tri(n-butyl)ammonium bis(undecahydrido-7,8-dicarbaundecaborate)ferrate(III), tri(n-butyl)ammonium bis(undecahydrido-7,8-dicarbaundecaborate)cobaltate(III), tri(n-butyl)ammonium bis(undecahydrido-7,8-dicarbaundecaborate)nickelate(III), tri(n-butyl)ammonium bis(undecahydrido-7,8-dicarbaundecaborate)cuprate(III), tri(n-butyl)ammonium bis(undecahydrido-7,8-dicarbaundecaborate)aurate(III), tri(n-butyl)ammonium bis(nonahydrido-7,8-dimethyl-7,8-dicarbaundecaborate)ferrate(III), tri(n-butyl)ammonium bis(nonahydrido-7,8-dimethyl-7,8-dicarbaundecaborate) chromate (III), tri(n-butyl)ammonium bis(tribromooctahydrido-7,8-dicarbaundecaborate)cobaltate(III), tris[tri(n-butyl)ammonium]bis(undecahydrido-7-carbaundecaborate) chromate(III), bis[tri(n-butyl)ammonium]bis(undecahydrido-7-carbaundecaborate)manganate(IV), bis[tri(n-butyl) ammonium]bis(undecahydrido-7-carbaundecaborate)cobaltate(III) and bis[tri(n-butyl)ammonium]bis(undecahydrido-7-carbaundecaborate)nickelate(IV).

[0268] Such compounds (B-4) which react with the Group 4 transition metal compound (A) to form an ion pair as above can be used as a mixture of two or more kinds.

[0269] As the component (B) for use in the present invention, a commercially available MAO/toluene solution using the aforesaid organoaluminum oxy-compound (B-3) and manufactured by Nippon Aluminum Alkyls, Ltd., or the like is preferably used.

[0270] In the preparation of the olefin polymerization catalyst for use in the present invention, a carrier can be used, when needed. The carrier is usually an inorganic or organic compound and is a granular or fine particulate solid. Examples of the inorganic compounds include porous oxides, inorganic chlorides, clay, clay minerals and ion-exchange layer compounds.

[0271] As the porous oxides, SiO_2, Al_2O_3, MgO, ZrO, TiO_2, B_2O_3, CaO, ZnO, BaO, ThO_2, and composites or mixtures thereof can be used, and for example, natural or synthetic zeolite, SiO_2—MgO, SiO_2—Al_2O_3, SiO_2—TiO_2, SiO_2—V_2O_5, SiO_2—Cr_2O_3 and SiO_2—TiO_2—MgO can be used.

[0272] The octene (co)polymer (ii) is preferably prepared by (co)polymerizing an α-olefin in the presence of such an olefin polymerization catalyst as above.

[0273] An embodiment of a process for preparing the octene (co)polymer (ii) is described below in detail.

[0274] In the present invention, polymerization reaction is carried out in a hydrocarbon medium. Examples of the hydrocarbon media include aliphatic hydrocarbons, such as propane, butane, pentane, hexane, heptane, octane, decane, dodecane and kerosine; alicyclic hydrocarbons, such as cyclopentane, cyclohexane and methylcyclopentane; aromatic hydrocarbons, such as benzene, toluene and xylene; halogenated hydrocarbons, such as ethylene chloride, chlorobenzene and dichloromethane; and petroleum fractions, such as gasoline, kerosine and gas oil. Further, olefins for use in the polymerization are also employable.

[0275] In the present invention, polymerization is carried out in the presence of such an olefin polymerization catalyst as above, and in the polymerization, the Group IV transition metal compound (A) is used in an amount of usually 10^{-8} to 1 gram atom/liter, preferably 10^{-7} to 10^{-1} gram atom/liter, in terms of a concentration of a transition metal atom in the polymerization reaction system.

[0276] The component (B-1) is used in such an amount that the molar ratio ((B-1)/M) of the component (B-1) to the transition metal atom (M) in the component (A) becomes usually 0.01 to 5000, preferably 0.05 to 2,000. The component (B-2) is used in such an amount that the molar ratio ((B-2)/M) of the component (B-2) to the transition metal atom (M) in the component (A) becomes usually 100 to 25000, preferably 500 to 10000. The component (B-3) is used in such an amount that the molar ratio ((B-3)/M) of an aluminum atom in the component (B-3) to the transition metal atom (M) in the component (A) becomes usually 10 to 5000, preferably 20 to 2000. The component (B-4) is used in such an amount that the molar ratio ((B-4)/M) of the component (B-4) to the transition metal atom (M) in the component (A) becomes usually 1 to 50, preferably 1 to 20.

[0277] The temperature for the polymerization of olefin using such an olefin polymerization catalyst is usually in the range of −50 to +200° C., preferably 0 to 180° C. The polymerization pressure is usually in the range of normal pressure to 10 MPa (gauge pressure), preferably normal pressure to 5 MPa (gauge pressure). The polymerization reaction can be carried out by any of a batch process, a semi-continuous process and a continuous process. Further, the polymerization can be carried out in two or more steps which are different in the reaction conditions. The molecular weight of the resulting olefin polymer can be controlled by allowing hydrogen to exist in the polymerization system or by changing the polymerization temperature.

[0278] The kinematic viscosity of the olefin polymer at 100° C. depends upon the molecular weight of the polymer. That is to say, as the molecular weight is increased, the viscosity becomes higher, and as the molecular weight is decreased, the viscosity becomes lower, so that the kinematic viscosity at 100° C. is adjusted by the above molecular weight control. Moreover, by removing a low-molecular weight component of the resulting polymer by a hitherto publicly known method such as vacuum distillation, the molecular weight distribution (Mw/Mn) of the resulting polymer can be adjusted to not more than 1.8.

[0279] In the present invention, a fatty acid ester (iii) having a kinematic viscosity at 100° C. of not more than 20 mm^2/s is contained. When the kinematic viscosity is in this range, it becomes possible to add the fatty acid ester in a sufficient amount for inhibiting swelling of a lubricating oil sealing agent when the kinematic viscosity of the whole lubricating oil composition is adjusted.

[0280] Although the fatty acid ester (iii) having a kinematic viscosity at 100° C. of not more than 20 mm^2/s is not specifically restricted, such a fatty acid ester consisting of only carbon, oxygen and hydrogen as described below is used.

[0281] The fatty acid ester is, for example, a monoester prepared from a monobasic acid and an alcohol; a diester prepared from a dibasic acid and an alcohol, or prepared from diol and a monobasic acid or an acid mixture; or a polyol ester prepared by the reaction of diol, triol (e.g., trimethylolpropane), tetraol (e.g., pentaerythritol), hexaol (e.g., dipentaerythritol) or the like with a monobasic acid or an acid mixture. Examples of such esters include tridecyl pelargonate, di-2-ethylhexyl adipate, di-2-ethylhexyl azelate, trimethylolpropane triheptanoate and pentaerythritol tetraheptanoate.

[0282] From the viewpoint of compatibility with a 1-decene (co)polymer or a 1-octene (co)polymer selected in the present invention, the alcohol part to constitute the ester is preferably an alcohol having two or more hydroxyl groups, and the fatty acid part is preferably a fatty acid of 8 or more carbon atoms. As the fatty acid, however, a fatty acid of 20 or less carbon atoms that is industrially readily obtainable is advantageous from the viewpoint of production cost. The fatty acid to constitute the ester may be used singly, and even an acid mixture of two or more kinds of fatty acids sufficiently exhibits the performance disclosed in the present invention. More specifically, trimethylolpropane lauric acid stearic acid mixed triester, diisodecyl adipate, etc. can be mentioned, and these are preferable from the viewpoint of compatibility with saturated hydrocarbon components, such as a 1-decene (co) polymer and a 1-octene (co)polymer, and the later-described stabilizers having a polar group, such as an antioxidant, an anti-corrosion agent, an anti-abrasion agent, a friction adjusting agent, a pour point depressant, a rust proof agent and an anti-foaming agent.

[0283] The lubricating oil composition of the present invention contains the decene (co)polymer (i) in an amount of 90 to 10% by mass, preferably 80 to 20% by mass, the octene (co)polymer (ii) in an amount of 5 to 85% by mass, and the ester (iii) in an amount of 5 to 15% by mass, with the proviso that the total amount of the decene (co)polymer (i), the octene (co)polymer (ii) and the ester (iii) is 100% by mass. When the decene (co)polymer (i), the octene (co)polymer (ii) and the ester (iii) are contained in the above ranges, a lubricating oil composition excellent in a balance between the shear stability and the low-temperature viscosity properties is obtained. By the addition of not less than 5% by mass of the ester, good adaptability to lubricating oil sealing materials such as resins or elastomers inside various internal combustion engines and industrial machines is obtained. Specifically, swelling of a lubricating oil sealing material can be inhibited. From the viewpoint of oxidation stability or heat resistance, the amount of the ester is preferably not more than 15% by mass, with the proviso that the total amount of the components (i) to (iii) is 100% by mass.

[0284] One characteristic of the lubricating oil composition of the present invention is that it contains boron atoms in an amount of not less than 5 ppm but not more than 75 ppm. A boron content of 1 ppm means that boron is contained in an amount of 1 mg in 1 kg of the composition. It has been found by the present invention that anti-micropitting performance of the lubricating oil composition is particularly enhanced by introducing boron in the composition in a specific concentration. It has been also found that performance of the later-described extreme pressure agent that is optionally added to the lubricating oil composition is sufficiently exhibited. The boron atoms in the lubricating oil composition are derived from the later-described boron compound. The content of the boron atoms in the lubricating oil composition is measured by ICP emission spectral analysis as in the later-described working examples.

[0285] As mechanism of exhibition of such effects, for example, the following hypothesis can be considered.

[0286] At a gear part in an industrial machine or the like, the surface convex portion of the frictional surface breaks through an oil film of lubricating oil under the high-load conditions (also referred to as “extreme pressure lubricating conditions”) to cause contact of metals, and because of increase in temperature of the frictional surface due to the contact of metals and further because of strong shear, the oil film is separated or removed from the frictional surface. As a result, friction and abrasion are increased, and this finally leads to seizing, resulting in a possibility of impossible lubrication.

[0287] (On that account, it is preferable to add the later-described extreme pressure agent capable of forming a reacted film having larger affinity for a metal under the extreme pressure lubricating conditions on the metal surface, to the lubricating oil composition of the present invention. It is presumed that the film formed by the extreme pressure agent on the metal surface is effective also for improvement in durability against separation of fine fragments from the gear tooth surface due to repeated compression stress received at the gear part in the high-load state, that is, micropitting.)

[0288] However, as described later, before the extreme pressure lubricating conditions in which the extreme pressure agent forms a film on the metal surface are reached, there is a possibility that hydrocarbon or other organic components to constitute the lubricating oil composition are carbonized by heating or shearing to thereby form a carbide film on the metal surface.

[0289] It is considered that the boron-containing compound interferes with a carbide or a carbide precursor prior to the formation of the above-mentioned carbide film and functions as a dispersing agent for preventing adhesion of a carbide to the metal surface. Further, it is presumed that boron has affinity for metals and therefore it bears a part of extreme pressure performance or anti-abrasion performance.

[0290] It is thought that with regard to the micropitting, that is, metal separation occurring particularly in the microscopic region, the boron-containing compound is greatly concerned in metals, and in the case of a boron atom content of not less than 5 ppm in the lubricating oil composition, the lubricating oil composition exhibits anti-micropitting performance. When long-term operation of an engine is taken into account, the boron atom content is preferably not less than 20 ppm. However, the boron-containing compound has an extremely high polarity relatively to saturated hydrocarbons such as a 1-decene (co)polymer and a 1-octene (co)polymer which are main components of the lubricating oil composition of the present invention, and therefore, from the viewpoint of compatibility, the content of boron atoms needs to be not more than 75 ppm, preferably not more than 50 ppm. It is thought that if the boron atom content exceeds the upper limit of this range, the boron-containing compound is aggregated because of poor compatibility, and it inhibits low-temperature viscosity properties of the lubricating oil composition, as an aggregate.

[0291] The boron-containing compound contained may be an organic compound or an inorganic compound. In the inorganic compounds, boron-containing acids, and their anhydrides, oxides and halides are included. In the organic boron compounds, amides and esters of boron are included. Further, borated acylated amine, a borated dispersing agent, borated epoxide and borated fatty acid ester of glycerol are also included.

[0292] Examples of the boron-containing compounds used include boron oxide; boron oxide hydrate; boron trioxide; boron trifluoride; boron tribromide; boron trichloride; and boron-containing acids, such as boronic acid (i.e., alkyl-B$(OH)_2$ or aryl-B$(OH)_2$), boric acid (i.e., H_3BO_3), tetraboric acid (i.e., H_2B4O_7), metaboric acid (i.e., HBO_2) and boron containing acid anhydride, and boron amides and various esters of such boron-containing acids. A complex of a trihalide of boron and ether, an organic acid or hydrocarbon is also employable. Examples of such complexes include boron trifluoride-triethyl ester, boron trifluoride-phosphoric acid, boron trichloride-chloroacetic acid, boron tribromide-dioxane, and boron trifluoride methyl ethyl ether.

[0293] From the viewpoint of compatibility with other components of the lubricating oil composition, the boron-containing compound is desirably an organic compound.

[0294] Examples of the boronic acids include methylboronic acid, phenylboronic acid, cyclohexylboronic acid, p-heptylphenylboronic acid and dodecylboronic acid. Examples of preferred boron-containing compounds include boron oxides, such as boron oxide, boron oxide hydrate and boron trioxide.

[0295] Examples of the boron containing acid esters include organic monoesters, organic diesters and organic triesters of boric acid and alcohols orphenols, such as methanol, ethanol, isopropanol, butanol, pentanol, hexanol, cyclohexanol, cyclopentanol, 1-octanol, 2-octanol, dodecanol, behenyl alcohol, oleyl alcohol, stearyl alcohol, benzyl alcohol, 2-butylcyclohexanol, ethylene glycol, propylene glycol, trimethylene glycol, 1,3-butanediol, 2,4-hexanediol, 1,2-cyclohexanediol, 1,3-octanediol, glycerol, pentaerythritol diethylene glycol, carbitol, cellosolve, triethylene glycol, tripropylene glycol, phenol, naphthol, p-butylphenol, o,p-diheptylphenol, n-cyclohexylphenol, 2,2-bis(p-hydroxyphenyl)propane, polyisobutene (molecular weight: 1,500)-substituted phenol, ethylene chlorohydrin, o-chlorophenol, m-nitrophenol, 6-bromooctanol and 7-keto-decanol. Lower alcohols, 1,2-glycol and 1,3-glycol, that is, alcohols having about 8 or less carbon atoms are useful for preparing the boron containing acid esters.

[0296] The process for preparing boron containing acid esters is already known, and for example, a process for preparing organic triborates using reaction of boron trichloride with 3 mol of alcohol or phenol or reaction of boron oxide with alcohol or phenol can be mentioned. As another process, direct esterification of tetraboric acid with 3 mol of alcohol or phenol can be mentioned. As a further process, formation of cyclic alkylene borate using direct esterification of, for example, boric acid with glycol can be mentioned.

[0297] In a certain embodiment, the boron-containing compound is a compound wherein an oil-soluble ash-free dispersing agent is borated.

[0298] The ash-free dispersing agent has basic nitrogen and/or at least one hydroxyl group in a molecule. By allowing basic nitrogen and/or at least one hydroxyl group in the ash-free dispersing agent to react with boron, a borated ash-free dispersing agent is obtained. Examples of preferred dispersing agents include alkenylsuccinic acid imide, alkenylsuccinic acid ester, alkenylsuccinic acid ester amide, Mannich base, hydrocarbyl polyamine and polymeric polyamine.

[0299] The alkenylsuccinic acid imide can be formed by a hitherto publicly known method, such as a method of heating alkenylsuccinic anhydride, acid, acid ester, acid halide or lower alkyl ester together with polyamine containing at least one primary amino group. The alkenylsuccinic anhydride can be readily formed by heating a mixture of an olefin and maleic anhydride up to a temperature of for example about 180 to 220° C. This olefin is a polymer or a copolymer of a lower monoolefin, such as ethylene, propylene, 1-butene, isobutene or a mixture thereof. The source of alkenyl group is, for example, polyisobutene having a gel permeation chromatography (GPC) number-average molecular weight of 10,000 or more, for example, about 500 to about 2,500, or about 800 to about 1,500. In a certain embodiment, the molecular weight of poyisobutylene is in the range of about 700 to about 5,000. The ratio of polyisobutylenesuccinic anhydride to amine is, for example, in the range of about 1.4 to about 3, or about 1.8 to about 2.2.

[0300] In the lubricating oil composition of the present invention, the above-mentioned boron-containing compounds may be used singly or in combination of two or more kinds.

[0301] The lubricating oil composition of the present invention preferably further contains one or more kinds of extreme pressure agents.

[0302] The extreme pressure agent in the present application is a component other than the components (i) to (iii), and is a general term for substances exerting an effect in preventing sieving. The extreme pressure agent is not specifically restricted, and examples thereof include sulfur-based extreme pressure agents, such as sulfides, sulfoxides, sulfones, thiophosphinates, thiocarbonates, sulfurized fats and oils, and sulfurized olefins; phosphoric acids, such as phosphoric acid ester, phosphorous acid ester, phosphoric acid ester amine salt and phosphorous acid ester amines; and halogen-based compounds, such as chlorinated hydrocarbon. These compounds may be used in combination of two or more kinds.

[0303] However, before the extreme pressure lubricating conditions are reached, there is a possibility that hydrocarbon or other organic components to constitute the lubricating oil composition are carbonized by heating or shearing to thereby form a carbide film on the metal surface. On this account, when the extreme pressure agent is used alone, there is a fear that the contact of the extreme pressure agent with the metal surface is inhibited by the carbide film and a satisfactory effect of the extreme pressure agent cannot be expected.

[0304] In the addition of the boron-containing compound to the lubricating oil composition, the boron-containing compound may be added alone. However, the lubricating oil composition of the present invention contains saturated hydrocarbons such as a 1-decene (co)polymer and a 1-octene (co) polymer as main components, and therefore, from the viewpoint of dispersibility of the boron-containing compound, it is preferable to add the boron-containing compound in a dissolved state in lubricating base oil such as mineral oil or 1-decene (co)polymer. In either method, the boron-containing compound is added so that the boron atom content may become not less than 5 ppm but not more than 75 ppm.

[0305] Specifically, it is more preferable that among so-called extreme pressure agent packages in which various components such as an extreme pressure component have been blended in advance, an extreme pressure agent package in which the boron-containing compound has been already added in a dissolved state is selected, and this package is added to the lubricating oil composition.

[0306] In the case where the boron-containing compound is added in a given amount as the extreme pressure agent package, it is preferable to select the extreme pressure agent package containing the boron-containing compound so that the amount of the extreme pressure agent package added may become 2 to 5% by mass based on 100% by mass of the components (i) to (iii) in the lubricating oil composition of the present invention. The extreme pressure agent component is introduced into the extreme pressure agent package in consideration of the concentration by which the extreme pressure agent component can be sufficiently dispersed in the lubricating oil composition, and therefore, if the amount of the extreme pressure agent package is less than 2% by mass, a satisfactory effect in the extreme pressure performance cannot be expected occasionally, from the viewpoint of restriction on the upper limit of the concentration. In contrast with this, if the amount thereof exceeds 5% by mass, the boron-containing compound is aggregated because of poor compatibility with the saturated hydrocarbon components such as a 1-decene (co)polymer and a 1-octent (co)polymer in the lubricating oil composition, and it sometimes inhibits low-temperature viscosity properties of the lubricating oil composition, as an aggregate.

[0307] Examples of preferred extreme pressure agents (packages) include Anglamol 98A available from LUBRIZOL CORPORATION and HITEC 1532 available from AFTON CHEMICAL CORPORATION. In particular, HITEC 307 available from AFTON CHEMICAL CORPORATION can be preferably used from the viewpoint of a balance between the boron-containing compound and other components.

[0308] The lubricating oil composition of the present invention may further contain additives, such as a viscosity index improver, an antioxidant, an anti-corrosion agent, an anti-abrasion agent, a friction adjusting gent, a pour point depressant, a rust proof agent and an anti-foaming agent.

[0309] As examples of the additives employable in the lubricating oil composition of the present invention, the following compounds can be given, and they can be used singly or in combination of two or more kinds.

[0310] Examples of detergent dispersants include metallic sulfonate, metallic phenate, metallic phosphanate and succinic acid imide. The detergent dispersant is usually used in an amount of 0 to 15% by mass based on 100% by mass of the total amount of the components (i) to (iii).

[0311] Examples of the anti-abrasion agents include inorganic or organic molybdenum compounds such as molybdenum disulfide, graphite, antimony sulfide and polytetrafluoroethylene. The anti-abrasion agent is used in an amount of 0 to 3% by mass based on 100% by mass of the total amount of the components (i) to (iii), when needed.

[0312] Example of the antioxidants include phenol-based compounds such as 2,6-di-tert-butyl-4-methylphenol, and amine-based compounds. The anti-abrasion agent is used in an amount of 0 to 3% by mass based on 100% by mass of the total amount of the components (i) to (iii), when needed.

[0313] Examples of the rust proof agents include various amine compounds, carboxylic acid metal salts, polyhydric alcohol esters, phosphorus compounds and sulfonates. The rust proof agent is used in an amount of 0 to 3% by mass based on 100% by mass of the total amount of the components (i) to (iii), when needed.

[0314] Examples of the anti-foaming agents include silicone-based compounds, such as dimethylsiloxane and a silica gel dispersion, alcohol-based compounds and ester-based compounds. The anti-foaming agent is used in an amount of 0 to 0.2% by mass based on 100% by mass of the total amount of the components (i) to (iii), when needed.

[0315] In addition to the above additives, a demulsifying agent, a colorant, an oiliness agent (oiliness improver), etc. can be used, when needed.

[0316] Since the lubricating oil composition of the present invention uses a combination of the specific decene (co)polymer (i) and the specific octene (co)polymer (ii), the composition is excellent in shear stability. The shear stability is evaluated by a KRL shear stability test based on the method described in Deutsche Industrie Normen DIN 52350-6. Specifically, the lubricating oil composition is allowed to stand at 60° C. for 20 hours under the shearing conditions (1450 rpm), and a kinematic viscosity decrease ratio, that is obtained from a kinematic viscosity at 40° C. after the test and a kinematic viscosity at 40° C. before the test, is determined. As this value is decreased, the shear stability of the rubricating oil composition becomes more excellent. When the kinematic viscosity of the lubricating oil composition of the present invention at 40° C. is adjusted to not less than 288 mm^2/s but not more than 352 mm^2/s, the kinematic viscosity decrease ratio of the lubricating oil composition at 40° C. after the KRL shear stability test, as measured in accordance with DIN 52350-6, is usually less than 10%, preferably less than 5%. A kinematic viscosity at 40° C. of not less than 288 mm^2/s but not more than 352 mm^2/s indicates that this lubricating oil composition has a viscosity grade (ISO 3448) of VG 320. The lubricating oil composition having this viscosity grade is particularly preferably used for gears for wind power generators.

[0317] As described above, the lubricating oil composition of the present invention contains the specific decene (co) polymer (i), the specific octene (co)polymer (ii) and the specific ester (iii) and contains boron atoms in a specific amount, and hence, the composition is excellent in shear stability, low-temperature viscosity properties and micropitting resistance and is also excellent in a balance among oxidation stability, heat resistance and temperature viscosity properties.

Uses

[0318] The lubricating oil composition of the present invention can be used as industrial lubricating oil (gear oil, hydraulic oil) or grease base oil. The lubricating oil composition of the present invention is excellent in shear stability, low-temperature viscosity properties and anti-micorpitting performance, as described above, and hence, the composition is particularly favorably used as lubricating oil for a wind power generator. If the lubricating oil composition is excellent in the shear stability and the anti-micropitting performance and if the composition is used for a gear for a wind power generator, the gear can be rendered maintenance-free. Moreover, also from the viewpoint of efficiency of energy conversion from wind force into electric power, it is preferable that the low-temperature viscosity properties are excellent.

[0319] The gear for a wind power generator of the present invention is lubricated with the above-mentioned lubricating oil composition.

[0320] Each of FIGS. **1** to **4** shows an example of a wind power generating apparatus having the gear for a wind power generator. The wind power generating apparatus is provided with a planetary gear (toothed wheel) type power transmission device, and specifically has a rotor nose **1**, an encasing opening **2**, a roller bearing **3**, a rotor carrier **4**, an azimuth bearing **5**, an external race **6**, an internal race **7**, a tow bin **8**, a driving stage **9**, a driven stage **10**, a generator **11**, a sun gear **12**, a hollow gear **13**, a hollow gear **14**, a power transmission device casing **15**, a planetary gear carrier **16**, a vibration damping device **17** and a fixed rail **18**. Functions and operations of these parts are the same as those disclosed in Japanese Patent Laid-Open Publication No. 337245/2000.

[0321] The lubricating oil composition of the present invention is favorably used for lubrication of the above gear part (toothed wheel part).

[0322] As previously described, the present invention relates to the following (1) to (7).

[0323] (1) A lubricating oil composition comprising the following (i) to (iii), having a kinematic viscosity at 40° C. of not less than 30 mm^2/s but not more than 750 mm^2/s and containing boron atoms in an amount of not less than 5 ppm but not more than 75 ppm,

[0324] (i) 90 to 10% by mass of a (co)polymer having a kinematic viscosity at 100° C. of not more than 45 mm^2/s, containing not less than 60% by mol of constituent units derived from 1-decene and having an acid value of less than 0.1 mgKOH/g,

[0325] (ii) 5 to 85% by mass of a (co)polymer having a kinematic viscosity at 100° C. of not less than 35 mm^2/s but not more than 1,500 mm^2/s, containing not less than 90% by mol of constituent units derived from 1-octene and having a molecular weight distribution, as measured by gel permeation chromatography, of not more than 1.8, and

[0326] (iii) 5 to 15% by mass of a fatty acid ester having a kinematic viscosity at 100° C. of not more than 20 mm^2/s,

[0327] with the proviso that the total amount of (i) to (iii) is 100% by mass.

[0328] Since the above lubricating oil composition uses a combination of the specific decene (co)polymer (i) and the specific octene (co)polymer, the composition is excellent in shear stability, low-temperature viscosity properties and temperature viscosity properties. Further, since the boron content is in the specific range, the lubricating oil composition is excellent in anti-micropitting performance and low-temperature viscosity properties.

[0329] (2) The lubricating oil composition as stated in (1), which further comprises one or more kinds of extreme pressure agents.

[0330] (3) The lubricating oil composition as stated in (1) or (2), wherein the kinematic viscosity of the lubricating oil composition at 40° C. is not less than 190 mm^2/s but not more than 750 mm^2/s.

[0331] When the kinematic viscosity at 40° C. is not more than 750 mm^2/s, the lubricating oil composition is excellent in fuel saving performance and energy conversion efficiency. When the kinematic viscosity at 40° C. is not less than 190 mm^2/s, the lubricating oil composition exhibits good lubricating properties and shear stability as gear oil at high temperatures and/or in a high-load state.

[0332] (4) The lubricating oil composition as stated in any one of (1) to (3), wherein the kinematic viscosity of the lubricating oil composition at 40° C. is not less than 288 mm^2/s but not more than 352 mm^2/s, and a kinematic viscosity decrease ratio of the lubricating oil composition at 40° C. after a KRL shear stability test, as measured in accordance with DIN 52350-6, is less than 10%.

[0333] When the lubricating oil composition is excellent in shear stability as above, the composition is favorably used for a gear for a wind power generator.

[0334] (5) The lubricating oil composition as stated in any one of (1) to (4), wherein the kinematic viscosity of the octene (co)polymer (ii) at 100° C. is not less than 150 mm^2/s but not more than 1,200 mm^2/s.

[0335] When the kinematic viscosity at 100° C. is not less than 150 mm^2/s, the lubricating oil composition exhibits excellent low-temperature viscosity properties. When the kinematic viscosity at 100° C. is not more than 1,200 mm^2/s, the shear stability of the lubricating oil composition is more excellent.

[0336] (6) The lubricating oil composition as stated in any one of (1) to (5), wherein the kinematic viscosity of the decene (co)polymer (i) at 100° C. is not more than 11 mm^2/s.

[0337] When the kinematic viscosity at 100° C. is not more than 11 mm^2/s, the low-temperature viscosity properties of the lubricating oil composition are more excellent.

[0338] (7) A gear for a wind power generator, which is lubricated with the lubricating oil composition as stated in any one of (1) to (6).

[0339] Such a gear for a wind power generator can be rendered maintenance-free and is excellent in efficiency of energy conversion from wind force into electric power.

EXAMPLES

[0340] The present invention is further described with reference to the following examples, but it should be construed that the present invention is in no way limited to those examples.

Average Molecular Weight, Molecular Weight Distribution

[0341] Number-average molecular weight (Mn) and molecular weight distribution (Mw/Mn) were measured in the following manner using GPC (Chromatopac C-R4A) manufactured by Shimadzu Corporation. As separation columns, TSKG6000H XL, G4000H XL, G3000H XL and G2000H XL were used. The column temperature was set at 40° C. As a mobile phase, tetrahydrofuran (available from Wako Pure Chemical Industries, Ltd.) was used. The developing rate was 0.8 ml/min, and the sample concentration was 0.2% by mass. The sample injection volume was 20 microliters, and as a detector, a differential refractometer was used. As standard polystyrene, polystyrene available from Tosoh Corporation was used.

Viscosity Properties

[0342] Kinematic viscosity at 100° C. and at 40° C. and viscosity index were measured and calculated by the method described in JIS K2283.

Shear Stability

[0343] Shear stability of a lubricating oil composition was evaluated in accordance with the method described in DIN 52350-6 using a KRL shear stability tester. The compounded oil was allowed to stand at 60° C. for 20 hours under the shearing conditions (1450 rpm). A kinematic viscosity at 40° C. was measured before and after the test, and a kinematic viscosity decrease ratio after the test was evaluated.

[0344] When the kinematic viscosity decrease ratio is less than 10% after a shear stress is given by the above method, the lubricating oil composition exhibits good durability. When the kinematic viscosity decrease ratio is less than 5%, the lubricating oil composition exhibits extremely excellent durability.

−30° C. Viscosity

[0345] −30° C. Viscosity was measured by a Brookfield viscometer in accordance with ASTM D2983. Taking error of measurement into account, −30° C. viscosities were classified into the following three categories.

[0346] High: more than 100,000 mPa·s

[0347] Medium: more than 80,000 mPa·s but not more than 100,000 mPa·s

[0348] Low: not more than 80,000 mPa·s

[0349] When the −30° C. viscosity is not more than 100,000 mPa·s, favorable lubricating properties can be maintained at extremely low temperatures down to about −40° C., and excellent fuel saving performance and energy conversion efficiency are exhibited. Particularly when the −30° C. viscosity is not more than 80,000 mPa·s, extremely excellent fuel saving performance and energy conversion efficiency are exhibited. If the −30° C. viscosity exceeds 100,000 mPa·s, fuel saving performance and energy conversion efficiency in an extremely low temperature environment become poor.

Micropitting Failure Load Stage

[0350] Failure load stage was measured by a FVA-54/7 micropitting test defined by Drive Technology Research Association (FVA). When the failure load stage is not less than 10, excellent anti-micropitting performance is exhibited.

(i) (Co)polymer Comprising 1-decene

[0351] As the decene (co)polymer (i) having a kinematic viscosity at 100° C. of not more than 45 mm^2/s and comprising 1-decene, poly-α-olefin (PAO) available from NESTE OIL CORPORATION, i.e., NEXBASE 2006 (kinematic viscosity at 100° C.: 6 mm^2/s), was used. The acid value of the (co)polymer, as measured in accordance with JIS K2501, was less than 0.1 mgKOH/g.

(ii) (Co)polymer Using 1-octene as Monomer

[0352] The octene (co)polymer (ii) using 1-octene as a monomer was prepared by the following polymerization example.

[0353] (Methyl)(p-tolyl)methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride used in the polymerization example was synthesized by the following process.

Synthesis of 6,6'-methyl(p-tolyl)fulvene

[0354] In a 200 ml three neck flask, 5.9 g (81.9 mmol) of lithium cyclopentadiene was placed in a nitrogen atmosphere, and 100 ml of dehydrated diethyl ether was added, followed by stirring. The resulting slurry was cooled with an ice bath, and 10.0 g (74.5 mmol) of 4'-methylacetophenone was dropwise added. Thereafter, the mixture was stirred at room temperature for 20 hours, and the resulting solution was quenched with a dilute hydrochloric acid aqueous solution. Then, 50 ml of hexane was added, and the resulting organic layer was washed with water and then dried over anhydrous magnesium sulfate. The solvent was distilled off, and the resulting viscous liquid was subjected to column chromatography separation using hexane to obtain an object of a red viscous liquid (yield: 9.8 g, 72%).

Synthesis of (methyl)(p-tolyl)cyclopentadienyl(octamethyloctahydrodibenzofluorenyl)methane

[0355] In a 200 ml three neck flask, 80 ml of tetrahydrofuran and 5.0 g (12.9 mmol) of octamethyloctahydrodibenzofluorene were mixed in a nitrogen atmosphere, and the resulting solution was cooled to −20° C. Then, to the solution was slowly dropwise added 8.45 ml of n-butyllithium (1.61M hexane solution, 13.5 mmol), followed by stirring at room temperature for 5 hours. Thereafter, this reaction solution was cooled to −20° C., and then 2.6 g (14.2 mmol) of 6,6'-methyl (p-tolyl)fulvene was slowly dropwise added. With slowly returning the temperature to room temperature, the reaction solution was stirred for 1 hour. This reaction solution was quenched with a dilute hydrochloric acid aqueous solution and then extracted with diethyl ether. The resulting organic layer was washed with a saturated sodium hydrogencarbonate aqueous solution and a saturated saline solution, and then dried over anhydrous magnesium sulfate. The solvent was distilled off, and the resulting solid was recrystallized with methanol to obtain a desired product of a white solid (yield: 7.2 g, 98%).

(iii) Ester

[0356] As the ester (iii) having a kinematic viscosity at 100° C. of not more than 20 mm^2/s, diisodecyl adipate (DIDA) available from Daihachi Chemical Industry Co., Ltd., which was a fatty acid ester, was used.

[0357] Extreme Pressure Agent Package

[0358] Extreme pressure agent A: As an extreme pressure agent package having a boron content of 1,260 ppm, a commercially available extreme pressure agent package (HITEC 307 available from AFTON CHEMICAL CORPORATION) was used.

[0359] Extreme pressure agent B: As an extreme pressure agent package having a boron content of less than 10 ppm, a commercially available extreme pressure agent package (HITEC 343 available from AFTON CHEMICAL CORPORATION) was used.

[0360] The boron content in the extreme pressure agent package was measured by ICP emission spectral analysis. The boron content in the lubricating oil composition was determined by converting the boron content in the extreme pressure agent package and the amounts of the components blended in the lubricating oil composition.

Polymerization Example 1

[0361] In a stainless steel autoclave having an internal volume of 2 liters and thoroughly purged with nitrogen, 1000 ml of 1-octene was placed, and the temperature in the system was raised to 65° C. Thereafter, hydrogen was fed to adjust the total pressure to 3 MPa-G. Next, 0.4 mmol of triisobutylaluminum, 0.0007 mmol of a heptane solution of di(p-tolyl) methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride and 0.35 mmol (in terms of Al concentration) of a hexane solution of methylaluminoxane (MMA0-3A, available from Tosoh Finechem Corporation) were forced into the autoclave with nitrogen, and polymerization was carried out at 70° C. for 60 minutes at a rotation speed of 400 rpm. A small amount of isopropanol was added to the system to terminate the polymerization. After release of pressure, to the solution taken out was added n-heptane, then the mixture was transferred into a separatory funnel, and 100 ml of 1N hydrochloric acid was added. The mixture was shaken and allowed to stand still for about 10 minutes, followed by performing oil-water separation. The organic layer was withdrawn and then washed with water. To the organic layer was added a small amount of sodium sulfate, and the mixture was allowed to stand still for about 10 minutes to dehydrate. Thereafter, the resulting solution was filtered to remove sodium sulfate, and then n-heptane and unreacted 1-octene were distilled off at 135° C. under reduced pressure (300 mmHg). The amount of the resulting transparent liquid polymer (1-octene polymer) was 83.7 g.

[0362] The molecular weight (Mn) of the resulting transparent liquid 1-octene polymer (polymer 1), as measured by gel permeation chromatography, was 6,450, and the distribution (Mw/Mn) was 1.67. The kinematic viscosity, as measured by the method described in JIS K2283, was 300 mm^2/s

at 100° C. and was 3,750 mm^2/s at 40° C. The viscosity index, as calculated from these kinematic viscosities, was 227.

Polymerization Example 2

[0363] In a stainless steel autoclave having an internal volume of 2 liters and thoroughly purged with nitrogen, 1000 ml of 1-octene was placed, and the temperature in the system was raised to 45° C. Thereafter, hydrogen was fed to adjust the total pressure to 3 MPa-G. Next, 0.4 mmol of triisobutylaluminum, 0.001 mmol of a heptane solution of di(p-tolyl)methylene (cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride and 0.50 mmol (in terms of Al concentration) of a hexane solution of methylaluminoxane (MA0-3A, available from Tosoh Finechem Corporation) were forced into the autoclave with nitrogen, and polymerization was carried out at 50° C. for 60 minutes at a rotation speed of 400 rpm. A small amount of isopropanol was added to the system to terminate the polymerization. After release of pressure, to the solution taken out was added n-heptane, then the mixture was transferred into a separatory funnel, and 100 ml of 1N hydrochloric acid was added. The mixture was shaken and allowed to stand still for about 10 minutes, followed by performing oil-water separation. The organic layer was withdrawn and then washed with water. To the organic layer was added a small amount of sodium sulfate, and the mixture was allowed to stand still for about 10 minutes to dehydrate. Thereafter, the resulting solution was filtered to remove sodium sulfate, and then n-heptane and unreacted 1-octene were distilled off at 135° C. under reduced pressure (300 mmHg). The amount of the resulting transparent liquid polymer (1-octene polymer) was 107.6 g.

[0364] The molecular weight (Mn) of the resulting transparent liquid 1-octene polymer (polymer 2), as measured by gel permeation chromatography, was 9,940, and the distribution (Mw/Mn) was 1.64. The kinematic viscosity, as measured by the method described in JIS K2283, was 640 mm^2/s at 100° C. and was 9,604 mm^2/s at 40° C. The viscosity index, as calculated from these kinematic viscosities, was 253.

Polymerization Example 3

[0365] In a stainless steel autoclave having an internal volume of 2 liters and thoroughly purged with nitrogen, 1000 ml of 1-octene was placed, and the temperature in the system was raised to 35° C. Thereafter, hydrogen was fed to adjust the total pressure to 3 MPa-G. Next, 0.4 mmol of triisobutylaluminum, 0.001 mmol of a heptane solution of di(p-tolyl)methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride and 0.50 mmol (in terms of Al concentration) of a hexane solution of methylaluminoxane (MMA0-3A, available from Tosoh Finechem Corporation) were forced into the autoclave with nitrogen, and polymerization was carried out at 40° C. for 60 minutes at a rotation speed of 400 rpm. A small amount of isopropanol was added to the system to terminate the polymerization. After release of pressure, to the solution taken out was added n-heptane, then the mixture was transferred into a separatory funnel, and 100 ml of 1N hydrochloric acid was added. The mixture was shaken and allowed to stand still for about 10 minutes, followed by performing oil-water separation. The organic layer was withdrawn and then washed with water. To the organic layer was added a small amount of sodium sulfate, and the mixture was allowed to stand still for about 10 minutes to dehydrate. Thereafter, the resulting solution was filtered to remove sodium sulfate, and then n-heptane and unreacted 1-octene were distilled off at 135° C. under reduced pressure (300 mmHg). The amount of the resulting transparent liquid polymer (1-octene polymer) was 108.8 g.

[0366] The molecular weight (Mn) of the resulting transparent liquid 1-octene polymer (polymer 3), as measured by gel permeation chromatography, was 11,430, and the distribution (Mw/Mn) was 1.66. The kinematic viscosity, as measured by the method described in JIS K2283, was 920 mm^2/s at 100° C. and was 14,720 mm^2/s at 40° C. The viscosity index, as calculated from these kinematic viscosities, was 269.

Polymerization Example 4

[0367] In a stainless steel autoclave having an internal volume of 2 liters and thoroughly purged with nitrogen, 300 ml of n-heptane and 700 ml of 1-hexene were placed, and the temperature in the system was raised to 75° C. Thereafter, hydrogen was fed to adjust the total pressure to 3 MPa-G. Next, 0.4 mmol of triisobutylaluminum, 0.0015 mmol of a heptane solution of di(p-tolyl)methylene(cyclopentadienyl)(octamethyloctahydrodibenzofluorenyl)zirconium dichloride and 0.75 mmol (in terms of Al concentration) of a hexane solution of methylaluminoxane (MMA0-3A, available from Tosoh Finechem Corporation) were forced into the autoclave with nitrogen, and polymerization was carried out at 80° C. for 60 minutes at a rotation speed of 400 rpm. A small amount of isopropanol was added to the system to terminate the polymerization. After release of pressure, to the solution taken out was added n-heptane, then the mixture was transferred into a separatory funnel, and 100 ml of 1N hydrochloric acid was added. The mixture was shaken and allowed to stand still for about 10 minutes, followed by performing oil-water separation. The organic layer was withdrawn and then washed with water. To the organic layer was added a small amount of sodium sulfate, and the mixture was allowed to stand still for about 10 minutes to dehydrate. Thereafter, the resulting solution was filtered to remove sodium sulfate, and then n-heptane and unreacted 1-hexene were distilled off at 135° C. under reduced pressure (300 mmHg). The amount of the resulting transparent liquid polymer (1-hexene polymer) was 333.0 g.

[0368] The molecular weight (Mn) of the resulting transparent liquid 1-hexene polymer (polymer 4), as measured by gel permeation chromatography, was 3,900, and the distribution (Mw/Mn) was 1.73. The kinematic viscosity, as measured by the method described in JIS K2283, was 319 mm^2/s at 100° C. and was 6,296 mm^2/s at 40° C. The viscosity index, as calculated from these kinematic viscosities, was 190.

Example 1

[0369] 33.2% by mass of the (co)polymer comprising 1-decene, 56.5% by mass of the polymer 1 obtained in Polymerization Example 1 as the (co)polymer using 1-octene as a monomer, 10.3% by mass of the ester and 2.7% by mass of the extreme pressure agent A were blended. The blending was controlled so that the kinematic viscosity at 40° C. would become 288 to 352 mm^2/s. Lubricating properties of the resulting lubricating oil composition are set forth in Table 1.

Example 2

[0370] 45.5% by mass of the (co)polymer comprising 1-decene, 44.2% by mass of the polymer 2 obtained in Polymerization Example 2 as the (co)polymer using 1-octene as a monomer, 10.3% by mass of the ester and 2.7% by mass of the extreme pressure agent A were blended. The blending was controlled so that the kinematic viscosity at 40° C. would become 288 to 352 mm^2/s. Lubricating properties of the resulting lubricating oil composition are set forth in Table 1.

Example 3

[0371] 49.7% by mass of the (co)polymer comprising 1-decene, 40.0% by mass of the polymer 3 obtained in Polymerization Example 3 as the (co)polymer using 1-octene as a monomer, 10.3% by mass of the ester and 2.7% by mass of the extreme pressure agent A were blended. The blending was controlled so that the kinematic viscosity at 40° C. would become 288 to 352 mm^2/s. Lubricating properties of the resulting lubricating oil composition are set forth in Table 1.

Example 4

[0372] 45.4% by mass of the (co)polymer comprising 1-decene, 44.2% by mass of the polymer 2 obtained in Polymerization Example 2 as the (co)polymer using 1-octene as a monomer, 10.4% by mass of the ester and 3.5% by mass of the extreme pressure agent A were blended. The blending was controlled so that the kinematic viscosity at 40° C. would become 288 to 352 mm^2/s. Lubricating properties of the resulting lubricating oil composition are set forth in Table 1.

Comparative Example 1

[0373] 32.3% by mass of the (co)polymer comprising 1-decene, 57.1% by mass of the polymer 1 obtained in Polymerization Example 1 as the (co)polymer using 1-octene as a monomer, 10.6% by mass of the ester and 6.0% by mass of the extreme pressure agent A were blended. The blending was controlled so that the kinematic viscosity at 40° C. would become 288 to 352 mm^2/s. Lubricating properties of the resulting lubricating oil composition are set forth in Table 2.

Comparative Example 2

[0374] 23.6% by mass of the (co)polymer comprising 1-decene, 66.1% by mass of Spectrasyn Ultra-150 available from ExxonMobil Chemical Company as the (co)polymer using 1-decene as a monomer, 10.3% by mass of the ester and 2.7% by mass of the extreme pressure agent A were blended. The blending was controlled so that the kinematic viscosity at 40° C. would become 288 to 352 mm^2/s. Lubricating properties of the resulting lubricating oil composition are set forth in Table 2.

Comparative Example 3

[0375] 34.0% by mass of the (co)polymer comprising 1-decene, 55.7% by mass of Spectrasyn Ultra-300 available from ExxonMobil Chemical Company as the (co)polymer using 1-decene as a monomer, 10.3% by mass of the ester and 2.7% by mass of the extreme pressure agent A were blended. The blending was controlled so that the kinematic viscosity at 40° C. would become 288 to 352 mm^2/s. Lubricating properties of the resulting lubricating oil composition are set forth in Table 2.

Comparative Example 4

[0376] 51.2% by mass of the (co)polymer comprising 1-decene, 38.5% by mass of Spectrasyn Ultra-1000 available from ExxonMobil Chemical Company as the (co)polymer using 1-decene as a monomer, 10.3% by mass of the ester and 2.7% by mass of the extreme pressure agent A were blended. The blending was controlled so that the kinematic viscosity at 40° C. would become 288 to 352 mm^2/s. Lubricating properties of the resulting lubricating oil composition are set forth in Table 2.

Comparative Example 5

[0377] Operations were carried out in accordance with Example 1, except that the extreme pressure agent A was replaced with the extreme pressure agent B. Lubricating properties of the resulting lubricating oil composition are set forth in Table 2.

TABLE 1

	Ex. 1	Ex. 2	Ex. 3	Ex. 4
Content of PAO (i) (% by mass)	33.2	45.5	49.7	45.4
Content of polymer (ii) (% by mass)	56.5	44.2	40.0	44.2
Content of ester (% by mass)	10.3	10.3	10.3	10.4
Content of extreme pressure agent (% by mass)	2.7	2.7	2.7	3.5
Content of boron (ppm)	33	33	33	44
40° C. Dynamic viscosity (mm^2/s)	325	326	329	328
−30° C. Viscosity (mPa · s)	medium	low	low	medium
Shear stability (dynamic viscosity decrease ratio, %)	2.3	5.8	9.8	5.8
Micropitting failure load stage	>10	>10	>10	>10

TABLE 2

	Comp. Ex. 1	Comp. Ex. 2	Comp. Ex. 3	Comp Ex. 4	Comp. Ex. 5
Content of PAO (i) (% by mass)	32.3	23.6	34.0	51.2	33.2
Content of polymer (ii) (% by mass)	57.1	66.1	55.7	38.5	56.5
Content of ester (% by mass)	10.6	10.3	10.3	10.3	10.3
Content of extreme pressure agent (% by mass)	6.0	2.7	2.7	2.7	2.7
Content of boron (ppm)	76	33	33	33	<1
40° C. Dynamic viscosity (mm^2/s)	326	315	320	319	325
−30° C. Viscosity (mPa · s)	high	medium	low	low	medium
Shear stability (dynamic viscosity decrease ratio, %)	2.3	10.4	17.9	51	2.3
Micropitting failure load stage	>10	>10	>10	>10	8

[0378] Polymerization Examples 1 to 3 are excellent in viscosity index, namely, temperature viscosity properties, as compared with the polymer using a monomer of 6 carbon atoms, i.e., 1-hexene, said polymer being shown in Polymerization Example 4. Accordingly, it is apparent that the low-temperature viscosity properties of the lubricating oil composition of the present invention are superior to those of the lubricating oil composition containing a polymer using 1-hexene as a monomer.

[0379] Examples 1 to 4 (Table 1) are excellent in −30° C. viscosity as compared with Comparative Example 1 (Table 2) having a boron content of more than 75 ppm.

[0380] Further, Examples 1 to 4 (Table 1) are excellent in shear stability as compared with Comparative Examples 2 to 4 (Table 2) in each of which a polymer using 1-decene as a monomer is contained in the lubricating oil composition.

[0381] Furthermore, Examples 1 to 4 (Table 1) are excellent in anti-micropitting performance as compared with Comparative Example 5 (Table 2) having a low boron content.

DESCRIPTION OF REFERENCE SIGNS

[0382] **1**: rotor nose
[0383] **2**: encasing opening
[0384] **3**: roller bearing
[0385] **4**: rotor carrier
[0386] **5**: azimuth bearing
[0387] **8**: tow bin
[0388] **9**: driving stage
[0389] **10**: driven stage
[0390] **11**: generator
[0391] **12**: sun gear
[0392] **13**: hollow gear
[0393] **14**: hollow gear
[0394] **15**: power transmission device casing
[0395] **16**: planetary gear carrier
[0396] **17**: vibration damping device
[0397] **18**: fixed rail

1. A lubricating oil composition comprising the following (i) to (iii), having a kinematic viscosity at 40° C. of not less than 30 mm^2/s but not more than 750 mm^2/s and containing boron atoms in an amount of not less than 5 ppm but not more than 75 ppm,

(i) 90 to 10% by mass of a (co)polymer having a kinematic viscosity at 100° C. of not more than 45 mm^2/s and having an acid value of less than 0.1 mgKOH/g, not less than 60% by mol of its constituent units being derived from 1-decene,

(ii) 5 to 85% by mass of a (co)polymer having a kinematic viscosity at 100° C. of not less than 35 mm^2/s but not more than 1,500 mm^2/s and having a molecular weight distribution, as measured by gel permeation chromatography, of not more than 1.8, not less than 90% by mol of its constituent units being derived from 1-octene, and

(iii) 5 to 15% by mass of a fatty acid ester having a kinematic viscosity at 100° C. of not more than 20 mm^2/s,

with the proviso that the total amount of (i) to (iii) is 100% by mass.

2. The lubricating oil composition as claimed in claim **1**, which further comprises one or more kinds of extreme pressure agents.

3. The lubricating oil composition as claimed in claim **1**, wherein the kinematic viscosity of the lubricating oil composition at 40° C. is not less than 190 mm^2/s but not more than 750 mm^2/s.

4. The lubricating oil composition as claimed in claim **1**, wherein the kinematic viscosity of the lubricating oil composition at 40° C. is not less than 288 mm^2/s but not more than 352 mm^2/s, and a kinematic viscosity decrease ratio of the lubricating oil composition at 40° C. after a KRL shear stability test, as measured in accordance with DIN 52350-6, is less than 10%.

5. The lubricating oil composition as claimed in claim **1**, wherein the kinematic viscosity of the (co)polymer (ii) at 100° C. is not less than 150 mm^2/s but not more than 1,200 mm^2/s.

6. The lubricating oil composition as claimed in claim **1**, wherein the kinematic viscosity of the (co)polymer (i) comprising 1-decene at 100° C. is not more than 11 mm^2/s.

7. A gear for a wind power generator, which is filled with the lubricating oil composition as claimed in claim **1**.

* * * * *