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(54) Title: ADDITION POLYMERIZATION METHOD, PREPOLYMERIZED CATALYTIC COMPONENT FOR ADDITION POLYMERIZATION, AND METHOD FOR PRODUCING ADDITION POLYMER

(57) Abstract: To provide a polymerization method which gives an addition polymer and a prepolymerized catalyst component for addition polymerization that contain less fine powders. A method for addition polymerization comprising the following steps: (1) a step of preparing a mixture by mixing a solvent with an organic compound (D) represented by Formula [2], (2) a step of addition polymerizing monomers in the mixture in the presence of a catalyst prepared by bringing a compound (A) selected from among transition metal compounds represented by Formula [1] and their μ-oxo type transition metal compound dimers, and an activating agent (B) into contact with each other, $L^1_aM^1X^1_b$ [1] R^1-O-(-R^2-O-)$_n$$Q^1$ [2]

DESCRIPTION

ADDITION POLYMERIZATION METHOD, PREPOLYMERIZED CATALYTIC COMPONENT FOR ADDITION POLYMERIZATION, AND METHOD FOR PRODUCING ADDITION POLYMER

Technical Field

[0001]

The present application claims the Paris Convention priority based on Japanese Patent Application No. 2011-046008 filed on March 3, 2011, the entire content of which is incorporated herein by reference.

The present invention relates to an addition polymerization method, a prepolymerized catalyst component for addition polymerization, and a method for producing an addition polymer.

Background Art

[0002]

Since addition polymers such as polypropylene and polyethylene are superior in mechanical properties, chemical resistance and the like, and also superior in balance between those properties and economical efficiency, they have been widely used for various molding fields.

[0003]

These addition polymers have recently been produced by polymerizing an olefin or the like using a catalyst comprising a transition metal compound and an organometallic component such as aluminoxane.

[0004]

It has been known that since these catalysts are soluble in a reaction system, when the catalyst is applied for polymerization accompanied by formation of addition polymer particles, it is possible to impart a fixed shape to an addition polymer to be formed by using specific particles as one of catalyst components. For example, Patent Document 1 describes modified particles obtained by using silica gel, an organozinc compound, trifluorophenols and water, and also describes that an olefin is polymerized using a catalyst comprising the particles, a transition metal compound and an organoaluminum.

[0005]

It has also been known that it is effective to perform a prepolymerization step as a prestage of a main polymerization step in the case of industrially carrying out polymerization which enables the production of an addition polymer in the form of particles. For example, Patent Document 2 describes a method in which an olefin is prepolymerized by a catalyst obtained by using specific particles as one of catalyst components to obtain a prepolymerized catalyst component for olefin polymerization, and then main polymerization is performed using the prepolymerized catalyst component.

Prior Art Document

Patent Document

[0006]

Patent Document 1: JP-A-2003-171413

Patent Document 2: JP-A-2002-293817

Summary of the Invention

Problems to be Solved by the Invention

[0007]

However, since an addition polymer and a prepolymerized catalyst component in the form of fine powders may be sometimes produced by these methods, they are not sufficiently satisfactory methods from the viewpoints of stable operability and productivity of the addition polymer. An object of the present invention is to provide a polymerization method which gives an addition polymer and a prepolymerized catalyst component for addition polymerization that contain less fine powders,

Means for Solving the Problems

[0008]

First, the present invention is directed to a method for addition polymerizing monomers capable of addition polymerization, the method comprises the following steps:

(1) a step of preparing a mixture by mixing a solvent for polymerization with an organic compound (D) represented by Formula [2],

(2) a step of addition polymerizing the monomers in the mixture in the presence of a catalyst prepared by bringing a compound (A) selected from among transition metal compounds represented by Formula [1] and their μ-oxo type transition metal compound dimers, and an activating agent (B) into contact with each other,

$$L^1_aM^1X^1_b \qquad [1]$$

wherein M^1 is a transition metal atom of Group 4; L^1 is a group having a cyclopentadiene type anionic frame or a group containing a hetero atom; X^1 is a halogen atom, a hydrocarbyloxy group, or a hydrocarbyl group other than groups having a cyclopentadiene type anionic frame; a is a number satisfying $0 < a \leq 3$; b is a number satisfying $0 < b \leq 3$; when a is more than 1, one L^1 may be linked to another L^1 either directly or by a group containing a carbon atom, a silicon atom, a nitrogen atom, an oxygen atom, a sulfur atom or a phosphorus atom,

$$R^1\text{-}O\text{-}(\text{-}R^2\text{-}O\text{-})_nQ^1 \qquad [2]$$

wherein R^1 is a hydrocarbyl group having 1 to 30 carbon atoms which may have a substituent; R^2 is an alkylene group having 1 to 20 carbon atoms which may have a substituent; Q^1 is a hydrogen atom, $\text{-}C(=O)OM^2$, $\text{-}R^3\text{-}C(=O)OM^2$, $\text{-}S(=O)_2OM^2$, $\text{-}R^3\text{-}S(=O)_2OM^2$, $\text{-}P(=O)(OH)(OM^2)$, $\text{-}P(=O)(OR^4)(OM^2)$ or $\text{-}P(=O)(OM^2)_2$; M^2 is a hydrogen atom or an alkali metal atom; R^3 is an alkylene group having 1 to 20 carbon atoms which may have a substituent; R^4 is a hydrocarbyl group having 1 to 20 carbon atoms which may have a substituent; n is a number of 1 to 100; when n is more than 1, R^2 groups may be the same or different.

[0009]

Second, the present invention is directed to a prepolymerized catalyst component for addition polymerization obtained by the above method when the above method is a prepolymerization method.

[0010]

Third, the present invention is directed to a method for producing an addition polymer, using the prepolymerized catalyst component for addition polymerization and if necessary, an organoaluminum compound (C).

[0011]

Fourth, the present invention is directed to a prepolymerized catalyst component for addition polymerization, wherein the cumulative weight fraction of a prepolymerized catalyst component for addition polymerization having a particle diameter of 60 μm or less is 7% or less.

Effects of the Invention

[0012]

According to method of the present invention, since it is possible to reduce the discharge of fine powders when an addition polymer and a prepolymerized catalyst component for addition polymerization are produced, polymerization can be performed stably and continuously.

Mode for Carrying Out the Invention

[0013]

The present invention will be described in detail below.

[0014]

Compound (A): transition metal compound or μ-oxo type transition metal compound dimer thereof

The compound (A) to be used in the present invention is selected from among transition metal compounds and their μ-oxo type transition metal compound dimers. Examples of the transition metal compound or μ-oxo type transition metal compound dimer thereof include a transition metal compound represented by Formula [1] described below or a μ-oxo type transition metal compound dimer thereof:

$$L^1_aM^1X^1_b \qquad [1]$$

wherein M^1 is a transition metal atom of Group 4; L^1 is a group having a cyclopentadiene type anionic frame or a group containing a hetero atom; X^1 is a halogen atom, a hydrocarbyloxy group, or a hydrocarbyl group other than groups having a cyclopentadiene type anionic frame; a is a number satisfying $0 < a \leq 3$; b is a number satisfying $0 < b \leq 3$; when a is more than 1, one L^1 may be linked to another L^1 either directly or by a group containing a carbon atom, a silicon atom, a nitrogen atom, an oxygen atom, a sulfur atom or a phosphorus atom.

[0015]

M^1 in Formula [1] is a titanium atom, a zirconium atom or a hafnium atom, and more preferably a zirconium atom.

[0016]

Examples of the group having a cyclopentadiene type anionic frame of L^1 in Formula [1] include an optionally substituted cyclopentadienyl group, an optionally substituted indenyl group and an optionally substituted fluorenyl group. Examples of L^1 include a cyclopentadienyl group, a methylcyclopentadienyl group, an ethylcyclopentadienyl group, a n-butylcyclopentadienyl group, a tert-butylcyclopentadienyl group, a 1,2-dimethylcyclopentadienyl group, a 1,3-dimethylcyclopentadienyl group, a 1-methyl-2-ethylcyclopentadienyl group, a 1-methyl-3-ethylcyclopentadienyl group, a 1-tert-butyl-2-methylcyclopentadienyl group, a 1-tert-butyl-3-methylcyclopentadienyl group, a 1-methyl-2-isopropylcyclopentadienyl group, a 1-methyl-3-isopropylcyclopentadienyl group, a 1-methyl-2-n-butylcyclopentadienyl group, a 1-methyl-3-n-butylcyclopentadienyl group, a η^5-1,2,3-trimethylcyclopentadienyl group, a η^5-1,2,4-trimethylcyclopentadienyl group, a tetramethylcyclopentadienyl group, a pentamethylcyclopentadienyl group, an indenyl group, a 4,5,6,7-tetrahydroindenyl group, a 2-methylindenyl group, a 3-methylindenyl group, a 4-methylindenyl group, a 5-methylindenyl group, a 6-methylindenyl group, a 7-methylindenyl group, a 2-tert-butylindenyl group, a 3-tert-butylindenyl group, a 4-tert-butylindenyl group, a 5-tert-butylindenyl group, a 6-tert-butylindenyl group, a 7-tert-butylindenyl group, a 2,3-dimethylindenyl group, a 4,7-dimethylindenyl group, a 2,4,7-trimethylindenyl group, a 2-methyl-4-isopropylindenyl group, a 4,5-benzindenyl group, a 2-methyl-4,5-benzindenyl group, a 4-phenylindenyl group, a 2-methyl-5-phenylindenyl group, a 2-methyl-4-phenylindenyl group, a 2-methyl-4-naphthylindenyl group, a fluorenyl group, a 2,7-dimethylfluorenyl group and a 2,7-di-tert-butylfluorenyl group.

[0017]

There is no particular limitation on hapticity η of the ligand contained in the group having a cyclopentadiene type anionic frame used in L^1 in Formula [1], and the hapticity η of the ligand may be any value which is available in the group having a cyclopentadiene type anionic frame. Examples of the hapticity of the ligand include five, four, three, two and one. The hapticity of the ligand is preferably five, three or one, and more preferably five or three.

[0018]

Examples of the hetero atom in the group containing a hetero atom of L^1 in Formula [1] include an oxygen atom, a sulfur atom, a nitrogen atom and a phosphorus atom, and the group is preferably an alkoxy group, an aryloxy group, a thioalkoxy group, a thioaryloxy group, an alkylamino group, an arylamino group, an alkylphosphino group, an arylphosphino group, an chelating ligand, or an aromatic heterocyclic group or aliphatic heterocyclic group containing an oxygen atom, a sulfur atom, an nitrogen atom and/or a phosphorus atom in its ring.

[0019]

Examples of the group containing a hetero atom of L^1 in Formula [1] include a methoxy group, an ethoxy group, a propoxy group, a butoxy group, a phenoxy group, a 2-methylphenoxy group, 2,6-dimethylphenoxy group, a 2,4,6-trimethylphenoxy group, a 2-ethylphenoxy group, a 4-n-propylphenoxy group, a 2-isopropylphenoxy group, a 2,6-diisopropylphenoxy group, a 4-sec-butylphenoxy group, a 4-tert-butylphenoxy group, a 2,6-di-sec-butylphenoxy group, a 2-tert-butyl-4-methylphenoxy group, a 2,6-di-tert-butylphenoxy group, a 4-methoxyphenoxy group, a 2,6-dimethoxyphenoxy group, a 3,5-dimethoxyphenoxy group, a 2-chlorophenoxy group, a 4-nitrosophenoxy group, a 4-nitrophenoxy group, a 2-aminophenoxy group, a 3-aminophenoxy group, a 4-aminothiophenoxy group, a 2,3,6-trichlorophenoxy group, a 2,4,6-trifluorophenoxy group, a thiomethoxy group, a dimethylamino group, a diethylamino group, a dipropylamino group, a diphenylamino group, an isopropylamino group, a tert-butylamino group, a pyrrolyl group, a dimethylphosphino group, a 2-(2-oxy-1-propyl)phenoxy group, catechol, resorcinol, 4-isopropylcatechol, 3-methoxycatechol, a 1,8-dihydroxynaphthyl group, a 1,2-dihydroxynaphthyl group, a 2,2'-biphenyldiol group, a 1,1'-bi-2-

naphthol group, a 2,2'-dihydroxy-6,6'-dimethylbiphenyl group, a 4,4',6,6'-tetra-tert-butyl-2,2'-methylenediphenoxy group and a 4,4',6,6'-tetramethyl-2,2'-isobutylidenediphenoxy group.

[0020]

Examples of the group containing a hetero atom also include a group represented by Formula [6]:

$R^7{}_3P=N-$ [6]

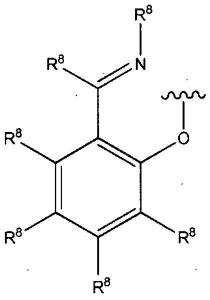
wherein R^7 each independently represents a hydrogen atom, a halogen atom or a hydrocarbyl group, or any two of R^7 groups are linked to one another to form a hydrocarbylene group.

[0021]

Examples of R^7 in Formula [6] include a hydrogen atom, a fluorine atom, a chlorine atom, a bromine atom, an iodine atom, a methyl group, an ethyl group, a n-propyl group, an isopropyl group, a n-butyl group, a tert-butyl group, a cyclopropyl group, a cyclobutyl group, a cycloheptyl group, a cyclohexyl group, a phenyl group, a 1-naphthyl group, a 2-naphthyl group and a benzyl group.

[0022]

Examples of the group containing a hetero atom also include a group represented by Formula [7]:



[7]

wherein R^8 group each independently represents a hydrogen atom, a halogen atom, a hydrocarbyl group, a halogenated hydrocarbyl group, a hydrocarbyloxy group, a silyl group or an amino group, or any two or more of R^8 groups are linked to one another to form a ring structure.

[0023]

Examples of R^8 in Formula [7] include a hydrogen atom, a fluorine atom, a chlorine atom, a bromine atom, an iodine atom, a phenyl group, a 1-naphthyl group, a 2-naphthyl group, a tert-butyl group, a 2,6-dimethylphenyl group, a 2-fluorenyl group, a 2-methylphenyl group, a 4-trifluoromethylphenyl group, a 4-methoxyphenyl group, a 4-pyridyl group, a cyclohexyl group, a 2-isopropylphenyl group, a benzyl group, a methyl group, a triethylsilyl group, a diphenylmethylsilyl group, a 1-methyl-1-phenylethyl group, a 1,1-dimethylpropyl group, a 2-chlorophenyl group and a pentafluorophenyl group.

[0024]

The chelating ligand of L^1 in Formula [1] means a ligand having a plurality of coordination sites, and examples of the ligand include acetylacetonate, diimine, oxazoline, bisoxazoline, terpyridine, acylhydrazone, diethylenetriamine, triethylenetetramine, porphyrin, crown ether and cryptate.

[0025]

Examples of the heterocyclic group of L^1 in Formula [1] include a pyridyl group, an N-substituted imidazolyl group and an N-substituted indazolyl group. Among them, a pyridyl group is preferred.

[0026]

When L^1 groups in Formula [1] are linked to each other by a residue containing a carbon atom, a silicon atom, a nitrogen atom, an oxygen atom, a sulfur atom or a phosphorus atom (that is, when groups having a cyclopentadiene type anionic frame are linked to each other by its residue, when groups containing a hetero atom are linked to each other by its residue, or when a group having a cyclopentadiene type anionic frame and a group containing a hetero atom are linked to each other by its residue), its residue is preferably a divalent residue in which the atom linked to L^1 is a carbon atom, a silicon atom, a nitrogen atom, an oxygen atom, a sulfur atom or a phosphorus atom, and the number of atoms linking two L^1 groups is 3 or less. Examples of its residue include alkylene groups such as a methylene group, an ethylene group and a propylene group; substituted alkylene groups such as a dimethylmethylene group (an isopropylidene group) and a diphenylmethylene group; substituted silylene groups such as a silylene group, a dimethylsilylene group, a diethylsilylene group, a diphenylsilylene group, a tetramethyldisilylene group and a dimethoxysilylene group; and hetero atoms such as a nitrogen atom, an oxygen atom, a sulfur atom and a phosphorus atom. Among them, a methylene group, an ethylene group, a dimethylmethylene group (isopropylidene group), a diphenylmethylene group, a dimethylsilylene group, a diethylsilylene group, a diphenylsilylene group or a dimethoxysilylene group is particularly preferred.

[0027]

Examples of the halogen atom of X^1 in Formula [1] include a fluorine atom, a chlorine atom, a bromine atom and an iodine atom. Examples of the hydrocarbyl group of X^1 include an alkyl group, an aralkyl group, an aryl group, and an alkenyl group, and among them, an alkyl group having 1 to 20 carbon atoms, an aralkyl group having 7 to 20 carbon atoms, an aryl group having 6 to 20 carbon atoms, or an alkenyl group having 3 to 20 carbon atoms is preferable.

[0028]

Examples of the alkyl group having 1 to 20 carbon atoms include a methyl group, an ethyl group, a n-propyl group, an isopropyl group, a n-butyl group, a sec-butyl group, a tert-butyl group, an isobutyl group, a n-pentyl group, a neopentyl group, an amyl group, a n-hexyl group, a n-octyl group, a n-decyl group, a n-dodecyl group, a n-pentadecyl group, and a n-eicosyl group, and among them, a methyl group, an ethyl group, an isopropyl group, a tert-butyl group, an isobutyl group or an amyl group is more preferable. Any of these alkyl groups may be substituted with a halogen atom such as a fluorine atom, a chlorine atom, a bromine atom, or an iodine atom. Examples of the alkyl group substituted with a halogen atom include a fluoromethyl group, a trifluoromethyl group, a chloromethyl group, a trichloromethyl group, a fluoroethyl group, a pentafluoroethyl group, a perfluoropropyl group, a perfluorobutyl group, a perfluorohexyl group, a perfluorooctyl group, a perchloropropyl group, a perchlorobutyl group, and a perbromopropyl group. These alkyl groups may have alkoxy groups such as a methoxy group and an ethoxy group; aryloxy groups such as a phenoxy group; or aralkyloxy groups such as a benzyloxy group as a substituent.

[0029]

Examples of the aralkyl group having 7 to 20 carbon atoms include a benzyl group, a (2-methylphenyl)methyl group, a (3-methylphenyl)methyl group, a (4-methylphenyl)methyl group, a (2,3-dimethylphenyl)methyl group, a (2,4-dimethylphenyl)methyl group, a (2,5-dimethylphenyl)methyl group, a (2,6-dimethylphenyl)methyl group, a (3,4-dimethylphenyl)methyl group, a (3,5-dimethylphenyl)methyl group, a (2,3,4-trimethylphenyl)methyl group, a (2,3,5-trimethylphenyl)methyl group, a (2,3,6-trimethylphenyl)methyl group, a (3,4,5-trimethylphenyl)methyl group, a (2,4,6-trimethylphenyl)methyl group, a (2,3,4,5-tetramethylphenyl)methyl group, a (2,3,4,6-tetramethylphenyl)methyl group, a (2,3,5,6-tetramethylphenyl)methyl group, a (pentamethylphenyl)methyl group, an(ethylphenyl)methyl group, a (n-propylphenyl)methyl group, an(isopropylphenyl)methyl group, a (n-butylphenyl)methyl group, a (sec-butylphenyl)methyl group, a (tert-butylphenyl)methyl group, a (n-pentylphenyl)methyl group, a (neopentylphenyl)methyl group, a (n-hexylphenyl)methyl group, a (n-octylphenyl)methyl group, a (n-decylphenyl)methyl group, a (n-dodecylphenyl)methyl group, a naphthylmethyl group, and an anthracenylmethyl group, and a benzyl group is more preferabe. These aralkyl groups may have halogen atoms such as a fluorine atom, a chlorine atom, a bromine atom, and an iodine atom; alkoxy groups such as a methoxy group and an ethoxy group; aryloxy groups such as a phenoxy group; or aralkyloxy groups such as a benzyloxy group as a substituent.

[0030]

Examples of the aryl group having 6 to 20 carbon atoms include a phenyl group, a 2-tolyl group, a 3-tolyl group, a 4-tolyl group, a 2,3-xylyl group, a 2,4-xylyl group, a 2,5-xylyl group, a 2,6-xylyl group, a 3,4-xylyl group, a 3,5-xylyl group, a 2,3,4-trimethylphenyl group, a 2,3,5-trimethylphenyl group, a 2,3,6-trimethylphenyl group, a 2,4,6-trimethylphenyl group, a 3,4,5-trimethylphenyl group, a 2,3,4,5-tetramethylphenyl group, a 2,3,4,6-tetramethylphenyl group, a 2,3,5,6-tetramethylphenyl group, a pentamethylphenyl group, an ethylphenyl group, a n-propylphenyl group, an isopropylphenyl group, a n-butylphenyl group, a sec-butylphenyl group, a tert-butylphenyl group, a n-pentylphenyl group, a neopentylphenyl group, a n-hexylphenyl group, a n-octylphenyl group, a n-decylphenyl group, a n-dodecylphenyl group, a n-tetradecylphenyl group, a naphthyl group, and an anthracenyl group, and a phenyl group is more preferabe. These aryl groups may have halogen atoms such as a fluorine atom, a chlorine atom, a bromine atom, and an iodine atom; alkoxy groups such as a methoxy group and an ethoxy group; aryloxy groups such as a phenoxy group; or aralkyloxy groups such as a benzyloxy group as a substituent.

[0031]

Examples of the alkenyl group having 3 to 20 carbon atoms include an allyl group, a metallyl group, a crotyl group and a 1,3-diphenyl-2-propenyl group, and among them, an allyl group or a metallyl group is more preferable.

[0032]

Examples of the hydrocarbyloxy group of X^1 in Formula [1] include an alkoxy group, an aralkyloxy group and an aryloxy group, and among them, an alkoxy group having 1 to 20 carbon atoms, an aralkyloxy group having 7 to 20 carbon atoms or an aryloxy group having 6 to 20 carbon atoms is more preferabe.

[0033]

Examples of the alkoxy group having 1 to 20 carbon atoms include a methoxy group, an ethoxy group, a n-propoxy group, an isopropoxy group, a n-butoxy group, a sec-butoxy group, a tert-butoxy group, a n-pentoxy group, a neopentoxy group, a n-hexoxy group, a n-octoxy group, a n-dodesoxy group, a n-pentadesoxy group, and a n-icosoxy group, and among them, a methoxy group, an ethoxy group, an isopropoxy group or a tert-butoxy group is preferabe. These alkoxy groups may have halogen atoms such as a fluorine atom, a chlorine atom, a bromine atom, and an iodine atom; alkoxy groups such as a methoxy group and an ethoxy group; aryloxy groups such as a phenoxy group; or aralkyloxy groups such as a benzyloxy group as a substituent.

[0034]

Examples of the aralkyloxy group having 7 to 20 carbon atoms include a benzyloxy group, a (2-methylphenyl)methoxy group, a (3-methylphenyl)methoxy group, a (4-methylphenyl)methoxy group, a (2,3-dimethylphenyl)methoxy group, a (2,4-dimethylphenyl)methoxy group, a (2,5-dimethylphenyl)methoxy group, a (2,6-dimethylphenyl)methoxy group, a (3,4-dimethylphenyl)methoxy group, a (3,5-dimethylphenyl)methoxy group, a (2,3,4-trimethylphenyl)methoxy group, a (2,3,5-trimethylphenyl)methoxy group, a (2,3,6-trimethylphenyl)methoxy group, a (2,4,5-trimethylphenyl)methoxy group, a (2,4,6-trimethylphenyl)methoxy group, a (3,4,5-trimethylphenyl)methoxy group, a (2,3,4,5-

tetramethylphenyl)methoxy group, a (2,3,4,6-tetramethylphenyl)methoxy group, a (2,3,5,6-tetramethylphenyl)methoxy group, a (pentamethylphenyl)methoxy group, an(ethylphenyl)methoxy group, a (n-propylphenyl)methoxy group, an(isopropylphenyl)methoxy group, a (n-butylphenyl)methoxy group, a (sec-butylphenyl)methoxy group, a (tert-butylphenyl)methoxy group, a (n-hexylphenyl)methoxy group, a (n-octylphenyl)methoxy group, a (n-decylphenyl)methoxy group, a naphthylmethoxy group, and anthracenylmethoxy group, and among them, a benzyloxy group is more preferabe. These aralkyloxy groups may have halogen atoms such as a fluorine atom, a chlorine atom, a bromine atom, and an iodine atom; alkoxy groups such as a methoxy group and an ethoxy group; aryloxy groups such as a phenoxy group; or aralkyloxy groups such as a benzyloxy group as a substituent.

[0035]

Examples of the aryloxy group having 6 to 20 carbon atoms include a phenoxy group, a 2-methylphenoxy group, a 3-methylphenoxy group, a 4-methylphenoxy group, a 2,3-dimethylphenoxy group, a 2,4-dimethylphenoxy group, a 2,5-dimethylphenoxy group, a 2,6-dimethylphenoxy group, a 3,4-dimethylphenoxy group, a 3,5-dimethylphenoxy group, a 2-tert-butyl-3-methylphenoxy group, a 2-tert-butyl-4-methylphenoxy group, a 2-tert-butyl-5-methylphenoxy group, a 2-tert-butyl-6-methylphenoxy group, a 2,3,4-trimethylphenoxy group, a 2,3,5-trimethylphenoxy group, a 2,3,6-trimethylphenoxy group, a 2,4,5-trimethylphenoxy group, a 2,4,6-trimethylphenoxy group, a 2-tert-butyl-3,4-dimethylphenoxy group, a 2-tert-butyl-3,5-dimethylphenoxy group, a 2-tert-butyl-3,6-dimethylphenoxy group, a 2,6-di-tert-butyl-3-methylphenoxy group, a 2-tert-butyl-4,5-dimethylphenoxy group, a 2,6-di-tert-butyl-4-methylphenoxy group, a 3,4,5-trimethylphenoxy group, a 2,3,4,5-tetramethylphenoxy group, a 2-tert-butyl-3,4,5-trimethylphenoxy group, a 2,3,4,6-tetramethylphenoxy group, a 2-tert-butyl-3,4,6-trimethylphenoxy group, a 2,6-di-tert-butyl-3,4-dimethylphenoxy group, a 2,3,5,6-tetramethylphenoxy group, a 2-tert-butyl-3,5,6-trimethylphenoxy group, a 2,6-di-tert-butyl-3,5-dimethylphenoxy group, a pentamethylphenoxy group, an ethylphenoxy group, a n-propylphenoxy group, an isopropylphenoxy group, a n-butylphenoxy group, a sec-butylphenoxy group, a tert-butylphenoxy group, a n-hexylphenoxy group, a n-octylphenoxy group, a n-decylphenoxy group, a n-tetradecylphenoxy group, a naphthoxy group and an anthrathenoxy group. These aryloxy groups may have halogen atoms such as a fluorine atom, a chlorine atom, a bromine atom, and an iodine atom; alkoxy groups such as a methoxy group and an ethoxy group; aryloxy groups such as a phenoxy group; or aralkyloxy groups such as a benzyloxy group as a substituent.

[0036]

X^1 in Formula [1] is more preferably a chlorine atom, a methyl group, an ethyl group, a n-propyl group, an isopropyl group, a n-butyl group, a methoxy group, an ethoxy group, a n-propoxy group, an isopropoxy group, a n-butoxy group, a trifluoromethoxy group, a phenyl group, a phenoxy group, a 2,6-di-tert-butylphenoxy group, a 3,4,5-trifluorophenoxy group, a pentafluorophenoxy group, a 2,3,5,6-tetrafluoro-4-pentafluorophenylphenoxy group or a benzyl group.

[0037]

a in Formula [1] is a number satisfying $0 < a \leq 3$, and b is a number satisfying $0 < b \leq 3$. a and b are appropriately selected according to the valance of M^1. When M^1 is a titanium atom, a zirconium atom, or a hafnium atom, a is preferably 2 and also b is preferably 2.

[0038]

Examples of the compound represented by Formula [1] in which a transition metal atom is a titanium atom include bis(cyclopentadienyl)titanium dichloride, bis(methylcyclopentadienyl)titanium dichloride, bis(ethylcyclopentadienyl)titanium dichloride, bis(n-butylcyclopentadienyl)titanium dichloride, bis(tert-butylcyclopentadienyl)titanium dichloride, bis(1,2-dimethylcyclopentadienyl)titanium dichloride, bis(1,3-dimethylcyclopentadienyl)titanium dichloride, bis(1-methyl-2-ethylcyclopentadienyl)titanium dichloride, bis(1-methyl-3-ethylcyclopentadienyl)titanium dichloride, bis(1-methyl-2-n-butylcyclopentadienyl)titanium dichloride, bis(1-methyl-3-n-butylcyclopentadienyl)titanium dichloride, bis(1-methyl-2-isopropylcyclopentadienyl)titanium dichloride, bis(1-methyl-3-isopropylcyclopentadienyl)titanium dichloride, bis(1-tert-butyl-2-methylcyclopentadienyl)titanium dichloride, bis(1-tert-butyl-3-methylcyclopentadienyl)titanium dichloride, bis(1,2,3-trimethylcyclopentadienyl)titanium dichloride, bis(1,2,4-trimethylcyclopentadienyl)titanium dichloride, bis(tetramethylcyclopentadienyl)titanium dichloride, bis(pentamethylcyclopentadienyl)titanium dichloride, bis(indenyl)titanium dichloride, bis(4,5,6,7-tetrahydroindenyl)titanium dichloride, bis(fluorenyl)titanium dichloride, bis(2-phenylindenyl)titanium dichloride,

[0039]

bis[2-(bis-3,5-trifluoromethylphenyl)indenyl]titanium dichloride, bis[2-(4-tert-butylphenyl)indenyl]titanium dichloride, bis[2-(4-trifluoromethylphenyl)indenyl]titanium dichloride, bis[2-(4-methylphenyl)indenyl]titanium dichloride, bis[2-(3,5-dimethylphenyl)indenyl]titanium dichloride, bis[2-(pentafluorophenyl)indenyl]titanium dichloride, cyclopentadienyl(pentamethylcyclopentadienyl)titanium dichloride, cyclopentadienyl(indenyl)titanium dichloride, cyclopentadienyl(fluorenyl)titanium dichloride, indenyl(fluorenyl)titanium dichloride, pentamethylcyclopentadienyl(indenyl)titanium dichloride, pentamethylcyclopentadienyl(fluorenyl)titanium dichloride, cyclopentadienyl(2-phenylindenyl)titanium dichloride, pentamethylcyclopentadienyl(2-phenylindenyl)titanium dichloride,

[0040]

dimethylsilylenebis(cyclopentadienyl)titanium dichloride, dimethylsilylenebis(2-methylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(3-methylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(2-n-butylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(3-n-butylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(2,3-dimethylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(2,4-dimethylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(2,5-dimethylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(3,4-dimethylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(2,3-ethylmethylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(2,4-ethylmethylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(2,5-ethylmethylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(3,5-ethylmethylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(2,3,4-trimethylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(2,3,5-trimethylcyclopentadienyl)titanium dichloride, dimethylsilylenebis(tetramethylcyclopentadienyl)titanium dichloride,

[0041]

dimethylsilylenebis(indenyl)titanium dichloride, dimethylsilylenebis(2-methylindenyl)titanium dichloride, dimethylsilylenebis(2-tert-butylindenyl)titanium dichloride, dimethylsilylenebis(2,3-dimethylindenyl)titanium dichloride, dimethylsilylenebis(2,4,7-trimethylindenyl)titanium dichloride, dimethylsilylenebis(2-methyl-4-isopropylindenyl)titanium dichloride, dimethylsilylenebis(4,5-benzindenyl)titanium dichloride, dimethylsilylenebis(2-methyl-4,5-benzindenyl)titanium dichloride, dimethylsilylenebis(2-phenylindenyl)titanium dichloride, dimethylsilylenebis(4-phenylindenyl)titanium dichloride, dimethylsilylenebis(2-methyl-4-phenylindenyl)titanium dichloride, dimethylsilylenebis(2-methyl-5-phenylindenyl)titanium dichloride, dimethylsilylenebis(2-methyl-4-naphthylindenyl)titanium dichloride, dimethylsilylenebis(4,5,6,7-tetrahydroindenyl)titanium dichloride,

[0042]

dimethylsilylene(cyclopentadienyl) (indenyl)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (indenyl)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (indenyl)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (indenyl)titanium dichloride, dimethylsilylene(cyclopentadienyl) (fluorenyl)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (fluorenyl)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (fluorenyl)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (indenyl)titanium dichloride, dimethylsilylene(indenyl) (fluorenyl)titanium dichloride, dimethylsilylenebis(fluorenyl)titanium dichloride, dimethylsilylene(cyclopentadienyl) (tetramethylcyclopentadienyl)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (fluorenyl)titanium dichloride,

[0043]

cyclopentadienyltitanium trichloride, pentamethylcyclopentadienyltitanium trichloride, cyclopentadienyl(dimethylamide)titanium dichloride, cyclopentadienyl(phenoxy)titanium dichloride, cyclopentadienyl(2,6-dimethylphenyl)titanium dichloride, cyclopentadienyl(2,6-diisopropylphenyl)titanium dichloride, cyclopentadienyl(2,6-di-tert-butylphenyl)titanium dichloride, pentamethylcyclopentadienyl(2,6-dimethylphenyl)titanium dichloride, pentamethylcyclopentadienyl(2,6-diisopropylphenyl)titanium dichloride, pentamethylcyclopentadienyl(2,6-tert-butylphenyl)titanium dichloride, indenyl(2,6-diisopropylphenyl)titanium dichloride, fluorenyl(2,6-diisopropylphenyl)titanium dichloride,

[0044]

dimethylsilylene(cyclopentadienyl) (2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (3-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (3,5-dimethyl-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (3-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (3,5-di-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (5-methyl-3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (3-tert-butyldimethylsilyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (5-methyl-3-trimethylsilyl-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (3-tert-butyl-5-methoxy-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (3-tert-butyl-5-chloro-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (3,5-diamyl-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(cyclopentadienyl) (1-naphthoxy-2-yl)titanium dichloride,

[0045]

dimethylsilylene(methylcyclopentadienyl) (2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (3-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (3,5-dimethyl-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (3-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (3,5-di-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (5-methyl-3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (3-tert-butyldimethylsilyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (5-methyl-3-trimethylsilyl-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (3-tert-butyl-5-methoxy-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (3-tert-butyl-5-chloro-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (3,5-diamyl-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(methylcyclopentadienyl) (1-naphthoxy-2-yl)titanium dichloride,

[0046]

dimethylsilylene(n-butylcyclopentadienyl) (2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (3-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (3,5-dimethyl-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (3-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (3,5-di-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (5-methyl-3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (3-tert-butyldimethylsilyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (5-methyl-3-trimethylsilyl-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (3-tert-butyl-5-methoxy-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (3-tert-butyl-5-chloro-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (3,5-diamyl-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(n-butylcyclopentadienyl) (1-naphthoxy-2-yl)titanium dichloride,

[0047]

dimethylsilylene(tert-butylcyclopentadienyl) (2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (3-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (3,5-dimethyl-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (3-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (3,5-di-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (5-methyl-3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (3-tert-butyldimethylsilyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (5-methyl-3-trimethylsilyl-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (3-tert-butyl-5-methoxy-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl)

(3-tert-butyl-5-chloro-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (3,5-diamyl-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(tert-butylcyclopentadienyl) (1-naphthoxy-2-yl)titanium dichloride,

[0048]

dimethylsilylene(tetramethylcyclopentadienyl) (2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (3-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (3,5-dimethyl-2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (3-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (3,5-di-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (5-methyl-3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (3-tert-butyldimethylsilyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (5-methyl-3-trimethylsilyl-2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (3-tert-butyl-5-methoxy-2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (3-tert-butyl-5-chloro-2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (3,5-diamyl-2-phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (3-phenyl-2-

phenoxy)titanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl) (1-naphthoxy-2-yl)titanium dichloride,

[0049]

dimethylsilylene(trimethylsilylcyclopentadienyl) (2-phenoxy)titanium dichloride, dimethylsilylene(trimethylsilylcyclopentadienyl) (3-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(trimethylsilylcyclopentadienyl) (3,5-dimethyl-2-phenoxy)titanium dichloride, dimethylsilylene(trimethylsilylcyclopentadienyl) (3-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(trimethylsilylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(trimethylsilylcyclopentadienyl) (3,5-di-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(trimethylsilylcyclopentadienyl) (5-methyl-3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(trimethylsilylcyclopentadienyl) (3-tert-butyldimethylsilyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(trimethylsilylcyclopentadienyl) (5-methyl-3-trimethylsilyl-2-phenoxy)titanium dichloride, dimethylsilylene(trimethylsilylcyclopentadienyl) (3-tert-butyl-5-methoxy-2-phenoxy)titanium dichloride, dimethylsilylene(trimethylsilylcyclopentadienyl) (3-tert-butyl-5-chloro-2-phenoxy)titanium dichloride, dimethylsilylene(trimethylsilylcyclopentadienyl) (3,5-diamyl-2-

phenoxy)titanium dichloride,

dimethylsilylene(trimethylsilylcyclopentadienyl) (3-phenyl-2-phenoxy)titanium dichloride,

dimethylsilylene(trimethylsilylcyclopentadienyl) (1-naphthoxy-2-yl)titanium dichloride,

[0050]

dimethylsilylene(indenyl) (2-phenoxy)titanium dichloride,

dimethylsilylene(indenyl) (3-methyl-2-phenoxy)titanium dichloride,

dimethylsilylene(indenyl) (3,5-dimethyl-2-phenoxy)titanium dichloride,

dimethylsilylene(indenyl) (3-tert-butyl-2-phenoxy)titanium dichloride,

dimethylsilylene(indenyl) (3-tert-butyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(indenyl) (3,5-di-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(indenyl) (5-methyl-3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(indenyl) (3-tert-butyldimethylsilyl-5-methyl-2-phenoxy)titanium dichloride,

dimethylsilylene(indenyl) (5-methyl-3-trimethylsilyl-2-phenoxy)titanium dichloride, dimethylsilylene(indenyl) (3-tert-butyl-5-methoxy-2-phenoxy)titanium dichloride, dimethylsilylene(indenyl) (3-tert-butyl-5-chloro-2-phenoxy)titanium dichloride, dimethylsilylene(indenyl) (3,5-diamyl-2-phenoxy)titanium dichloride, dimethylsilylene(indenyl) (3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(indenyl) (1-naphthoxy-2-yl)titanium dichloride,

[0051]

dimethylsilylene(fluorenyl) (2-phenoxy)titanium dichloride,

dimethylsilylene(fluorenyl) (3-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (3,5-dimethyl-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (3-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (3-tert-butyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (3,5-di-tert-butyl-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (5-methyl-3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (3-tert-butyldimethylsilyl-5-methyl-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (5-methyl-3-trimethylsilyl-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (3-tert-butyl-5-methoxy-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (3-tert-butyl-5-chloro-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (3,5-diamyl-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (3-phenyl-2-phenoxy)titanium dichloride, dimethylsilylene(fluorenyl) (1-naphthoxy-2-yl)titanium dichloride,

[0052]

(tert-butylamide)tetramethylcyclopentadienyl-1,2-ethanediyltitanium dichloride, (methylamide)tetramethylcyclopentadienyl-1,2-ethanediyltitanium dichloride, (ethylamide)tetramethylcyclopentadienyl-1,2-ethanediyltitanium dichloride, (tert-butylamide)tetramethylcyclopentadienyldimethylsilanetitanium dichloride, (benzylamide)tetramethylcyclopentadienyldimethylsilanetitanium dichloride, (phenyl phosphide)tetramethylcyclopentadienyldimethylsilanetitanium dichloride, (tert-butylamide)indenyl-1,2-ethanediyltitanium dichloride, (tert-butylamide)tetrahydroindenyl-1,2-ethanediyltitanium dichloride, (tert-butylamide)fluorenyl-1,2-ethanediyltitanium dichloride, (tert-butylamide)indenyldimethylsilanetitanium dichloride, (tert-butylamide)tetrahydroindenyldimethylsilanetitanium dichloride, (tert-butylamide)fluorenyldimethylsilanetitanium dichloride,

[0053]

(dimethylaminomethyl)tetramethylcyclopentadienyltitanium(III)dichloride, (dimethylaminoethyl)tetramethylcyclopentadienyltitanium(III) dichloride, (dimethylaminopropyl)tetramethylcyclopentadienyltitanium(III) dichloride, (N-pyrrolidinylethyl)tetramethylcyclopentadienyltitanium dichloride, (B-dimethylaminoborabenzene)cyclopentadienyltitanium dichloride, cyclopentadienyl(9-mesitylboraanthracenyl)titanium dichloride,

[0054]

2,2'-thiobis[4-methyl-6-tert-butylphenoxy]titanium dichloride, 2,2'-thiobis[4-methyl-6-(1-methylethyl)phenoxy]titanium dichloride, 2,2'-thiobis(4,6-dimethylphenoxy)titanium dichloride, 2,2'-thiobis(4-methyl-6-tert-butylphenoxy)titanium dichloride, 2,2'-methylenebis(4-methyl-6-tert-butylphenoxy)titanium dichloride, 2,2'-ethylenebis(4-methyl-6-tert-butylphenoxy)titanium dichloride, 2,2'-sulfinylbis(4-methyl-6-tert-butylphenoxy)titanium dichloride, 2,2'-(4,4',6,6'-tetra-tert-butyl-1,1'-biphenoxy)titanium dichloride, (di-tert-butyl-1,3-propanediamide)titanium dichloride, (dicyclohexyl-1,3-propanediamide)titanium dichloride,

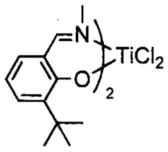
[0055]

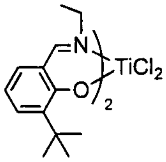
[bis(trimethylsilyl)-1,3-propanediamide]titanium dichloride, [bis(tert-butyldimethylsilyl)-1,3-propanediamide]titanium dichloride, [bis(2,6-dimethylphenyl)-1,3-propanediamide]titanium dichloride, [bis(2,6-

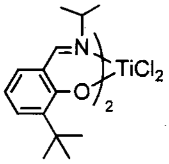
diisopropylphenyl)-1,3-propanediamide]titanium dichloride, [bis(2,6-di-tert-butylphenyl)-1,3-propanediamide]titanium dichloride, [bis(triisopropylsilyl)naphthalenediamide]titanium dichloride, [bis(trimethylsilyl)naphthalenediamide]titanium dichloride, [bis(tert-butyldimethylsilyl)naphthalenediamide]titanium dichloride, [hydrotris(3,5-dimethylpyrazolyl)borate]titanium trichloride, [hydrotris(3,5-diethylpyrazolyl)borate]titanium trichloride, [hydrotris(3,5-di-tert-butylpyrazolyl)borate]titanium trichloride, [tris(3,5-dimethylpyrazolyl)methyl]titanium trichloride, [tris(3,5-diethylpyrazolyl)methyl]titanium trichloride, and [tris(3,5-di-tert-butylpyrazolyl)methyl]titanium trichloride, and also include compounds in which "(2-phenoxy)" in the above-mentioned compounds is replaced by "(3-phenyl-2-phenoxy)", "(3-trimethylsilyl-2-phenoxy)", or "(3-tert-butyldimethylsilyl-2-phenoxy)", and compounds in which "dimethylsilylene" in the above-mentioned compounds is replaced by "methylene", "ethylene", "dimethylmethylene (isopropylidene)", "diphenylmethylene", "diethylsilylene", "diphenylsilylene", or "dimethoxysilylene". Examples of the compound represented by Formula [1] in which a transition metal atom is a zirconium atom or a hafnium atom include compounds in which "titanium" in the above-mentioned compound is replaced by "zirconium" or "hafnium", respectively.

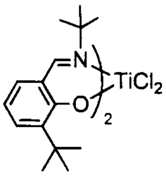
[0056]

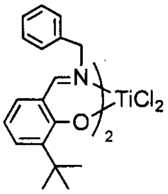
Examples of the transition metal compound represented by Formula [1] also include the following compounds.

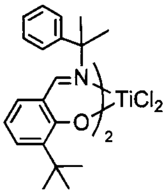


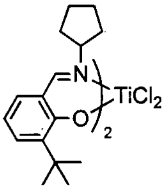


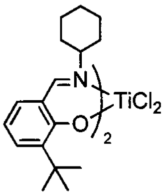


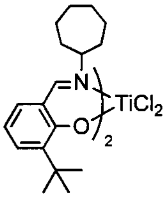


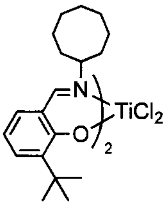


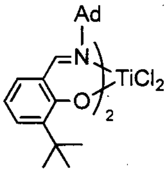












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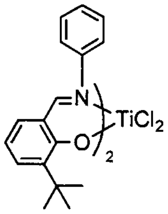

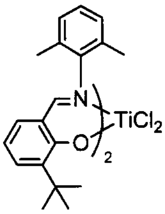

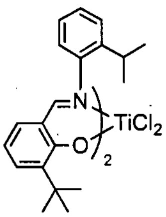

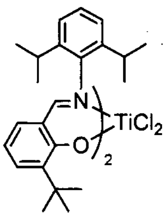

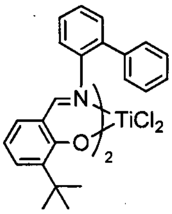

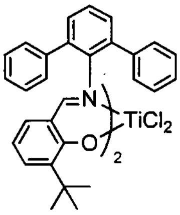

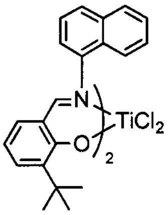

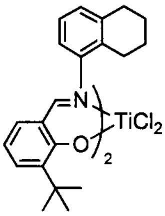

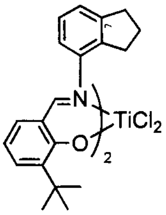

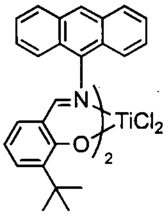

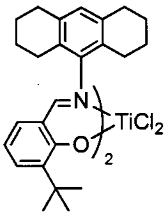

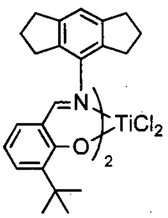

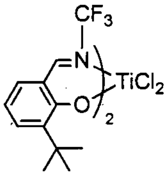

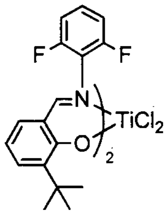

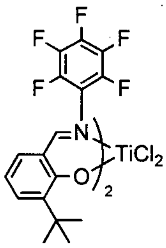

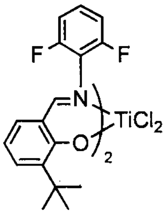

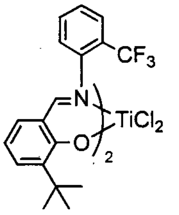

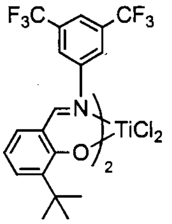

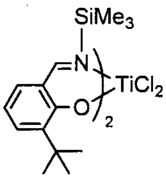

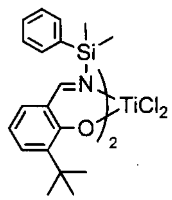

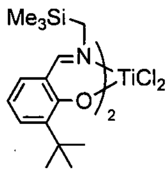

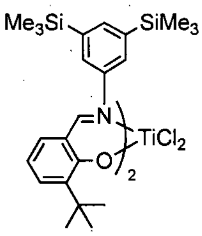

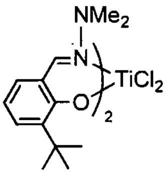

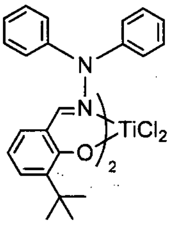

[0057]

Examples of the transition metal compound further include, in addition to these compounds, compounds in which the titanium atom of the above-mentioned compounds is replaced by a zirconium atom or a hafnium atom.

[0058]

Examples of the transition metal compound further include compounds in which “dichloride” in the above-mentioned compounds is replaced by “difluoride”, “dibromide”, “diiodide”, “dimethyl, “diethyl”, “diisopropyl”, “diphenyl”, “dibenzyl”, “dimethoxide”, “diethoxide”, “di(n-propoxide)”, “di(isopropoxide)”, “diphenoxide”, or “di(pentafluorophenoxide)”, and compounds in which “trichloride” in the above-mentioned compounds is replaced by “trifluoride”, “tribromide”, “triiodide”, “trimethyl”, “triethyl”, “triisopropyl”, “triphenyl”, “tribenzyl”, “trimethoxide”, “triethoxide”, “tri(n-propoxide)”, “tri(isopropoxide)”, “triphenoxide”, or “tri(pentafluorophenoxide)”.

[0059]

These transition metal compounds may be used alone, or two or more kinds of them may be used in combination.

[0060]

Among the above-mentioned transition metal compounds, the compounds (A) to be used in the present invention are preferably compounds in which M^1 is zirconium, and transition metal compounds which have at least one group having a cyclopentadiene type anionic frame as L^1 in Formula [1]. Among them, zirconium compounds, which have two groups having a cyclopentadiene type anionic frame as L^1 in Formula [1], L^1 groups being linked to each other by a group containing a carbon atom, a silicon atom, an oxygen atom, a sulfur atom or a phosphorus atom, are particularly preferable.

[0061]

The transition metal compound represented by Formula [1] can be produced by the production methods described in JP-A-6-340684, JP-A-7-258321, and International Publication No. WO 95/00562 and the like.

[0062]

Organic Compound (D)

The organic compound (D) to be used in the present invention is specifically an organic compound represented by Formula [2]:

$$R^1\text{-}O\text{-}(\text{-}R^2\text{-}O\text{-})_nQ^1 \qquad [2]$$

wherein R^1 is a hydrocarbyl group having 1 to 30 carbon atoms which may have a substituent; R^2 is an alkylene group having 1 to 20 carbon atoms which may have a substituent; Q^1 is a hydrogen atom, $\text{-}C(=O)OM^2$, $\text{-}R^3\text{-}C(=O)OM^2$, $\text{-}S(=O)_2OM^2$, $\text{-}R^3\text{-}S(=O)_2OM^2$, $\text{-}P(=O)(OH)(OM^2)$, $\text{-}P(=O)(OR^4)(OM^2)$ or $\text{-}P(=O)(OM^2)_2$; M^2 is a hydrogen atom or an alkali metal atom; R^3 is an alkylene group having 1 to 20 carbon atoms which may have a substituent; R^4 is a hydrocarbyl group having 1 to 20 carbon atoms which may have a substituent; n is a number of 1 to 100; when n is more than 1, R^2 groups may be the same or different.

[0063]

Examples of the hydrocarbyl group having 1 to 30 carbon atoms which may have a substituent of R^1 include an alkyl group having 1 to 30 carbon atoms which may have a substituent, an aralkyl group having 7 to 30 carbon atoms which may have a substituent, an aryl group having 6 to 30 carbon atoms which may have a substituent and the like.

[0064]

Examples of the alkyl group having 1 to 30 carbon atoms which may have a substituent of R^1 include an alkyl group having 1 to 30 carbon atoms, an alkyl group having 1 to 30 carbon atoms which has a halogen atom as a substituent, an alkyl group having 1 to 30 carbon atoms which has a substituted silyl group as a substituent, an alkyl group having 1 to 30 carbon atoms which has a substituted amino group as a substituent, an alkyl group having 1 to 30 carbon atoms which has a hydrocarbyloxy group as a substituent and the like.

[0065]

Examples of the alkyl group having 1 to 30 carbon atoms of R^1 include a methyl group, an ethyl group, a n-propyl group, an isopropyl group, a n-butyl group, a sec-butyl group, a tert-butyl group, a n-pentyl group, a neopentyl group, an isopentyl group, a n-hexyl group, a n-heptyl group, a n-octyl group, a n-nonyl group, a n-decyl group, a n-undecyl group, a n-dodecyl group, a n-tridecyl group, a n-tetradecyl group, a n-pentadecyl group, a n-hexadecyl group, a n-heptadecyl group, a n-octadecyl group, a n-nonadecyl group, a n-eicosyl group, a n-heneicosyl group, a n-docosyl group, a n-tricosyl group, a n-tetracosyl group, a n-pentacosyl group, a n-hexacosyl group, a n-heptacosyl group, a n-octacosyl group, a n-nonacosyl group, a n-triacontyl group and the like.

[0066]

Examples of the alkyl group having 1 to 30 carbon atoms which has a halogen atom as a substituent of R^1 include a fluoromethyl group, a difluoromethyl group, a trifluoromethyl group, a chloromethyl group, a dichloromethyl group, a trichloromethyl group, a bromomethyl group, a dibromomethyl group, a tribromomethyl group, an iodomethyl group, a diiodomethyl group, a triiodomethyl group, a fluoroethyl group, a difluoroethyl group, a trifluoroethyl group, a tetrafluoroethyl group, a pentafluoroethyl group, a chloroethyl group, a dichloroethyl group, a trichloroethyl group, a tetrachloroethyl group, a pentachloroethyl group, a bromoethyl group, a dibromoethyl group, a tribromoethyl group, a tetrabromoethyl group, a pentabromoethyl group, a perfluoropropyl group, a perfluorobutyl group, a perfluoropentyl group, a perfluorohexyl group, a perfluorooctyl group, a perfluorododecyl group, a perfluoropentadecyl group, a perfluoroeicosyl group, a perfluorotriacontyl group, a perchloropropyl group, a perchlorobutyl group, a perchloropentyl group, a perchlorohexyl group, a perchlorooctyl group, a perchlorododecyl group, a perchloropentadecyl group, a perchloroeicosyl group, a perchlorotriacontyl group, a perbromopropyl group, a perbromobutyl group, a perbromopentyl group, a perbromohexyl group, a perbromooctyl group, a perbromododecyl group, a perbromopentadecyl group, a perbromoeicosyl group, a perbromotriacontyl group and the like.

[0067]

Examples of the alkyl group having 1 to 30 carbon atoms which has a substituted silyl group as a substituent of R^1 include a trimethylsilylmethyl group, a trimethylsilylethyl group, a trimethylsilylpropyl group, a trimethylsilylbutyl group, a bis(trimethylsilyl)methyl group, a bis(trimethylsilyl)ethyl group, a bis(trimethylsilyl)propyl group, a bis(trimethylsilyl)butyl group, a triphenylsilylmethyl group and the like.

[0068]

Examples of the alkyl group having 1 to 30 carbon atoms which has a substituted amino group as a substituent of R^1 include a dimethylaminomethyl group, a dimethylaminoethyl group, a dimethylaminopropyl group, a dimethylaminobutyl group, a bis(dimethylamino)methyl group, a bis(dimethylamino)ethyl group, a bis(dimethylamino)propyl group, a bis(dimethylamino)butyl group, a phenylaminomethyl group, a diphenylaminomethyl group and the like.

[0069]

Examples of the alkyl group having 1 to 30 carbon atoms which has a hydrocarbyloxy group as a substituent of R^1 include a methoxymethyl group, an ethoxymethyl group, a n-propoxymethyl group, an isopropoxymethyl group, a n-butoxymethyl group, a sec-butoxymethyl group, a tert-butoxymethyl group, a phenoxymethyl group, a methoxyethyl group, an ethoxyethyl group, a n-propoxyethyl group, an isopropoxyethyl group, a n-butoxyethyl group, a sec-butoxyethyl group, a tert-butoxyethyl group, a phenoxyethyl group, a methoxy-n-propyl group, an ethoxy -n-propyl group, a n-propoxy-n-propyl group, an isopropoxy -n-propyl group, a n-butoxy-n-propyl group, a sec-butoxy-n-propyl group, a tert-butoxy-n-propyl group, a phenoxy-n-propyl group, a methoxyisopropyl group, an ethoxyisopropyl group, a n-propoxyisopropyl group, an isopropoxyisopropyl group, a n-butoxyisopropyl group, a sec-butoxyisopropyl group, a tert-butoxyisopropyl group, a phenoxyisopropyl group and the like.

[0070]

Examples of the aralkyl group having 7 to 30 carbon atoms which may have a substituent of R^1 include an aralkyl group having 7 to 30 carbon atoms, an aralkyl group having 7 to 30 carbon atoms which has a halogen atom as a substituent and the like.

[0071]

Examples of the aralkyl group having 7 to 30 carbon atoms of R^1 include a benzyl group, a (2-methylphenyl)methyl group, a (3-methylphenyl)methyl group, a (4-methylphenyl)methyl group, a (2,3-dimethylphenyl)methyl group, a (2,4-dimethylphenyl)methyl group, a (2,5-dimethylphenyl)methyl group, a (2,6-dimethylphenyl)methyl group, a (3,4-dimethylphenyl)methyl group, a (4,6-dimethylphenyl)methyl group, a (2,3,4-trimethylphenyl)methyl group, a (2,3,5-trimethylphenyl)methyl group, a (2,3,6-trimethylphenyl)methyl group, a (3,4,5-trimethylphenyl)methyl group, a (2,4,6-trimethylphenyl)methyl group, a (2,3,4,5-tetramethylphenyl)methyl group, a (2,3,4,6-tetramethylphenyl)methyl group, a (2,3,5,6-tetramethylphenyl)methyl group, a (pentamethylphenyl)methyl group, an (ethylphenyl)methyl group, a (n-propylphenyl)methyl group, an (isopropylphenyl)methyl group, a (n-butylphenyl)methyl group, a (sec-butylphenyl)methyl group, a (tert-butylphenyl)methyl group, a (n-pentylphenyl)methyl group, a (neopentylphenyl)methyl group, a (n-hexylphenyl)methyl group, a (n-octylphenyl)methyl group, a (n-decylphenyl)methyl group, a (n-decylphenyl)methyl group, a (n-tetradecylphenyl)methyl group, a naphthylmethyl group, an anthracenylmethyl group, a phenylethyl group, a phenylpropyl group, a phenylbutyl group, a diphenylmethyl group, a diphenylethyl group, a diphenylpropyl group, a diphenylbutyl group and the like.

[0072]

Examples of the aralkyl group having 7 to 30 carbon atoms which has a halogen atom as a substituent of R^1 include a 2-fluorobenzyl group, a 3-fluorobenzyl group, a 4-fluorobenzyl group, a 2-chlorobenzyl group, a 3-chlorobenzyl group, a 4-chlorobenzyl group, a 2-bromobenzyl group, a 3-bromobenzyl group, a 4-bromobenzyl group, a 2-iodobenzyl group, a 3-iodobenzyl group, a 4-iodobenzyl group and the like.

[0073]

Examples of the aryl group having 6 to 30 carbon atoms which may have a substituent of R^1 include an aryl group having 6 to 30 carbon atoms, an aryl group having 6 to 30 carbon atoms which has a halogen atom as a substituent, an aryl group having 6 to 30 carbon atoms which has a substituted silyl group as a substituent, an aryl group having 6 to 30 carbon atoms which has a substituted amino group as a substituent, an aryl group having 6 to 30 carbon atoms which has a hydrocarbyloxy group as a substituent and the like.

[0074]

Examples of the aryl group having 6 to 30 carbon atoms of R^1 include a phenyl group, a 2-tolyl group, a 3-tolyl group, a 4-tolyl group, a 2,3-xylyl group, a 2,4-xylyl group, a 2,5-xylyl group, a 2,6-xylyl group, a 3,4-xylyl group, a 3,5-xylyl group, a 2,3,4-trimethylphenyl group, a 2,3,5-trimethylphenyl group, a 2,3,6-trimethylphenyl group, a 2,4,6-trimethylphenyl group, a 3,4,5-trimethylphenyl group, a 2,3,4,5-

tetramethylphenyl group, a 2,3,4,6-tetramethylphenyl group, a 2,3,5,6-tetramethylphenyl group, a pentamethylphenyl group, an ethylphenyl group, a diethylphenyl group, a triethylphenyl group, a n-propylphenyl group, an isopropylphenyl group, a n-butylphenyl group, a sec-butylphenyl group, a tert-butylphenyl group, a n-pentylphenyl group, a neopentylphenyl group, a n-hexylphenyl group, a n-octylphenyl group, a n-decylphenyl group, a n-dodecylphenyl group, a n-tetradecylphenyl group, a naphthyl group, an anthracenyl group and the like.

[0075]

Examples of the aryl group having 6 to 30 carbon atoms which has a halogen atom as a substituent of R^1 include a 2-fluorophenyl group, a 3-fluorophenyl group, a 4-fluorophenyl group, a 2-chlorophenyl group, a 3-chlorophenyl group, a 4-chlorophenyl group, a 2-bromophenyl group, a 3-bromophenyl group, a 4-bromophenyl group, a 2-iodophenyl group, a 3-iodophenyl group, a 4-iodophenyl group and the like.

[0076]

Examples of the aryl group having 6 to 30 carbon atoms which has a substituted silyl group as a substituent of R^1 include a trimethylsilylphenyl group, a bis(trimethylsilyl)phenyl group and the like.

[0077]

Examples of the aryl group having 6 to 30 carbon atoms which has a substituted amino group as a substituent of R^1 include a dimethylaminophenyl group, a bis(dimethylamino)phenyl group, a diphenylaminophenyl group and the like.

[0078]

Examples of the aryl group having 6 to 30 carbon atoms which has a hydrocarbyloxy group as a substituent of R^1 include a methoxyphenyl group, an ethoxyphenyl group, a n-propoxyphenyl group, an isopropoxyphenyl group, a n-butoxyphenyl group, a sec-butoxyphenyl group, a tert-butoxyphenyl group, a phenoxyphenyl group and the like.

[0079]

R^1 in Formula [2] is preferably an alkyl group having 1 to 30 carbon atoms which may have a substituent, and more preferably an alkyl group having 1 to 30 carbon atoms.

[0080]

R^2 represents an alkylene group having 1 to 20 carbon atoms which may have a substituent, and R^2 groups may be the same or different.

[0081]

Examples of the alkylene group having 1 to 20 carbon atoms which may have a substituent of R^2 include an alkylene group having 1 to 20 carbon atoms ,an alkylene group having 1 to 20 carbon atoms which has a halogen atom as a substituent, an alkylene group having 1 to 20 carbon atoms which has a substituted silyl group as a substituent, an alkylene group having 1 to 20 carbon atoms which has a substituted amino group as a substituent, an alkylene group having 1 to 20 carbon atoms which has a hydrocarbyloxy group as a substituent and the like.

[0082]

Examples of the alkylene group having 1 to 20 carbon atoms of R^2 include a methylene group, an ethylene group, a propylene group, a butylene group, a pentylene group, a hexylene group, a heptylene group, an octylene group, a nonylene group, a decylene group, a undecylene group, a dodecylene group, a tridecylene group, a tetradecylene group, a pentadecylene group, a hexadecylene group, a heptadecylene group, an octadecylene group, a nonadecylene group, an eicosylene group, a methylmethylene group, a dimethylmethylene group, an ethylmethylene group, an ethylmethylmethylene group, a diethylmethylene group, a propylenemethylene group, a methylethylene group, a dimethylethylene group, a trimethylethylene group, a tetramethylethylene group and the like.

[0083]

Examples of the alkylene group having 1 to 20 carbon atoms which has a halogen atom as a substituent of R^2 include a fluoromethylene group, a difluoromethylene group, a chloromethylene group, a dichloromethylene group, a bromomethylene group, a dibromomethylene group, a iodomethylene group, a diiodomethylene group, a fluoroethylene group, a difluoroethylene group, a trifluoroethylene group, a tetrafluoroethylene group, a chloroethylene group, a dichloroethylene group, a trichloroethylene group, a tetrachloroethylene group, a bromoethylene group, a dibromoethylene group, a tribromoethylene group, a tetrabromoethylene group, a perfluoropropylene group, a perfluorobutylene group, a perfluoropentylene group, a perfluorohexylene group, a perfluorooctylene group, a perfluorododecylene group, a perfluoropentadecylene group, a perfluoroeicosylene group, a perchloropropylene group, a perchlorobutylene group, a perchloropentylene group, a perchlorohexylene group, a perchlorooctylene group, a perchlorododecylene group, a perchloropentadecylene group, a perchloroeicosylene group, a perbromopropylene group, a perbromobutylene group, a perbromopentylene group, a perbromohexylene group, a perbromooctylene group, a perbromododecylene group, a perbromopentadecylene group, a perbromoeicosylene group and the like.

[0084]

Examples of the alkylene group having 1 to 30 carbon atoms which has a substituted silyl group as a substituent of R^2 include a trimethylsilylmethylene group, a trimethylsilylethylene group, a trimethylsilylpropylene group, a trimethylsilylbutylene group, a bis(trimethylsilyl)methylene group, a bis(trimethylsilyl)ethylene group, a bis(trimethylsilyl)propylene group, a bis(trimethylsilyl)butylene group, a triphenylsilylmethyl group and the like.

[0085]

Examples of the alkylene group having 1 to 30 carbon atoms which has a substituted amino group as a substituent of R^2 include a dimethylaminomethylene group, a dimethylaminoethylene group, a dimethylaminopropylene group, a dimethylaminobutylene group, a bis(dimethylamino)methylene group, a bis(dimethylamino)ethylene group, a bis(dimethylamino)propylene group, a bis(dimethylamino)butylene group, a phenylaminomethylene group, a diphenylaminomethylene group and the like.

[0086]

Examples of the alkylene group having 1 to 20 carbon atoms which has a hydrocarbyloxy group as a substituent of R^2 include a methoxymethylene group, an ethoxy methylene group, a npropoxymethylene group, an isopropoxy methylene group, a n-butoxymethylene group, a sec-butoxymethylene group, a tert-butoxymethylene group, a phenoxymethylene group, a methoxyethylene group, an ethoxy ethylene group, a n-propoxyethylene group, an isopropoxyethylene group, a n-butoxyethylene group, a sec-butoxyethylene group, a tert-butoxyethylene group, a phenoxyethylene group, a methoxy-n-propylene group, an ethoxy-n-propylene group, a n-propoxy-n-propylene group, an isopropoxy-n-propylene group, a n-butoxy-n-propylene group, a sec-butoxy-n-propylene group, a tert-butoxy-n-propylene group, a phenoxy-n-propylene group, a methoxyisopropylene group, an ethoxy isopropylene group, a n-propoxyisopropylene group, an isopropoxy isopropylene group, a n-butoxyisopropylene group, a sec-butoxyisopropylene group, a tert-butoxyisopropylene group, a phenoxyisopropylene group and the like.

[0087]

R^2 in Formula [2] is preferably an alkylene group having 1 to 20 carbon atoms, more preferably a methylene group, an ethylene group, a propylene group, a butylene group, a pentylene group, or a hexylene group, and still more preferably an ethylene group.

[0088]

Q^1 represents a hydrogen atom, $-C(=O)OM^2$, $-R^3-C(=O)OM^2$, $-S(=O)_2OM^2$, $-R^3-S(=O)_2OM^2$, $-P(=O)(OH)(OM^2)$, $-P(=O)(OR^4)(OM^2)$, or $-P(=O)(OM^2)_2$.

[0089]

Q^1 is preferably a hydrogen atom, $-C(=O)OM^2$, $-R^3-C(=O)OM^2$, $-S(=O)_2OM^2$, or $-R^3-S(=O)_2OM^2$, and more preferably hydrogen atom, $-R^3$-

$C(=O)OM^2$, or $-S(=O)_2OM^2$.

[0090]

In the above exemplification, M^2 represents an alkali metal atom.

[0091]

M^2 is preferably a sodium atom or a potassium atom, and more preferably a sodium atom.

[0092]

In the above exemplification, R^3 represents an alkylene group having 1 to 20 carbon atoms which may have a substituent.

[0093]

Examples of the alkylene group having 1 to 20 carbon atoms which may have a substituent of R^3 include the same alkylene groups as those exemplified in R^2.

[0094]

R^3 is preferably an alkylene group having 1 to 20 carbon atoms, more preferably a methylene group, an ethylene group, a propylene group, a butylene group, a pentylene group, or a hexylene group, and still more preferably a methylene group.

[0095]

R^4 represents a hydrocarbyl group having 1 to 20 carbon atoms which may have a substituent.

[0096]

Examples of the hydrocarbyl group having 1 to 20 carbon atoms which may have a substituent of R^4 include an alkyl group having 1 to 20 carbon atoms which may have a substituent, an aralkyl group having 7 to

20 carbon atoms which may have a substituent, an aryl group having 6 to 20 carbon atoms which may have a substituent and the like.

[0097]

Examples of the alkyl group having 1 to 20 carbon atoms which may have a substituent of R^4 include an alkyl group having 1 to 20 carbon atoms, an alkyl group having 1 to 20 carbon atoms which has a halogen atom as a substituent, an alkyl group having 1 to 20 carbon atoms which has a substituted silyl group as a substituent, an alkyl group having 1 to 20 carbon atoms which has a substituted amino group as a substituent, an alkyl group having 1 to 20 carbon atoms which has a hydrocarbyloxy group as a substituent and the like.

[0098]

Examples of the alkyl group having 1 to 20 carbon atoms of R^4 include a methyl group, an ethyl group, a n-propyl group, an isopropyl group, a n-butyl group, a sec-butyl group, a tert-butyl group, a n-pentyl group, a neopentyl group, an isopentyl group, a n-hexyl group, a n-heptyl group, a n-octyl group, a n-nonyl group, a n-decyl group, a n-undecyl group, a n-dodecyl group, a n-tridecyl group, a n-tetradecyl group, a n-pentadecyl group, a n-hexadecyl group, a n-heptadecyl group, a n-octadecyl group, a n-nonadecyl group, a n-eicosyl group and the like.

[0099]

Examples of the alkyl group having 1 to 20 carbon atoms which has a halogen atom as a substituent of R^4 include a fluoromethyl group, a difluoromethyl group, a trifluoromethyl group, a chloromethyl group, a dichloromethyl group, a trichloromethyl group, a bromomethyl group, a dibromomethyl group, a tribromomethyl group, an iodomethyl group, a diiodomethyl group, a triiodomethyl group, a fluoroethyl group, a difluoroethyl group, a trifluoroethyl group, a tetrafluoroethyl group, a pentafluoroethyl group, a chloroethyl group, a dichloroethyl group, a trichloroethyl group, a tetrachloroethyl group, a pentachloroethyl group, a bromoethyl group, a dibromoethyl group, a tribromoethyl group, a tetrabromoethyl group, a pentabromoethyl group, a perfluoropropyl group, a perfluorobutyl group, a perfluoropentyl group, a perfluorohexyl group, a perfluorooctyl group, a perfluorododecyl group, a perfluoropentadecyl group, a perfluoroeicosyl group, a perchloropropyl group, a perchlorobutyl group, a perchloropentyl group, a perchlorohexyl group, a perchlorooctyl group, a perchlorododecyl group, a perchloropentadecyl group, a perchloroeicosyl group, a perbromopropyl group, a perbromobutyl group, a perbromopentyl group, a perbromohexyl group, a perbromooctyl group, a perbromododecyl group, a perbromopentadecyl group, a perbromoeicosyl group and the like.

[0100]

Examples of the alkyl group having 1 to 20 carbon atoms which has a substituted silyl group as a substituent of R^4 include a trimethylsilylmethyl group, a trimethylsilylethyl group, a trimethylsilylpropyl group, a trimethylsilylbutyl group, a bis(trimethylsilyl)methyl group, a bis(trimethylsilyl)ethyl group, a bis(trimethylsilyl)propyl group, a bis(trimethylsilyl)butyl group, a triphenylsilylmethyl group and the like.

[0101]

Examples of the alkyl group having 1 to 20 carbon atoms which has a substituted amino group of R^4 as a substituent include a dimethylaminomethyl group, a dimethylaminoethyl group, a dimethylaminopropyl group, a dimethylaminobutyl group, a bis(dimethylamino)methyl group, a bis(dimethylamino)ethyl group, a bis(dimethylamino)propyl group, a bis(dimethylamino)butyl group, a phenylaminomethyl group, a diphenylaminomethyl group and the like.

[0102]

Examples of the alkyl group having 1 to 20 carbon atoms which has a hydrocarbyloxy group as a substituent of R^4 include a methoxymethyl group, an ethoxymethyl group, a n-propoxymethyl group, an isopropoxymethyl group, a n-butoxymethyl group, a sec-butoxymethyl group, a tert-butoxymethyl group, a phenoxymethyl group, a methoxyethyl group, an ethoxyethyl group, a n-propoxyethyl group, an isopropoxyethyl group, a n-butoxyethyl group, a sec-butoxyethyl group, a tert-butoxyethyl group, a phenoxyethyl group, a methoxy-n-propyl group, an ethoxy-n-propyl group, a n-propoxy-n-propyl group, an isopropoxy-n-propyl group, a n-butoxy-n-propyl group, a sec-butoxy-n-propyl group, a tert-butoxy-n-propyl group, a phenoxy-n-propyl group, a methoxyisopropyl group, an ethoxyisopropyl group, a n-propoxyisopropyl group, an isopropoxyisopropyl group, a n-butoxyisopropyl group, a sec-butoxyisopropyl group, a tert-butoxyisopropyl group, a phenoxyisopropyl group and the like.

[0103]

Examples of the aralkyl group having 7 to 20 carbon atoms which may have a substituent of R^4 include an aralkyl group having 7 to 20 carbon atoms, an aralkyl group having 7 to 20 carbon atoms which has a halogen atom as a substituent and the like.

[0104]

Examples of the aralkyl group having 7 to 20 carbon atoms of R^4 include a benzyl group, a (2-methylphenyl)methyl group, a (3-methylphenyl)methyl group, a (4-methylphenyl)methyl group, a (2,3-dimethylphenyl)methyl group, a (2,4-dimethylphenyl)methyl group, a (2,5-dimethylphenyl)methyl group, a (2,6-dimethylphenyl)methyl group, a (3,4-dimethylphenyl)methyl group, a (4,6-dimethylphenyl)methyl group, a (2,3,4-trimethylphenyl)methyl group, a (2,3,5-trimethylphenyl)methyl group, a (2,3,6-trimethylphenyl)methyl group, a (3,4,5-trimethylphenyl)methyl group, a (2,4,6-trimethylphenyl)methyl group, a (2,3,4,5-tetramethylphenyl)methyl group, a (2,3,4,6-tetramethylphenyl)methyl group, a (2,3,5,6-tetramethylphenyl)methyl group, a (pentamethylphenyl)methyl group, an (ethylphenyl)methyl group, a (n-propylphenyl)methyl group, an (isopropylphenyl)methyl group, a (n-butylphenyl)methyl group, a (sec-butylphenyl)methyl group, a (tert-butylphenyl)methyl group, a (n-pentylphenyl)methyl group, a (neopentylphenyl)methyl group, a (n-hexylphenyl)methyl group, a (n-octylphenyl)methyl group, a (n-decylphenyl)methyl group, a (n-decylphenyl)methyl group, a (n-tetradecylphenyl)methyl group, a naphthylmethyl group, an anthracenylmethyl group, a phenylethyl group, a phenylpropyl group, a phenylbutyl group, a diphenylmethyl group, a diphenylethyl group, a diphenylpropyl group, a diphenylbutyl group and the like.

[0105]

Examples of the aralkyl group having 7 to 20 carbon atoms which has a halogen atom as a substituent of R^4 include a 2-fluorobenzyl group, a 3-fluorobenzyl group, a 4-fluorobenzyl group, a 2-chlorobenzyl group, a 3-chlorobenzyl group, a 4-chlorobenzyl group, a 2-bromobenzyl group, a 3-bromobenzyl group, a 4-bromobenzyl group, a 2-iodobenzyl group, a 3-iodobenzyl group, a 4-iodobenzyl group and the like.

[0106]

Examples of the aryl group having 6 to 20 carbon atoms of R^4 include a phenyl group, a 2-tolyl group, a 3-tolyl group, a 4-tolyl group, a 2,3-xylyl group, a 2,4-xylyl group, a 2,5-xylyl group, a 2,6-xylyl group, a 3,4-xylyl group, a 3,5-xylyl group, a 2,3,4-trimethylphenyl group, a 2,3,5-trimethylphenyl group, a 2,3,6-trimethylphenyl group, a 2,4,6-trimethylphenyl group, a 3,4,5-trimethylphenyl group, a 2,3,4,5-tetramethylphenyl group, a 2,3,4,6-tetramethylphenyl group, a 2,3,5,6-tetramethylphenyl group, a pentamethylphenyl group, an ethylphenyl group, a n-propylphenyl group, an isopropylphenyl group, a n-butylphenyl group, a sec-butylphenyl group, a tert-butylphenyl group, a n-pentylphenyl group, a neopentylphenyl group, a n-hexylphenyl group, a n-octylphenyl group, a n-decylphenyl group, a n-dodecylphenyl group, a n-tetradecylphenyl group, a naphthyl group, an anthracenyl group and the like.

[0107]

Examples of the aryl group having 6 to 20 carbon atoms which is substituted with a halogen atom of R^4 include a 2-fluorophenyl group, a 3-fluorophenyl group, a 4-fluorophenyl group, a 2-chlorophenyl group, a 3-chlorophenyl group, a 4-chlorophenyl group, a 2-bromophenyl group, a 3-bromophenyl group, a 4-bromophenyl group, a 2-iodophenyl group, a 3-

iodophenyl group, a 4-iodophenyl group and the like.

[0108]

R^4 is preferably an alkyl group having 1 to 20 carbon atoms, more preferably an alkyl group having 1 to 5 carbon atoms, and still more preferably a methyl group, an ethyl group, a n-propyl group or an isopropyl group.

[0109]

n represents a number of 1 to 100.

[0110]

n is preferably a number of 1 to 50, more preferably a number of 1 to 30, and still more preferably a number of 1 to 12.

[0111]

Examples of the organic compound (D) include polyoxyethylene lauryl ether, polyoxyethylene lauryl ether acetic acid, sodium polyoxyethylene lauryl ether acetate, potassium polyoxyethylene lauryl ether acetate, ammonium polyoxyethylene lauryl ether acetate, triethanolamine polyoxyethylene lauryl ether acetate, polyoxyethylene lauryl ether sulfuric acid, sodium polyoxyethylene lauryl ether sulfate, potassium polyoxyethylene lauryl ether sulfate, ammonium polyoxyethylene lauryl ether sulfate, triethanolamine polyoxyethylene lauryl ether sulfate, polyoxyethylene lauryl ether phosphoric acid, sodium polyoxyethylene lauryl ether phosphate, potassium polyoxyethylene lauryl ether phosphate, ammonium polyoxyethylene lauryl ether phosphate, triethanolamine polyoxyethylene lauryl ether phosphate and the like.

[0112]

The organic compound (D) is preferably polyoxyethylene lauryl ether, polyoxyethylene lauryl ether acetic acid, sodium polyoxyethylene lauryl ether acetate, polyoxyethylene lauryl ether sulfuric acid, or sodium polyoxyethylene lauryl ether sulfate, and more preferably sodium polyoxyethylene lauryl ether acetate.

[0113]

Activating Agent (B)

The activating agent (B) to be used in the present invention may be an agent which can activate the compound (A). When a catalyst for addition polymerization is applied to polymerization which forms addition polymer particles (e.g., slurry polymerization, gas phase polymerization, bulk polymerization, etc.), it is preferred to use specific particles as one components of a catalyst for addition polymerization because addition polymers having a fixed shape are produced. It is preferable to use, as the specific particles, modified particles of the following (I), (II) or (III):

[0114]

(I): modified particles produced by bringing (a), (b), (c) and (d) described below into contact with each other,

(a): a compound represented by Formula [3],

$M^3L^2_2$ [3]

(b): a compound represented by Formula [4],

$R^5_{t-1}TH$ [4]

(c): a compound represented by Formula [5], and

$R^6_{u-2}T'H_2$ [5]

(d): inorganic oxide particles or organic polymer particles, wherein M^3 is a Group 12 element; L^2 is a hydrogen atom, a halogen atom or a hydrocarbyl group, and two L^2 groups may be the same or different; R^5 is an electron withdrawing group or a group containing an electron withdrawing group; when t-1 is more than 1, R^5 groups may be the same or different; R^6 is a hydrocarbyl group or a halogenated hydrocarbyl group ;T is a Group 15 element or a Group 16 element; t is a number corresponding to the valence of T; T' is a Group 15 element or a Group 16 element; u is a number corresponding to the valence of T'.

[0115]

(II): modified particles produced by bringing inorganic oxide particles or organic polymer particles (d) into contact with an aluminoxane (e).

[0116]

(III): modified particles produced by bringing inorganic oxide particles or organic polymer particles (d), the aluminoxane (e) and the transition metal compound (A) into contact with one another.

These are further described below sequentially.

[0117]

M^3 in Formula [3] represents a Group 12 element of the Periodic Table of the elements (the 1989 IUPAC revised edition of inorganic chemical nomenclature). Examples of M^3 include a zinc atom, a cadmium atom and a mercury atom. M^3 is particularly preferably a zinc atom.

[0118]

In Formula [3], L^2 is a hydrogen atom, a halogen atom, or a hydrocarbyl group. Examples of the halogen atom of L^2 include a fluorine atom, a chlorine atom, a bromine atom, and an iodine atom. The hydrocarbyl group of L^2 is preferably an alkyl group, an aryl group, or an aralkyl group.

[0119]

The alkyl group of the hydrocarbyl group of L^2 is preferably an alkyl group having 1 to 20 carbon atoms, and examples thereof include a methyl group, an ethyl group, a n-propyl group, an isopropyl group, a n-butyl group, a sec-butyl group, a tert-butyl group, an isobutyl group, a n-pentyl group, a neopentyl group, a n-hexyl group, a n-heptyl group, a n-octyl group, a n-decyl group, a n-dodecyl group, a n-pentadecyl group and a n-eicosyl group, and a methyl group, an ethyl group, an isopropyl group, a tert-butyl group or an isobutyl group is more preferabe.

[0120]

Any of these alkyl groups may be substituted with a halogen atom such as a fluorine atom, a chlorine atom, a bromine atom or an iodine atom. Examples of the alkyl group having 1 to 20 carbon atoms which is substituted with a halogen atom can include a fluoromethyl group, a difluoromethyl group, a trifluoromethyl group, a fluoroethyl group, a difluoroethyl group, a trifluoroethyl group, a tetrafluoroethyl group, a pentafluoroethyl group, a perfluoropropyl group, a perfluorobutyl group, a perfluoropentyl group, a perfluorohexyl group, a perfluorooctyl group, a perfluorododecyl group, a perfluoropentadecyl group, a perfluoroeicosyl group, a 1H,1H-perfluoropropyl group, a 1H,1H-perfluorobutyl group, a 1H,1H-perfluoropentyl group, a 1H,1H-perfluorohexyl group, a 1H,1H-perfluorooctyl group, a 1H,1H-perfluorododecyl group, a 1H,1H-perfluoropentadecyl group, a 1H,1H-perfluoroeicosyl group, and alkyl groups in which “fluoro” of these alkyl groups is replaced by “chloro”, “bromo” or “iodo”. Any of these alkyl groups may be substituted with an alkoxy group such as a methoxy group or an ethoxy group; an aryloxy group such as a phenoxy group; or an aralkyloxy group such as a benzyloxy group.

[0121]

The aryl group of the hydrocarbyl group of L^2 is preferably an aryl group having 6 to 20 carbon atoms, and examples thereof include a phenyl group, a 2-tolyl group, a 3-tolyl group, a 4-tolyl group, a 2,3-xylyl group, a 2,4-xylyl group, a 2,5-xylyl group, a 2,6-xylyl group, a 3,4-xylyl group, a 3,5-xylyl group, a 2,3,4-trimethylphenyl group, a 2,3,5-trimethylphenyl group, a 2,3,6-trimethylphenyl group, a 2,4,6-trimethylphenyl group, a 3,4,5-trimethylphenyl group, a 2,3,4,5-tetramethylphenyl group, a 2,3,4,6-tetramethylphenyl group, a 2,3,5,6-tetramethylphenyl group, a pentamethylphenyl group, an ethylphenyl group, a n-propylphenyl group, an isopropylphenyl group, a n-butylphenyl group, a sec-butylphenyl group, a tert-butylphenyl group, an isobutylphenyl group, a n-pentylphenyl group, a neopentylphenyl group, a n-hexylphenyl group, a n-octylphenyl group, a n-decylphenyl group, a n-dodecylphenyl group, a n-tetradecylphenyl group, a naphthyl group and an anthracenyl group, and more preferably a phenyl group. Any of these aryl groups may be substituted with a halogen atom such as a fluorine atom, a chlorine atom, a bromine atom or an iodine atom; an alkoxy group such as a methoxy group or an ethoxy group; an aryloxy group such as a phenoxy group; or an aralkyloxy group such as a benzyloxy group.

[0122]

The aralkyl group of the hydrocarbyl group of L^2 is preferably an aralkyl group having 7 to 20 carbon atoms, and examples thereof can include a benzyl group, a (2-methylphenyl)methyl group, a (3-methylphenyl)methyl group, a (4-methylphenyl)methyl group, a (2,3-dimethylphenyl)methyl group, a (2,4-dimethylphenyl)methyl group, a (2,5-dimethylphenyl)methyl group, a (2,6-dimethylphenyl)methyl group, a (3,4-dimethylphenyl)methyl group, a (3,5-dimethylphenyl)methyl group, a (2,3,4-trimethylphenyl)methyl group, a (2,3,5-trimethylphenyl)methyl group, a (2,3,6-trimethylphenyl)methyl group, a (3,4,5-trimethylphenyl)methyl group, a (2,4,6-trimethylphenyl)methyl group, a (2,3,4,5-tetramethylphenyl)methyl group, a (2,3,4,6-tetramethylphenyl)methyl group, a (2,3,5,6-tetramethylphenyl)methyl group, a (pentamethylphenyl)methyl group, an (ethylphenyl)methyl group, a (n-propylphenyl)methyl group, an (isopropylphenyl)methyl group, a (n-butylphenyl)methyl group, a (sec-butylphenyl)methyl group, a (tert-butylphenyl)methyl group, an (isobutylphenyl)methyl group, a (n-pentylphenyl)methyl group, a (neopentylphenyl)methyl group, a (n-hexylphenyl)methyl group, a (n-octylphenyl)methyl group, a (n-decylphenyl)methyl group, a naphthylmethyl group and an anthracenylmethyl group. Among them, a benzyl group is more preferred. Any of these aralkyl groups may be substituted with a halogen atom such as a fluorine atom, a chlorine atom, a bromine atom or an iodine atom; an alkoxy group such as a methoxy group or an ethoxy group; an aryloxy group such as a phenoxy group; or an aralkyloxy group such as a benzyloxy group.

[0123]

L^2 in Formula [3] is preferably a hydrogen atom, an alkyl group or an aryl group, more preferably a hydrogen atom or an alkyl group, and particularly preferably an alkyl group. Two L^2 may be the same as or different from each other.

[0124]

T in Formula [4] represents a Group 15 element or a Group 16 element of the Periodic Table of the elements (the 1989 IUPAC revised edition of inorganic chemical nomenclature). Examples of the Group 15 element include a nitrogen atom, a phosphorus atom and the like; and examples of the Group 16 element include an oxygen atom, a sulfur atom and the like. T is preferably a nitrogen atom or an oxygen atom, and particularly preferably an oxygen atom.

t in Formula [4] represents the valence of T, and t is 3 when T is the Group 15 element and t is 2 when T is the Group 16 element.

[0125]

T' in Formula [5] represents a Group 15 element or a Group 16 element of the Periodic Table of the elements (the 1989 IUPAC revised edition of inorganic chemical nomenclature). Examples of the Group 15 element include a nitrogen atom, a phosphorus atom and the like; and examples of the Group 16 element include an oxygen atom, a sulfur atom and the like. T' is preferably a nitrogen atom or an oxygen atom, and particularly preferably an oxygen atom.

u in Formula [5] represents the valence of T', and u is 3 when T' is the Group 15 element and u is 2 when T' is the Group 16 element.

[0126]

R^2 in Formula [4] represents an electron withdrawing group or a group containing an electron withdrawing group and, when there are more than one R^2, they may be the same or different. A substituent constant σ of Hammett's rule is known as an indicator of electron withdrawing properties, and examples of the electron withdrawing group include functional groups having a positive substituent constant σ of Hammett's rule.

[0127]

Examples of the electron withdrawing group include a fluorine atom, a chlorine atom, a bromine atom, an iodine atom, a cyano group, a nitro group, a carbonyl group, a sulfone group, a phenyl group and the like. Examples of the group containing an electron withdrawing group include a halogenated alkyl group, a halogenated aryl group, a (halogenated alkyl)aryl group, a cyanated aryl group, a nitrated aryl group, an ester group (an alkoxycarbonyl group, an aralkyloxycarbonyl group, or an aryloxycarbonyl group) and the like.

[0128]

Examples of the halogenated alkyl group include a fluoromethyl group, a chloromethyl group, a bromomethyl group, an iodomethyl group, a difluoromethyl group, a dichloromethyl group, a dibromomethyl group, a diiodomethyl group, a trifluoromethyl group, a trichloromethyl group, a tribromomethyl group, a triiodomethyl group, a 2,2,2-trifluoroethyl group, a 2,2,2-trichloroethyl group, a 2,2,2-tribromoethyl group, a 2,2,2-triiodoethyl group, a 2,2,3,3,3-pentafluoropropyl group, a 2,2,3,3,3-pentachloropropyl group, a 2,2,3,3,3-pentabromopropyl group, a 2,2,3,3,3-pentaiodopropyl group, a 2,2,2-trifluoro-1-trifluoromethylethyl group, a 2,2,2-trichloro-1-trichloromethylethyl group, a 2,2,2-tribromo-1-tribromomethylethyl group, a 2,2,2-triiodo-1-triiodomethylethyl group, a 1,1-bis(trifluoromethyl)-2,2,2-trifluoroethyl group, a 1,1-bis(trichloromethyl)-2,2,2-trichloroethyl group, a 1,1-bis(tribromomethyl)-2,2,2-tribromoethyl group, a 1,1-bis(triiodomethyl)-2,2,2-triiodoethyl group and the like.

[0129]

Examples of the halogenated aryl group include a 2-fluorophenyl group, a 3-fluorophenyl group, a 4-fluorophenyl group, a 2,4-difluorophenyl group, a 2,6-difluorophenyl group, a 3,4-difluorophenyl group, a 3,5-difluorophenyl group, a 2,4,6-trifluorophenyl group, a 3,4,5-trifluorophenyl group, a 2,3,5,6-tetrafluorophenyl group, a pentafluorophenyl group, a 2,3,5,6-tetrafluoro-4-trifluoromethylphenyl group, a 2,3,5,6-tetrafluoro-4-pentafluorophenylphenyl group, a perfluoro-1-naphthyl group, a perfluoro-2-naphthyl group, a 2-chlorophenyl group, a 3-chlorophenyl group, a 4-chlorophenyl group, a 2,4-dichlorophenyl group, a 2,6-dichlorophenyl group, a 3,4-dichlorophenyl group, a 3,5-dichlorophenyl group, a 2,4,6-trichlorophenyl group, a 3,4,5-trichlorophenyl group, a 2,3,5,6-tetrachlorophenyl group, a pentachlorophenyl group, a 2,3,5,6-tetrachloro-4-trichloromethylphenyl group, a 2,3,5,6-tetrachloro-4-pentachlorophenylphenyl group, a perchloro-1-naphthyl group, a perchloro-2-naphthyl group, a 2-bromophenyl group, a 3-bromophenyl group, a 4-bromophenyl group, a 2,4-dibromophenyl group, a 2,6-dibromophenyl group, a 3,4-dibromophenyl group, a 3,5-dibromophenyl group, a 2,4,6-tribromophenyl group, a 3,4,5-tribromophenyl group, a 2,3,5,6-

tetrabromophenyl group, a pentabromophenyl group, a 2,3,5,6-tetrabromo-4-tribromomethylphenyl group, a 2,3,5,6-tetrabromo-4-pentabromophenylphenyl group, a perbromo-1-naphthyl group, a perbromo-2-naphthyl group, a 2-iodophenyl group, a 3-iodophenyl group, a 4-iodophenyl group, a 2,4-diiodophenyl group, a 2,6-diiodophenyl group, a 3,4-diiodophenyl group, a 3,5-diiodophenyl group, a 2,4,6-triiodophenyl group, a 3,4,5-triiodophenyl group, a 2,3,5,6-tetraiodophenyl group, a pentaiodophenyl group, a 2,3,5,6-tetraiodo-4-triiodomethylphenyl group, a 2,3,5,6-tetraiodo-4-pentaiodophenylphenyl group, a periodo-1-naphthyl group, a periodo-2-naphthyl group and the like.

[0130]

Examples of the (halogenated alkyl)aryl group include a 2-(trifluoromethyl)phenyl group, a 3-(trifluoromethyl)phenyl group, a 4-(trifluoromethyl)phenyl group, a 2,6-bis(trifluoromethyl)phenyl group, a 3,5-bis(trifluoromethyl)phenyl group, a 2,4,6-tris(trifluoromethyl)phenyl group, a 3,4,5-tris(trifluoromethyl)phenyl group and the like.

[0131]

Examples of the cyanated aryl group include a 2-cyanophenyl group, a 3-cyanophenyl group, a 4-cyanophenyl group and the like.

[0132]

Examples of the nitrated aryl group include a 2-nitrophenyl group, a 3-nitrophenyl group, a 4-nitrophenyl group and the like.

[0133]

Examples of the ester group include a methoxycarbonyl group, an ethoxycarbonyl group, a normal propoxycarbonyl group, an isopropoxycarbonyl group, a phenoxycarbonyl group, a trifluoromethoxycarbonyl group, a pentafluorophenoxycarbonyl group and the like.

[0134]

R^5 is preferably a halogenated hydrocarbyl group, and more preferably a halogenated alkyl group or a halogenated aryl group. R^5 is still more preferably a fluoromethyl group, a difluoromethyl group, a trifluoromethyl group, a 2,2,2-trifluoroethyl group, a 2,2,3,3,3-pentafluoropropyl group, a 2,2,2-trifluoro-1-trifluoromethylethyl group, a 1,1-bis(trifluoromethyl)-2,2,2-trifluoroethyl group, a 2-fluorophenyl group, a 3-fluorophenyl group, a 4-fluorophenyl group, a 2,4-difluorophenyl group, a 2,6-difluorophenyl group, a 3,4-difluorophenyl group, a 3,5-difluorophenyl group, a 2,4,6-trifluorophenyl group, a 3,4,5-trifluorophenyl group, a 2,3,5,6-tetrafluorophenyl group, a pentafluorophenyl group, a 2,3,5,6-tetrafluoro-4-trifluoromethylphenyl group, a 2,3,5,6-tetrafluoro-4-pentafluorophenylphenyl group, a perfluoro-1-naphthyl group, a perfluoro-2-naphthyl group, a chloromethyl group, a dichloromethyl group, a trichloromethyl group, a 2,2,2-trichloroethyl group, a 2,2,3,3,3-pentachloropropyl group, a 2,2,2-trichloro-1-trichloromethylethyl group, a 1,1-bis(trichloromethyl)-2,2,2-trichloroethyl group, a 4-chlorophenyl group, a 2,6-dichlorophenyl group, a 3,5-dichlorophenyl group, a 2,4,6-trichlorophenyl group, a 3,4,5-trichlorophenyl group or a pentachlorophenyl group, particularly preferably a fluoroalkyl group or a fluoroaryl group, and most preferably a trifluoromethyl group, a 2,2,2-trifluoro-1-trifluoromethylethyl group, a 1,1-bis(trifluoromethyl)-2,2,2-trifluoroethyl group, a 3,5-difluorophenyl group, a 3,4,5-trifluorophenyl group or a pentafluorophenyl group.

[0135]

R^6 in Formula [5] represents a hydrocarbyl group or a halogenated hydrocarbyl group. The hydrocarbyl group in R^6 is preferably an alkyl group, an aryl group or an aralkyl group, and the same hydrocarbyl group as that described as L^2 in Formula [5] is used. Examples of the halogenated hydrocarbyl group in R^6 include a halogenated alkyl group, a halogenated aryl group, a (halogenated alkyl)aryl group and the like, and the same halogenated alkyl group, halogenated aryl group and (halogenated alkyl)aryl group as those listed as examples of the electron withdrawing group in R^5 of Formula [4] are used.

[0136]

R^6 in Formula [5] is preferably a halogenated hydrocarbyl group, and more preferably a fluorinated hydrocarbyl group.

[0137]

Examples of the compound (a) to be used to obtain the modified particles (I) include, when M^3 is a zinc atom, dialkyl zinc such as dimethyl zinc, diethyl zinc, dipropyl zinc, di-n-butyl zinc, diisobutyl zinc or di-n-hexyl zinc; diaryl zinc such as diphenyl zinc, dinaphthyl zinc or bis(pentafluorophenyl)zinc; dialkenyl zinc such as diallyl zinc; halogenated alkyl zinc such as bis(cyclopentadienyl) zinc, methyl zinc chloride, ethyl zinc chloride, propyl zinc chloride, n-butyl zinc chloride, isobutyl zinc chloride, n-hexyl zinc chloride, methyl zinc bromide, ethyl zinc bromide, propyl zinc bromide, n-butyl zinc bromide, isobutyl zinc bromide, n-hexyl zinc bromide, methyl zinc iodide, ethyl zinc iodide, propyl zinc iodide, n-butyl zinc iodide, isobutyl zinc iodide or n-hexyl zinc iodide; and halogenated zinc such as zinc fluoride, zinc chloride, zinc bromide or zinc iodide.

[0138]

The compound (a) is preferably dialkyl zinc, more preferably dimethyl zinc, diethyl zinc, dipropyl zinc, di-n-butyl zinc, diisobutyl zinc or di-n-hexyl zinc, and particularly preferably dimethyl zinc or diethyl zinc.

[0139]

Examples of the amines of the compound (b) include di(fluoromethyl)amine, di(chloromethyl)amine, di(bromomethyl)amine, di(iodomethyl)amine, bis(difluoromethyl)amine, bis(dichloromethyl)amine, bis(dibromomethyl)amine, bis(diiodomethyl)amine, bis(trifluoromethyl)amine, bis(trichloromethyl)amine, bis(tribromomethyl)amine, bis(triiodomethyl)amine, bis(2,2,2-trifluoroethyl)amine, bis(2,2,2-trichloroethyl)amine, bis(2,2,2-tribromoethyl)amine, bis(2,2,2-triiodoethyl)amine, bis(2,2,3,3,3-pentafluoropropyl)amine, bis(2,2,3,3,3-pentachloropropyl)amine, bis(2,2,3,3,3-pentabromopropyl)amine, bis(2,2,3,3,3-pentaiodopropyl)amine, bis(2,2,2-trifluoro-1-trifluoromethylethyl)amine, bis(2,2,2-trichloro-1-trichloromethylethyl)amine, bis(2,2,2-tribromo-1-tribromomethylethyl)amine, bis(2,2,2-triiodo-1-triiodomethylethyl)amine, bis(1,1-bis(trifluoromethyl)-2,2,2-trifluoroethyl)amine, bis(1,1-bis(trichloromethyl)-2,2,2-trichloroethyl)amine, bis(1,1-bis(tribromomethyl)-2,2,2-tribromoethyl)amine, bis(1,1-bis(triiodomethyl)-2,2,2-triiodoethyl)amine, bis(2-fluorophenyl)amine, bis(3-fluorophenyl)amine, bis(4-fluorophenyl)amine, bis(2-chlorophenyl)amine, bis(3-chlorophenyl)amine, bis(4-chlorophenyl)amine, bis(2-bromophenyl)amine, bis(3-bromophenyl)amine, bis(4-bromophenyl)amine, bis(2-iodophenyl)amine, bis(3-iodophenyl)amine, bis(4-iodophenyl)amine, bis(2,6-difluorophenyl)amine, bis(3,5-difluorophenyl)amine, bis(2,6-dichlorophenyl)amine, bis(3,5-dichlorophenyl)amine, bis(2,6-dibromophenyl)amine, bis(3,5-dibromophenyl)amine, bis(2,6-diiodophenyl)amine, bis(3,5-diiodophenyl)amine, bis(2,4,6-trifluorophenyl)amine, bis(2,4,6-trichlorophenyl)amine, bis(2,4,6-tribromophenyl)amine, bis(2,4,6-triiodophenyl)amine, bis(3,4,5-trifluorophenyl)amine, bis(3,4,5-trichlorophenyl)amine, bis(3,4,5-tribromophenyl)amine, bis(3,4,5-triiodophenyl)amine, bis(pentafluorophenyl)amine, bis(pentachlorophenyl)amine, bis(pentabromophenyl)amine, bis(pentaiodophenyl)amine, bis(2-(trifluoromethyl)phenyl)amine, bis(3-(trifluoromethyl)phenyl)amine, bis(4-(trifluoromethyl)phenyl)amine, bis(2,6-di(trifluoromethyl)phenyl)amine, bis(3,5-di(trifluoromethyl)phenyl)amine, bis(2,4,6-tri(trifluoromethyl)phenyl)amine, bis(3,4,5-tri(trifluoromethyl)phenyl)amine, bis(2-cyanophenyl)amine, (3-cyanophenyl)amine, bis(4-cyanophenyl)amine, bis(2-nitrophenyl)amine, bis(3-nitrophenyl)amine, bis(4-nitrophenyl)amine and the like. Further examples of the compound (b) include phosphine compounds in which a nitrogen atom is substituted with a phosphorus atom. Those phosphine compounds are compounds in which amine of the above examples is replaced by phosphine.

[0140]

Examples of the alcohols of the compound (b) include fluoromethanol, chloromethanol, bromomethanol, iodomethanol, difluoromethanol, dichloromethanol, dibromomethanol, diiodomethanol, trifluoromethanol, trichloromethanol, tribromomethanol, triiodomethanol, 2,2,2-trifluoroethanol, 2,2,2-trichloroethanol, 2,2,2-tribromoethanol, 2,2,2-triiodoethanol, 2,2,3,3,3-pentafluoropropanol, 2,2,3,3,3-pentachloropropanol, 2,2,3,3,3-pentabromopropanol, 2,2,3,3,3-pentaiodopropanol, 2,2,2-trifluoro-1-trifluoromethylethanol, 2,2,2-trichloro-1-trichloromethylethanol, 2,2,2-tribromo-1-tribromomethylethanol, 2,2,2-triiodo-1-triiodomethylethanol, 1,1-bis(trifluoromethyl)-2,2,2-trifluoroethanol, 1,1-bis(trichloromethyl)-2,2,2-trichloroethanol, 1,1-bis(tribromomethyl)-2,2,2-tribromoethanol, 1,1-bis(triiodomethyl)-2,2,2-triiodoethanol and the like. Further examples of the compound (b) include thiol compounds in which an oxygen atom is substituted with a sulfur atom. Those thiol compounds are compounds in which methanol of the above examples is replaced by methanethiol, ethanol is replaced by ethanethiol, and propanol is replaced by propanethiol.

[0141]

Examples of the phenols of the compound (b) include 2-fluorophenol, 3-fluorophenol, 4-fluorophenol, 2,4-difluorophenol, 2,6-difluorophenol, 3,4-difluorophenol, 3,5-difluorophenol, 2,4,6-trifluorophenol, 3,4,5-trifluorophenol, 2,3,5,6-tetrafluorophenol, pentafluorophenol, 2,3,5,6-tetrafluoro-4-trifluoromethylphenol, 2,3,5,6-tetrafluoro-4-pentafluorophenylphenol, perfluoro-1-naphthol, perfluoro-2-naphthol, 2-chlorophenol, 3-chlorophenol, 4-chlorophenol, 2,4-dichlorophenol, 2,6-dichlorophenol, 3,4-dichlorophenol, 3,5-dichlorophenol, 2,4,6-trichlorophenol, 3,4,5-trichlorophenol, 2,3,5,6-tetrachlorophenol, pentachlorophenol, 2,3,5,6-tetrachloro-4-trichloromethylphenol, 2,3,5,6-tetrachloro-4-pentachlorophenylphenol, perchloro-1-naphthol, perchloro-2-naphthol, 2-bromophenol, 3-bromophenol, 4-bromophenol, 2,4-dibromophenol, 2,6-dibromophenol, 3,4-dibromophenol, 3,5-dibromophenol, 2,4,6-tribromophenol, 3,4,5-tribromophenol, 2,3,5,6-tetrabromophenol, pentabromophenol, 2,3,5,6-tetrabromo-4-tribromomethylphenol, 2,3,5,6-tetrabromo-4-pentabromophenylphenol, perbromo-1-naphthol, perbromo-2-naphthol, 2-iodophenol, 3-iodophenol, 4-iodophenol, 2,4-diiodophenol, 2,6-diiodophenol, 3,4-diiodophenol, 3,5-diiodophenol, 2,4,6-triiodophenol, 3,4,5-triiodophenol, 2,3,5,6-tetraiodophenol, pentaiodophenol, 2,3,5,6-tetraiodo-4-triiodomethylphenol, 2,3,5,6-tetraiodo-4-pentaiodophenylphenol, periodo-1-naphthol, periodo-2-naphthol, 2-(trifluoromethyl)phenol, 3-(trifluoromethyl)phenol, 4-(trifluoromethyl)phenol, 2,6-bis(trifluoromethyl)phenol, 3,5-bis(trifluoromethyl)phenol, 2,4,6-tris(trifluoromethyl)phenol, 3,4,5-tris(trifluoromethyl)phenol, 2-cyanophenol, 3-cyanophenol, 4-cyanophenol, 2-nitrophenol, 3-nitrophenol, 4-nitrophenol and the like. Further examples of the compound (b) include thiophenol compounds in which an oxygen atom is substituted with a sulfur atom. Those thiophenol compounds are compounds in which phenol of the above examples is replaced by thiophenol.

[0142]

Preferably, the amine of the compound (b) is bis(trifluoromethyl)amine, bis(2,2,2-trifluoroethyl)amine, bis(2,2,3,3,3-pentafluoropropyl)amine, bis(2,2,2-trifluoro-1-trifluoromethylethyl)amine, bis(1,1-bis(trifluoromethyl)-2,2,2-trifluoroethyl)amine or bis(pentafluorophenyl)amine, the alcohol of the compound (b) is trifluoromethanol, 2,2,2-trifluoroethanol, 2,2,3,3,3-pentafluoropropanol, 2,2,2-trifluoro-1-trifluoromethylethanol or 1,1-bis(trifluoromethyl)-2,2,2-trifluoroethanol, and the phenol of the compound (b) is 2-fluorophenol, 3-fluorophenol, 4-fluorophenol, 2,6-difluorophenol, 3,5-difluorophenol, 2,4,6-trifluorophenol, 3,4,5-trifluorophenol, pentafluorophenol, 2-(trifluoromethyl)phenol, 3-(trifluoromethyl)phenol, 4-(trifluoromethyl)phenol, 2,6-bis(trifluoromethyl)phenol, 3,5-bis(trifluoromethyl)phenol, 2,4,6-tris(trifluoromethyl)phenol or 3,4,5-tris(trifluoromethyl)phenol.

[0143]

The compound (b) is more preferably bis(trifluoromethyl)amine, bis(pentafluorophenyl)amine, trifluoromethanol, 2,2,2-trifluoro-1-trifluoromethylethanol, 1,1-bis(trifluoromethyl)-2,2,2-trifluoroethanol, 2-fluorophenol, 3-fluorophenol, 4-fluorophenol, 2,6-difluorophenol, 3,5-difluorophenol, 2,4,6-trifluorophenol, 3,4,5-trifluorophenol, pentafluorophenol, 4-(trifluoromethyl)phenol, 2,6-bis(trifluoromethyl)phenol or 2,4,6-tris(trifluoromethyl)phenol, and still more preferably 3,5-difluorophenol, 3,4,5-trifluorophenol, pentafluorophenol or 1,1-bis(trifluoromethyl)-2,2,2-trifluoroethanol.

[0144]

Examples of the compound (c) include water, hydrogen sulfide, amine and an aniline compound. Examples of the amine include alkylamines such as methylamine, ethylamine, n-propylamine, isopropylamine, n-butylamine, sec-butylamine, tert-butylamine, isobutylamine, n-pentylamine, neopentylamine, isopentylamine, n-hexylamine, n-octylamine, n-decylamine, n-dodecylamine, n-pentadecylamine and n-eicosylamine; aralkylamines such as allylamine, cyclopentadienylamine and benzylamine; fluoromethylamine, difluoromethylamine, trifluoromethylamine, 2,2,2-trifluoroethylamine, 2,2,3,3,3-pentafluoropropylamine, 2,2,2-trifluoro-1-trifluoromethylethylamine, 1,1-bis(trifluoromethyl)-2,2,2-trifluoroethylamine, perfluoropropylamine, perfluorobutylamine, perfluoropentylamine, perfluorohexylamine, perfluorooctylamine, perfluorododecylamine, perfluoropentadecylamine, perfluoroeicosylamine; and halogenated alkylamines in which “fluoro” of these amines are replaced by “chloro”, “bromo” or “iodo”.

[0145]

Examples of the aniline compound of the compound (c) include aniline, naphthylamine, anthracenylamine, 2-tolylamine, 3-tolylamine, 4-tolylamine, 2,3-xylylamine, 2,4-xylylamine, 2,5-xylylamine, 2,6-xylylamine, 3,4-xylylamine, 3,5-xylylamine, 2,3,4-trimethylaniline, 2,3,5-trimethylaniline, 2,3,6-trimethylaniline, 2,4,6-trimethylaniline, 3,4,5-trimethylaniline, 2,3,4,5-tetramethylaniline, 2,3,4,6-tetramethylaniline, 2,3,5,6-tetramethylaniline, pentamethylaniline, 2-ethylaniline, 3-ethylaniline, 4-ethylaniline, 2,3-diethylaniline, 2,4-diethylaniline, 2,5-diethylaniline, 2,6-diethylaniline, 3,4-diethylaniline, 3,5-diethylaniline, 2,3,4-triethylaniline, 2,3,5-triethylaniline, 2,3,6-triethylaniline, 2,4,6-triethylaniline, 3,4,5-triethylaniline, 2,3,4,5-tetraethylaniline, 2,3,4,6-

tetraethylaniline, 2,3,5,6-tetraethylaniline, pentaethylaniline, and alkylaniline in which "ethyl" of these compounds is replaced by "n-propyl", "isopropyl", "n-butyl", "sec-butyl", "tert-butyl", "n-pentyl", "neopentyl", "n-hexyl", "n-octyl", "n-decyl", "n-dodecyl" or "n-tetradecyl"; halogenated anilines such as 2-fluoroaniline, 3-fluoroaniline, 4-fluoroaniline, 2,6-difluoroaniline, 3,5-difluoroaniline, 2,4,6-trifluoroaniline, 3,4,5-trifluoroaniline and pentafluoroaniline; 2-(trifluoromethyl)aniline, 3-(trifluoromethyl)aniline, 4-(trifluoromethyl)aniline, 2,6-di(trifluoromethyl)aniline, 3,5-di(trifluoromethyl)aniline, 2,4,6-tri(trifluoromethyl)aniline, and (halogenated alkyl)aniline in which "fluoro" of these compounds is replaced by "chloro", "bromo" or "iodo".

[0146]

The compound (c) is preferably water, hydrogen sulfide, methylamine, ethylamine, n-propylamine, isopropylamine, n-butylamine, sec-butylamine, tert-butylamine, isobutylamine, n-octylamine, aniline, 2,6-xylylamine, 2,4,6-trimethylaniline, naphthylamine, anthracenylamine, benzylamine, trifluoromethylamine, pentafluoroethylamine, perfluoropropylamine, perfluorobutylamine, perfluoropentylamine, perfluorohexylamine, perfluorooctylamine, perfluorododecylamine, perfluoropentadecylamine, perfluoroeicosylamine, 2-fluoroaniline, 3-fluoroaniline, 4-fluoroaniline, 2,6-difluoroaniline, 3,5-difluoroaniline, 2,4,6-trifluoroaniline, 3,4,5-trifluoroaniline, pentafluoroaniline, 2-(trifluoromethyl)aniline, 3-(trifluoromethyl)aniline, 4-(trifluoromethyl)aniline, 2,6-bis(trifluoromethyl)aniline, 3,5-bis(trifluoromethyl)aniline or 2,4,6-tris(trifluoromethyl)aniline, particularly preferably water, trifluoromethylamine, perfluorobutylamine, perfluorooctylamine, perfluoropentadecylamine, 2-fluoroaniline, 3-fluoroaniline, 4-fluoroaniline, 2,6-difluoroaniline, 3,5-difluoroaniline, 2,4,6-trifluoroaniline, 3,4,5-trifluoroaniline, pentafluoroaniline, 2-(trifluoromethyl)aniline, 3-(trifluoromethyl)aniline, 4-(trifluoromethyl)aniline, 2,6-bis(trifluoromethyl)aniline, 3,5-bis(trifluoromethyl)aniline or 2,4,6-tris(trifluoromethyl)aniline, and most preferably water or pentafluoroaniline.

[0147]

Examples of (d) include inorganic oxide particles and organic polymer particles. Among them, porous particles having a uniform particle diameter, which are commonly used as a carrier, are preferable. From the viewpoint of the particle size distribution of the obtained addition polymer, regarding the particle size distribution of (d), the volume-based geometric standard deviation of the particle diameter of (d) is preferably 2.5 or less, more preferably 2.0 or less, and still more preferably 1.7 or less.

[0148]

Any inorganic oxide or a mixture of any inorganic oxide may be used as the inorganic oxide particles of (d). Examples of the inorganic oxide include SiO_2, Al_2O_3, MgO, ZrO_2, TiO_2, B_2O_3, CaO, ZnO, BaO, and ThO_2, and a mixture thereof, 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. These inorganic oxides are preferably SiO_2 and/or Al_2O_3, and particularly preferably SiO_2 (i.e., silica). The above inorganic oxide may contain a small amount of carbonates, sulfates, nitrates, and oxide components, such as Na_2CO_3, K_2CO_3, $CaCO_3$, $MgCO_3$, Na_2SO_4,

$Al_2(SO_4)_3$, $BaSO_4$, KNO_3, $Mg(NO_3)_2$, $Al(NO_3)_3$, Na_2O, K_2O, and Li_2O.

[0149]

The inorganic oxide is preferably dried to substantially remove moisture, and a drying method is preferably a method of drying by heating. The drying temperature is usually a temperature within a range from 100 to 1,500°C, preferably from 100 to 1,000°C, and still more preferably from 200 to 800°C, for the inorganic oxide in which moisture cannot be visually confirmed. There is no particular limitation on the drying time, and it is preferably from 10 minutes to 50 hours, and more preferably from 1 hour to 30 hours. Examples of the method of drying by heating include a method in which drying is carried out by circulating a dried inert gas (for example, nitrogen or argon, etc.) at a given flow rate during heating, or a method in which drying is carried out by heating under reduced pressure.

[0150]

Usually, hydroxyl groups are formed and present on the surface of an inorganic oxide. A modified inorganic oxide produced by substituting active hydrogen of surface hydroxyl groups with various substituents may be used as the inorganic oxide. Examples of the modified inorganic oxide include inorganic oxides subjected to a contact treatment with trialkylchlorosilane such as trimethylchlorosilane or tert-butyldimethylchlorosilane; triarylchlorosilane such as triphenylchlorosilane; dialkyldichlorosilane such as dimethyldichlorosilane; diaryldichlorosilane such as diphenyldichlorosilane; alkyltrichlorosilane such as methyltrichlorosilane; aryltrichlorosilane such as phenyltrichlorosilane; trialkylalkoxysilane such as trimethylmethoxysilane; triarylalkoxysilane such as triphenylmethoxysilane; dialkyldialkoxysilane such as dimethyldimethoxysilane; diaryldialkoxysilane such as diphenyldimethoxysilane; alkyltrialkoxysilane such as methyltrimethoxysilane; aryltrialkoxysilane such as phenyltrimethoxysilane; tetraalkoxysilane such as tetramethoxysilane; alkyldisilazane such as 1,1,1,3,3,3-hexamethyldisilazane; tetrachlorosilane; alcohol such as methanol or ethanol; phenol; dialkyl magnesium such as dibutyl magnesium, butylethyl magnesium or butyloctyl magnesium; and alkyl lithium such as butyl lithium.

[0151]

Further examples include inorganic oxides produced by subjecting inorganic oxides which have brought into contact with trialkyl aluminum to a contact treatment with dialkylamine such as diethylamine or diphenylamine; alcohol such as methanol or ethanol; or phenol.

[0152]

The strength of the inorganic oxide per se may be sometimes increased by the hydrogen bond of hydroxyl groups to each other. In that case, if all active hydrogens of surface hydroxyl groups are substituted with various substituents, a decrease in particle strength may sometimes occur. Therefore, it is not necessarily required to substitute all active hydrogens of surface hydroxyl groups of the inorganic oxide, and the substitution rate of the surface hydroxyl group may be appropriately determined. There is no particular limitation on a method of changing the substitution rate of the surface hydroxyl group. Examples of the method include a method of changing the amount of the compound to be used in the contact treatment.

[0153]

There is no particular limitation on the average particle diameter of inorganic oxide particles, and the average particle diameter is usually from 1 to 5,000 µm, preferably from 5 to 1,000 µm, more preferably from 10 to 500 µm, and still more preferably from 10 to 100 µm. The pore volume is preferably 0.1 ml/g or more, and more preferably from 0.3 to 10 ml/g. The specific surface area is preferably from 10 to 1,000 m^2/g, and more preferably from 100 to 500 m^2/g.

[0154]

Any organic polymer or a mixture of any organic polymer may be used as the organic polymer particles (d). The organic polymer is preferably a polymer which has a functional group containing active hydrogen, or a non-proton-donating Lewis basic functional group.

[0155]

There is no particular limitation on the functional group containing active hydrogen as long as the functional group has active hydrogen. Examples of the functional group include a primary amino group, a secondary amino group, an imino group, an amide group, a hydrazide group, an amidino group, a hydroxy group, a hydroperoxy group, a carboxyl group, a formyl group, a carbamoyl group, a sulfonic acid group, a sulfinic acid group, a sulfenic acid group, a thiol group, a thioformyl group, a pyrrolyl group, an imidazolyl group, a piperidyl group, an indazolyl group and a carbazolyl group. The functional group is preferably a primary amino group, a secondary amino group, an imino group, an amide group, an imide group, a hydroxy group, a formyl group, a carboxyl group, a sulfonic acid group or a thiol group, and particularly preferably a primary amino group, a secondary amino group, an amide group or a hydroxy group. These groups may be substituted with a halogen atom or a hydrocarbyl group having 1 to 20 carbon atoms.

[0156]

There is no particular limitation on the non-proton-donating Lewis basic functional group, as long as it is a functional group which has a Lewis basic moiety having no active hydrogen atom. Examples of the functional group include a pyridyl group, an N-substituted imidazolyl group, an N-substituted indazolyl group, a nitrile group, an azide group, an N-substituted imino group, an N,N-substituted amino group, an N,N-substituted aminooxy group, an N,N,N-substituted hydrazino group, a nitroso group, a nitro group, a nitrooxy group, a furyl group, a carbonyl group, a thiocarbonyl group, an alkoxy group, an alkyloxycarbonyl group, an N,N-substituted carbamoyl group, a thioalkoxy group, a substituted sulfinyl group, a substituted sulfonyl group, and a substituted sulfonic acid group. The functional group is preferably a heterocyclic group, and more preferably an aromatic heterocyclic group having an oxygen atom and/or a nitrogen atom in the ring. The functional group is particularly preferably a pyridyl group, an N-substituted imidazolyl group, or an N-substituted indazolyl group, and most preferably a pyridyl group. These groups may be substituted with a halogen atom, or a hydrocarbyl group having 1 to 20 carbon atoms.

[0157]

There is no particular limitation on the content of the functional group having active hydrogen or non-proton-donating Lewis basic functional group in the organic polymer. The content thereof is preferably from 0.01 to 50 mmol/g, and more preferably from 0.1 to 20 mmol/g, expressed by a molar amount of a functional group per gram of the organic polymer.

[0158]

Examples of a method for producing the organic polymer, which has a functional group having active hydrogen or a non-proton-donating Lewis basic functional group, include a method of homopolymerizing a monomer which has a functional group having active hydrogen or a non-proton-donating Lewis basic functional group, and one or more polymerizable unsaturated groups, or a method of copolymerizing the said monomer with the other monomer having a polymerizable unsaturated group. It is preferred to copolymerize together with a crosslinking polymerizable monomer having two or more polymerizable unsaturated groups at this time.

[0159]

Examples of the polymerizable unsaturated group include alkenyl groups such as a vinyl group and an allyl group; and alkynyl groups such as an ethyne group. Examples of the monomer which has a functional group having active hydrogen and one or more polymerizable unsaturated groups include a vinyl group-containing primary amine, a vinyl group-containing secondary amine, a vinyl group-containing amide compound, and a vinyl group-containing hydroxy compound. Examples of the monomer include N-(1-ethenyl)amine, N-(2-propenyl)amine, N-(1-ethenyl)-N-methylamine, N-(2-propenyl)-N-methylamine, 1-ethenylamide, 2-propenylamide, N-methyl-(1-ethenyl)amide, N-methyl-(2-propenyl)amide, vinyl alcohol, 2-propen-1-ol, and 3-buten-1-ol. Examples of the monomer which has a functional group having a Lewis basic moiety with no active hydrogen atom and one or more polymerizable unsaturated groups include vinylpyridine, vinyl(N-substituted) imidazole, and vinyl(N-substituted) indazole.

[0160]

Examples of the other monomer having a polymerizable unsaturated group include ethylene, α-olefin, an aromatic vinyl compound and a cyclic olefin compound. Examples of the monomer include ethylene, propylene, 1-butene, 1-hexene, 4-methyl-1-pentene, styrene, norbornene and dicyclopentadiene. Ethylene or styrene is preferred. Two or more kinds of these monomers may be used. Examples of the crosslinking polymerizable monomer having two or more polymerizable unsaturated groups include divinylbenzene and the like.

[0161]

There is no particular limitation on the average particle diameter of the organic polymer particles, and the average particle diameter is usually from 1 to 5,000 μm, preferably from 5 to 1,000 μm, and more preferably from 10 to 500 μm. There is no particular limitation on the pore volume, and the pore volume is preferably 0.1 ml/g or more, and more preferably 0.3 to 10 ml/g. There is no particular limitation on the specific surface area, and the specific surface area is preferably from 10 to 1,000 m^2/g, and more preferably from 50 to 500 m^2/g.

[0162]

These organic polymer particles are preferably dried to substantially remove moisture, and more preferably dried by heating. The drying temperature of the organic polymer whose moisture cannot be visually confirmed is usually from 30 to 400°C, preferably from 50 to 200°C, and more preferably from 70 to 150°C. There is no particular limitation on the heating time, and the heating time is preferably from 10 minutes to 50 hours, and more preferably from 1 hour to 30 hours. Examples of the method of drying the organic polymer particles by heating include a method in which organic polymer particles are dried by circulating a dried inert gas (e.g., nitrogen, argon, etc.) at a given flow rate while heating, or a method in which organic polymer particles are dried by heating under reduced pressure.

[0163]

There is no particular limitation on the order of bringing (a), (b), (c) and (d) described above into contact with each other so as to obtain modified particle (I) of the present invention, and examples of the order include the following orders:

<1> an order in which a contact product of (a) and (b) is brought into contact with (c) to obtain a contact product and the obtained contact product is brought into contact with (d);

<2> an order in which a contact product of (a) and (b) is brought into contact with (d) to obtain a contact product and the obtained contact product is brought into contact with (c);

<3> an order in which a contact product of (a) and (c) is brought into contact with (b) to obtain a contact product and the obtained contact product is brought into contact with (d);

<4> an order in which a contact product of (a) and (c) is brought into contact

with (d) to obtain a contact product and the obtained contact product is brought into contact with (b);

<5> an order in which a contact product of (a) and (d) is brought into contact with (b) to obtain a contact product and the obtained contact product is brought into contact with (c);

<6> an order in which a contact product of (a) and (d) is brought into contact with (c) to obtain a contact product and the obtained contact product is brought into contact with (b);

<7> an order in which a contact product of (b) and (c) is brought into contact with (a) to obtain a contact product and the obtained contact product is brought into contact with (d);

<8> an order in which a contact product of (b) and (c) is brought into contact with (d) to obtain a contact product and the obtained contact product is brought into contact with (a);

<9> an order in which a contact product of (b) and (d) is brought into contact with (a) to obtain a contact product and the obtained contact product is brought into contact with (c);

<10> an order in which a contact product of (b) and (d) is brought into contact with (c) to obtain a contact product and the obtained contact product is brought into contact with (a);

<11> an order in which a contact product of (c) and (d) is brought into contact with (a) to obtain a contact product and the obtained contact product is brought into contact with (b); and

<12> an order in which a contact product of (c) and (d) is brought into contact with (b) to obtain a contact product and the obtained contact product

is brought into contact with (a).

The contact order is preferably <1>, <2>, <3>, <5>, <11> or <12> described above. The contact order is particularly preferably <2> or <5>.

[0164]

Such a contact treatment is preferably carried out under an inert gas atmosphere. The treatment temperature is usually from -100 to 300°C, and preferably from -80 to 200°C. The treatment time is usually from 1 minute to 200 hours, and preferably from 10 minutes to 100 hours. Such a treatment may be carried out using a solvent, or these compounds may be directly brought into contact with each other without using a solvent.

[0165]

A solvent which is inert to (a), (b), (c), (d) described above and contact products thereof is used as the solvent. However, as described above, when the respective compounds are brought into contact with each other in a stepwise manner, a solvent capable of reacting with a certain compound in a certain stage can be used in another stage if the solvent does not react with each compound in another stage. That is, the solvent to be used in each step is the same or different. Examples of the solvent include nonpolar solvents such as an aliphatic hydrocarbyl solvent and an aromatic hydrocarbyl solvent; and polar solvents such as a halide solvent, an ether-based solvent, an alcohol-based solvent, a phenol-based solvent, a carbonyl-based solvent, a phosphoric acid derivative, a nitrile-based solvent, a nitro compound, an amine-based solvent and a sulfur compound. Examples thereof include aliphatic hydrocarbyl solvents such as butane, pentane, hexane, heptane, octane, 2,2,4-trimethylpentane and cyclohexane; aromatic hydrocarbyl solvents such as benzene, toluene and xylene; halide solvents such as dichloromethane, difluoromethane, chloroform, 1,2-dichloroethane, 1,2-dibromoethane, 1,1,2-trichloro-1,2,2-trifluoroethane, tetrachloroethylene, chlorobenzene, bromobenzene and o-dichlorobenzene; ether-based solvents such as dimethyl ether, diethyl ether, diisopropyl ether, di-n-butyl ether, methyl-tert-butyl-ether, anisole, 1,4-dioxane, 1,2-dimethoxyethane, bis(2-methoxyethyl)ether, tetrahydrofuran and tetrahydropyran; alcohol-based solvents such as methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, 2-methyl-1-propanol, 3-methyl-1-butanol, cyclohexanol, benzyl alcohol, ethylene glycol, propylene glycol, 2-methoxyethanol, 2-ethoxyethanol, diethylene glycol, triethylene glycol and glycerin; phenol-based solvents such as phenol and p-cresol; carbonyl-based solvents such as acetone, ethyl methyl ketone, cyclohexanone, acetic anhydride, ethyl acetate, butyl acetate, ethylene carbonate, propylene carbonate, N,N-dimethylformamide, N,N-dimethylacetamide and N-methyl-2-pyrrolidone; phosphoric acid derivatives such as hexamethylphosphoric acid triamide and triethyl phosphate; nitrile-based solvents such as acetonitrile, propionitrile, succinonitrile and benzonitrile; nitro compounds such as nitromethane and nitrobenzene; amine-based solvents such as pyridine, piperidine and morpholine; and sulfur compounds such as dimethyl sulfoxide and sulfolane.

[0166]

When a contact product (f) produced by bringing the compounds (a), (b) and (c) into contact with each other is brought into contact with the particles (d), namely, when the contact product (f) is produced in the respective methods of <1>, <3> and <7> described above, a solvent (s1) is preferably the above aliphatic hydrocarbyl solvent, aromatic hydrocarbyl solvent or ether-based solvent.

[0167]

When the contact product (f) is brought into contact with the particles (d), a solvent (s2) is preferably a polar solvent. The E_T^N value (C. Reichardt, "Solvents and Solvents Effects in Organic Chemistry", 2nd ed., VCH Verlag (1988)) or the like is known as an indicator which represents the polarity of the solvent, and a solvent satisfying the range of the following inequality expression: $0.8 \geq E_T^N \geq 0.1$ is particularly preferred. Examples of the polar solvent include dichloromethane, dichlorodifluoromethanechloroform, 1,2-dichloroethane, 1,2-dibromoethane, 1,1,2-trichloro-1,2,2-trifluoroethane, tetrachloroethylene, chlorobenzene, bromobenzene, o-dichlorobenzene, dimethyl ether, diethyl ether, diisopropyl ether, di-n-butyl ether, methyl-tert-butyl ether, anisole, 1,4-dioxane, 1,2-dimethoxyethane, bis(2-methoxyethyl)ether, tetrahydrofuran, tetrahydropyran, methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, 2-methyl-1-propanol, 3-methyl-1-butanol, cyclohexanol, benzyl alcohol, ethylene glycol, propylene glycol, 2-methoxyethanol, 2-ethoxyethanol, diethylene glycol, triethylene glycol, acetone, ethyl methyl ketone, cyclohexanone, acetic anhydride, ethyl acetate, butyl acetate, ethylene carbonate, propylene carbonate, N,N-dimethylformamide, N,N-dimethylacetamide, N-methyl-2-pyrrolidone, hexamethylphosphoric acid triamide, triethyl phosphate, acetonitrile, propionitrile, succinonitrile, benzonitrile, nitromethane, nitrobenzene, ethylenediamine, pyridine, piperidine , morpholine, dimethyl sulfoxide and sulfolane. The solvent (s2) is more preferably dimethyl ether, diethyl ether, diisopropyl ether, di-n-butyl ether, methyl-tert-butyl ether, anisole, 1,4-dioxane, 1,2-dimethoxyethane, bis(2-methoxyethyl)ether, tetrahydrofuran, tetrahydropyran, methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, 2-methyl-1-propanol, 3-methyl-1-butanol, cyclohexanol, benzylalcohol, ethylene glycol, propylene glycol, 2-methoxyethanol, 2-ethoxyethanol, diethylene glycol or triethylene glycol, particularly preferably di-n-butyl ether, methyl-tert-butyl ether, 1,4-dioxane, tetrahydrofuran, methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, 2-methyl-1-propanol, 3-methyl-1-butanol or cyclohexanol, and most preferably tetrahydrofuran, methanol, ethanol, 1-propanol or 2-propanol.

[0168]

It is also possible to use, as the solvent (s2), a mixed solvent of the polar solvent and a hydrocarbyl solvent. The aliphatic hydrocarbyl solvent and the aromatic hydrocarbyl solvent listed above are used as the hydrocarbyl solvent. Examples of the mixed solvent of the polar solvent and the hydrocarbyl solvent can include a hexane/methanol mixed solvent, a hexane/ethanol mixed solvent, a hexane/1-propanol mixed solvent, a hexane/2-propanol mixed solvent, a heptane/methanol mixed solvent, a heptane/ethanol mixed solvent, a heptane/1-propanol mixed solvent, a heptane/2-propanol mixed solvent, a toluene/methanol mixed solvent, a toluene/ethanol mixed solvent, a toluene/1-propanol mixed solvent, a toluene/2-propanol mixed solvent, a xylene/methanol mixed solvent, a xylene/ethanol mixed solvent, a xylene/1-propanol mixed solvent and a xylene/2-propanol mixed solvent. The mixed solvent is preferably a hexane/methanol mixed solvent, a hexane/ethanol mixed solvent, a heptane/methanol mixed solvent, a heptane/ethanol mixed solvent, a toluene/methanol mixed solvent, a toluene/ethanol mixed solvent, a xylene/methanol mixed solvent or a xylene/ethanol mixed solvent. The mixed solvent is more preferably a hexane/methanol mixed solvent, a hexane/ethanol mixed solvent, a toluene/methanol mixed solvent or a toluene/ethanol mixed solvent. The mixed solvent is most preferably a toluene/ethanol mixed solvent. The content of an ethanol fraction in the toluene/ethanol mixed solvent is preferably within a range from 10 to 50% by volume, and more preferably from 15 to 30% by volume.

[0169]

It is also possible to use a hydrocarbyl solvent as the solvent (s1) and the solvent (s2) in a method in which the contact product (f) produced by bringing (a), (b) and (c) into contact with each other is brought into contact with (d), namely, in the respective methods of <1>, <3> and <7> described above. In this case, the shorter the time until the produced contact product (f) is brought into contact with the particles (d) after bringing (a), (b) and (c) into contact with each other, the better. The time is preferably from 0 to 5 hours, more preferably from 0 to 3 hours, and most preferably from 0 to 1 hour. When the contact product (f) is brought into contact with the particles (d), the temperature at that time is usually from -100°C to 40°C, preferably from -20°C to 20°C, and most preferably from -10°C to 10°C.

[0170]

In the case of <2>, <5>, <6>, <8>, <9>, <10>, <11> and <12>

described above, any of the above nonpolar solvent and the above polar solvent can be used. The nonpolar solvent is preferred. The reason is considered that, since a contact product of (a) and (c), or a contact product produced by bringing a contact product of (a) and (b) into contact with (c) commonly exhibits low solubility in a nonpolar solvent, in a case where (d) exists in a reaction system when these contact products are formed, the thus formed contact products are precipitated on a surface of (d) and are more likely to be immobilized.

[0171]

There is no particular limitation on the amount of the respective compounds of (a), (b) and (c) described above to be brought into contact with each other. When a molar ratio of the amount of the respective compounds to be brought into contact with each other is assumed to be a molar ratio (a) : (b) : (c) = 1 : y : z, it is preferred that y and z substantially satisfy Formula (1) described below:

$$|m - y - 2z| < 1 \qquad (1)$$

wherein m in Formula (1) represents the valence of M^3.

y in Formula (1) is preferably a number of 0.01 to 1.99, more preferably a number of 0.10 to 1.80, still more preferably a number of 0.20 to 1.50, and most preferably a number of 0.30 to 1.00. Preferable range of z in Formula (1) is determined by m, y and Formula (1).

[0172]

Compounds (a) and (d) are each used in such an amount that the number of typical metal atoms derived from the compound (a) contained in the modified particles (I) is preferably 0.05 mmol or more, and more preferably from 0.1 to 20 mmol, expressed by the molar number of the typical metal atoms per gram of the produced modified particles (I).

[0173]

In order to allow the reaction to proceed more quickly, a heating step at a higher temperature is preferably added after the contact treatment described above. In the heating step, a solvent having a high boiling point is preferably used so as to achieve a higher temperature. In the case of carrying out the heating step, the solvent used in the contacting step may be replaced by another solvent having a higher boiling point.

[0174]

In the modified particles (I), as a result of the contact treatment, the compounds (a), (b), (c) and/or (d) as raw materials may remain as an unreacted product. However, the produced modified particles (I) are preferably washed so as to remove the unreacted product from the produced modified particles (I). A solvent to be used to wash the modified particles (I) may be the same or different used to produce the modified particles (I). The modified particles (I) are preferably washed under an inert gas atmosphere. The washing temperature is usually from -100 to 300°C, and preferably from -80 to 200°C. The washing time is usually from 1 minute to 200 hours, and preferably from 10 minutes to 100 hours.

[0175]

It is preferred to remove the solvent in a state where the modified particles (I) in the solvent are sedimented and amorphous or fine particles float at the upper portion of slurry, in the case of the above washing treatment, so as to obtain modified particles (I) having a uniform particle diameter and shape.

[0176]

After the contact treatment or the washing treatment, the solvent is preferably distilled off from the product, followed by drying the product at a temperature of 0°C or higher under reduced pressure for 1 hour to 24 hours. The drying treatment is more preferably carried out at a temperature of 0°C to 200°C for 1 hour to 24 hours, still more preferably at a temperature of 10°C to 200°C for 1 hour to 24 hours, particularly preferably at a temperature of 10°C to 160°C for 2 hours to 18 hours, and most preferably at a temperature of 15°C to 160°C for 4 hours to 18 hours.

[0177]

Examples of a method for producing the modified particles (I) is described in more detail below by way of a case where M^3 is a zinc atom, the compound (b) is 3,4,5-trifluorophenol, the compound (c) is water, and (d) is silica. Using tetrahydrofuran as a solvent, a solution of diethyl zinc in hexane is added thereto to form a mixture (1). After cooling the mixture (1) to 3°C, 3,4,5-trifluorophenol is added dropwise to the mixture (1) in an equimolar amount based on the amount of diethyl zinc to form a mixture (2), followed by stirring the mixture (2) at room temperature for 10 minutes to 24 hours. Furthermore, water is added dropwise to the mixture (2) in a 0.5-fold molar amount based on the amount of diethyl zinc to form a mixture (3), followed by stirring the mixture (3) at room temperature for 10 minutes to 24 hours. Then, the solvent is distilled off from the mixture (3) and a product is obtained. The product is dried at 120°C under reduced pressure for 8 hours and a solid component (1) is obtained. Tetrahydrofuran and silica are added to the solid component (1) to form a mixture (4), followed by stirring the mixture (4) at 40°C for 2 hours, thereby a solid component (2) is obtained. The solid component (2) is washed with tetrahydrofuran and then dried at 120°C under reduced pressure for 8 hours. Thus, the modified particles (I) of the present invention can be produced.

[0178]

A cyclic aluminoxane having a structure represented by Formula $\{-Al(E^2)-O-\}_e$ and/or a linear aluminoxane having a structure represented by Formula $E^3\{-Al(E^3)-O-\}_cAlE^3_2$ is/are preferably used as the aluminoxane (e) used in the preparation of modified particles (II). In the formulas, E^2 and E^3 each represent a hydrocarbyl group, each E^2 may be the same or different, each E^3 may be the same or different, e represents a number of 2 or more, and c represents a number of 1 or more.

The hydrocarbyl group in E^2 or E^3 is preferably a hydrocarbyl group having 1 to 8 carbon atoms, and more preferably an alkyl group.

[0179]

Examples of E^2 and E^3 include alkyl groups such as a methyl group, an ethyl group, a normal propyl group, an isopropyl group, a normal butyl group, an isobutyl group, a normal pentyl group and a neopentyl group. e is a number of 2 or more, and c is a number of 1 or more. E^2 and E^3 are preferably methyl groups or isobutyl groups, e is from 2 to 40, and c is from 1 to 40.

[0180]

The above aluminoxane can be produced by various methods. There is no particular limitation on the methods, and the aluminoxane may be produced in accordance with a known method. For example, the aluminoxane can be produced by bringing a solution, prepared by dissolving trialkyl aluminum (e.g., trimethyl aluminum, etc.) in a proper organic solvent (benzene, aliphatic hydrocarbyl, etc.), into contact with water. Further examples of the method include a method in which the aluminoxane is produced by bringing a metal salt containing crystal water (e.g., copper sulfate hydrate, etc.) into contact with trialkyl aluminum (e.g., trimethyl aluminum, etc.). It is considered that the aluminoxane produced by such a method is usually a mixture of a cyclic aluminoxane and a linear aluminoxane.

[0181]

The particles (d) used to obtain the modified particles (II) are particles similar to (d) used in the modified particles (I).

[0182]

The modified particles (II) can be produced by bringing the aluminoxane (e) into contact with the particles (d) using any method. Specifically, the modified particles (II) can be produced by dispersing the particles (d) in a solvent and adding the aluminoxane (e) thereto.

In this case, any solvent described in the description of the method for producing the modified particles (I) can be used, and the solvent is preferably a solvent which does not react with the aluminoxane (e), and more preferably a solvent which dissolves the aluminoxane (e). Specifically, aromatic hydrocarbyl solvents such as toluene and xylene, or aliphatic hydrocarbyl solvents such as hexane, heptane and octane are preferred, and toluene or xylene is more preferred.

[0183]

The contacting temperature and contacting time used in the case of bringing the aluminoxane (e) into contact with the particles (d) can be optionally selected. The temperature is usually from -100°C to 200°C, preferably from -50°C to 150°C, and still more preferably from -20°C to 120°C. Particularly in an initial stage of the reaction, these are preferably reacted at a low temperature so as to suppress heat generation. The amounts of the aluminoxane (e) and the particles (d), which are brought into contact with each other, are optional amounts. The amount of the aluminoxane (e) is usually from 0.01 to 100 mmol, preferably from 0.1 to 20 mmol, and still more preferably from 1 to 10 mmol, expressed by aluminum atoms per gram of the particles (d).

[0184]

Modified particles (III) are produced by further using the compound (A) in the preparation of the modified particles (II).

The transition metal compound represented by Formula [4] or its μ-oxo type transition metal compound dimer is used as the compound (A).

The modified particles (III) can be produced by brining the aluminoxane (e), the particles (d) and the compound (A) into contact with each other using any method. It is preferable that the aluminoxane (e), the particles (d) and the compound (A) be brought into contact with each other in a solvent. It is also possible to use, as the solvent, any solvent described above. The solvent is preferably a solvent which does not react with the aluminoxane (e) and the compound (A), and more preferably a solvent which dissolves the aluminoxane (e) and the compound (A). Specifically, aromatic hydrocarbyl solvents such as toluene and xylene, or aliphatic hydrocarbyl solvents such as hexane, heptane and octane are preferred, and toluene or xylene is more preferred.

[0185]

The contacting temperature and contacting time used in the case of bringing the aluminoxane (e), the particles (d) and the compound (A) into contact with each other are optionally selected. The temperature is usually from -100°C to 200°C, preferably from -50°C to 150°C, and still more preferably from -20°C to 120°C. Particularly in an initial stage of the reaction, these are preferably reacted at a low temperature so as to suppress heat generation. The amounts of the aluminoxane (e), the particles (d) and the compound (A), which are brought into contact with each other, are optional amounts. The amount of the aluminoxane (e) is usually from 0.01 to 100 mmol, preferably from 0.1 to 20 mmol, and still more preferably from 1 to 10 mmol, expressed by aluminum atoms per gram of the particles (d). The amount of the compound (A) is usually from 0.1 to 1,000 μmol, preferably from 1 to 500 μmol, and still more preferably from 10 to 200 μmol, expressed by the transition metal atoms per gram of the particles (d).

[0186]

Organoaluminum Compound (C)

The organoalminium compound (C) to be used in the present invention is a known organoalminium compound. It is preferably an organoalminium compound represented by Formula [8]:

$R^9{}_dAlY_{3-d}$ [8]

wherein each R^9 independendetly represents a hydrocarbyl group; each Y independetly represents a hydrogen atom, a halogen atom, an alkoxy group, an aralkyloxy group or an aryloxy group; and d is a number satisfying $0 < d \leq 3$.

[0187]

R^9 in Formula [8] is preferably a hydrocarbyl group having 1 to 24 carbon atoms, and more preferably an alkyl group having 1 to 24 carbon atoms. Examples of R6 include a methyl group, an ethyl group, a n-propyl group, a n-butyl group, an isobutyl group, a n-hexyl group, a 2-methylhexyl group, a n-octyl group and the like, and preferably an ethyl group, a n-butyl group, an isobutyl group, a n-hexyl group or a n-octyl group.

[0188]

Examples of a case where Y is a halogen atom include a fluorine atom, a chlorine atom, a bromine atom and an iodine atom, and preferably a chlorine atom.

The alkoxy group in Y is preferably an alkoxy group having 1 to 24 carbon atoms, and examples of the aokoxy group include a methoxy group, an ethoxy group, a n-propoxy group, an isopropoxy group, a n-butoxy group, a sec-butoxy group, a tert-butoxy group, a n-pentoxy group, a neopentoxy group, a n-hexoxy group, a n-octoxy group, a n-dodesoxy group, a n-pentadesoxy group, a n-icosoxy group and the like, and preferably a methoxy group, an ethoxy group or a tert-butoxy group.

[0189]

The aryloxy group in Y is preferably an aryloxy group having 6 to 24 carbon atoms, and examples of the aryloxy group include a phenoxy group, a 2-methylphenoxy group, a 3-methylphenoxy group, a 4-methylphenoxy group, a 2,3-dimethylphenoxy group, a 2,4-dimethylphenoxy group, a 2,5-dimethylphenoxy group, a 2,6-dimethylphenoxy group, a 3,4-dimethylphenoxy group, a 3,5-dimethylphenoxy group, a 2,3,4-trimethylphenoxy group, a 2,3,5-trimethylphenoxy group, a 2,3,6-trimethylphenoxy group, a 2,4,5-trimethylphenoxy group, a 2,4,6-trimethylphenoxy group, a 3,4,5-trimethylphenoxy group, a 2,3,4,5-tetramethylphenoxy group, a 2,3,4,6-tetramethylphenoxy group, a 2,3,5,6-tetramethylphenoxy group, a pentamethylphenoxy group, an ethylphenoxy group, a n-propylphenoxy group, an isopropylphenoxy group, a n-butylphenoxy group, a sec-butylphenoxy group, a tert-butylphenoxy group, a n-hexylphenoxy group, a n-octylphenoxy group, a n-decylphenoxy group, a n-tetradecylphenoxy group, a naphthoxy group, an anthrathenoxy group and the like.

[0190]

The aralkyloxy group in Y is preferably an aralkyloxy group having 7 to 24 carbon atoms, and examples ot the aralkyloxy group include a benzyloxy group, a (2-methylphenyl)methoxy group, a (3-methylphenyl)methoxy group, a (4-methylphenyl)methoxy group, a (2,3-dimethylphenyl)methoxy group, a (2,4-dimethylphenyl)methoxy group, a (2,5-dimethylphenyl)methoxy group, a (2,6-dimethylphenyl)methoxy group, a (3,4-dimethylphenyl)methoxy group, a (3,5-dimethylphenyl)methoxy group, a (2,3,4-trimethylphenyl)methoxy group, a (2,3,5-trimethylphenyl)methoxy group, a (2,3,6-trimethylphenyl)methoxy group, a (2,4,5-trimethylphenyl)methoxy group, a (2,4,6-trimethylphenyl)methoxy group, a (3,4,5-trimethylphenyl)methoxy group, a (2,3,4,5-

tetramethylphenyl)methoxy group, a (2,3,5,6-tetramethylphenyl)methoxy group, a (pentamethylphenyl)methoxy group, an (ethylphenyl)methoxy group, a (n-propylphenyl)methoxy group, an (isopropylphenyl)methoxy group, a (n-butylphenyl)methoxy group, a (sec-butylphenyl)methoxy group, a (tert-butylphenyl)methoxy group, a (n-hexylphenyl)methoxy group, a (n-octylphenyl)methoxy group, a (n-decylphenyl)methoxy group, a (n-tetradecylphenyl)methoxy group, a naphthylmethoxy group, an anthracenylmethoxy group and the like, and preferably a benzyloxy group.

[0191]

Examples of the organoaluminum compound represented by Formula [8] include trialkyl aluminums such as trimethyl aluminum, triethyl aluminum, tri-n-propyl aluminum, tri-n-butyl aluminum, triisobutyl aluminum, tri-n-hexyl aluminum and tri-n-octyl aluminum; dialkyl aluminum chlorides such as dimethyl aluminum chloride, diethyl aluminum chloride, di-n-propyl aluminum chloride, di-n-butyl aluminum chloride, diisobutyl aluminum chloride and di-n-hexyl aluminum chloride; alkyl aluminum dichlorides such as methyl aluminum dichloride, ethyl aluminum dichloride, n-propyl aluminum dichloride, n-butyl aluminum dichloride, isobutyl aluminum dichloride and n-hexyl aluminum dichloride; dialkyl aluminum hydrides such as dimethyl aluminum hydride, diethyl aluminum hydride, di-n-propyl aluminum hydride, di-n-butyl aluminum hydride, diisobutyl aluminum hydride and di-n-hexyl aluminum hydride; alkyl(dialkoxy) aluminums such as methyl(dimethoxy) aluminum, methyl(diethoxy) aluminum and methyl(di-tert-butoxy) aluminum; dialkyl(alkoxy) aluminums such as dimethyl(methoxy) aluminum, dimethyl(ethoxy) aluminum and dimethyl(tert-butoxy) aluminum; alkyl(diaryloxy) aluminums such as methyl(diphenoxy) aluminum, methylbis(2,6-diisopropylphenoxy) aluminum and methylbis(2,6-diphenylphenoxy) aluminum; and dialkyl(aryloxy) aluminums such as dimethyl(phenoxy) aluminum, dimethyl(2,6-diisopropylphenoxy) aluminum and dimethyl(2,6-diphenylphenoxy) aluminum.

[0192]

Among them, the organoaluminum compound is preferably trialkyl aluminum, more preferably trimethyl aluminum, triethyl aluminum, tri-n-butyl aluminum, triisobutyl aluminum, tri-n-hexyl aluminum or tri-n-octyl aluminum, and particularly preferably triisobutyl aluminum or tri-n-octyl aluminum.

These organoaluminum compounds may be used alone, or two or more kinds of them may be used in combination.

[0193]

Electron-Donating Compound (E)

In the production of the prepolymerized catalyst component for addition polymerization and addition polymer of the present invention, an electron-donating compound (E) may be brought into contact with a catalyst or a prepolymerized catalyst component for addition polymerization. The electron-donating compound (E) is preferably a compound containing a nitrogen atom, a phosphorus atom, an oxygen atom or a sulfur atom, and examples thereof include an oxygen-containing compound, a nitrogen-containing compound, a phosphorus-containing compound, and a sulfur-containing compound, and among them, an oxygen-containing compound or a nitrogen-containing compound is preferabe. Examples of the oxygen-containing compound include alkoxysilicons, ethers, ketones, aldehydes, carboxylic acids, esters of organic acid or inorganic acid, acid amides of organic acid or inorganic acid, acid anhydrides and the like, and among them, alkoxysilicons or ethers are preferable. Examples of the nitrogen-containing compound include amines, nitriles, isocyanates and the like, and amines are preferable.

[0194]

The alkoxysilicon is preferably an alkoxysilane represented by Formula $R^{10}_{r}Si(OR^{11})_{4-r}$, wherein each R^{10} independently represents a hydrocarbyl group having 1 to 20 carbon atoms, a hydrogen atom, or a hetero atom-containing substituent; each R^{11} independently represents a hydrocarbyl group having 1 to 20 carbon atoms; r represents a number satisfying $0 \leq r < 4$.

[0195]

When R^{10} and R^{11} are hydrocarbyl groups, examples thereof include linear alkyl groups such as a methyl group, an ethyl group, a propyl group, a butyl group, and a pentyl group; branched alkyl groups such as an isopropyl group, a sec-butyl group, a tert-butyl group, and a tert-amyl group; cycloalkyl groups such as a cyclopentyl group and a cyclohexyl group; cycloalkenyl groups such as a cyclopentenyl group; aryl groups such as a phenyl group and a tolyl group; and the like. When R^{10} is a hetero atom-containing substituent, examples of the hetero atom include an oxygen atom, a nitrogen atom, a sulfur atom, and a phosphorus atom. Examples thereof include a dimethylamino group, a methylethylamino group, a diethylamino group, an ethyl-n-propylamino group, a di-n-propylamino group, a pyrrolyl group, a pyridyl group, a pyrrolidinyl group, a piperidyl group, a perhydroindolyl group, a perhydroisoindolyl group, a perhydroquinolyl group, a perhydroisoquinolyl group, a perhydrocarbazolyl group, a perhydroacridinyl group, a furyl group, a pyranyl group, a perhydrofuryl group, a thienyl group and the like.

R^{10} and R^{11} are preferably alkyl groups, and also r is preferably a number satisfying $4 > r \geq 2$.

[0196]

Examples of the alkoxysilicons include tetramethoxysilane, methyltrimethoxysilane, ethyltrimethoxysilane, normal propyltrimethoxysilane, isopropyltrimethoxysilane, normal butyltrimethoxysilane, isobutyltrimethoxysilane, sec-butyltrimethoxysilane, tert-butyltrimethoxysilane, normal pentyltrimethoxysilane, tert-amyltrimethoxysilane, dimethyldimethoxysilane, diethyldimethoxysilane, di-normal butyldimethoxysilane, diisobutyldimethoxysilane, di-tert-butyldimethoxysilane, methylethyldimethoxysilane, methyl-normal-propyldimethoxysilane, methyl-normal-butyldimethoxysilane, methylisobutyldimethoxysilane, tert-butylmethyldimethoxysilane, tert-butylethyldimethoxysilane, tert-butyl-normal-propyldimethoxysilane, tert-butylisopropyldimethoxysilane, tert-butyl-normal-butyldimethoxysilane, tert-butylisobutyldimethoxysilane, tert-amylmethyldimethoxysilane, tert-amylethyldimethoxysilane, tert-amyl-normal-propyldimethoxysilane, tert-amyl-normal-butyldimethoxysilane, isobutylisopropyldimethoxysilane, dicyclobutyldimethoxysilane, cyclobutylmethyldimethoxysilane, cyclobutylethyldimethoxysilane, cyclobutylisopropyldimethoxysilane, cyclobutyl-normal-butyldimethoxysilane, cyclobutylisobutyldimethoxysilane, cyclobutyl-tert-butyldimethoxysilane, dicyclopentyldimethoxysilane, cyclopentylmethyldimethoxysilane, cyclopentyl-normal-propyldimethoxysilane, cyclopentylisopropyldimethoxysilane, cyclopentyl-normal-butyldimethoxysilane, cyclopentylisobutyldimethoxysilane, cyclopentyl-tert-butyldimethoxysilane, dicyclohexyldimethoxysilane, cyclohexylmethyldimethoxysilane, cyclohexylethyldimethoxysilane, cyclohexyl-normal-propyldimethoxysilane, cyclohexylisopropyldimethoxysilane, cyclohexyl-normal-butyldimethoxysilane, cyclohexylisobutyldimethoxysilane, cyclohexyl-tert-butyldimethoxysilane, cyclohexylcyclopentyldimethoxysilane, cyclohexylphenyldimethoxysilane, diphenyldimethoxysilane, phenylmethyldimethoxysilane, phenylethyldimethoxysilane, phenyl-normal-propyldimethoxysilane, phenylisopropyldimethoxysilane, phenyl-normal-butyldimethoxysilane, phenylisobutyldimethoxysilane, phenyl-tert-butyldimethoxysilane, phenylcyclopentyldimethoxysilane, 2-norbornanemethyldimethoxysilane, bis(perhydroquinolino)dimethoxysilane, bis(perhydroisoquinolino)dimethoxysilane, (perhydroquinolino) (perhydroisoquinolino)dimethoxysilane, (perhydroquinolino)methyldimethoxysilane, (perhydroisoquinolino)methyldimethoxysilane, (perhydroquinolino)ethyldimethoxysilane, (perhydroisoquinolino)ethyldimethoxysilane, (perhydroquinolino) (n-propyl)dimethoxysilane, (perhydroisoquinolino) (n-propyl)dimethoxysilane, (perhydroquinolino) (tert-butyl)dimethoxysilane, (perhydroisoquinolino) (tert-butyl)dimethoxysilane, trimethylmethoxysilane, triethylmethoxysilane, tri-normal-propylmethoxysilane, triisopropylmethoxysilane, tri-normal-butylmethoxysilane, triisobutylmethoxysilane, tri-tert-butylmethoxysilane and the like. Examples thereof include compounds in which methoxy of these compounds is replaced by ethoxy, propoxy, normal butoxy, isobutoxy, tert-butoxy or phenoxy. Dialkyldialkoxysilane or trialkylmonoalkoxysilane is preferred, and trialkylmonoalkoxysilane is more preferred.

[0197]

Examples of the ethers include a dialkyl ether, an alkylaryl ether, a diaryl ether, a diether compound, cyclic ethers and cyclic diethers.

[0198]

Examples of the ether include dimethyl ether, diethyl ether, di-normal-propyl ether, diisopropyl ether, di-normal-butyl ether, diisobutyl ether, di-tert-butyl ether, dicyclohexyl ether, diphenyl ether, methyl ethyl ether, methyl-normal-propyl ether, methyl isopropyl ether, methyl-normal-butyl ether, methyl isobutyl ether, methyl-tert-butyl ether, methyl cyclohexyl ether, methylphenyl ether, ethylene oxide, propylene oxide, oxetane, tetrahydrofuran, 2,5-dimethyltetrahydrofuran, tetrahydropyran, 1,2-dimethoxyethane, 1,2-diethoxyethane, 1,2-diisobutoxyethane, 2,2-dimethoxypropane, 1,3-dimethoxypropane, 2,2-diisobutyl-1,3-dimethoxypropane, 2-isopropyl-2-isopentyl-1,3-dimethoxypropane, 2,2-bis(cyclohexylmethyl)-1,3-dimethoxypropane, 2-isopropyl-2-3,7-dimethyloctyl-1,3-dimethoxypropane, 2,2-diisopropyl-1,3-dimethoxypropane, 2-isopropyl-2-cyclohexylmethyl-1,3-dimethoxypropane, 2,2-dicyclohexyl-1,3-

dimethoxypropane, 2-isopropyl-2-isobutyl-1,3-dimethoxypropane, 2,2-diisopropyl-1,3-dimethoxypropane, 2,2-dipropyl-1,3-dimethoxypropane, 2-isopropyl-2-cyclohexyl-1,3-dimethoxypropane, 2-isopropyl-2-cyclopentyl-1,3-dimethoxypropane, 2,2-dicyclopentyl-1,3-dimethoxypropane, 2-heptyl-2-pentyl-1,3-dimethoxypropane, 1,2-dimethoxybenzene, 1,3-dimethoxybenzene, 1,4-dimethoxybenzene, 1,3-dioxolane, 1,4-dioxane, 1,3-dioxane and the like. The ether is preferably diethyl ether, di-normal-butyl ether, methyl-normal-butyl ether, methylphenyl ether, tetrahydrofuran, 1,3-dioxane, 1,4-dioxane or 1,3-dioxolane, and more preferably diethyl ether, di-normal-butyl ether or tetrahydrofuran.

[0199]

Examples of the carboxylic acid ester include mono or polyvalent carboxylic acid esters, and examples of the carboxylic acid ester include a saturated aliphatic carboxylic acid ester, an unsaturated aliphatic carboxylic acid ester, an alicyclic carboxylic acid ester and an aromatic carboxylic acid ester. Examples of the carboxylic acid ester include methyl acetate, ethyl acetate, normal butyl acetate, isobutyl acetate, tert-butyl acetate, phenyl acetate, methyl propionate, ethyl propionate, ethyl butyrate, ethyl valerate, ethyl acrylate, methyl methacrylate, methyl benzoate, ethyl benzoate, normal butyl benzoate, isobutyl benzoate, tert-butyl benzoate, methyl toluate, ethyl toluate, methyl anisate, ethyl anisate, dimethyl succinate, diethyl succinate, di-normal-butyl succinate, dimethyl malonate, diethyl malonate, di-normal-butyl malonate, dimethyl maleate, dibutyl maleate, diethyl itaconate, di-normal-butyl itaconate, monoethyl phthalate, dimethyl phthalate, methylethyl phthalate, diethyl phthalate, di-normal-propyl phthalate, diisopropyl phthalate, di-normal-butyl phthalate, diisobutyl phthalate, di-tert-butyl phthalate, dipentyl phthalate, di-n-hexyl phthalate, diheptyl phthalate, di-normal-octyl phthalate, di(2-ethylhexyl)phthalate, diisodecyl phthalate, dicyclohexyl phthalate, diphenyl phthalate, dimethyl isophthalate, diethyl isophthalate, di-normal-butyl isophthalate, diisobutyl isophthalate, di-tert-butyl isophthalate, dimethyl terephthalate, diethyl terephthalate, di-normal-butyl terephthalate, diisobutyl terephthalate, di-tert-butyl terephthalate and the like. The carboxylic acid ester is preferably methyl acetate, ethyl acetate, methyl benzoate, ethyl benzoate, dimethyl phthalate, diethyl phthalate, di-normal-butyl phthalate, diisobutyl phthalate, dimethyl terephthalate or diethyl terephthalate, and still more preferably methyl benzoate, dimethyl phthalate, diethyl phthalate, diisobutyl phthalate or dimethyl terephthalate.

[0200]

Examples of amines include trihydrocarbylamine, and examples thereof include trimethylamine, triethylamine, tripropylamine, trinormal butylamine, triisobutylamine, trihexylamine, trioctylamine, tridodecylamine, and triphenylamine. Triethylamine or trioctylamine is preferred.

[0201]

The alkoxysilicons, the ethers or the amines are preferably used as the electron-donating compound (E). The amines are more preferably used. These electron-donating compounds (E) may be used alone, or two or more kinds of them may be used in combination.

[0202]

The method for producing an addition polymer of the present invention includes the following two methods.

The first method is a method for producing an addition polymer, which includes the following steps:

(1) a step of preparing a mixture by mixing a solvent for polymerization with the organic compound (D), and

(2) a step of addition polymerizing monomers capable of addition polymerization in the mixture in the presence of a catalyst to produce an addition polymer.

The second method is a method for producing an addition polymer, which includes the following steps:

(1) a step of preparing a mixture by mixing a solvent for polymerization with the organic compound (D),

(2) a step of prepolymerizing monomers capable of addition polymerization in the mixture in the presence of a catalyst to obtain a prepolymerized catalyst component for addition polymerization, and

(3) a step of addition polymerizing monomers capable of addition polymerization in the presence of the prepolymerized catalyst component for addition polymerization to produce an addition polymer.

[0203]

Examples of the solvent for polymerization include aliphatic hydrocarbon solvents such as butane, pentane, hexane, heptane, and octane; aromatic hydrocarbon solvents such as benzene and toluene; and halogenated hydrocarbon solvents such as dichloromethane. The monomer per se can also be used as a solvent for polymerization, and examples of the monomer include ethylene, propylene, 1-butene, and 1-hexene.

[0204]

The present invention includes the step of preparing a mixture by mixing a solvent for polymerization with the organic compound (D) represented by Formula [2].

[0205]

The amount of the organic compound (D) based on the solvent for polymerization is usually within a range from 0.1 to 1,000 mg/L, more preferably from 1 to 100 mg/L, and still more preferably from 1.5 to 40 mg/L.

[0206]

The solvent for polymerization and the organic compound (D) may be mixed in a polymerization reactor or, after mixing the solvent for polymerization with the organic compound (D) in advance, the mixture may be charged into a polymerization reactor. The organic compound (D) may be mixed with the solvent after introducing the monomer into the solvent for polymerization.

A preferable method is a method in which a mixture is prepared by mixing the solvent for polymerization and the organic compound (D), a catalyst is prepared by mixing the compound (A), the activating agent (B) and if necessary, the organic compound (C) with the mixture, and then a monomer is introduced into the mixture and polymerization is performed by the catalyst.

[0207]

There is no particular limitation on the method of mixing the solvent for polymerization with the organic compound (D), and examples thereof include a method in which the solvent for polymerization and the organic compound (D) are mixed as they are; a method in which the organic compound (D) is once mixed with the same solvent as the solvent for polymerization, and then the mixture is mixed with the solvent for polymerization; a method in which the organic compound (D) is once mixed with a solvent different from the solvent for polymerization, and then the mixture is mixed with the solvent for polymerization; a method in which the organic compound (D) is added to the solvent for polymerization, together with an inert gas flow; and a method in which the organic compound (D) is added to the solvent for polymerization, together with a monomer gas flow.

[0208]

In the case of mixing the solvent for polymerization with the organic compound (D), the solvent for polymerization preferably has been stirred.

[0209]

There is no particular limitation on the temperature at which the solvent for polymerization is mixed with the organic compound (D), and it is usually from 0°C to 150°C, preferably from 5°C to 100°C, and more preferably from 10°C to 80°C.

[0210]

The mixture prepared by mixing the solvent for polymerization with the organic compound (D) can be used for polymerization immediately after its preparation, or a mixture is prepared by mixing the solvent for polymerization with the organic compound (D) and then the mixture can be used for polymerization after a while. In the latter case, the mixture is preferably used for polymerization within 5 hours, more preferably 3 hours, and still more preferably 1 hour after preparation of the mixture.

[0211]

Catalyst

The catalyst in the present invention is obtained by bringing the compound (A), the activating agent (B) and if necessary, the organoaluminum compound (C) into contact with one another. With respect to the ratio of amounts of the respective components to be brought into contact with one another, the amount of the compound (A) based on the amount of the activating agent (B) is usually from 0.1 to 1,000 μmol/g, preferably from 1 to 500 μmol/g, and more preferably from 10 to 300 μmol/g. The amount of the organoaluminum compound (C) based on the amount of the compound (A) is usually from 0.01 to 10,000 mmol/g, preferably from 0.1 to 1,000 mmol/g, and more preferably from 0.5 to 200 mmol/g.

[0212]

There is no particular limitation on a method of bringing the compound (A), the activating agent (B) and the organoaluminum compound (C) into contact with one another, and examples of the method include methods of <13> to <15> described below:

<13> a method in which a contact product produced by bringing the above respective components into contact with one another is supplied in a polymerization reactor;

<14> a method in which the above respective components are separately supplied in a polymerization reactor and these components are brought into contact with one another in the polymerization reactor; and

<15> a method in which any components of the above respective components are brought into contact with one another to obtain a precontact product

before supplying the components in a polymerization reactor, and the precontact product is brought into contact with the remaining component(s) in the polymerization reactor.

The contact method is preferably <14> described above. The compound (A) may be supplied into a polymerization reactor in a powder or a slurry state of being suspended in a solvent.

A preferred method is that the respective components, a contact product obtained by bringing the respective components into contact with one another, or a precontact product is supplied into a polymerization reactor in which a mixture obtained by mixing the solvent for polymerization with the organic compound (D) is present.

[0213]

There is no particular limitation on the order of bringing the compound (A), the activating agent (B) and the organoaluminum compound (C) into contact with one another, and examples of the order include orders of <16> to <21> described below:

<16> an order in which a contact product produced by bringing the compound (A) into contact with the activating agent (B) is brought into contact with the organoaluminum compound (C);

<17> an order in which a contact product produced by bringing the compound (A) into contact with the organoaluminum compound (C) is brought into contact with the activating agent (B);

<18> an order in which a contact product produced by bringing the activating agent (B) into contact with the organoaluminum compound (C) is brought into contact with the compound (A);

<19> an order in which a contact product produced by bringing the compound (A) into contact with the organoaluminum compound (C) is brought into contact with a contact product produced by bringing the activating agent (B) into contact with the organoaluminum compound (C);

<20> an order in which a contact product produced by bringing the compound (A) into contact with the activating agent (B) is brought into contact with a contact product produced by bringing the activating agent (B) into contact with the organoaluminum compound (C); and

<21> an order in which a contact product produced by bringing the compound (A) into contact with the organoaluminum compound (C) is brought into contact with a contact product produced by bringing the compound (A) into contact with the activating agent (B).

The contact order is preferably <16> or <17> described above.

[0214]

When the respective components are brought into contact with one another, a solvent may be used. It is preferred to use a solvent since an active spot can be efficiently formed. The solvent to be used at the time of contact may be a solvent which does not deactivate an active spot to be produced. The solvent is more preferably a solvent which dissolves the compound (A). Examples of the solvent include aliphatic hydrocarbon solvents such as butane, pentane, hexane, and octane; aromatic hydrocarbon solvents such as benzene, toluene, and xylene; halogenated hydrocarbon solvents such as dichloromethane; and polar solvents such as ethers, esters, and ketones. When the catalyst is used for polymerization accompanied by formation of particles (for example, slurry polymerization, bulk polymerization, etc.), a preferred solvent is a solvent which does not dissolve the produced addition polymer. Specifically, aliphatic hydrocarbon solvents are preferable. The monomer may be present at the time of contact.

[0215]

In the case of bringing the respective components into contact with one another, the temperature can be optionally set. The temperature is usually from -50°C to 100°C, preferably from -30°C to 80°C, and more preferably from -10°C to 60°C. The contact time can be optionally set. It is usually from substantially 0 minute to 24 hours, preferably from 1 minute to 12 hours, and more preferably from 3 minutes to 10 hours. When the components are charged continuously, the contact time is 0.

[0216]

It is preferred to bring the respective components into contact with one another while stirring them.

[0217]

Addition Polymerization Method using Catalyst

An addition polymerization method using a catalyst is a method of addition polymerizing monomers using a catalyst in a solvent. Examples of the addition polymerization method using a catalyst include a solution polymerization method in which monomers are polymerized in a solvent, a slurry polymerization method (a suspension polymerization method), and a bulk polymerization method in which monomers as a solvent are polymerized, and among them, a slurry polymerization method or a bulk polymerization method is preferable.

[0218]

The addition polymerization using a catalyst can be carried out in a batch manner or a continuous manner. The polymerization time of the addition polymerization using a catalyst is commonly determined according to the kind of the objective addition polymer and the polymerization reactor to be used, and is usually from 1 minute to 20 hours.

[0219]

The addition polymerization using a catalyst can be carried out according to publicly-known methods and conditions. A preferred method thereof is a method in which monomers, a solvent and, optionally, other materials to be supplied are continuously or intermittently supplied to a polymerization reactor, and then the produced addition polymer is continuously or intermittently extracted from the polymerization reactor. It is possible to use, as the polymerization reactor, a loop reactor, a reactor equipped with a stirrer, and a reactor in which a plurality of reactors equipped with a stirrer, each having a different type and a different polymerization reaction condition, are connected in series, in parallel or a combination thereof.

[0220]

The solvent for polymerization, the polymerization temperature and the polymerization pressure are selected so that the solvent and at least a portion of the monomer can be maintained in a liquid phase and the monomer can be brought into contact with the catalyst. The polymerization temperature is usually from about -50°C to about 150°C. The polymerization pressure is usually from about 0.001 MPa to about 10

MPa.

[0221]

In the addition polymerization using a catalyst, a catalyst or a component to be used so as to obtain the catalyst, and a monomer can be supplied to the polymerization reactor in any order by publicly-known methods. Examples of a method of supplyng them to the polymerization reactor include (1) a method in which a catalyst or components to be used so as to obtain the catalyst, and monomers are simultaneously supplied to the polymerization reactor containing a mixture obtained by mixing a solvent for polymerization with the organic compound (D), and (2) a method in which a catalyst or components to be used so as to obtain the catalyst, and monomers are successively supplied to the polymerization reactor containing a mixture obtained by mixing a solvent for polymerization with the organic compound (D).

[0222]

It is possible to control the molecular weight of the produced addition polymer by the polymerization temperature or a molecular weight modifier such as hydrogen.

[0223]

Prepolymerization Method and Prepolymerized Catalyst Component for Addition Polymerization

Among the addition polymerization method of the present invention, a method of prepolymerizing monomers capable of addition polymerization using a catalyst to obtain a prepolymerized catalyst component for addition polymerization is particularly referred to as a prepolymerization method.

The prepolymerized catalyst component for addition polymerization may be sometimes referred to as a prepolymerized catalyst component.

[0224]

The prepolymerization can be carried out in the same method and conditions as in the above-mentioned addition polymerization. The prepolymerization can be carried out in a batch manner or a continuous manner. Furthermore, the prepolymerization can be carried out in two or more stages, each having a different reaction condition. Generally, the prepolymerization time is appropriately determined according to the kind of the objective olefin polymer and a reactor, and can be within a range from 1 minute to 20 hours. The prepolymerization temperature is preferably from -50°C to 100°C, more preferably from -30°C to 80°C, and still more preferably from -10°C to 60°C. The temperature may be changed during the prepolymerization. The pressure at the time of the prepolymerization is usually from 0.001 MPa to 5 MPa, and preferably from 0.01 MPa to 2 MPa.

[0225]

In the prepolymerization of the present invention, a catalyst, components to be used so as to obtain the catalyst, and monomers can be supplied to the polymerization reactor by the same method as in the above-mentioned addition polymerization. The method is preferably a method of supplying the powdery compound (A) to the polymerization reactor.

[0226]

In the prepolymerization, it is also possible to adjust the molecular weight of a prepolymer by a molecular weight modifier such as hydrogen.

[0227]

In the present invention, since a prepolymerized catalyst component is prepared in the presence of a solvent for polymerization, the prepolymerized catalyst component may be used for main polymerization, together with the solvent for polymerization. The prepolymerized catalyst component is separated from the solvent for polymerization and the prepolymerized catalyst component separated is subjected to a treatment such as demonomerization, solvent distillation, filtration, washing, and drying to obtain a prepolymerized catalyst component in a solid state, and the resultant prepolymerized catalyst component may be used for the main polymerization.

[0228]

The smaller the content of fine powders in the prepolymerized catalyst component, the better from the viewpoint of enhancing operation stability of the main polymerization using the prepolymerized catalyst component. The content of fine powders in the prepolymerized catalyst component is evaluated, for example, by the content of particles having a diameter of 60 μm or less. The content of particles having a diameter of 60 μm or less in the prepolymerized catalyst component is preferably from 7.0% or less, more preferably from 5.0% or less, still more preferably from 2.0% or less, and particularly preferably from 0.8% or less. The content of fine powders in the prepolymerized catalyst component can be adjusted, for example, by the median diameter or SPAN of the activating agent (B) to be used in the prepolymerization, and the kind of the organic compound (D) to be mixed with the solvent for polymerization.

[0229]

The prepolymerization is carried out so that the amount of the polymer produced by prepolymerization can be within a range from 0.1 to 1,000 g, preferably from 0.5 to 500 g, more preferably from 1 to 100 g per gram of the component (B). The amount of the polymer produced by prepolymerization is sometimes referred to as the degree of prepolymerization.

[0230]

Main Polymerization

In the present invention, addition polymerization using a prepolymerized catalyst component is referred to as main polymerization. In the main polymerization, the prepolymerized catalyst component obtained may be used as it is as a catalyst for addition polymerization, and a contact product of the prepolymerized catalyst component and the organoaluminum compound may be used as a catalyst for addition polymerization. From the viewpoint of being superior in polymerization activity, the latter case is preferable. As the organoaluminum compound for the latter case, the organoaluminum compound exemplified as the compound (C) is used. When the organoaluminum compound is used in such a manner, the amount thereof to be used is usually from 1 to 10,000 mol/mol, preferably from 10 to 5,000 mol/mol, and more preferably from 30 to 1,000 mol/mol, based on the compound (A).

[0231]

When the prepolymerized catalyst component and the organoaluminum compound (C) are used after bringing them into contact with each other, the prepolymerized catalyst component and the organoaluminum compound (C) can be supplied into a polymerization reactor in any order, or they may be supplied into a polymerization reactor after bringing them into contact with each other in advance.

[0232]

There is no particular limitation on a method of supplying the prepolymerized catalyst component and organoaluminum compound (C) to a reactor for catalyst preparation, or a polymerization reactor for the main polymerization. Examples of the method include a method in which the prepolymerized catalyst component and the organoaluminum compound (C) are supplied in a solid state; a method in which the prepolymerized catalyst component and the organoaluminum compound (C) are supplied in the state of being dissolved, suppended or slurried in a hydrocarbyl solvent from which components causing deactiviation of a catalyst component such as moisture and oxygen have been sufficiently removed. Examples of the hydrocarbyl solvent include aliphatic hydrocarbyl solvents such as butane, pentane, hexane, heptane, and octane; aromatic hydrocarbyl solvents such as benzene and toluene; and halogenated hydrocarbyl solvents such as dichloromethane, and among them, aliphatic hydrocarbyl solvents or aromatic hydrocarbyl solvents are preferable, and aliphatic hydrocarbyl solvents are more preferable.

[0233]

There is no particular limitation on a polymerization method in the main polymerization. Examples of the method include (1) a gas phase polymerization method of polymerizing gaseous monomers, (2) a solution polymerization method of polymerizing monomers in a solvent, or a slurry polymerization method (suspension polymerization method), and (3) a bulk polymerization method of polymerizing liquid monomers in the solvent consisting of the liquid monomers. The main polymerization can be carried out in a batch manner or a continuous manner. Furthermore, the main prepolymerization can be carried out in two or more stages, each having a different reaction condition. The polymerization time of the main polymerization is appropriately determined according to the kind of the objective addition polymer and the polymerization reactor to be used, and is usually within a range from 1 minute to 20 hours.

[0234]

When the main polymerization is solution polymerization, slurry polymerization, or bulk polymerization, the polymerization can be carried out according to the publicly-known methods and conditions. A preferred method thereof is a method in which monomers, a diluent and, optionally, other materials to be supplied are continuously or intermittently supplied to a polymerization reactor, and then the produced addition polymer is continuously or intermittently extracted from the polymerization reactor. Examples of the polymerization reactor include a loop reactor, a reactor equipped with a stirrer, and a reactor in which a plurality of reactors equipped with a stirrer, each having a different type and a different polymerization reaction condition, are connected in series, in parallel or a combination thereof.

[0235]

When the main polymerization is gas phase polymerization, the gas phase polymerization can be carried out according to the publicly-known methods and conditions. The reactor for the gas phase polymerization is a fluidized bed type reaction vessel, and preferably a fluidized bed type reaction vessel with an enlarged portion. The reactor may have a stirring blade in the reaction vessel.

Examples of a method in which the prepolymerized catalyst component is supplied to a reactor include a method in which the catalyst component is supplied in a state free from moisture usually together with an inert gas such as nitrogen or argon, hydrogen or ethylene; or a method in which the catalyst component is supplied in a state of a solution prepared by dissolving the prepolymerized catalyst component in a solvent or in a state of a slurry prepared by diluting the prepolymerized catalyst component with the solvent.

[0236]

When the main polymerization is gas phase polymerization, there is no particular limitation on the polymerization temperature of the gas phase polymerization, as long as the polymerization temperature is lower than the melting temperature of a addition polymer to be produced, and it is preferably from 0°C to 150°C, and particularly preferably from 30°C to 100°C. For the purpose of controlling the melt fluidity of the addition polymer to be produced, hydrogen may be added as a molecular weight modifier. When gaseous monomers are polymerized, the polymerization may be carried out in an inert gas.

[0237]

Monomers capable of Addition Polymerization

The method for producing an addition polymer of the present invention is a method for producing an addition polymer comprising addition polymerizing monomers capable of addition polymerization in the presence of the above catalyst or the prepolymerized catalyst for addition polymerization.

Examples of the monomer in the method for producing an addition polymer of the present invention include an olefin, a diolefin, a cyclic olefin, an alkenylaromatic hydrocarbyl and a polar monomer, each having 2 to 20 carbon atoms. Two or more kinds of monomers can be used.

[0238]

Examples of the monomer include olefins such as ethylene, propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, 5-methyl-1-hexene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene and vinylcyclohexane; diolefins such as 1,5-hexadiene, 1,4-hexadiene, 1,4-pentadiene, 1,7-octadiene, 1,8-nonadiene, 1,9-decadiene, 4-methyl-1,4-hexadiene, 5-methyl-1,4-hexadiene, 7-methyl-1,6-octadiene, 5-ethylidene-2-norbornene, dicyclopentadiene, 5-vinyl-2-norbornene, norbornadiene, 5-methylene-2-norbornene, 1,5-cyclooctadiene, 5,8-endomethylenehexahydronaphthalene, 1,3-butadiene, isoprene, 1,3-hexadiene, 1,3-octadiene, 1,3-cyclooctadiene and 1,3-cyclohexadiene; cyclic olefins such as norbornene, 5-methyl-2-norbornene, 5-ethyl-2-norbornene, 5-butyl-2-norbornene, 5-phenyl-2-norbornene, 5-benzyl-2-norbornene, tetracyclododecene, tricyclodecene, tricycloundecene, pentacyclopentadecene, pentacyclohexadecene, 8-methyltetracyclododecene, 8-ethyltetracyclododecene, 5-acetyl-2-norbornene, 5-acetyloxy-2-norbornene, 5-methoxycarbonyl-2-norbornene, 5-

ethoxycarbonyl-2-norbornene, 5-methyl-5-methoxycarbonyl-2-norbornene, 5-cyano-2-norbornene, 8-methoxycarbonyltetracyclododecene, 8-methyl-8-tetracyclododecene and 8-cyanotetracyclododecene; alkenylaromatic hydrocarbyls such as styrene, alkenylbenzenes (e.g., 2-phenylpropylene, 2-phenylbutene, and 3-phenylpropylene), alkylstyrenes (e.g., p-methylstyrene, m-methylstyrene, o-methylstyrene, p-ethylstyrene, m-ethylstyrene, o-ethylstyrene, α-methylstyrene, 2,4-dimethylstyrene, 2,5-dimethylstyrene, 3,4-dimethylstyrene, 3,5-dimethylstyrene, 3-methyl-5-ethylstyrene, 1,1-diphenylethylene, p-tertiary butylstyrene, and p-secondary butylstyrene), bisalkenylbenzenes (e.g., divinylbenzene) and alkenylnaphthalene (e.g., 1-vinylnaphthalene); α,β-unsaturated carboxylic acids (e.g., acrylic acid, methacrylic acid, fumaric acid, maleic anhydride, itaconic acid, itaconic anhydride, and bicyclo(2,2,1)-5-heptene-2,3-dicarboxylic acid), metal salts of metals such as sodium, potassium, lithium, zinc, magnesium and calcium of the α,β-unsaturated carboxylic acid; and α,β-unsaturated carboxylic acid esters (e.g., methyl acrylate, ethyl acrylate, n-propyl acrylate, isopropyl acrylate, tert-butyl acrylate, 2-ethylhexyl acrylate, methyl methacrylate, ethyl methacrylate, n-propyl methacrylate, isopropyl methacrylate, n-butyl methacrylate, and isobutyl methacrylate), unsaturated dicarboxylic acids (e.g., maleic acid and itaconic acid), vinyl esters (e.g., vinyl acetate, vinyl propionate, vinyl caproate, vinyl caprate, vinyl laurate, vinyl stearate, and vinyl trifluoroacetate) and polar monomers such as unsaturated carboxylic acid glycidyl esters (e.g., glycidyl acrylate, glycidyl methacrylate, and itaconic acid monoglycidyl ester).

[0239]

The monomer is preferably an olefin having 2 to 20 carbon atoms. Examples of the olefin include ethylene, propylene, 1-butene, 1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene, 4-methyl-1-pentene, 4-methyl-2-pentene, vinylcyclohexane and the like. The monomer to be used in the prepolymerization is particularly preferably ethylene.

[0240]

When two or more kinds of monomers are used, ethylene and α-olefin are preferably used. The combination of monomers to be used is preferably a combination of ethylene and propylene, a combination of ethylene and 1-butene, a combination of ethylene and 1-hexene, a combination of ethylene and 1-octene, a combination of ethylene, 1-butene and 1-hexene, a combination of ethylene, 1-butene and 1-octene, a combination of ethylene, 1-hexene and 1-octene, a combination of propylene and 1-butene, a combination of propylene and 1-hexene, or a combination of propylene and 1-octene.

[0241]

The addition polymer obtained by the production method of the present invention is particularly preferably a copolymer of ethylene and α-olefin, and among them, a copolymer of ethylene and α-olefin having a polyethylene crystal structure is preferable. The α-olefin is preferably an α-olefin having 3 to 8 carbon atoms, and examples thereof include 1-butene, 1-hexene, and 1-octene.

Examples

[0242]

The present invention will be described in more detail below by way of examples and comparative examples, but the present invention is not limited thereto. The properties of addition polymers in examples were measured by the following methods.

[0243]

(1) Elemental Analysis

Zn: After adding a sample in an aqueous sulfuric acid solution (1M), ultrasonic waves were applied to the aqueous sulfuric acid solution containing the sample thereby extracting a metal component in the aqueous sulfuric acid solution from the sample. Zn in a liquid portion of the aqueous sulfuric acid solution containing the metal component was quantitatively determined by ICP spectrometry.

F: A sample was combusted in a flask filled with oxygen and the generated combustion gas was absorbed in an aqueous sodium hydroxide solution (10%), and then F in the obtained aqueous solution was quantitatively determined using an ionic electrode method.

[0244]

(2) MFR: MFR is a melt flow rate (unit: g/10 minutes) measured at 190°C under a load of 21.18 N (2.16 kg) in accordance with the method defined in JIS K7210-1995.

[0245]

(3) Swell ratio = SR: A value which is obtained by dividing a strand diameter obtained at the time of the measurement of MFR by an inner diameter (2.095 mm) of a die used for measureing MFR.

[0246]

(4) Cumulative Weight Fraction of Particles having Particle Diameter of Prepolymerized Catalyst Component for Addition Polymerization of 60 µm or less.

A prepolymerized catalyst component for addition polymerization was dispersed in a dry state and the particle size distribution thereof was measured under the following conditions using a laser diffraction particle size distribution measurement apparatus HELOS&RODOS system manufactured by SYMPATEC GmbH. The amount of a prepolymerized catalyst component for addition polymerization having a particle diameter of 60 µm or less was calculated expressed by volume.

[0247]

(Measuring Conditions)

Range : R4 1.8 to 350 µm
Trigger conditions : Reference duration 2 seconds
: Time base 100 ms
: Start channel 15 ≥ 0.5%
: Stop 2s, channel 15 ≤ 0.5%, 10s real time
Dispersion conditions : RODOS (dry gas flow type dispersing unit) direct charge type
: Feeder VIBRI
: Feed 50%
: Injector 4 mm
: Dispersion pressure 1.5 bars

[0248]

[Example 1]

(1) Production of Activating Agent (B)

In the same manner as in the preparation of component (A) of Example 1(1) and Example 1 (2) of JP-A-2009-79180, modified particles were produced. As a result of elemental analysis, Zn content was 11% by weight and F content was 6.4% by weight.

[0249]

(2) Prepolymerization

After vacuum drying, a nitrogen-substituted autoclave (inner volume of 3 L) equipped with a stirrer was evacuated, and then charged with 480 g of butane and 1.0 ml of a solution of sodium polyoxyethylene lauryl acetate (n = 10) (KAOAKYPO RLM-100NV, manufactured by Kao Corporation) (D) in hexane, the concentration of which had been adjusted to 10 mg/ml. Then, 192 mg (360 μmol) of ethylenebis(indenyl)zirconium diphenoxide (A) was added to the autoclave, followed by stirring at 50°C for 1 hour and further cooling to 30°C. Then, 1 g of ethylene was added and 7.0 g of the activating agent (B) obtained in the above (1) was added. Furthermore, 3.5 ml (3.5 mmol) of a solution of triisobutyl aluminum (C) in hexane, the concentration of which had been adjusted to 1.0 mmol/ml, was added, and prepolymerization was initiated. First, while feeding ethylene at a rate of 0.13 g/minute, prepolymerization was carried out at 30°C for 30 minutes. While feeding an ethylene/hydrogen mixed gas (hydrogen concentration: 0.193 mol%) at a rate of 0.81 g/minute as a substitute for ethylene, the temperature was raised to 50°C over 30 minutes and then prepolymerization was carried out at 50°C for 2 hours. The monomer and butane were purged, and then a prepolymerized catalyst component for addition polymerization was recovered. The recovered amount was 131.7 g, and the degree of polymerization per activating agent (B) was 18.7 g/g. The particle size distribution of the prepolymerized catalyst component was measured, and as a result, the amount of particles having a particle diameter of 60 μm or less contained in the prepolymerized catalyst component was 1.9%.

[0250]

[Example 2]

(1) Prepolymerization

Prepolymerization was carried out in the same manner as in Example 1(2), except that 200 mg (374 μmol) of ethylenebis(indenyl)zirconium diphenoxide (A) and 2.0 ml of a solution of sodium polyoxyethylene lauryl acetate (n = 10) (KAOAKYPO RLM-100NV, manufactured by Kao Corporation) (D) in hexane, the concentration of which had been adjusted to 10 mg/ml, were added. The recovered amount was 131.2 g and the degree of polymerization per activating agent (B) was 18.7 g/g. The particle size distribution of the prepolymerized catalyst component was measured, and as a result, the amount of particles having a particle diameter of 60 μm or less contained in the prepolymerized catalyst component was 0.8%.

(2) Main Polymerization

After vacuum drying, an argon-substituted autoclave (inner volume of 5 L) equipped with a stirrer was evacuated, and hydrogen was added at a partial pressure of 0.037 MPa, and then 154 g of hexene-1 and 1,046 g of butane were supplied into the autoclave and the temperature was raised to

70°C. Then, ethylene was added so that the partial pressure thereof might become 1.6 MPa, and the system was stabilized. As a result of gas chromatographic analysis, the gas composition in the system was as follows: hydrogen = 1.86 mol%. A solution (2.0 ml) of triisobutyl aluminum in hexane, the concentration of which had been adjusted to 1 mmol/ml, was supplied thereto. Then, a solution (1.0 ml) of triethylamine in toluene, the concentration of which had been adjusted to 0.1 mmol/ml, was added thereto. Furthermore, 381.3 mg of the prepolymerized catalyst component obtained in the above (1) was supplied. While feeding an ethylene/hydrogen mixed gas (hydrogen: 0.295 mol%) so as to maintain the entire pressure at a given value, polymerization was carried out at 70°C for 3 hours. As a result, 72 g of an olefin polymer was obtained. The activity of polymerization per activating agent (B) was 3,500 g/g. The obtained olefin polymer exhibited an MFR of 2.12, and an SR of 1.45.

[0251]

[Example 3]

(1) Prepolymerization

Prepolymerization was carried out in the same manner as in Example 1(2), except that 195mg (365 µmol) of ethylenebis(indenyl)zirconium diphenoxide (A) and 5.0 ml of a solution of sodium polyoxyethylene lauryl acetate (n = 10) (KAOAKYPO RLM-100NV, manufactured by Kao Corporation) (D) in hexane, the concentration of which had been adjusted to 10 mg/ml, were added. The recovered amount was 129.2 g and the degree of polymerization per activating agent (B) was 18.2 g/g. The particle size distribution of the prepolymerized catalyst component was measured, and as a result, the amount of particles having a particle diameter of 60 μm or less contained in the prepolymerized catalyst component was 1.5%.

(2) Main Polymerization

After vacuum drying, an argon-substituted autoclave (inner volume of 5 L) equipped with a stirrer was evacuated, and hydrogen was added at a partial pressure of 0.037 MPa, and then 154 g of hexene-1 and 1,046 g of butane were supplied into the autoclave and the temperature was raised to 70°C. Then, ethylene was added so that the partial pressure thereof might become 1.6 MPa, and the system was stabilized. As a result of gas chromatographic analysis, the gas composition in the system was as follows: hydrogen = 1.86 mol%. A solution (2.0 ml) of triisobutyl aluminum in hexane, the concentration of which had been adjusted to 1 mmol/ml, was supplied thereto. Then, a solution (1.0 ml) of triethylamine in toluene, the concentration of which had been adjusted to 0.1 mmol/ml, was added thereto. Furthermore, 374.2 mg of the prepolymerized catalyst component obtained in the above (1) was supplied. While feeding an ethylene/hydrogen mixed gas (hydrogen 0.300 mol%) so as to maintain the entire pressure at a given value, polymerization was carried out at 70°C for 3 hours. As a result, 98.5 g of an olefin polymer was obtained. The activity of polymerization per activating agent (B) was 4,780 g/g. The obtained olefin polymer exhibited an MFR of 1.19, and an SR of 1.39.

[0252]

[Example 4]

(1) Prepolymerization

Prepolymerization was carried out in the same manner as in Example 1(2), except that 205mg (384 μmol) of ethylenebis(indenyl)zirconium diphenoxide (A) and 30.0 ml of a solution of sodium polyoxyethylene lauryl acetate (n = 10) (KAOAKYPO RLM-100NV, manufactured by Kao Corporation) (D) in hexane, the concentration of which had been adjusted to 10 mg/ml, were added. The recovered amount was 143.9 g and the degree of polymerization per activating agent (B) was 20.4 g/g. The particle size distribution of the prepolymerized catalyst component was measured, and as a result, the amount of particles having a particle diameter of 60 μm or less contained in the prepolymerized catalyst component was 0.4%.

(2) Main Polymerization

After vacuum drying, an argon-substituted autoclave (inner volume of 5 L) equipped with a stirrer was evacuated, and hydrogen was added at a partial pressure of 0.037 MPa, and then 154 g of hexene-1 and 1,046 g of butane were supplied into the autoclave and the temperature was raised to 70°C. Then, ethylene was added so that the partial pressure thereof might become 1.6 MPa, and the system was stabilized. As a result of gas chromatographic analysis, the gas composition in the system was as follows: hydrogen = 1.92 mol%. A solution (2.0 ml) of triisobutyl aluminum in hexane, the concentration of which had been adjusted to 1 mmol/ml, was supplied thereto. Then, a solution (1.0 ml) of triethylamine in toluene, the concentration of which had been adjusted to 0.1 mmol/ml, was added thereto. Furthermore, 409.8 mg of the prepolymerized catalyst component obtained in the above (1) was supplied. While feeding an ethylene/hydrogen mixed gas (hydrogen 0.300 mol%) so as to maintain the entire pressure at a given value, polymerization was carried out at 70°C for 3 hours. As a result, 3.7 g of an olefin polymer was obtained. The activity of polymerization per activating agent (B) was 200 g/g. The obtained olefin polymer exhibited an MFR of 5.

[0253]

[Example 5]

(1) Prepolymerization

Prepolymerization was carried out in the same manner as in Example 1(2), except that 195 mg (365 μmol) of ethylenebis(indenyl)zirconium diphenoxide (A) and 5.0 ml of a solution of sodium polyoxyethylene lauryl sulfate (n = 2) (EMAL 270J, manufactured by Kao Corporation) (D) in hexane, the concentration of which had been adjusted to 10 mg/ml, were added. The recovered amount was 123.0 g and the degree of polymerization per activating agent (B) was 17.5 g/g. The particle size distribution of the prepolymerized catalyst component was measured, and as a result, the amount of particles having a particle diameter of 60 μm or less contained in the prepolymerized catalyst component was 5.3%.

(2) Main Polymerization

After vacuum drying, an argon-substituted autoclave (inner volume of 5 L) equipped with a stirrer was evacuated, and hydrogen was added at a partial pressure of 0.037 MPa, and then 154 g of hexene-1 and 1,046 g of butane were supplied into the autoclave and the temperature was raised to 70°C. Then, ethylene was added so that the partial pressure thereof might become 1.6 MPa, and the system was stabilized. As a result of gas chromatographic analysis, the gas composition in the system was as follows: hydrogen = 1.86 mol%. A solution (2.0 ml) of triisobutyl aluminum in hexane, the concentration of which had been adjusted to 1 mmol/ml, was supplied thereto. Then, a solution (1.0 ml) of triethylamine in toluene, the concentration of which had been adjusted to 0.1 mmol/ml, was added thereto. Furthermore, 355.0 mg of the prepolymerized catalyst component obtained in the above (1) was supplied. While feeding an ethylene/hydrogen mixed gas (hydrogen 0.309 mol%) so as to maintain the entire pressure at a given value, polymerization was carried out at 70°C for 3 hours. As a result, 21.3 g of an olefin polymer was obtained. The activity of polymerization per activating agent (B) was 1,000 g/g. The obtained olefin polymer exhibited an MFR of 6.54, and an SR of 1.52.

[0254]

[Example 6]

(1) Prepolymerization

Prepolymerization was carried out in the same manner as in Example 1(2), except that 191 mg (359 μmol) of ethylenebis(indenyl)zirconium diphenoxide (A) and 5.0 ml of a solution of polyoxyethylene lauryl ether (EMALGEN 108, manufactured by Kao Corporation) (D) in hexane, the concentration of which had been adjusted to 10 mg/ml, were added. The recovered amount was 123.2 g and the degree of polymerization per activating agent (B) was 17.5 g/g. The particle size distribution of the prepolymerized catalyst component was measured, and as a result, the amount of particles having a particle diameter of 60 μm or less contained in the prepolymerized catalyst component was 2.4%.

(2) Main Polymerization

After vacuum drying, an argon-substituted autoclave (inner volume of 5 L) equipped with a stirrer was evacuated, and hydrogen was added at a partial pressure of 0.037 MPa, and then 154 g of hexene-1 and 1,046 g of butane were supplied into the autoclave and the temperature was raised to 70°C. Then, ethylene was added so that the partial pressure thereof might become 1.6 MPa, and the system was stabilized. As a result of gas chromatographic analysis, the gas composition in the system was as follows: hydrogen = 1.89 mol%. A solution (2.0 ml) of triisobutyl aluminum in hexane, the concentration of which had been adjusted to 1 mmol/ml, was supplied thereto. Then, a solution (1.0 ml) of triethylamine in toluene, the concentration of which had been adjusted to 0.1 mmol/ml, was added thereto. Furthermore, 352.1 mg of the prepolymerized catalyst component obtained in the above (1) was supplied. While feeding an ethylene/hydrogen mixed gas (hydrogen 0.309 mol%) so as to maintain the entire pressure at a given value, polymerization was carried out at 70°C for 3 hours. As a result, 26.8 g of an olefin polymer was obtained. The activity of polymerization per activating agent (B) was 1,300 g/g. The obtained olefin polymer exhibited an MFR of 4.14, and an SR of 1.48.

[Comparative Example 1]

(1) Prepolymerization

Prepolymerization was carried out in the same manner as in Example 1(2), except that 198mg (372 μmol) of ethylenebis(indenyl)zirconium diphenoxide (A) and 7.0 g of the activating agent (B) obtained in Example 1(1) were added, and the organic compound (D) was not added. The recovered amount was 130.3 g and the degree of polymerization per activating agent (B) was 18.6 g/g. The particle size distribution of the prepolymerized catalyst component was measured, and as a result, the amount of particles having a particle diameter of 60 μm or less contained in the prepolymerized catalyst component was 11.0%.

(2) Main Polymerization

After vacuum drying, an argon-substituted autoclave (inner volume of 5 L) equipped with a stirrer was evacuated, and hydrogen was added at a partial pressure of 0.037 MPa, and then 154 g of hexene-1 and 1,046 g of butane were supplied into the autoclave and the temperature was raised to 70°C. Then, ethylene was added so that the partial pressure thereof might become 1.6 MPa, and the system was stabilized. As a result of gas chromatographic analysis, the gas composition in the system was as follows: hydrogen = 1.86 mol%. A solution (2.0 ml) of triisobutyl aluminum in hexane, the concentration of which had been adjusted to 1 mmol/ml, was supplied thereto. Then, a solution (1.0 ml) of triethylamine in toluene, the concentration of which had been adjusted to 0.1 mmol/ml, was added thereto. Furthermore, 378.3 mg of the prepolymerized catalyst component obtained in the above (1) was supplied. While feeding an ethylene/hydrogen mixed gas (hydrogen 0.301 mol%) so as to maintain the entire pressure at a given value, polymerization was carried out at 70°C for 3 hours. As a result, 76.0 g of an olefin polymer was obtained. The activity of polymerization per activating agent (B) was 3,720 g/g. The obtained olefin polymer exhibited an MFR of 2.44, and an SR of 1.45.

CLAIMS

1. A method for addition polymerizing monomers capable of addition polymerization, wherein the method comprises the following steps: (1) a step of preparing a mixture by mixing a solvent for polymerization with an organic compound (D) represented by Formula [2],
(2) a step of addition polymerizing said monomers in the mixture in the presence of a catalyst prepared by bringing a compound (A) selected from among transition metal compounds represented by Formula [1] and their μ-oxo type transition metal compound dimers, and an activating agent (B) into contact with each other,

$$L^1_aM^1X^1_b \qquad [1]$$

wherein M^1 is a transition metal atom of Group 4; L^1 is a group having a cyclopentadiene type anionic frame or a group containing a hetero atom; X^1 is a halogen atom, a hydrocarbyloxy group, or a hydrocarbyl group other than groups having a cyclopentadiene type anionic frame; a is a number satisfying $0 < a \leq 3$; b is a number satisfying $0 < b \leq 3$; when a is more than 1, one L^1 may be linked to another L^1 either directly or by a group containing a carbon atom, a silicon atom, a nitrogen atom, an oxygen atom, a sulfur atom or a phosphorus atom,

$$R^1\text{-}O\text{-}(\text{-}R^2\text{-}O\text{-})_nQ^1 \qquad [2]$$

wherein R^1 is a hydrocarbyl group having 1 to 30 carbon atoms which may have a substituent; R^2 is an alkylene group having 1 to 20 carbon atoms which may have a substituent; Q^1 is a hydrogen atom, $-C(=O)OM^2$, $-R^3-C(=O)OM^2$, $-S(=O)_2OM^2$, $-R^3-S(=O)_2OM^2$, $-P(=O)(OH)(OM^2)$, -

P(=O)(OR^4)(OM^2) or -P(=O)(OM^2)$_2$; M^2 is a hydrogen atom or an alkali metal atom; R^3 is an alkylene group having 1 to 20 carbon atoms which may have a substituent; R^4 is a hydrocarbyl group having 1 to 20 carbon atoms which may have a substituent; n is a number of 1 to 100; when n is more than 1, R^2 groups may be the same or different.

2. A method for addition polymerizing monomers capable of addition polymerization, wherein the method comprises the following steps: (1) a step of preparing a mixture by mixing a solvent for polymerization with an organic compound (D) represented by Formula [2], (2) a step of addition polymerizing said monomers in the mixture in the presence of a catalyst prepared by bringing a compound (A) selected from among transition metal compounds represented by Formula [1] and their μ-oxo type transition metal compound dimers, an activating agent (B) and an organoaluminum compound (C) into contact with one another,

$$L^1_a M^1 X^1_b \qquad [1]$$

wherein M^1 is a transition metal atom of Group 4; L^1 is a group having a cyclopentadiene type anionic frame or a group containing a hetero atom; X^1 is a halogen atom, a hydrocarbyloxy group, or a hydrocarbyl group other than groups having a cyclopentadiene type anionic frame; a is a number satisfying $0 < a \leq 3$; b is a number satisfying $0 < b \leq 3$; when a is more than 1, one L^1 may be linked to another L^1 either directly or by a group containing a carbon atom, a silicon atom, a nitrogen atom, an oxygen atom, a sulfur atom or a phosphorus atom,

$$R^1\text{-}O\text{-}(\text{-}R^2\text{-}O\text{-})_n Q^1 \qquad [2]$$

wherein R^1 is a hydrocarbyl group having 1 to 30 carbon atoms which may have a substituent; R^2 is an alkylene group having 1 to 20 carbon atoms which may have a substituent; Q^1 is a hydrogen atom, $-C(=O)OM^2$, $-R^3-C(=O)OM^2$, $-S(=O)_2OM^2$, $-R^3-S(=O)_2OM^2$, $-P(=O)(OH)(OM^2)$, $-P(=O)(OR^4)(OM^2)$ or $-P(=O)(OM^2)_2$; M^2 is a hydrogen atom or an alkali metal atom; R^3 is an alkylene group having 1 to 20 carbon atoms which may have a substituent; R^4 is a hydrocarbyl group having 1 to 20 carbon atoms which may have a substituent; n is a number of 1 to 100; when n is more than 1, R^2 groups may be the same or different.

3. The method according to claim 1 or 2, wherein the activating agent (B) is a modified particle produced by bringing the following (a), (b), (c) and (d) into contact with one another,

(a): a compound represented by Formula [3],

$M^3L^2_2$ [3]

(b): a compound represented by Formula [4],

$R^5_{t-1}TH$ [4]

(c): a compound represented by Formula [5],

$R^6_{u-2}T'H_2$ [5]

(d): an inorganic oxide particle or an organic polymer particle,

wherein M^3 is a Group 12 element; L^2 is a hydrogen atom, a halogen atom or a hydrocarbyl group, and two L^2 groups may be the same or different; R^5 is an electron withdrawing group or a group containing an electron withdrawing group; and when t-1 is more than 1, R^5 groups may be the same or different; R^6 is a hydrocarbyl group or a halogenated hydrocarbyl group; T is a Group 15 element or a Group 16 element; t is a number corresponding to the valence of T; T' is a Group 15 element or a Group 16 element; u is a number corresponding to the valence of T'.

4. The method according to any one of claims 1 to 3, wherein Q^1 in Formula [2] is a hydrogen atom, $-C(=O)OM^2$, $-R^3-C(=O)OM^2$, $-S(=O)_2OM^2$, or $-R^3-S(=O)_2OM^2$, M^2 is a hydrogen atom or an alkali metal atom, and R^3 is an alkylene group having 1 to 20 carbon atoms which may have a substituent.

5. The method according to claim 4, wherein Q^1 in Formula [2] is a hydrogen atom, $-R^3-C(=O)OM^2$, or $-S(=O)_2OM^2$.

6. The method according to any one of claims 1 to 5, wherein the method is carried out in a slurry.

7. A method for producing an addition polymer, the method comprising addition polymerizing monomers capable of addition polymerization, by the method according to any one of claims 1 to 6.

8. A method for producing a prepolymerized catalyst component for addition polymerization, the method comprising producing a prepolymerized catalyst component for addition polymerization, by the method according to any one of claims 1 to 6.

9. A prepolymerized catalyst component for addition polymerizing,

wherein the prepolymerized catalyst component is obtained by the method according to claim 8, and wherein the cumulative weight fraction of a prepolymerized catalyst component having a particle diameter of 60 µm or less is 7% or less.

10. A method for producing an addition polymer, the method comprising polymerizing monomers capable of addition polymerization in the presence of the prepolymerized catalyst component for addition polymerization obtained by the method according to claim 8.

11. A method for producing an addition polymer, the method comprising polymerizing monomers capable of addition polymerization in the presence of a mixture of the prepolymerized catalyst component for addition polymerization obtained by the method according to claim 8, and an organoaluminum compound (C).

12. The method according to claim 10 or 11, wherein the monomers are ethylene and an α-olefin.

INTERNATIONAL SEARCH REPORT

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PCT/JP2012/056042

A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl. C08F4/6592(2006.01)i, C08F10/00(2006.01)i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl. C08F4/6592, C08F10/00

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Published examined utility model applications of Japan 1922-1996
Published unexamined utility model applications of Japan 1971-2012
Registered utility model specifications of Japan 1996-2012
Published registered utility model applications of Japan 1994-2012

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	JP 09-157320 A (MITSUI PETROCHEM IND CO LTD) 1997.06.17, Claims,【0016】-【0026】,【0037】,【0049】,【0051】, Examples (No Family)	1,2,4-12 3
X Y	JP 2003-201309 A (TOSOH CORP) 2003.07.18, Claims,【0015】-【0028】,【0030】,【0031】,【0036】, Examples (No Family)	1,2,4-12 3
X Y	JP 2001-048912 A (MITSUI CHEM INC) 2001.02.20, Claims,【0013】-【0032】,【0039】-【0042】,【0053】,【0060】,【0062】, Examples (No Family)	1,2,4-12 3

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

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C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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X Y	JP 2006-206768 A (MITSUI CHEM INC) 2006.08.10, Claims,【0035】-【0051】,【0061】-【0065】,【0071】,【0083】, Examples (No Family)	1,2,4-12 3
X Y A	JP 10-330413 A (MITSUI CHEM INC) 1998.12.15, Claims,【0016】-【0037】,【0052】,【0065】, Examples (No Family)	1,2,4,5,7-12 3 6
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