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(54) **OIL COMPOSITION COMPRISING A VISCOSITY INDEX IMPROVER**

ÖLZUSAMMENSETZUNG MIT EINEM VISKOSITÄTSINDEXVERBESSERER

COMPOSITION D'HUILE COMPRENANT UN AMÉLIORATEUR D'INDICE DE VISCOSITÉ

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**EP 2 899 255 B1**

## Description

**[0001]** The present invention relates to an oil composition comprising a base oil and a viscosity index improver, the viscosity index improver consisting of a hydrogenated product of a block copolymer containing a constitutional unit derived from farnesene, and a process for producing the oil composition.

**[0002]** Hydrogenated products of block copolymers constituted of a polymer block containing a constitutional unit derived from an aromatic vinyl compound and a polymer block containing a constitutional unit derived from a conjugated diene exhibit properties similar to those of vulcanized rubbers without further subjecting the hydrogenated products to vulcanization, i.e., are excellent in damping property, flexibility, rubber elasticity and weather resistance, and therefore have been extensively used in the applications such as sundries, parts for automobiles, various industrial parts, etc.

**[0003]** The hydrogenated products of block copolymers are produced, for example, by subjecting a block copolymer obtained by sequentially polymerizing an aromatic vinyl compound and a conjugated diene such as isoprene and butadiene to hydrogenation (for example, refer to PTL1 and PTL2).

**[0004]** In addition, PTL3 describes that the hydrogenated products of block copolymers constituted of a polymer block containing a constitutional unit derived from an aromatic vinyl compound and a polymer block containing a constitutional unit derived from a conjugated diene are used as a viscosity index improver for lubricating oils.

**[0005]** Meanwhile, PTL4 and PTL5 describe a polymer of  $\beta$ -farnesene, but fail to make a sufficient study on practical properties thereof.

**[0006]** Further, EP 2 444 475 A1 describes a lubricant additive, and US 2011/0207642 A1 describes a lubricating oil composition.

## Citation List

**[0007]**

PTL1: JP 2777239B

PTL2: JP 2010-090267A

PTL3: JP 2009-532513A

PTL4: JP 2012-502135A

PTL5: JP 2012-502136A

**[0008]** The viscosity index improver described in PTL3 is excellent in an effect of improving a viscosity index and a high-temperature high-shear viscosity of lubricating oils. However, it has been still demanded to develop different kinds of viscosity index improvers that are more excellent in the above effect.

**[0009]** The present invention aims at providing an oil composition, comprising a base oil and a viscosity index improver, the oil composition being enhanced in viscosity index and high-temperature high-shear viscosity, and the viscosity index improver being excellent in an effect of improving a viscosity index and a high-temperature high-shear viscosity of oils, and a process for producing the oil composition.

**[0010]** The present invention relates to the following aspects [1] and [2].

[1] An oil composition comprising a base oil and a viscosity index improver, the viscosity index improver consisting of a hydrogenated product of a block copolymer comprising a polymer block (A) containing a constitutional unit (a) derived from an aromatic vinyl compound as a main component and a polymer block (B) containing a constitutional unit (b) derived from a conjugated diene as a main component, a content of a constitutional unit (b1) derived from farnesene in a whole amount of the constitutional unit (b) derived from the conjugated diene being from 40 to 100% by mass, and a content of a constitutional unit (b2) derived from a conjugated diene other than farnesene in a whole amount of the constitutional unit (b) derived from the conjugated diene being from 0 to 60% by mass, and 50 mol% or more of carbon-carbon double bonds in the constitutional unit (b) derived from the conjugated diene being hydrogenated, wherein a mass ratio of a whole amount of the constitutional unit (a) derived from the aromatic vinyl compound and a whole amount of the constitutional unit (b) derived from the conjugated diene [(a)/(b)] is 10/90 to 50/50, wherein the content of the viscosity index improver in the oil composition is from 0.1 to 10% by mass.

[2] A process for producing the oil composition according to the above aspect [1], comprising a process for producing the viscosity index improver comprising:

a polymerization step of obtaining the block copolymer comprising the polymer block (A) containing the constitutional unit (a) derived from the aromatic vinyl compound as a main component and the polymer block (B)

containing the constitutional unit (b) derived from the conjugated diene as a main component by anionic polymerization; and  
 a hydrogenation step of hydrogenating 50 mol% or more of carbon-carbon double bonds in the constitutional unit (b) derived from the conjugated diene; and mixing a base oil with the viscosity index improver.

**[0011]** According to the present invention, it is possible to provide an oil composition that is enhanced in viscosity index and high-temperature high-shear viscosity, the viscosity index improver thereof being excellent in an effect of improving a viscosity index and a high-temperature high-shear viscosity of oils, and a process for producing the oil composition.

[Viscosity Index Improver]

**[0012]** The viscosity index improver consists of a hydrogenated product of a block copolymer comprising a polymer block (A) containing a constitutional unit (a) derived from an aromatic vinyl compound as a main component and a polymer block (B) containing a constitutional unit (b) derived from a conjugated diene as a main component (the copolymer may also be hereinafter referred to merely as a "copolymer", and the hydrogenated product of the copolymer may also be hereinafter referred to merely as a "hydrogenated copolymer"), in which a content of a constitutional unit (b1) derived from farnesene in a whole amount of the constitutional unit (b) derived from the conjugated diene is from 40 to 100% by mass, and a content of a constitutional unit (b2) derived from a conjugated diene other than farnesene in a whole amount of the constitutional unit (b) derived from the conjugated diene is from 0 to 60% by mass, and 50 mol% or more of carbon-carbon double bonds in the constitutional unit (b) derived from the conjugated diene are hydrogenated.

**[0013]** The viscosity index improver is constituted of a hydrogenated product of a block copolymer including a polymer block (A) containing the constitutional unit (a) derived from the aromatic vinyl compound as a main component and a polymer block (B) containing the constitutional unit (b) derived from the conjugated diene as a main component (hereinafter also referred to merely as a "hydrogenated block copolymer").

<Constitutional Unit (a) Derived From Aromatic Vinyl Compound>

**[0014]** The aforementioned copolymer contains the constitutional unit (a) derived from the aromatic vinyl compound. Examples of the aromatic vinyl compound include styrene,  $\alpha$ -methylstyrene, 2-methylstyrene, 3-methylstyrene, 4-methylstyrene, 4-propylstyrene, 4-t-butylstyrene, 4-cyclohexylstyrene, 4-dodecylstyrene, 2,4-dimethylstyrene, 2,4-diisopropylstyrene, 2,4,6-trimethylstyrene, 2-ethyl-4-benzylstyrene, 4-(phenylbutyl)styrene, 1-vinylnaphthalene, 2-vinylnaphthalene, vinylanthracene, N,N-diethyl-4-aminoethylstyrene, vinylpyridine, 4-methoxystyrene, monochlorostyrene, dichlorostyrene and divinylbenzene. These aromatic vinyl compounds may be used alone or in combination of any two or more thereof. Of these aromatic vinyl compounds, from the viewpoint of improving a viscosity index and a high-temperature high-shear viscosity of oils, preferred are styrene,  $\alpha$ -methylstyrene and 4-methylstyrene, and more preferred is styrene.

**[0015]** Meanwhile, in the present invention, compounds containing both of an aromatic group and a conjugated diene bond in a molecule thereof should be excluded from the conjugated diene as the component from which the constitutional unit (b) is derived, and should be included in the aromatic vinyl compound as the component from which the constitutional unit (a) is derived. However, the content of the compounds containing both of an aromatic group and a conjugated diene bond in a molecule thereof in the constitutional unit (a) is preferably not more than 10% by mass, more preferably not more than 5% by mass, and still more preferably not more than 1% by mass.

<Constitutional Unit (b) Derived From Conjugated Diene>

**[0016]** The aforementioned copolymer includes the constitutional unit (b) derived from the conjugated diene. The copolymer contains a constitutional unit (b1) derived from farnesene as the constitutional unit (b), and may further contain a constitutional unit (b2) derived from a conjugated diene other than farnesene as the constitutional unit (b).

**[0017]** The content of the constitutional unit (b1) derived from farnesene in a whole amount of the constitutional unit (b) derived from the conjugated diene is from 40 to 100% by mass, preferably from 50 to 100% by mass, still more preferably from 55 to 100% by mass, even still more preferably from 60 to 100% by mass, further even still more preferably from 90 to 100% by mass, and further even still more preferably from 95 to 100% by mass.

**[0018]** Also, the content of the constitutional unit (b2) derived from the conjugated diene other than farnesene in a whole amount of the constitutional unit (b) derived from the conjugated diene is from 0 to 60% by mass, preferably from 0 to 50% by mass, still more preferably from 0 to 10% by mass, and even still more preferably from 0 to 5% by mass.

**[0019]** The farnesene may be industrially produced from sugars as a raw material which are extracted from plants such as canes by using microorganisms. Thus, the viscosity index improver of the present invention can be produced

with a less burden on environments by using the farnesene as a raw material.

(Constitutional Unit (b1) Derived From Farnesene)

**[0020]** The farnesene as the component from which the constitutional unit (b1) is derived may be either  $\alpha$ -farnesene or  $\beta$ -farnesene represented by the following formula (I), or  $\alpha$ -farnesene and  $\beta$ -farnesene may be used in combination with each other to form the constitutional unit (b1).

**[0021]** The content of a constitutional unit derived from  $\beta$ -farnesene in the constitutional unit (b1) is preferably not less than 60% by mass, more preferably not less than 80% by mass, still more preferably not less than 90% by mass, even still more preferably not less than 99% by mass, and further even still more preferably 100% by mass, from the viewpoints of facilitating production of the copolymer and improving a viscosity index and a high-temperature high-shear viscosity of oils.



(Constitutional Unit (b2) Derived From Conjugated Diene Other Than Farnesene)

**[0022]** The aforementioned copolymer may contain a constitutional unit (b2) derived from a conjugated diene other than farnesene. Examples of the conjugated diene other than farnesene as the component from which the constitutional unit (b2) is derived include butadiene, isoprene, 2,3-dimethylbutadiene, 2-phenylbutadiene, 1,3-pentadiene, 2-methyl-1,3-pentadiene, 1,3-hexadiene, 1,3-octadiene, 1,3-cyclohexadiene, 2-methyl-1,3-octadiene, 1,3,7-octatriene, myrcene and chloroprene. These conjugated dienes other than farnesene may be used alone or in combination of any two or more thereof. Of these conjugated dienes, preferred is at least one compound selected from the group consisting of butadiene, isoprene and myrcene, and more preferred is at least one compound selected from the group consisting of butadiene and isoprene.

<Other Constitutional Unit (c)>

**[0023]** The aforementioned copolymer may also contain the other constitutional unit (c) in addition to the constitutional units (a), (b1) and (b2).

**[0024]** Examples of compounds as a component from which the other constitutional unit (c) is derived include unsaturated hydrocarbon compounds such as propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, 1-hexene, 1-octene, 1-decene, 1-undecene, 1-dodecene, 1-tridecene, 1-tetradecene, 1-pentadecene, 1-hexadecene, 1-heptadecene, 1-octadecene, 1-nonadecene and 1-eicosene; functional group-containing unsaturated compounds such as acrylic acid, methacrylic acid, methyl acrylate, methyl methacrylate, acrylonitrile, methacrylonitrile, maleic acid, fumaric acid, crotonic acid, itaconic acid, 2-acryloylethanesulfonic acid, 2-methacryloylethanesulfonic acid, 2-acrylamide-2-methylpropanesulfonic acid, 2-methacrylamide-2-methylpropanesulfonic acid, vinylsulfonic acid, vinyl acetate and methyl vinyl ether; and the like. These other constitutional units (c) may be used alone or in combination of any two or more thereof.

<Copolymer>

**[0025]** The copolymer contains the aforementioned constitutional unit (a) derived from the aromatic vinyl compound and the aforementioned constitutional unit (b) derived from the conjugated diene. Meanwhile, the copolymer may further contain the aforementioned constitutional unit (c).

**[0026]** The total content of the constitutional unit (a) and the constitutional unit (b) in the copolymer is preferably not less than 60% by mass, more preferably not less than 80% by mass, still more preferably not less than 90% by mass, and even still more preferably not less than 99% by mass, from the viewpoint of improving a viscosity index and a high-temperature high-shear viscosity of oils.

**[0027]** In addition, from the same viewpoint as described above, the content of the constitutional unit (c) in a whole amount of the constitutional units constituting the copolymer is preferably not more than 40% by mass, more preferably not more than 20% by mass, still more preferably not more than 10% by mass, and even still more preferably not more than 1% by mass.

**[0028]** The content of the constitutional unit (a) in the copolymer is preferably from 1 to 99% by mass, more preferably from 2 to 90% by mass, still more preferably from 3 to 85% by mass, even still more preferably from 5 to 80% by mass,

further even still more preferably from 10 to 50% by mass, and further even still more preferably from 15 to 45% by mass, from the viewpoint of improving a viscosity index and a high-temperature high-shear viscosity of oils.

**[0029]** The mass ratio of a whole amount of the constitutional unit (a) to a whole amount of the constitutional unit (b) [(a)/(b)] is 10/90 to 50/50, preferably from 15/85 to 45/55, from the viewpoint of improving a viscosity index and a high-temperature high-shear viscosity of oils.

(Block Copolymer)

**[0030]** The copolymer is in the form of a block copolymer including a polymer block (A) containing the constitutional unit (a) derived from the aromatic vinyl compound as a main component and a polymer block (B) containing the constitutional unit (b) derived from the conjugated diene as a main component.

**[0031]** The expression "as a main component" as used herein means that the content of the constitutional unit (a) derived from the aromatic vinyl compound on the basis of a total mass of the polymer block (A) is not less than 50% by mass, preferably not less than 70% by mass, and more preferably not less than 90% by mass, and also means that the content of the constitutional unit (b) derived from the conjugated diene on the basis of a total mass of the polymer block (B) is not less than 50% by mass, preferably not less than 65% by mass, and more preferably not less than 80% by mass.

**[0032]** The polymer block (B) contains from 40 to 100% by mass of the constitutional unit (b1) derived from farnesene and from 60 to 0% by mass of the constitutional unit (b2) derived from the conjugated diene other than farnesene. The preferred specific examples and content ratios of the constitutional units (b1) and (b2) are the same as described above.

**[0033]** Furthermore, the polymer block (b) may also contain a small amount of a constitutional unit derived from any other copolymerizable monomer unless the object and effects of the present invention are adversely affected. Examples of the other copolymerizable monomer include anion-polymerizable copolymerizable monomers, e.g., aromatic vinyl compounds such as styrene,  $\alpha$ -methylstyrene, o-methylstyrene, m-methylstyrene, p-methylstyrene, 1,3-dimethylstyrene, diphenylethylene, 1-vinylnaphthalene, 4-propylstyrene, 4-cyclohexylstyrene, 4-dodecylstyrene, 2-ethyl-4-benzylstyrene and 4-(phenylbutyl)styrene. These other copolymerizable monomers may be used alone or in combination of any two or more thereof. In the case where the polymer block contains the constitutional unit derived from the other copolymerizable monomer, the constitutional unit may be bonded thereto either in a random form or in a tapered form.

**[0034]** In the case where the polymer block (b) contains the constitutional unit derived from the other copolymerizable monomer, the content of the constitutional unit derived from the other copolymerizable monomer in the polymer block is preferably not more than 35% by mass, more preferably not more than 10% by mass, and still more preferably not more than 5% by mass.

**[0035]** The bonding configuration of each of the polymer block (A) and the polymer block (B) is not particularly limited, and may be either a linear configuration, a branched configuration, a radial configuration or a combination of any two or more of these configurations. Of these bonding configurations, when representing the polymer block (A) and the polymer block (B) by A and B, respectively, from the viewpoint of improving a viscosity index and a high-temperature high-shear viscosity of oils, preferred are the bonding configuration in which the respective polymer blocks are linearly bonded to each other, and which is represented by  $(A-B)_l$ ,  $A-(B-A)_m$  or  $B-(A-B)_n$  wherein l, m and n are each independently an integer of 1 or more, and the bonding configuration in which the respective polymer blocks are radially bonded to each other, and which is represented by  $(A-B)_pX$  or  $(B-A)_qX$  wherein p and q are each independently an integer of 3 or more, and X is a residue of a coupling reagent.

**[0036]** Of these copolymers having the above bonding configurations, from the viewpoints of improving a viscosity index and a high-temperature high-shear viscosity of oils, preferred are a di-block copolymer having the bonding configuration represented by A-B or a tri-block copolymer having the bonding configuration represented by A-B-A.

**[0037]** In addition, in the case where the block copolymer contains two or more polymer blocks (A) or two or more polymer blocks (B), the two or more polymer blocks (A) or (B) may be respectively polymer blocks containing the same constitutional unit or polymer blocks containing different kinds of constitutional units from each other. For example, in the two polymer blocks (A) present in the tri-block copolymer represented by [A-B-A], the respective aromatic vinyl compounds contained therein may be constituted of either the same kind of compound or different kinds of compounds.

**[0038]** The peak top molecular weight of the polymer block (A) is preferably not less than 10,000 and not more than 60,000, and more preferably not less than 15,000 and not more than 50,000, from the viewpoints of attaining a good effect of improving a viscosity index of oils and facilitating production of an oil composition. When the peak top molecular weight of the polymer block (A) is less than 10,000, the effect of improving a viscosity index of oils tends to be deteriorated. On the other hand, when the peak top molecular weight of the polymer block (A) is more than 60,000, the resulting block copolymer tends to be deteriorated in solubility in oils, and therefore there is a possibility that no effect of improving a viscosity index of oils is attained. Meanwhile, when the block copolymer contains the two or more polymer blocks (A), the peak top molecular weight of at least one of the polymer blocks (A) preferably falls within the above-specified range.

**[0039]** Also, it is more preferred that the aforementioned block polymer contains at least one polymer block selected from the group consisting of the polymer block (A) and a polymer block (A') each containing the constitutional unit (a)

derived from the aromatic vinyl compound as a main component, and the polymer block (B) containing the constitutional unit (b) derived from the conjugated diene as a main component, in which the bonding configuration of these polymer blocks in the block copolymer is A-B or A-B-A', and the peak top molecular weight of the polymer block (A') is not less than 1,000 and less than 10,000. The peak top molecular weight of the polymer block (A') is still more preferably not less than 1,000 and not more than 8,000. When the peak top molecular weight of the polymer block (A') is less than 10,000, the obtained hydrogenated block copolymer can be prevented from forming a gel-like network in an oil composition, and the oil composition obtained therefrom can be prevented from suffering from increase in kinetic viscosity thereof as measured at a temperature of each of 40°C and 100°C. As a result, the use of the oil composition as a lubricating oil is facilitated.

**[0040]** The peak top molecular weight of each of the polymer blocks (A) and (A') can be determined by sampling a part of a reaction solution during synthesis of the block copolymer and measuring a peak top molecular weight of the respective polymer blocks of the block copolymer in the sampled reaction solution by the method described below in Examples.

<Hydrogenated Copolymer>

**[0041]** The hydrogenated copolymer constituting the viscosity index improver is produced by hydrogenating the aforementioned copolymer in which 50 mol% or more of carbon-carbon double bonds in the constitutional unit (b) derived from the conjugated diene are hydrogenated, namely the hydrogenated copolymer has a hydrogenation rate of 50 mol% or more.

**[0042]** The hydrogenation rate as used herein is the value represented by the following formula:

$$\text{Hydrogenation rate} = (1 - M2/M1) \times 100 \text{ (mol\%)}$$

wherein M1 is a molar number of double bonds derived from the conjugated diene which are contained per 1 mole of the copolymer; and M2 is a molar number of double bonds derived from the conjugated diene which are contained per 1 mole of the hydrogenated copolymer.

**[0043]** The hydrogenation rate is preferably not less than 60 mol%, and more preferably not less than 70 mol%, from the viewpoint of improving a heat resistance and a shear stability of oils as well as a viscosity index and a high-temperature high-shear viscosity of oils. Meanwhile, the hydrogenation rate may be determined by the method described below in Examples.

**[0044]** The peak top molecular weight (Mp) of the hydrogenated copolymer is preferably from 4,000 to 1,500,000, more preferably from 9,000 to 1,200,000, still more preferably from 20,000 to 1,100,000, and most preferably from 100,000 to 800,000, from the viewpoints of facilitating production of an oil composition and improving a viscosity index of oils.

**[0045]** Meanwhile, the peak top molecular weight (Mp) as used in the present specification means the value measured by the method described below in Examples.

**[0046]** The molecular weight distribution (Mw/Mn) of the hydrogenated copolymer is preferably from 1 to 4, more preferably from 1 to 3, and still more preferably from 1 to 2. When the molecular weight distribution of the hydrogenated copolymer falls within the above-specified range, the resulting hydrogenated block copolymer can exhibit a less variation in viscosity thereof.

[Process for Producing Hydrogenated Block Copolymer]

**[0047]** The hydrogenated block copolymer may be suitably produced by a process including a polymerization step of obtaining a block copolymer including a polymer block (A) containing the constitutional unit (a) derived from the aromatic vinyl compound as a main component and a polymer block (B) containing the constitutional unit (b) derived from the conjugated diene as a main component by anionic polymerization; and a hydrogenation step of hydrogenating 50 mol% or more of carbon-carbon double bonds in the constitutional unit (b) derived from the conjugated diene.

<Polymerization Step>

**[0048]** The block copolymer may be produced by any suitable polymerization methods such as a solution polymerization method or the methods described in JP 2012-502135A and JP 2012-502136A, in particular, is preferably produced by the solution polymerization method. In the present invention, anionic polymerization method is used. In the anionic polymerization method, the aromatic vinyl compound, and farnesene and/or the conjugated diene other than farnesene

are preferably sequentially added in the presence of a solvent and an anionic polymerization initiator as well as, if required, a Lewis base as an optional component, thereby obtaining the block copolymer.

[0049] Examples of the anionic polymerization initiator include alkali metals such as lithium, sodium and potassium; alkali earth metals such as beryllium, magnesium, calcium, strontium and barium; lanthanoid-based rare earth metals such as lanthanum and neodymium; and compounds containing the above alkali metals, alkali earth metals or lanthanoid-based rare earth metals. Of these anionic polymerization initiators, preferred are the alkali metals, the compounds containing the alkali metals, and organic alkali metal compounds.

[0050] Specific examples of the organic alkali metal compounds include organic lithium compounds such as methyl lithium, ethyl lithium, n-butyl lithium, sec-butyl lithium, t-butyl lithium, hexyl lithium, phenyl lithium, stilbene lithium, dilithiomethane, dilithionaphthalene, 1,4-dilithiobutane, 1,4-dilithio-2-ethyl cyclohexane and 1,3,5-trilithiobenzene; and sodium naphthalene and potassium naphthalene, etc. Among these organic alkali metal compounds, preferred are organic lithium compounds; more preferred are n-butyl lithium and sec-butyl lithium; and still more preferred is sec-butyl lithium. Meanwhile, the organic alkali metal compound may be reacted with a secondary amine such as diisopropylamine, dibutylamine, dihexylamine and dibenzylamine to use the compound in the form of an organic alkali metal amide.

[0051] The amount of the organic alkali metal compound used for the polymerization may vary depending upon a molecular weight of the resulting block copolymer, and is usually in the range of from 0.01 to 3% by mass on the basis of a total amount of the aromatic vinyl compound, and farnesene and/or the conjugated diene other than farnesene.

[0052] The solvent used in the polymerization step is not particularly limited unless it adversely affects the anionic polymerization reaction. Examples of the solvent used in the polymerization step include saturated aliphatic hydrocarbons such as n-pentane, isopentane, n-hexane, n-heptane and isooctane; saturated alicyclic hydrocarbons such as cyclopentane, cyclohexane and methyl cyclopentane; and aromatic hydrocarbons such as benzene, toluene and xylene. These solvents may be used alone or in combination of any two or more thereof. The amount of the solvent used in the polymerization step is not particularly limited.

[0053] The Lewis base acts for controlling a microstructure of each of the constitutional unit (b1) derived from farnesene and the constitutional unit (b2) derived from the conjugated diene other than farnesene. Examples of the Lewis base include ether compounds such as dibutyl ether, diethyl ether, tetrahydrofuran, dioxane and ethylene glycol diethyl ether; pyridine; tertiary amines such as N,N,N',N'-tetramethyl ethylenediamine and trimethylamine; alkali metal alkoxides such as potassium-t-butoxide; and phosphine compounds. The amount of the Lewis base, if used, is usually preferably in the range of from 0.01 to 1,000 mol equivalent on the basis of 1 mol of the anionic polymerization initiator.

[0054] The temperature used in the above polymerization reaction is usually from -80 to 150°C, preferably from 0 to 100°C and more preferably from 10 to 90°C. The polymerization reaction may be carried out by either a batch method or a continuous method. The respective monomers, i.e., the aromatic vinyl compound, and farnesene and/or the conjugated diene other than farnesene may be supplied to the polymerization reaction solution in a continuous or intermittent manner such that the abundance ratio of each of the aromatic vinyl compound, and farnesene and/or the conjugated diene other than farnesene in the polymerization reaction system falls within a specific range, or the aromatic vinyl compound, and farnesene and/or the conjugated diene other than farnesene may be sequentially polymerized such that the ratio of the respective monomers in the polymerization reaction solution is controlled to a specific range, whereby it is possible to produce the block copolymer.

[0055] The polymerization reaction may be stopped by adding an alcohol such as methanol and isopropanol as a terminating reagent to the reaction system. The resulting polymerization reaction solution may be poured into a poor solvent such as methanol to precipitate the block copolymer. Alternatively, there may be used the method in which the polymerization reaction solution is rinsed with water, and then the obtained reaction product is separated from the water layer and dried to isolate the block copolymer therefrom.

{Modified Copolymer}

[0056] In the polymerization step, the block copolymer can be obtained in the form of an unmodified block copolymer as described above. However, the block copolymer may also be in the form of a modified block copolymer obtained by the following method.

[0057] The above block copolymer may be modified prior to the below-mentioned hydrogenation step. Examples of a functional group that may be introduced into the block copolymer include an amino group, an alkoxy silyl group, a hydroxyl group, an epoxy group, a carboxyl group, a carbonyl group, a mercapto group, an isocyanate group and an acid anhydride group.

[0058] As the method of modifying the block copolymer, there may be used, for example, the method in which before adding the terminating reagent, a coupling reagent such as tin tetrachloride, tetrachlorosilane, dimethyldichlorosilane, dimethyldiethoxysilane, tetramethoxysilane, tetraethoxysilane, 3-aminopropyltriethoxysilane, tetraglycidyl-1,3-bisaminomethylcyclohexane and 2,4-tolylene diisocyanate which are capable of reacting with an active end of the polymer chain, a chain end-modifying reagent such as 4,4'-bis(diethylamino)benzophenone and N-vinylpyrrolidone, or the other

modifying reagent as described in JP 2011-132298A is added to the polymerization reaction system. Furthermore, the isolated copolymer may be grafted with maleic anhydride or the like.

**[0059]** The site of the block polymer into which the functional group is introduced may be either a chain end or a side chain of the polymer. In addition, these functional groups may be used alone or in combination of any two or more thereof. The modifying reagent is usually preferably used in an amount of from 0.01 to 10 mol equivalent on the basis of the anionic polymerization initiator used in the polymerization step.

#### <Hydrogenation Step>

**[0060]** When the block copolymer obtained by the above method is subjected to hydrogenation step, it is possible to produce the hydrogenated block copolymer. As the hydrogenation method, there may be used conventionally known methods. For example, a solution prepared by dissolving the block copolymer in a solvent that has no adverse influence on the hydrogenation reaction is subjected to hydrogenation reaction in the presence of a hydrogenation catalyst. Examples of the hydrogenation catalyst include Ziegler-based catalysts; metal-supported catalysts obtained by supporting a metal such as nickel, platinum, palladium, ruthenium and rhodium on a carrier such as carbon, silica and diatomaceous earth; and organic metal complexes containing a metal such as cobalt, nickel, palladium, rhodium and ruthenium. In the hydrogenation step, the hydrogenation reaction may be carried out by adding the hydrogenation catalyst to the polymerization reaction solution containing the block copolymer obtained by the above method for producing the block copolymer. In the present invention, there is preferably used palladium carbon formed by supporting palladium on carbon.

**[0061]** In the hydrogenation reaction, a hydrogen pressure used therein is preferably from 0.1 to 20 MPa, the reaction temperature is preferably from 100 to 200°C, and the reaction time is preferably from 1 to 20 h.

**[0062]** The hydrogenation rate of carbon-carbon double bonds in the polymer block (B) contained in the block copolymer is preferably not less than 50 mol%, more preferably not less than 60 mol%, and still more preferably not less than 70 mol%, from the viewpoint of improving a heat resistance and a shear stability of oils as well as a viscosity index and a high-temperature high-shear viscosity of oils. Meanwhile, the hydrogenation rate may be calculated by the method described below in Examples.

#### [Oil Composition]

**[0063]** The oil composition of the present invention includes a base oil and the aforementioned viscosity index improver.

**[0064]** The oil composition may be suitably used as an engine oil, an automatic transmission oil, a gear lube oil and a hydraulic pressure oil.

**[0065]** The content of the viscosity index improver in the oil composition is from 0.1 to 10% by mass, preferably from 0.1 to 7% by mass, still more preferably from 0.2 to 5% by mass, and even still more preferably from 0.5 to 5% by mass.

#### <Base Oil>

**[0066]** Examples of the base oil include fuel oils such as intermediate fraction fuels, synthetic or natural lubricating oils, unrefined oils and industrial oils. The base oil may be at least one compound selected from the group consisting of a paraffin-based compound, a naphthene-based compound and an aromatic compound. In addition, the base oil may also be at least one oil selected from the group consisting of a natural oil and an artificially synthesized oil.

#### <Other Additives>

**[0067]** The oil composition of the present invention may also contain other additives such as a rust preventive, an antioxidant, a surfactant, a pour point depressant, a cleaning dispersant, a metal deactivator, a defoamer, a friction controller, an extreme pressure reagent and one or more additional viscosity index improvers.

#### <Method of Producing Oil Composition>

**[0068]** The oil composition of the present invention may be produced by methods conventionally known in the art.

**[0069]** For example, the oil composition of the present invention may be produced by mixing the aforementioned base oil with the aforementioned viscosity index improver. The mixing of these components may be carried out using conventionally known mixers.

**[0070]** The mixing is preferably carried out while heating. The heating temperature is preferably from 80 to 180°C.

**[0071]** In the case where the oil composition of the present invention is applied to an engine oil, an automatic transmission oil, a gear lube oil, a hydraulic pressure oil or the like, the kinetic viscosity thereof as measured at 40°C is preferably in the range of from 40 to 120 mm<sup>2</sup>/sec, and more preferably from 50 to 70 mm<sup>2</sup>/sec. In addition, from the



same viewpoint, the kinetic viscosity of the oil composition of the present invention as measured at 100°C is preferably in the range of from 6 to 25 mm<sup>2</sup>/sec, and more preferably from 8 to 15 mm<sup>2</sup>/sec. When the kinetic viscosity of the oil composition lies within the above-specified range, the oil composition is capable of reducing a fuel consumption in vehicles when used in the above applications.

## Examples

**[0072]** The present invention will be described in more detail below by referring to the following examples. It should be noted, however, that the following examples are only illustrative and not intended to limit the invention thereto. Meanwhile,  $\beta$ -farnesene (purity: 97.6% by mass; available from Amyris Biotechnologies Inc.) was purified using a 3 Å molecular sieve and distilled under a nitrogen atmosphere to remove hydrocarbon-based impurities such as zingiberene, bisabolene, farnesene epoxide, farnesol isomers, E,E-farnesol, squalene, ergosterol and several kinds of dimers of farnesene therefrom, and the thus purified  $\beta$ -farnesene was used in the following polymerization.

### (1) Measurement of Molecular Weight Distribution (Mw/Mn) and Peak Top Molecular Weight (Mp)

**[0073]** The weight-average molecular weight (Mw) and the molecular weight distribution (Mw/Mn) of each of the hydrogenated block copolymers produced in the respective Examples and Comparative Examples were measured by GPC (gel permeation chromatography) in terms of a molecular weight of polystyrene as a reference standard substance. Also, the peak top molecular weight (Mp) of the hydrogenated block copolymer was determined from a position of a peak top of the molecular weight distribution (Mw/Mn). The measuring devices and conditions used in the above measurement are as follows.

- Apparatus: GPC device "GPC8020" available from Tosoh Corp.
- Separating column: "TSKgelG4000HXL" available from Tosoh Corp.
- Detector: "RI-8020" available from Tosoh Corp.
- Eluent: Tetrahydrofuran
- Eluent flow rate: 1.0 mL/min
- Sample concentration: 5 mg/10 mL
- Column temperature: 40°C

### (2) Method of Measuring Hydrogenation Rate

**[0074]** In the respective Examples and Comparative Examples, the block copolymer and the hydrogenated block copolymer obtained after hydrogenating the block copolymer were respectively dissolved in a deuterated chloroform solvent, and each of the resulting solutions was subjected to <sup>1</sup>H-NMR measurement at 50°C using "Lambda-500" available from JOEL Ltd. The hydrogenation rate of the polymer block (B) in the hydrogenated block copolymer was calculated from the peak of protons contained in carbon-carbon double bonds observed in the range of from 4.5 to 6.0 ppm in the resulting spectrum, according to the following formula.

**[0075]** Hydrogenation Rate = {1 - (number of moles of carbon-carbon double bonds contained per 1 mole of hydrogenated block copolymer)/(number of moles of carbon-carbon double bonds contained per 1 mole of block copolymer)} x 100 (mol%)

### (3) Kinetic Viscosity

**[0076]** The kinetic viscosity was measured at a temperature of each of 40°C and 100°C according to JIS K2283.

### (4) Viscosity Index

**[0077]** The viscosity index was measured according to JIS K2283.

### (5) High-Temperature High-Shear Viscosity

**[0078]** The high-temperature high-shear viscosity was measured at a temperature of 150°C according to ASTM D4683.

### [Example 1]

**[0079]** A pressure reaction vessel previously purged with nitrogen and then dried was charged with 62.4 kg of cy-

clohexane as a solvent and 51.0 g of sec-butyl lithium (in the form of a 10.5% by mass cyclohexane solution) as an anionic polymerization initiator. The contents of the reaction vessel were heated to 50°C, and then 2.34 kg of styrene (1) was added thereto, followed by polymerizing the contents of the reaction vessel for 1 h. Successively, 10.9 kg of  $\beta$ -farnesene was added to the reaction vessel, followed by polymerizing the contents of the reaction vessel for 2 h. Furthermore, 2.34 kg of styrene (2) was added to the reaction vessel, followed by polymerizing the contents of the reaction vessel for 1 h, thereby obtaining a reaction solution containing a polystyrene-poly( $\beta$ -farnesene)-polystyrene triblock copolymer. Added into the reaction solution was palladium carbon (amount of palladium supported: 5% by mass) as a hydrogenation catalyst which was used in an amount of 5% by mass on the basis of the block copolymer, and the block copolymer was subjected to hydrogenation reaction under a hydrogen pressure of 2 MPa at a temperature of 150°C for 10 h. The obtained reaction mixture was allowed to stand for cooling and pressure releasing, and then subjected to filtration to remove the palladium carbon therefrom. The resulting filtrate was concentrated and further vacuum-dried, thereby obtaining a hydrogenated product of the polystyrene-poly( $\beta$ -farnesene)-polystyrene triblock copolymer (hereinafter referred to as a "hydrogenated block copolymer (A1)"). The formulations of various raw materials used above and the results of measurement of various properties of the thus obtained hydrogenated block copolymer (A1) are shown in Table 1 below.

**[0080]** The resulting hydrogenated block copolymer (A1) and a paraffin-based oil "DIANA FRESIA S-32" available from Idemitsu Kosan Co., Ltd., were used as a viscosity index improver and a base oil, respectively, and the hydrogenated block copolymer (A1) and the base oil were formulated as shown in Table 2. These components were mixed in a pressure reaction vessel purged with nitrogen at a temperature of 120°C at a rotating speed of 350 rpm for 6 h, thereby obtaining an oil composition. The thus obtained oil composition was subjected to the above evaluation. The results are shown in Table 2.

#### [Example 2]

**[0081]** A pressure reaction vessel previously purged with nitrogen and then dried was charged with 62.4 kg of cyclohexane as a solvent and 40.6 g of sec-butyl lithium (in the form of a 10.5% by mass cyclohexane solution) as an anionic polymerization initiator. The contents of the reaction vessel were heated to 50°C, and then 13.0 kg of  $\beta$ -farnesene was added thereto, followed by polymerizing the contents of the reaction vessel for 2 h. Successively, 2.59 kg of styrene was added to the reaction vessel, followed by polymerizing the contents of the reaction vessel for 1 h, thereby obtaining a reaction solution containing a poly( $\beta$ -farnesene)-polystyrene diblock copolymer. Added into the reaction solution was palladium carbon (amount of palladium supported: 5% by mass) as a hydrogenation catalyst which was used in an amount of 5% by mass on the basis of the block copolymer, and the block copolymer was subjected to hydrogenation reaction under a hydrogen pressure of 2 MPa at a temperature of 150°C for 10 h. The obtained reaction mixture was allowed to stand for cooling and pressure releasing, and then subjected to filtration to remove the palladium carbon therefrom. The resulting filtrate was concentrated and further vacuum-dried, thereby obtaining a hydrogenated product of the poly( $\beta$ -farnesene)-polystyrene diblock copolymer (hereinafter referred to as a "hydrogenated block copolymer (A2)"). The formulations of various raw materials used above and the results of measurement of various properties of the thus obtained hydrogenated block copolymer (A2) are shown in Table 1 below.

**[0082]** The subsequent procedure was conducted in the same manner as in Example 1 except that the resulting hydrogenated block copolymer (A2) and the base oil were formulated as shown in Table 2, thereby obtaining an oil composition. The thus obtained oil composition was subjected to the above evaluation. The results are shown in Table 2.

#### [Example 3]

**[0083]** The same procedure as in Example 2 was repeated except that the respective components were formulated as shown in Table 1, thereby obtaining a hydrogenated block copolymer (A3) and an oil composition and subjecting the resulting products to the above evaluation. The results are shown in Tables 1 and 2.

#### [Example 4]

**[0084]** The same procedure as in Example 1 was repeated except that the respective components were formulated as shown in Table 1, thereby obtaining a hydrogenated block copolymer (A4) and an oil composition and subjecting the resulting products to the above evaluation. The results are shown in Tables 1 and 2.

#### [Example 5]

**[0085]** A pressure reaction vessel previously purged with nitrogen and then dried was charged with 62.4 kg of cyclohexane as a solvent and 118.4 g of sec-butyl lithium (in the form of a 10.5% by mass cyclohexane solution) as an

anionic polymerization initiator. The contents of the reaction vessel were heated to 50°C, and then a mixture of 5.43 kg of  $\beta$ -farnesene and 4.32 kg of butadiene was added thereto, followed by polymerizing the contents of the reaction vessel for 2 h. Successively, 5.85 kg of styrene was added to the reaction vessel, followed by polymerizing the contents of the reaction vessel for 1 h, thereby obtaining a reaction solution containing a poly( $\beta$ -farnesene/butadiene)-polystyrene diblock copolymer. Added into the reaction solution was palladium carbon (amount of palladium supported: 5% by mass) as a hydrogenation catalyst which was used in an amount of 5% by mass on the basis of the block copolymer, and the block copolymer was subjected to hydrogenation reaction under a hydrogen pressure of 2 MPa at a temperature of 150°C for 10 h. The obtained reaction mixture was allowed to stand for cooling and pressure releasing, and then subjected to filtration to remove the palladium carbon therefrom. The resulting filtrate was concentrated and further vacuum-dried, thereby obtaining a hydrogenated product of the poly( $\beta$ -farnesene/butadiene)-polystyrene diblock copolymer (hereinafter referred to as a "hydrogenated block copolymer (A5)"). The formulations of various raw materials used above and the results of measurement of various properties of the thus obtained hydrogenated block copolymer (A5) are shown in Table 1 below.

**[0086]** The subsequent procedure was conducted in the same manner as in Example 1 except that the resulting hydrogenated block copolymer (A5) and the base oil were formulated as shown in Table 2, thereby obtaining an oil composition. The thus obtained oil composition was subjected to the above evaluation. The results are shown in Table 2.

[Example 6]

**[0087]** The same procedure as in Example 5 was repeated except that the hydrogenated block copolymer (A5) and the base oil were formulated as shown in Table 2, thereby obtaining an oil composition. The thus obtained oil composition was subjected to the above evaluation. The results are shown in Table 2.

[Example 7]

**[0088]** A pressure reaction vessel previously purged with nitrogen and then dried was charged with 62.4 kg of cyclohexane as a solvent and 104.9 g of sec-butyl lithium (in the form of a 10.5% by mass cyclohexane solution) as an anionic polymerization initiator. The contents of the reaction vessel were heated to 50°C, and then a mixture of 4.88 kg of  $\beta$ -farnesene and 4.88 kg of isoprene was added thereto, followed by polymerizing the contents of the reaction vessel for 2 h. Successively, 5.85 kg of styrene was added to the reaction vessel, followed by polymerizing the contents of the reaction vessel for 1 h, thereby obtaining a reaction solution containing a poly( $\beta$ -farnesene/isoprene)-polystyrene diblock copolymer. Added into the reaction solution was palladium carbon (amount of palladium supported: 5% by mass) as a hydrogenation catalyst which was used in an amount of 5% by mass on the basis of the block copolymer, and the block copolymer was subjected to hydrogenation reaction under a hydrogen pressure of 2 MPa at a temperature of 150°C for 10 h. The obtained reaction mixture was allowed to stand for cooling and pressure releasing, and then subjected to filtration to remove the palladium carbon therefrom. The resulting filtrate was concentrated and further vacuum-dried, thereby obtaining a hydrogenated product of the poly( $\beta$ -farnesene/isoprene)-polystyrene diblock copolymer (hereinafter referred to as a "hydrogenated block copolymer (A6)"). The formulations of various raw materials used above and the results of measurement of various properties of the thus obtained hydrogenated block copolymer (A6) are shown in Table 1 below.

**[0089]** The subsequent procedure was conducted in the same manner as in Example 1 except that the resulting hydrogenated block copolymer (A6) and the base oil were formulated as shown in Table 2, thereby obtaining an oil composition. The thus obtained oil composition was subjected to the above evaluation. The results are shown in Table 2.

[Example 8]

**[0090]** The same procedure as in Example 7 was repeated except that the hydrogenated block copolymer (A6) and the base oil were formulated as shown in Table 2, thereby obtaining an oil composition. The thus obtained oil composition was subjected to the above evaluation. The results are shown in Table 2.

[Comparative Example 1]

**[0091]** A pressure reaction vessel previously purged with nitrogen and then dried was charged with 62.4 kg of cyclohexane as a solvent and 98.6 g of sec-butyl lithium (in the form of a 10.5% by mass cyclohexane solution) as an anionic polymerization initiator. The contents of the reaction vessel were heated to 50°C, and then 8.36 kg of isoprene was added thereto, followed by polymerizing the contents of the reaction vessel for 2 h. Successively, 5.34 kg of styrene was added to the reaction vessel, followed by polymerizing the contents of the reaction vessel for 1 h, thereby obtaining a reaction solution containing a polyisoprene-polystyrene diblock copolymer. Added into the reaction solution was pal-

ladium carbon (amount of palladium supported: 5% by mass) as a hydrogenation catalyst which was used in an amount of 5% by mass on the basis of the block copolymer, and the block copolymer was subjected to hydrogenation reaction under a hydrogen pressure of 2 MPa at a temperature of 150°C for 10 h. The obtained reaction mixture was allowed to stand for cooling and pressure releasing, and then subjected to filtration to remove the palladium carbon therefrom. The resulting filtrate was concentrated and further vacuum-dried, thereby obtaining a hydrogenated product of the polyisoprene-polystyrene diblock copolymer (hereinafter referred to as a "hydrogenated block copolymer (B1)"). The formulations of various raw materials used above and the results of measurement of various properties of the thus obtained hydrogenated block copolymer (B1) are shown in Table 1 below.

**[0092]** The subsequent procedure was conducted in the same manner as in Example 1 except that the resulting hydrogenated block copolymer (B1) was used as a viscosity index improver, thereby obtaining an oil composition. The thus obtained oil composition was subjected to the above evaluation. The results are shown in Table 2.

[Comparative Example 2]

**[0093]** The same procedure as in Comparative Example 1 was repeated except that the hydrogenated block copolymer (B1) and the base oil were formulated as shown in Table 2, thereby obtaining an oil composition. The thus obtained oil composition was subjected to the above evaluation. The results are shown in Table 2.

[Comparative Example 3]

**[0094]** The same procedure as in Comparative Example 1 was repeated except that the respective components were formulated as shown in Table 1, thereby obtaining a hydrogenated block copolymer (B2) and an oil composition and subjecting the resulting products to the above evaluation. The results are shown in Tables 1 and 2.

[Comparative Example 4]

**[0095]** A pressure reaction vessel previously purged with nitrogen and then dried was charged with 62.4 kg of cyclohexane as a solvent, 79.4 g of sec-butyl lithium (in the form of a 10.5% by mass cyclohexane solution) as an anionic polymerization initiator and 375 g of tetrahydrofuran as a Lewis base. The contents of the reaction vessel were heated to 50°C, and then 0.47 kg of styrene (1) was added thereto, followed by polymerizing the contents of the reaction vessel for 1 h. Successively, a mixture of 6.86 kg of isoprene and 6.86 kg of butadiene was added to the reaction vessel, followed by polymerizing the contents of the reaction vessel for 2 h. Further, 1.40 kg of styrene (2) was added to the reaction vessel, followed by polymerizing the contents of the reaction vessel for 1 h, thereby obtaining a reaction solution containing a polystyrene-poly(isoprene/butadiene)-polystyrene triblock copolymer. Added into the reaction solution was palladium carbon (amount of palladium supported: 5% by mass) as a hydrogenation catalyst which was used in an amount of 5% by mass on the basis of the block copolymer, and the block copolymer was subjected to hydrogenation reaction under a hydrogen pressure of 2 MPa at a temperature of 150°C for 10 h. The obtained reaction mixture was allowed to stand for cooling and pressure releasing, and then subjected to filtration to remove the palladium carbon therefrom. The resulting filtrate was concentrated and further vacuum-dried, thereby obtaining a hydrogenated product of the polystyrene-poly(isoprene/butadiene)-polystyrene triblock copolymer (hereinafter referred to as a "hydrogenated block copolymer (B3)"). The formulations of various raw materials used above and the results of measurement of various properties of the thus obtained hydrogenated block copolymer (B3) are shown in Table 1 below.

**[0096]** The subsequent procedure was conducted in the same manner as in Example 1 except that the resulting hydrogenated block copolymer (B3) and the base oil were formulated as shown in Table 2, thereby obtaining an oil composition. The thus obtained oil composition was subjected to the above evaluation. The results are shown in Table 2.

[Comparative Example 5]

**[0097]** Only the base oil, i.e., the paraffin-based oil "DIANA FRESIA S-32" available from Idemitsu Kosan Co., Ltd., was used as such in Comparative Example 5, and subjected to the above evaluation. The results are shown in Table 2.

TABLE 1

	Hydrogenated block copolymers						Hydrogenated block copolymers		
	A1	A2	A3	A4	A5	A6	B1	B2	B3
Amounts used [kg]									
Cyclohexane	62.4	62.4	62.4	62.4	62.4	62.4	62.4	62.4	62.4
sec-Butyl lithium	0.0510	0.0406	0.1080	0.1124	0.1184	0.1049	0.0986	0.0921	0.0794
Styrene (1)	2.34	2.59	5.85	0.77	5.85	5.85	5.34	5.14	0.47
Styrene (2)	2.34			5.39					1.40
$\beta$ -Farnesene	10.9	13.0	9.75	9.44	5.43	4.88			
Butadiene					4.32				6.86
Isoprene						4.88	8.36	8.56	6.86
Tetrahydrofuran									0.375
(b1)/(b) [% by mass] (*1)	100	100	100	100	56	50	0	0	0
(a)/(b) [mass ratio] (*2)	30/70	17/83	37.5/62.5	39.5/60.5	37.5/62.5	37.5/62.5	39/61	37.5/62.5	12/88
Polymer skeleton (*3)	St-F-St	F-St	F-St	St-F-St	F/Bd-St	F/IP-St	IP-St	IP-St	St-IP/Bd-St
Properties									
Peak top molecular weight (Mp)	228,000	244,000	92,000	91,700	109,200	118,500	122,000	137,000	165,000
Peak top molecular weight of styrene block (1)	28,800	40,000	33,900	30,100	35,000	31,000	34,000	35,000	3,500
Peak top molecular weight of styrene block (2)	28,800	-	-	4,300	-	-	-	-	11,000
Molecular weight distribution (Mw/Mn)	1.13	1.13	1.06	1.04	1.05	1.03	1.03	1.05	1.06
Hydrogenation rate [%] (*4)	90.6	91.5	91.7	91.8	98.7	99.0	99.4	99.1	87.0
<p>Note (*1): (b1)/(b): Content of a constitutional unit (b1) derived from farnesene in a whole amount of a constitutional unit (b) derived from a conjugated diene.</p> <p>(*2): (a)/(b): Mass ratio of a whole amount of a constitutional unit (a) to a whole amount of a constitutional unit (b).</p> <p>(*3): St-F-St: Polystyrene-poly(<math>\beta</math>-farnesene)-polystyrene triblock copolymer.</p> <p>F-St: Poly(<math>\beta</math>-farnesene)-polystyrene diblock copolymer.</p> <p>F/Bd-St: Poly(<math>\beta</math>-farnesene/butadiene)-polystyrene diblock copolymer.</p> <p>F/IP-St: Poly(<math>\beta</math>-farnesene/isoprene)-polystyrene diblock copolymer.</p> <p>IP-St: Polyisoprene-polystyrene diblock copolymer.</p> <p>St-IP/Bd-St: Polystyrene-polyisoprene/polybutadiene-polystyrene triblock copolymer.</p> <p>(*4): Hydrogenation rate of carbon-carbon double bonds in a constitutional unit (b) derived from a conjugated diene.</p>									

TABLE 2

	Examples								Comparative Examples				
	1	2	3	4	5	6	7	8	1	2	3	4	5
Base oil													
DIANA FRESIA S-32 (mass%)	99	99	98	98	98.5	98	98.5	98	99	98	99	99	100
Viscosity index improver													
Hydrogenated block copolymer (A1) (mass%)	1												
Hydrogenated block copolymer (A2) (mass%)		1											
Hydrogenated block copolymer (A3) (mass%)			2										
Hydrogenated block copolymer (A4) (mass%)				2									
Hydrogenated block copolymer (A5) (mass%)					1.5	2							
Hydrogenated block copolymer (A6) (mass%)							1.5	2					
Hydrogenated block copolymer (B1) (mass%)									1	2			
Hydrogenated block copolymer (B2) (mass%)											1		
Hydrogenated block copolymer (B3) (mass%)												1	
Evaluation													
Kinetic viscosity (at 40°C) (mm <sup>2</sup> /s)	114.7	64.0	54.7	55.0	55.8	65.5	56.0	65.9	56.7	229.0	61.9	57.8	31.2
Kinetic viscosity (at 100°C) (mm <sup>2</sup> /s)	23.5	10.6	9.2	9.6	9.4	10.8	9.3	10.7	9.2	31.6	10.0	8.6	5.4
Viscosity index (VI)	237	155	147	160	152	156	148	152	144	179	147	84	105
High-temperature high-shear viscosity (HTHS) (mPa•s)	2.8	2.8	2.8	2.8	2.8	3.3	2.8	3.2	2.7	4.1	2.8	2.8	2.0

[0098] From the results shown in Tables 1 and 2, it was confirmed that the hydrogenated block copolymers obtained in Examples 1 to 8 were excellent in an effect of improving a viscosity index and a high-temperature high-shear viscosity of oils even though they were viscosity index improvers produced from a plant-derived raw material ( $\beta$ -farnesene), and it was also confirmed that the hydrogenated block copolymers of these Examples were excellent in an effect of improving a viscosity index of oils to an extent identical to or higher than those attained by using the hydrogenated block copolymers obtained in Comparative Examples 1 to 4. Furthermore, the hydrogenated block copolymers obtained in Examples 2 to 8 exhibited a sufficiently reduced kinetic viscosity as measured at a temperature of each of 40°C and 100°C, and therefore also excellent in an effect of saving a fuel consumption in automobiles.

## Claims

1. An oil composition comprising a base oil and a viscosity index improver, the viscosity index improver consisting of a hydrogenated product of a block copolymer comprising a polymer block (A) containing a constitutional unit (a) derived from an aromatic vinyl compound as a main component and a polymer block (B) containing a constitutional unit (b) derived from a conjugated diene as a main component, a content of a constitutional unit (b1) derived from farnesene in a whole amount of the constitutional unit (b) derived from the conjugated diene being from 40 to 100% by mass, and a content of a constitutional unit (b2) derived from a conjugated diene other than farnesene in a whole amount of the constitutional unit (b) derived from the conjugated diene being from 0 to 60% by mass, and 50 mol% or more of carbon-carbon double bonds in the constitutional unit (b) derived from the conjugated diene being hydrogenated, wherein a mass ratio of a whole amount of the constitutional unit (a) derived from the aromatic vinyl compound and a whole amount of the constitutional unit (b) derived from the conjugated diene [(a)/(b)] is 10/90 to 50/50, wherein the content of the viscosity index improver in the oil composition is from 0.1 to 10% by mass.
2. The oil composition according to claim 1, wherein the farnesene is  $\beta$ -farnesene.
3. The oil composition according to claim 1 or 2, wherein a peak top molecular weight (Mp) of the viscosity index improver is from 4,000 to 1,500,000, wherein the peak top molecular weight (Mp) is determined from a position of a peak top of the molecular weight distribution (Mw/Mn) measured by gel permeation chromatography in terms of a molecular weight of polystyrene as a reference standard substance.
4. The oil composition according to any one of claims 1 to 3, wherein a molecular weight distribution (Mw/Mn) of the viscosity index improver is from 1 to 4, wherein the molecular weight distribution (Mw/Mn) is measured by gel permeation chromatography in terms of a molecular weight of polystyrene as a reference standard substance.
5. The oil composition according to any one of claims 1 to 4, wherein the aromatic vinyl compound is styrene.
6. The oil composition according to any one of claims 1 to 5, wherein the conjugated diene other than farnesene is at least one compound selected from the group consisting of isoprene, butadiene and myrcene.
7. The oil composition according to any one of claims 1 to 6, wherein 50 mol% or more of carbon-carbon double bonds in the polymer block (B) are hydrogenated.
8. A process for producing the oil composition as claimed in any one of claims 1 to 7, comprising a process for producing the viscosity index improver comprising:
  - a polymerization step of obtaining the block copolymer comprising the polymer block (A) containing the constitutional unit (a) derived from the aromatic vinyl compound as a main component and the polymer block (B) containing the constitutional unit (b) derived from the conjugated diene as a main component by anionic polymerization; and
  - a hydrogenation step of hydrogenating 50 mol% or more of carbon-carbon double bonds in the constitutional unit (b) derived from the conjugated diene; and mixing a base oil with the viscosity index improver.

## Patentansprüche

1. Ölzusammensetzung, umfassend ein Grundöl und einen Viskositätsindexverbesserer, wobei der Viskositätsindexverbesserer aus einem hydrierten Produkt eines Blockcopolymers, umfassend einen Polymerblock (A), enthaltend einen Baustein (a), abgeleitet von einer aromatischen Vinylverbindung, als einen Hauptbestandteil, und einen Polymerblock (B), enthaltend einen Baustein (b), abgeleitet von einem konjugierten Dien, als einen Hauptbestandteil, besteht,  
 ein Gehalt eines Bausteins (b1), abgeleitet von Farnesen, in einer Gesamtmenge des Bausteins (b), abgeleitet von dem konjugierten Dien, von 40 bis 100 Massen-% beträgt und ein Gehalt eines Bausteins (b2), abgeleitet von einem konjugierten Dien, das von Farnesen verschieden ist, in einer Gesamtmenge des Bausteins (b), abgeleitet von dem konjugierten Dien, von 0 bis 60 Massen-% beträgt, und  
 50 Mol-% oder mehr der Kohlenstoff-Kohlenstoff-Doppelbindungen in dem Baustein (b), abgeleitet von dem konjugierten Dien, hydriert sind,  
 wobei ein Massenverhältnis einer Gesamtmenge des Bausteins (a), abgeleitet von der aromatischen Vinylverbindung, und einer Gesamtmenge des Bausteins (b), abgeleitet von dem konjugierten Dien, [(a)/(b)] 10/90 bis 50/50 beträgt, wobei der Gehalt des Viskositätsindexverbesserers in der Ölzusammensetzung von 0,1 bis 10 Massen-% beträgt.
2. Ölzusammensetzung nach Anspruch 1, wobei das Farnesen  $\beta$ -Farnesen ist.
3. Ölzusammensetzung nach Anspruch 1 oder 2, wobei ein Peakspitzenmolekulargewicht (Mp) des Viskositätsindexverbesserers von 4.000 bis 1.500.000 beträgt, wobei das Peakspitzenmolekulargewicht (Mp) aus einer Position einer Peakspitze der Molekulargewichtsverteilung (Mw/Mn), gemessen durch Gelpermeationschromatographie, bezogen auf ein Molekulargewicht von Polystyrol als eine Referenzstandardverbindung, bestimmt wird.
4. Ölzusammensetzung nach einem der Ansprüche 1 bis 3, wobei eine Molekulargewichtsverteilung (Mw/Mn) des Viskositätsindexverbesserers von 1 bis 4 beträgt, wobei die Molekulargewichtsverteilung (Mw/Mn) durch Gelpermeationschromatographie, bezogen auf ein Molekulargewicht von Polystyrol als eine Referenzstandardverbindung, gemessen wird.
5. Ölzusammensetzung nach einem der Ansprüche 1 bis 4, wobei die aromatische Vinylverbindung Styrol ist.
6. Ölzusammensetzung nach einem der Ansprüche 1 bis 5, wobei das konjugierte Dien, das von Farnesen verschieden ist, mindestens eine Verbindung, ausgewählt aus der Gruppe, bestehend aus Isopren, Butadien und Myrcen, ist.
7. Ölzusammensetzung nach einem der Ansprüche 1 bis 6, wobei 50 Mol-% oder mehr der Kohlenstoff-Kohlenstoff-Doppelbindungen in dem Polymerblock (B) hydriert sind.
8. Verfahren zur Herstellung der Ölzusammensetzung nach einem der Ansprüche 1 bis 7, umfassend ein Verfahren zur Herstellung des Viskositätsindexverbesserers, umfassend:  
 einen Polymerisationsschritt des Erhaltens des Blockcopolymers, umfassend den Polymerblock (A), enthaltend den Baustein (a), abgeleitet von der aromatischen Vinylverbindung, als einen Hauptbestandteil, und den Polymerblock (B), enthaltend den Baustein (b), abgeleitet von dem konjugierten Dien, als einen Hauptbestandteil, durch anionische Polymerisation; und  
 einen Hydrierschritt des Hydrierens von 50 Mol-% oder mehr der Kohlenstoff-Kohlenstoff-Doppelbindungen in dem Baustein (b), abgeleitet von dem konjugierten Dien;  
 und Mischen eines Grundöls mit dem Viskositätsindexverbesserer.

## Revendications

1. Composition d'huile comprenant une huile de base et un agent améliorant l'indice de viscosité, l'agent améliorant l'indice de viscosité consistant en le produit hydrogéné d'un copolymère séquencé comprenant une séquence polymérique (A) contenant une unité constitutive (a) dérivée d'un composé vinylaromatique comme composant principal et une séquence polymérique (B) contenant une unité constitutive (b) dérivée d'un diène conjugué comme composant principal,  
 une teneur d'une unité constitutive (b1) dérivée du farnésène par rapport à l'entièreté de l'unité constitutive (b)



dérivée du diène conjugué se situant dans l'intervalle allant de 40 à 100% en masse et une teneur d'une unité constitutive (b2) dérivée d'un diène conjugué autre que le farnésène par rapport à l'entièreté de l'unité constitutive (b) dérivée du diène conjugué se situant dans l'intervalle allant de 0 à 60% en masse, et 50% en moles ou plus des doubles liaisons carbone-carbone dans l'unité constitutive (b) dérivée du diène conjugué étant hydrogénées,

où le rapport massique de l'entièreté de l'unité constitutive (a) dérivée du composé vinylaromatique et de l'entièreté de l'unité constitutive (b) dérivée du diène conjugué [(a)/(b)] se situe dans l'intervalle allant de 10/90 à 50/50, où la teneur en agent améliorant l'indice de viscosité dans la compositions d'huile se situe dans l'intervalle allant de 0,1 à 10% en masse.

2. Composition d'huile selon la revendication 1, où le farnésène est le  $\beta$ -farnésène.
3. Composition d'huile selon la revendication 1, où le sommet du pic de poids moléculaire (Mp) de l'agent améliorant l'indice de viscosité se situe dans l'intervalle allant de 4000 à 1 500 000, où le sommet du pic de poids moléculaire (Mp) est déterminé à partir du sommet du pic de la distribution de poids moléculaire (Mw/Mn) mesurée par chromatographie de perméation sur gel en termes de poids moléculaire du polystyrène comme substance standard de référence.
4. Composition d'huile selon l'une quelconque des revendications 1 à 3, où la distribution de poids moléculaire (Mw/Mn) de l'agent améliorant l'indice de viscosité se situe dans l'intervalle allant de 1 à 4, où la distribution de poids moléculaire (Mw/Mn) est mesurée par chromatographie de perméation sur gel en termes de poids moléculaire du polystyrène comme substance standard de référence.
5. Composition d'huile selon l'une quelconque des revendications 1 à 4, où le composé vinylaromatique est le styrène.
6. Composition d'huile selon l'une quelconque des revendications 1 à 5, où le diène conjugué autre que le farnésène est au moins un composé choisi parmi le groupe consistant en l'isoprène, le butadiène et le myrcène.
7. Composition d'huile selon l'une quelconque des revendications 1 à 6, où 50% en moles ou plus des doubles liaisons carbone-carbone dans la séquence polymérique (B) sont hydrogénées.
8. Procédé de production de la composition d'huile selon l'une quelconque des revendications 1 à 7, comprenant un procédé de production de l'agent améliorant l'indice de viscosité, comprenant :
  - une étape de polymérisation pour obtenir le copolymère séquencé comprenant la séquence polymérique (A) contenant l'unité constitutive (a) dérivée d'un composé vinylaromatique comme composant principal et la séquence polymérique (B) contenant l'unité constitutive (b) dérivée du diène conjugué comme composant principal, par polymérisation anionique, et
  - une étape d'hydrogénation de 50% en moles ou plus des doubles liaisons carbone-carbone dans l'unité constitutive (b) dérivée du diène conjugué, et
  - le mélange d'une huile de base à l'agent améliorant l'indice de viscosité.

**REFERENCES CITED IN THE DESCRIPTION**

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