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-9 APR 2003

TITLE

OLEFIN POLYMERIZATION CATALYST AND PROCESS FOR OLEFIN
POLYMERIZATION USING THE OLEFIN POLYMERIZATION CATALYST

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FIELD OF THE INVENTION

The present invention relates to a novel olefin
polymerization catalyst and a process for olefin
polymerization using the olefin polymerization catalyst.

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BACKGROUND OF THE INVENTION

Polyolefins generally have excellent mechanical
properties, so that they are used in many fields such as
fields of various molded products. With the
diversification of property requirements for the
polyolefins, polyolefins of various properties have been
desired in recent years, and besides increase of
productivity of the polyolefins has been also desired.

As catalysts for producing polyolefins, titanium
catalysts comprising a titanium compound and an
organoaluminum compound and vanadium catalysts comprising
a vanadium compound and an organoaluminum compound have
been hitherto known.

In addition, Ziegler catalysts comprising a
metallocene compound such as zirconocene and an
organoaluminum oxy-compound (aluminoxane) are known as



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-9 APR 2003

catalysts capable of producing polyolefins with a high polymerization activity.

Recently, a transition metal compound having a ligand of diimine structure has been proposed as a novel olefin polymerization catalyst (see International Patent Publication No. 9623010).

As a novel olefin polymerization catalyst, further, a transition metal compound having a salicylaldoimine ligand has been recently proposed by the present applicant in Japanese Patent Application No. 132706/1998. This catalyst exhibits high olefin polymerization activity at a temperature of about 25°C. In the production of polyolefins using an industrial process, however, a polymerization catalyst exhibiting higher polymerization activity at high temperatures is desired because higher temperatures are advantageous in that the reaction heat can be easily removed and the viscosity of the reaction solution can be lowered to thereby enhance productivity.

OBJECT OF THE INVENTION

The present invention has been made under such circumstances as mentioned above, and it is an object of the invention to provide an olefin polymerization catalyst which exhibits a high polymerization activity at

-9 APR 2003

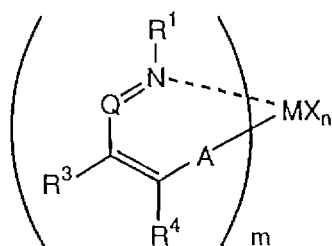
high temperatures. It is another object of the invention to provide a process for olefin polymerization using the catalyst having such excellent properties.

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SUMMARY OF THE INVENTION

The olefin polymerization catalyst according to the present invention comprises a transition metal compound which is represented by the following formula (I) and in which the net charge parameter of the central metal M is not more than 2.00:

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(I)

wherein M is a transition metal atom of Group 4 to Group 5 of the periodic table,

15

m is an integer of 1 to 5,

Q is a nitrogen atom or a carbon atom having a substituent group R²,

A is an oxygen atom, a sulfur atom, a selenium atom or a nitrogen atom having a substituent group R⁵,

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R¹ is an aliphatic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an

-7 APR 2003

alicyclic hydrocarbon group, or an alicyclic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group,

R^2 to R^5 may be the same or different, they are each
5 a hydrogen atom, a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group, a sulfur-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound
10 residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, two or more of them may be bonded to each other to form a ring, and when m is 2 or greater, at least one of groups indicated by R^2 to R^5 contained in one ligand and at least one of
15 groups indicated by R^2 to R^5 contained in another ligand may be bonded to each other,

when m is 2 or greater, R^1s , R^2s , R^3s , R^4s , or R^5s may be the same or different,

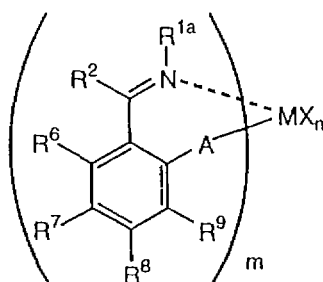
n is a number satisfying a valence of M , and

20 X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a
25 heterocyclic compound residual group, a silicon-

-9 APR 2003

containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

The transition metal compound represented by the above formula (I) includes a transition metal compound which is represented by the following formula (I-a) and in which the net charge parameter of the central metal M is in the range of 1.70 to 2.00:



(I-a)

wherein M, m, A, n and X have the same meanings as those of M, m, A, n and X in the formula (I),

R^{1a} is an aliphatic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group,

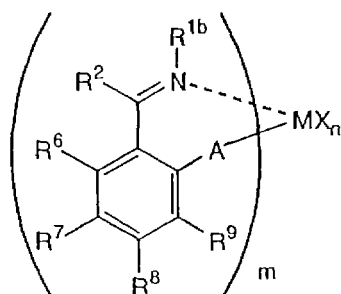
R^2 and R^5 to R^9 may be the same or different, they are each a hydrogen atom, a hydrocarbon group, an oxygen-

-9 APR 2003

containing group, a nitrogen-containing group, a sulfur-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound
 5 residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, two or more of them may be bonded to each other to form a ring, and when m is 2 or greater, at least one of groups indicated by R² and R⁵ to R⁹ contained in one ligand and at least
 10 one of groups indicated by R² and R⁵ to R⁹ contained in another ligand may be bonded to each other, and

when m is 2 or greater, R^{1a}s, R²s, R⁵s, R⁶s, R⁷s, R⁸s, or R⁹s may be the same or different.

The transition metal compound represented by the
 15 formula (I) also includes a transition metal compound which is represented by the following formula (I-b) and in which the net charge parameter of the central metal M is in the range of 1.50 to 1.89:



(I-b)

-9 APR 2003

wherein M, m, A, n and X have the same meanings as those of M, m, A, n and X in the formula (I),

5 R^{1b} is an alicyclic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group,

R^2 and R^5 to R^9 may be the same or different, they are each a hydrogen atom, a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group, a sulfur-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, two or more
10 of them may be bonded to each other to form a ring, and when m is 2 or greater, at least one of groups indicated by R^2 and R^5 to R^9 contained in one ligand and at least one of groups indicated by R^2 and R^5 to R^9 contained in
15 another ligand may be bonded to each other, and

20 when m is 2 or greater, R^{1bs} , R^{2s} , R^{5s} , R^{6s} , R^{7s} , R^{8s} , or R^{9s} may be the same or different.

The transition metal compound represented by the formula (I-b) is preferably a compound wherein A is -O-.

25 The transition metal compound (A-1) represented by the formula (I), (I-a) or (I-b) may be used as an olefin

-9 APR 2003

polymerization catalyst in combination with at least one compound (B) selected from:

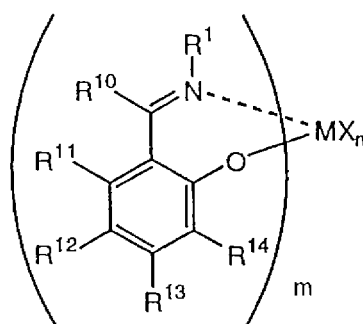
(B-1) an organometallic compound,

(B-2) an organoaluminum oxy-compound, and

5 (B-3) a compound which reacts with the transition metal compound to form an ion pair.

Another embodiment of the present invention is an olefin polymerization catalyst comprising a transition metal compound represented by the following formula (II):

10



(II)

wherein M, R¹, n and X have the same meanings as those of M, R¹, n and X in the formula (I),

15

m is 1 or 2,

R¹⁰ and R¹¹ to R¹³ may be the same or different and are each a hydrogen atom, a hydrocarbon group, a hydrocarbon-substituted silyl group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group,

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-9 APR 2003

R^{14} is a hydrocarbon group or a hydrocarbon-substituted silyl group,

when m is 2, at least one of groups indicated by R^{10} to R^{14} contained in one ligand and at least one of groups indicated by R^{10} to R^{14} contained in another ligand may be bonded to each other, and

when m is 2, R^1s , $R^{10}s$, $R^{11}s$, $R^{12}s$, $R^{13}s$, or $R^{14}s$ may be the same or different.

A preferred embodiment of the transition metal compound represented by the formula (II) is a compound wherein R^1 is an aliphatic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group and has not less than total 5 carbon atoms, or a compound wherein R^1 is an alicyclic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group and has not less than total 7 carbon atoms.

In the formula (II), R^{12} is preferably an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group.

The transition metal compound (A-2) represented by the formula (II) may be used as an olefin polymerization catalyst in combination with at least one compound (B) selected from:

(B-1) an organometallic compound,

(B-2) an organoaluminum oxy-compound, and

(B-3) a compound which reacts with the transition metal compound to form an ion pair.

The process for olefin polymerization according to the present invention comprises polymerizing an olefin in the presence of the olefin polymerization catalyst mentioned above.

BRIEF DESCRIPTION OF THE DRAWING

Fig. 1 is an explanatory view showing steps for preparing the olefin polymerization catalyst according to the present invention.

DETAILED DESCRIPTION OF THE INVENTION

The olefin polymerization catalyst according to the invention and the process for olefin polymerization using the catalyst are described in detail hereinafter.

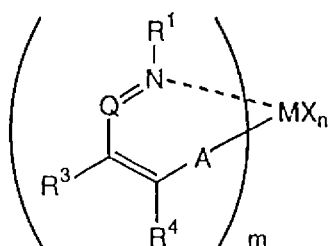
The meaning of the term "polymerization" used herein is not limited to "homopolymerization" but may comprehend "copolymerization". Also, the meaning of the term "polymer" used herein is not limited to "homopolymer" but may comprehend "copolymer".

The olefin polymerization catalyst according to the invention comprises a transition metal compound represented by the formula (I) described below.

-9 APR 2003

(A-1) Transition metal compound

The transition metal compound (A-1) is represented by the following formula (I).



(I)

N---M generally indicates coordination, but in the invention, it sometimes indicates no coordination.

In the formula (I), M is a transition metal atom of Group 4 to Group 5 of the periodic table, specifically titanium, zirconium, hafnium, vanadium, niobium, tantalum or the like, preferably titanium, zirconium or hafnium, particularly preferably zirconium.

m is an integer of 1 to 5, preferably an integer of 1 to 3, more preferably 1 or 2.

Q is a nitrogen atom (-N=) or a carbon atom having a substituent group R² (-C(R²)=), preferably a carbon atom having a substituent group R².

A is an oxygen atom (-O-), a sulfur atom (-S-), a selenium atom (-Se-) or a nitrogen atom having a

-9 APR 2003

substituent group R^5 ($-N(R^5)-$), more preferably an oxygen atom.

The carbon atom directly bonded to the nitrogen atom forms a part of an alicyclic structure.

5 R^1 is an aliphatic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group, or an alicyclic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group.

10 The term "aliphatic hydrocarbon group" used in the definition of R^1 means that a carbon atom which is directly bonded to the nitrogen atom in the formula (I) is an aliphatic skeleton moiety even when the aliphatic hydrocarbon group is substituted with an aromatic
15 hydrocarbon group or an alicyclic hydrocarbon group. Likewise, the term "alicyclic hydrocarbon group" used in the definition of R^1 means that a carbon atom which is directly bonded to the nitrogen atom in the formula (I) is an alicyclic skeleton moiety even when the alicyclic
20 hydrocarbon group is substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group.

Examples of the groups indicated by R^1 include aliphatic hydrocarbon groups of 1 to 30 carbon atoms, such as methyl, ethyl, n-propyl, isopropyl, n-butyl,
25 isobutyl, sec-butyl, tert-butyl, n-pentyl, tert-amyl,

-9 APR 2003

1,2-dimethylpropyl, 1-ethylpropyl, isoamyl, 1-methylbutyl,
2-methylbutyl, neopentyl, n-hexyl, 1,3-dimethylbutyl,
3,3-dimethylbutyl, n-heptyl, 1-ethylpentyl, 1-methylhexyl,
n-octyl, 1,5-dimethylhexyl, 2-ethylhexyl, 1-methylheptyl,
5 tert-octyl, n-nonyl, n-decyl, n-undecyl, n-dodecyl, n-
tridecyl, n-tetradecyl, n-pentadecyl, n-hexadecyl, n-
heptadecyl and n-octadecyl; alicyclic hydrocarbon groups
which may be substituted with aliphatic hydrocarbon
groups and have 3 to 30 carbon atoms, such as cyclopropyl,
10 cyclobutyl, cyclopentyl, cyclohexyl, 2-methylcyclohexyl,
2,2-dimethylcyclohexyl, 2,3-dimethylcyclohexyl, 2,6-
dimethylcyclohexyl, 2,2,6,6-tetramethylcyclohexyl and
adamantyl; aliphatic hydrocarbon groups substituted with
alicyclic hydrocarbon groups and having 6 to 30 carbon
15 atoms, such as cyclopropylmethyl, cyclobutylmethyl,
cyclopentylmethyl and cyclohexylmethyl; and aliphatic
hydrocarbon groups substituted with aromatic hydrocarbon
groups and having 4 to 30 carbon atoms, such as benzyl.

R^1 is desirably an aliphatic hydrocarbon group which
20 may be substituted with an aromatic hydrocarbon group or
an alicyclic hydrocarbon group and has 1 to 30 carbon
atoms, preferably 5 to 30 carbon atoms, more preferably 5
to 20 carbon atoms, or an alicyclic hydrocarbon group
which may be substituted with an aromatic hydrocarbon
25 group or an aliphatic hydrocarbon group and has 3 to 30

-9 APR 2003

carbon atoms, preferably 5 to 30 carbon atoms, more preferably 5 to 20 carbon atoms.

R^2 to R^5 may be the same or different and are each a hydrogen atom, a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group, a sulfur-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group.

Examples of the hydrocarbon groups include straight-chain or branched alkyl groups of 1 to 30 carbon atoms, preferably 1 to 20 carbon atoms, such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, neopentyl and n-hexyl; straight-chain or branched alkenyl groups of 2 to 30 carbon atoms, preferably 2 to 20 carbon atoms, such as vinyl, allyl and isopropenyl; straight-chain or branched alkynyl groups of 2 to 30 carbon atoms, preferably 2 to 20 carbon atoms, such as ethynyl and propargyl; cyclic saturated hydrocarbon groups of 3 to 30 carbon atoms, preferably 3 to 20 carbon atoms, such as cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl and adamantyl; cyclic unsaturated hydrocarbon groups of 5 to 30 carbon atoms, such as cyclopentadienyl, indenyl and fluorenyl; aryl groups of 6 to 30 carbon

-9 APR 2003

atoms, preferably 6 to 20 carbon atoms, such as phenyl, benzyl, naphthyl, biphenyl, terphenyl, phenanthryl and anthryl; and alkyl-substituted aryl groups, such as tolyl, isopropylphenyl, t-butylphenyl, dimethylphenyl and
5 di-t-butylphenyl.

In the above hydrocarbon groups, a halogen atom may be substituted for the hydrogen atom. Examples of the hydrocarbon groups wherein a halogen atom is substituted for the hydrogen atom include halogenated hydrocarbon
10 groups of 1 to 30 carbon atoms, preferably 1 to 20 carbon atoms, such as trifluoromethyl, pentafluorophenyl and chlorophenyl.

The above hydrocarbon groups may be substituted with other hydrocarbon groups. Examples of the hydrocarbon
15 groups substituted with other hydrocarbon groups include aryl group-substituted alkyl groups, such as benzyl and cumyl.

Of the above groups, preferable are straight-chain or branched alkyl groups of 1 to 30 carbon atoms,
20 preferably 1 to 20 carbon atoms, such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, t-butyl, neopentyl and n-hexyl; aryl groups of 6 to 30 carbon atoms, preferably 6 to 20 carbon atoms, such as phenyl, naphthyl, biphenyl, terphenyl, phenanthryl
25 and anthryl; and substituted aryl groups wherein the

-9 APR 2003

above aryl groups are substituted with 1 to 5 substituent groups such as halogen atoms, alkyl groups of 1 to 30 carbon atoms, preferably 1 to 20 carbon atoms, alkoxy groups of 1 to 30 carbon atoms, preferably 1 to 20 carbon atoms, aryl groups of 6 to 30 carbon atom, preferably 6 to 20 carbon atoms, and aryloxy groups of 6 to 30 carbon atoms, preferably 6 to 20 carbon atoms.

The oxygen-containing group is a group containing 1 to 5 oxygen atoms therein. In the oxygen-containing groups, the later-described heterocyclic compound residual group is not included, and a group containing a nitrogen atom, a sulfur atom, a phosphorus atom, a halogen atom or a silicon atom each of which is directly bonded to the oxygen atom is not included either.

Examples of the oxygen-containing groups include a hydroxyl group, an alkoxy group, an aryloxy group, an arylalkoxy group, an acetoxy group, a carbonyl group, an ester group, an ether group, an acyl group, a carboxyl group, a carbonato group, a peroxy group and a carboxylic anhydride group. Of these, preferable are a hydroxyl group, an alkoxy group, an aryloxy group, an arylalkoxy group, an acetoxy group, a carbonyl group and the like. Specific examples of the preferred oxygen-containing groups include a hydroxyl group; alkoxy groups, such as methoxy, ethoxy, n-propoxy, isopropoxy, n-butoxy,

-9 APR 2003

isobutoxy and tert-butoxy; aryloxy groups, such as phenoxy, methylphenoxy, 2,6-dimethylphenoxy, 2,4,6-trimethylphenoxy and naphthoxy; arylalkoxy groups, such as phenylmethoxy and phenylethoxy; an acetoxymethyl group; and
5 a carbonyl group. When the oxygen-containing group contains carbon atom, the number of carbon atoms is desired to be in the range of 1 to 30, preferably 1 to 20.

The nitrogen-containing group is a group containing 1 to 5 nitrogen atoms therein, and the later-described
10 heterocyclic compound residual group is not included in the nitrogen-containing groups. Examples of the nitrogen-containing groups include an amino group, an imino group, an amido group, an imido group, a nitro group, a hydrazino group, a hydrazono group, a nitroso
15 group, a cyano group, an isocyano group, a cyanic ester group, an amidino group, a diazo group and an ammonium salt group derived from an amino group. Of these, preferable are an amino group, an imino group, an amido group, an imido group, a nitro group and the like.
20 Specific examples of the preferred nitrogen-containing groups include amino groups, such as methylamino, dimethylamino, diethylamino, dipropylamino, dibutylamino, dicyclohexylamino, phenylamino, diphenylamino, ditolylamino, dinaphthylamino and methylphenylamino;
25 imino groups, such as methylimino, ethylimino,

-9 APR 2003

propylimino, butylimino and phenylimino; amido groups, such as acetamido, N-methylacetamido and N-methylbenzamido; imido groups, such as acetimido and benzimido; and a nitro group. When the nitrogen-containing group contains carbon atom, the number of carbon atoms is desired to be in the range of 1 to 30, preferably 1 to 20.

The sulfur-containing group is a group containing 1 to 5 sulfur atoms therein, and the later-described heterocyclic compound residual group is not included in the sulfur-containing groups. Examples of the sulfur-containing groups include a sulfonato group, a sulfinato group, an alkylthio group, an arylthio group, a mercapto group, a thioester group, a dithioester group, a thioacyl group, a thioether group, a thiocyanic ester group, an isothiocyanic ester group, a sulfonic ester group, a sulfonamido group, a thiocarboxyl group, a dithiocarboxyl group, a sulfo group, a sulfonyl group, a sulfinyl group and a sulfenyl group. Of these, preferable are a sulfonato group, a sulfinato group, an alkylthio group, an arylthio group and the like. Specific examples of the preferred sulfur-containing groups include sulfonato groups, such as methylsulfonato, trifluoromethanesulfonato, phenylsulfonato, benzylsulfonato, p-toluenesulfonato,

-9 APR 2003

trimethylbenzenesulfonato, triisobutylbenzenesulfonato,
p-chlorobenzenesulfonato and pentafluorobenzenesulfonato;
sulfinato groups, such as methylsulfinato,
phenylsulfinato, benzylsulfinato, p-toluenesulfinato,
5 trimethylbenzenesulfinato and
pentafluorobenzenesulfinato; alkylthio groups, such as
methylthio and ethylthio; and arylthio groups, such as
phenylthio, methylphenylthio and naphthylthio. When the
sulfur-containing group contains carbon atom, the number
10 of carbon atoms is desired to be in the range of 1 to 30,
preferably 1 to 20.

The boron-containing group is a group containing 1
to 5 boron atoms therein, and the later-described
heterocyclic compound residual group is not included in
15 the boron-containing groups. Examples of the boron-
containing groups include a boranediyl group, a
boranetriyl group, a diboranyl group and a group
represented by BR_4 (R is hydrogen, an alkyl group, an
aryl group which may have a substituent group, a halogen
20 atom or the like).

The aluminum-containing group is a group containing
1 to 5 aluminum atoms therein. Examples of the aluminum-
containing groups include those having 1 or 2 hydrocarbon
groups of 1 to 30 carbon atoms, preferably 1 to 20 carbon
25 atoms, as substituent groups, and those represented by

-7 APR 2003

AlR_4 (R is a hydrogen atom, an alkyl group, an aryl group which may have a substituent group, a halogen atom or the like).

The phosphorus-containing group is a group
5 containing 1 to 5 phosphorus atoms therein, and the later-described heterocyclic compound residual group is not included in the phosphorus-containing groups. Examples of the phosphorus-containing groups include a phosphoryl group, a thiophosphoryl group, a phosphine
10 group, a phosphite group, a phosphonic acid group and a phosphinic acid group. Of these, preferable are a phosphine group, a phosphite group, a phosphonic acid group, a phosphinic acid group and the like. Specific examples of the preferred phosphorus-containing groups
15 include trialkylphosphine groups, such as trimethylphosphine, tributylphosphine and tricyclohexylphosphine; triarylphosphine groups, such as triphenylphosphine and tritolylphosphine; phosphite group (phosphido groups), such as methylphosphite,
20 ethylphosphite and phenylphosphite; a phosphonic acid group; and a phosphinic acid group.

The halogen-containing group is, for example, a group containing at least one atom selected from fluorine, chlorine, bromine and iodine. Specific examples of the
25 halogen-containing groups include fluorine-containing

groups, such as PF_6 and BF_4 ; chlorine-containing groups, such as ClO_4 and SbCl_6 ; and iodine-containing groups, such as IO_4 .

The heterocyclic compound residual group is a group
5 having a ring structure containing at least one hetero
atom. The hetero atom is, for example, oxygen, nitrogen,
sulfur, phosphorus or boron. The ring structure is, for
example, a ring of 3 to 18 members, preferably a ring of
4 to 7 members, more preferably a ring of 5 or 6 members.
10 Examples of the heterocyclic compound residual groups
include residual groups of nitrogen-containing compounds
such as pyrrole, pyridine, pyrimidine, quinoline and
triazine; residual groups of oxygen-containing compounds
such as furan and pyran; residual groups of sulfur-
15 containing compounds such as thiophene; and groups
wherein these heterocyclic compound residual groups are
further substituted with substituent groups such as alkyl
groups of 1 to 30 carbon atoms, preferably 1 to 20 carbon
atoms, and alkoxy groups of 1 to 30 carbon atoms,
20 preferably 1 to 20 carbon atoms.

The silicon-containing group is a group containing 1
to 5 silicon atoms therein. Examples of the silicon-
containing groups include a silyl group, a siloxy group,
a hydrocarbon-substituted silyl group, a hydrocarbon-
25 substituted siloxy group, a hydrocarbon-substituted silyl

-9 APR 2003

ether group, a silicon-substituted alkyl group and a silicon-substituted aryl group. Of these, preferable are a hydrocarbon-substituted silyl group, a hydrocarbon-substituted silyl ether group, a silicon-substituted alkyl group, a silicon-substituted aryl group and the like. Specific examples of the preferred silicon-containing groups include hydrocarbon-substituted silyl groups, such as methylsilyl, dimethylsilyl, trimethylsilyl, ethylsilyl, diethylsilyl, triethylsilyl, tripropylsilyl, tricyclohexylsilyl, phenylsilyl, diphenylsilyl, triphenylsilyl, dimethyl-t-butylsilyl, dimethylphenylsilyl, methyldiphenylsilyl, tritolylsilyl, trinaphthylsilyl and dimethyl(pentafluorophenyl)silyl; hydrocarbon-substituted silyl ether groups, such as trimethylsilyl ether; silicon-substituted alkyl groups, such as trimethylsilylmethyl; and silicon-substituted aryl groups, such as trimethylsilylphenyl. Of these, hydrocarbon-substituted silyl groups are particularly preferable. Of the hydrocarbon-substituted silyl groups, preferable are methylsilyl, dimethylsilyl, trimethylsilyl, ethylsilyl, diethylsilyl, triethylsilyl, dimethylphenylsilyl and triphenylsilyl, and particularly preferable are trimethylsilyl, triethylsilyl, triphenylsilyl and dimethylphenylsilyl. When the silicon-containing group contains carbon atom, the number

-9 APR 2003

of carbon atoms is desired to be in the range of 1 to 30, preferably 1 to 20.

Examples of the germanium-containing groups include groups wherein silicon is replaced with germanium in the
5 above-mentioned silicon-containing groups.

Examples of the tin-containing groups include groups wherein silicon is replaced with tin in the above-mentioned silicon-containing groups.

R^4 is preferably an oxygen-containing group, such as
10 an alkoxy group, an aryloxy group or a hydroxyl group, a nitrogen-containing group, such as an amino group, an imino group, an amido group, an imido group or a nitro group, or a sulfur-containing group, such as an alkylthio group or an arylthio group; it is more preferably an
15 alkoxy group, an aryloxy group or an amino group; and it is particularly preferably an alkoxy group.

The oxygen-containing group, nitrogen-containing group, sulfur-containing group, boron-containing group, aluminum-containing group and phosphorus-containing group
20 mentioned above are each preferably a group whose characteristic atomic group is directly bonded to a nitrogen atom or a carbon atom.

Two or more groups of R^1 to R^5 , preferably neighboring groups, may be bonded to each other to form
25 an aliphatic ring, an aromatic ring or a hydrocarbon ring

-9 APR 2003

containing a hetero atom such as a nitrogen atom. These rings may further have a substituent group. In the present invention, it is particularly preferable that the substituent groups R^3 and R^4 are bonded to form an aromatic ring.

When m is 2 or greater in the formula (I), one group of R^2 to R^5 contained in one ligand and one group of R^2 to R^5 contained in another ligand may be bonded to each other.

When m is 2 or greater, R^1s , R^2s , R^3s , R^4s , or R^5s may be the same or different.

n is a number satisfying a valence of M , specifically, an integer of 2 to 4, preferably 2.

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group.

Examples of the hydrocarbon groups, the oxygen-containing group, the sulfur-containing group, the boron-containing groups, the aluminum-containing groups, the phosphorus-containing groups, the halogen-containing

-9 APR 2003

groups, the heterocyclic compound residual groups, the silicon-containing groups, the germanium-containing groups and the tin-containing groups include the same groups as previously described with respect to R^2 to R^5 in the formula (I).

The halogen atom is fluorine, chlorine, bromine or iodine.

Examples of the nitrogen-containing groups include an amino group; alkylamino groups, such as methylamino, dimethylamino, diethylamino, dipropylamino, dibutylamino and dicyclohexylamino; and arylamino or alkylarylamino groups, such as phenylamino, diphenylamino, ditolylamino, dinaphthylamino and methylphenylamino.

Of the above groups, a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group or a nitrogen-containing group is preferable as X, and a hydrogen atom, a halogen atom or a hydrocarbon group is more preferable as X.

When n is 2 or greater, plural groups indicated by X may be the same or different, and plural groups indicated by X may be bonded to each other to form a ring.

In the transition metal compound represented by the formula (I), the net charge parameter of the central metal M is not more than 2.00, preferably in the range of 1.40 to 2.00.

-9 APR 2003

The "net charge parameter" (sometimes abbreviated to "NC" hereinafter) of the central metal M referred to herein is a value of a Mulliken charge. The Mulliken charge is an electric charge of the central metal and is obtained in the following manner. The substituent groups indicated by X in the formula (I) are all replaced with methyl groups to give a complex, and the structure of the complex is optimized using the semiempirical molecular orbital method PM3 (tm). From the thus optimized structure, one methyl group is removed to obtain a structure (I'). The program used herein is SPARTANrel1.5.0.3. Using the structure (I'), an RHF wave function is found out by the nonempirical molecular orbital method program Gaussian98, to obtain a Mulliken charge as an electric charge of the central metal. For the inner shell of the central metal, an effective inner shell potential of Hay and Wadt was used as the basis function, for the valence shell of the central metal, their double zeta basis function was used, and for other atoms, 3-21G was used. The thus obtained Mulliken charge value is defined as a net charge parameter. Also in case of the compound represented by the formula (I-a), (I-b) or (II) described below, the net charge parameter is obtained in the same manner as described above.

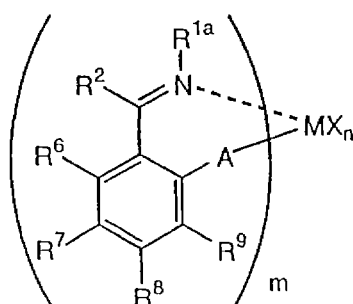
-9 APR 2003

The NC is not more than 2.0. When R¹ is an aliphatic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group, the NC is preferably in the range of 1.70 to 2.0.

5 When R¹ is an alicyclic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group, the NC is preferably in the range of 1.50 to 1.89.

When the NC of the central metal M is not more than
10 2.0, the transition metal compound used as an olefin polymerization catalyst exhibits excellent polymerization activity, especially at high temperatures.

The transition metal compound represented by the formula (I) includes a transition metal compound
15 represented by the following formula or (I-b).



(I-a)

In the above formula, M, m, A, n and X have the same
20 meanings as those of M, m, A, n and X in the formula (I).

-9 APR 2003

R^{1a} is an aliphatic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group. Examples of such groups include aliphatic hydrocarbon groups of 1 to 30 carbon atoms, such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, n-pentyl, tert-amyl, 1,2-dimethylpropyl, 1-ethylpropyl, isoamyl, 1-methylbutyl, 2-methylbutyl, neopentyl, n-hexyl, 1,3-dimethylbutyl, 3,3-dimethylbutyl, n-heptyl, 1-ethylpentyl, 1-methylhexyl, n-octyl, 1,5-dimethylhexyl, 2-ethylhexyl, 1-methylheptyl, tert-octyl, n-nonyl, n-decyl, n-undecyl, n-dodecyl, n-tridecyl, n-tetradecyl, n-pentadecyl, n-hexadecyl, n-heptadecyl and n-octadecyl; aliphatic hydrocarbon groups substituted with aromatic hydrocarbon groups and having 6 to 30 carbon atoms, such as benzyl; and aliphatic hydrocarbon groups substituted with alicyclic hydrocarbon groups and having of 4 to 30 carbon atoms, such as cyclopropylmethyl, cyclobutylmethyl, cyclopentylmethyl and cyclohexylmethyl.

R^{1a} desirably is a group having 1 to 30 carbon atoms, preferably 5 to 30 carbon atoms, more preferably 5 to 20 carbon atoms.

R^2 and R^5 to R^9 may be the same or different and are each a hydrogen atom, a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group, a sulfur-

-9 APR 2003

containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group. Examples of such groups include the same groups as previously described with respect to R^2 to R^5 in the formula (I).

Two or more groups of R^2 and R^5 to R^9 , preferably neighboring groups, may be bonded to each other to form an aliphatic ring, an aromatic ring or a hydrocarbon ring containing a hetero atom such as a nitrogen atom. These rings may further have a substituent group.

When m is 2 or greater in the formula (I-a), at least one of groups indicated by R^2 and R^5 to R^9 contained in one ligand and at least one of groups indicated by R^2 and R^5 to R^9 contained in another ligand may be bonded to each other.

When m is 2 or greater, $R^{1a}s$, R^2s , R^5s , R^6s , R^7s , R^8s , or R^9s may be the same or different.

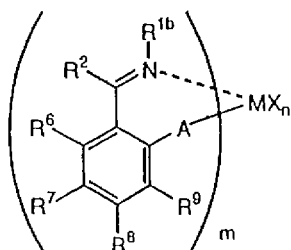
In the transition metal compound represented by the formula (I-a), the net charge parameter of the central metal M is in the range of 1.70 to 2.00, preferably 1.895 to 1.912.

When the NC of the central metal M is in the above range, the transition metal compound used as an olefin

-9 APR 2003

polymerization catalyst exhibits excellent polymerization activity, especially at high temperatures.

(I-b)



In the above formula, M, m, A, n and X have the same meanings as those of M, m, A, n and X in the formula (I).

R^{1b} is an alicyclic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group. Examples of such groups include alicyclic hydrocarbon groups which may be substituted with aliphatic hydrocarbon groups and have 3 to 30 carbon atoms, preferably 5 to 30 carbon atoms, more preferably 5 to 20 carbon atoms, such as cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, 2-methylcyclohexyl, 2,2-dimethylcyclohexyl, 2,6-dimethylcyclohexyl, 2,2,6,6-tetramethylcyclohexyl, adamantyl, cyclopropylmethyl, cyclobutylmethyl, cyclopentylmethyl and cyclohexylmethyl.

R^2 and R^5 to R^9 may be the same or different and are each a hydrogen atom, a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group, a sulfur-containing group, a boron-containing group, an aluminum-

-9 APR 2003

containing group, a phosphorus-containing group, a
halogen-containing group, a heterocyclic compound
residual group, a silicon-containing group, a germanium-
containing group or a tin-containing group. Examples of
5 such groups include the same groups as previously
described with respect to R^2 to R^5 in the formula (I).

Two or more groups of R^2 and R^5 to R^9 , preferably
neighboring groups, may be bonded to each other to form
an aliphatic ring, an aromatic ring or a hydrocarbon ring
10 containing a hetero atom such as a nitrogen atom. These
rings may further have a substituent group.

When m is 2 or greater in the formula (I-b), at
least one of groups indicated by R^2 and R^5 to R^9 contained
in one ligand and at least one of groups indicated by R^2
15 and R^5 to R^9 contained in another ligand may be bonded to
each other.

When m is 2 or greater, $R^{1b}s$, R^2s , R^5s , R^6s , R^7s , R^8s ,
or R^9s may be the same or different.

In the transition metal compound represented by the
20 formula (I-b), the net charge parameter of the central
metal M is preferably in the range of 1.50 to 1.89, more
preferably in the range of 1.80 to 1.89.

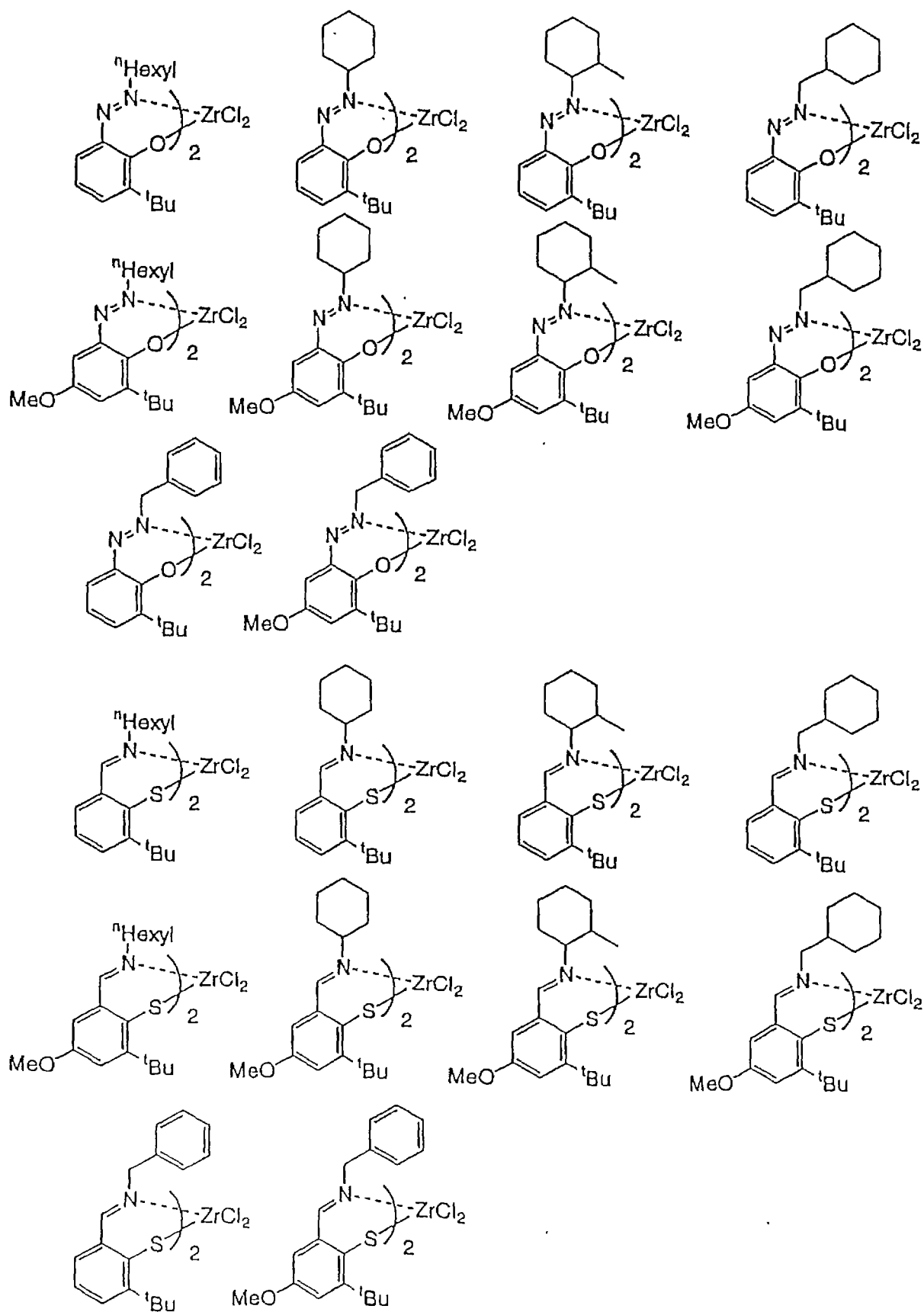
When the NC of the central metal M is in the above
range, the transition metal compound used as an olefin

-9 APR 2003

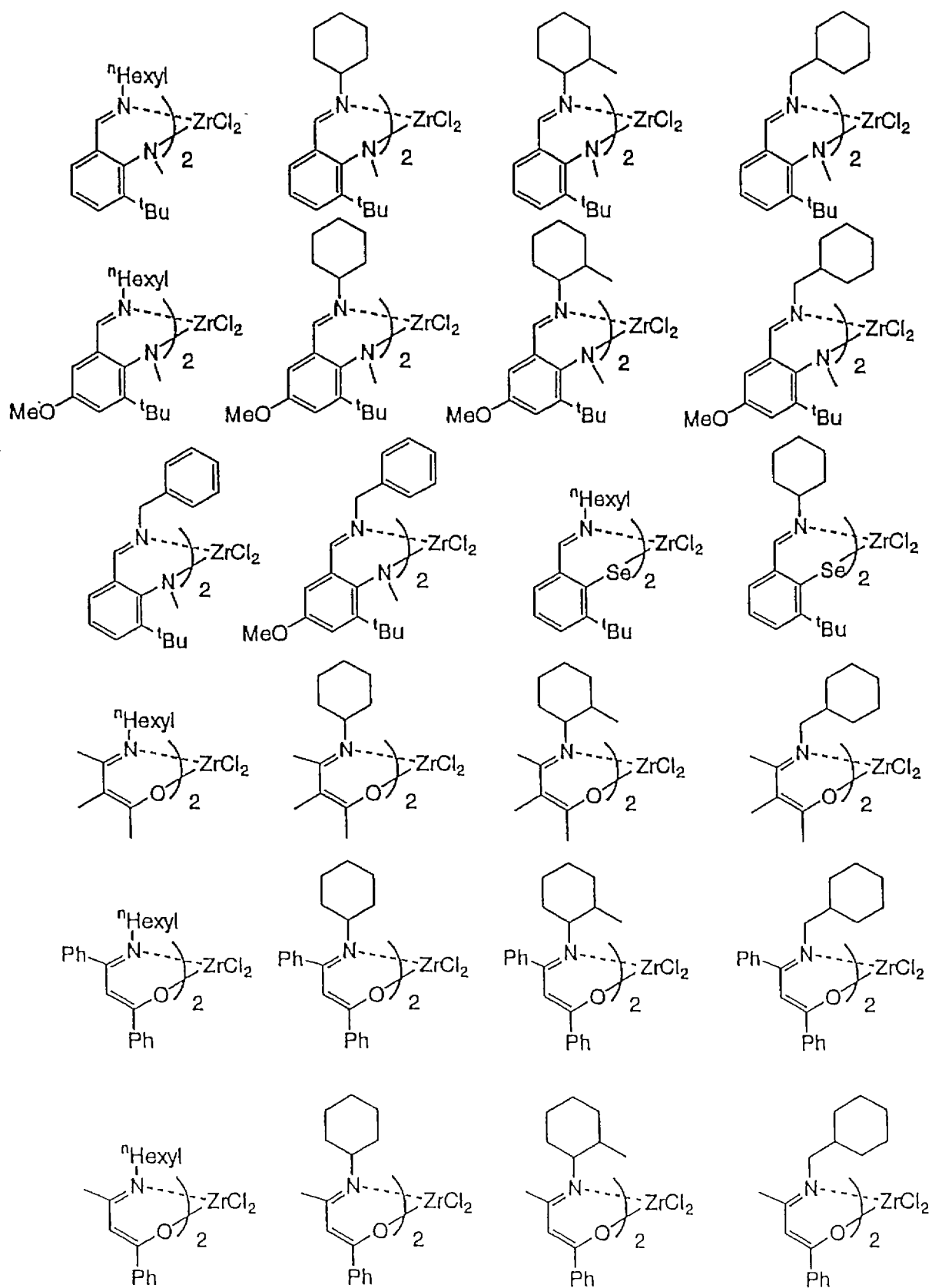
polymerization catalyst exhibits excellent polymerization activity, especially at high temperatures.

Examples of the transition metal compounds represented by the formula (I) are given below, but the
5 compounds of the formula (I) are not limited to those examples. In the following examples, the compounds represented by the formula (I-a) or (I-b) are included. Of the following examples, the compounds wherein, in the formula (I), Q is a carbon atom having a substituent
10 group R^2 , A is an oxygen atom, and R^3 and R^4 are bonded to each other to form an aromatic ring include the compounds represented by the formula (II) described later.

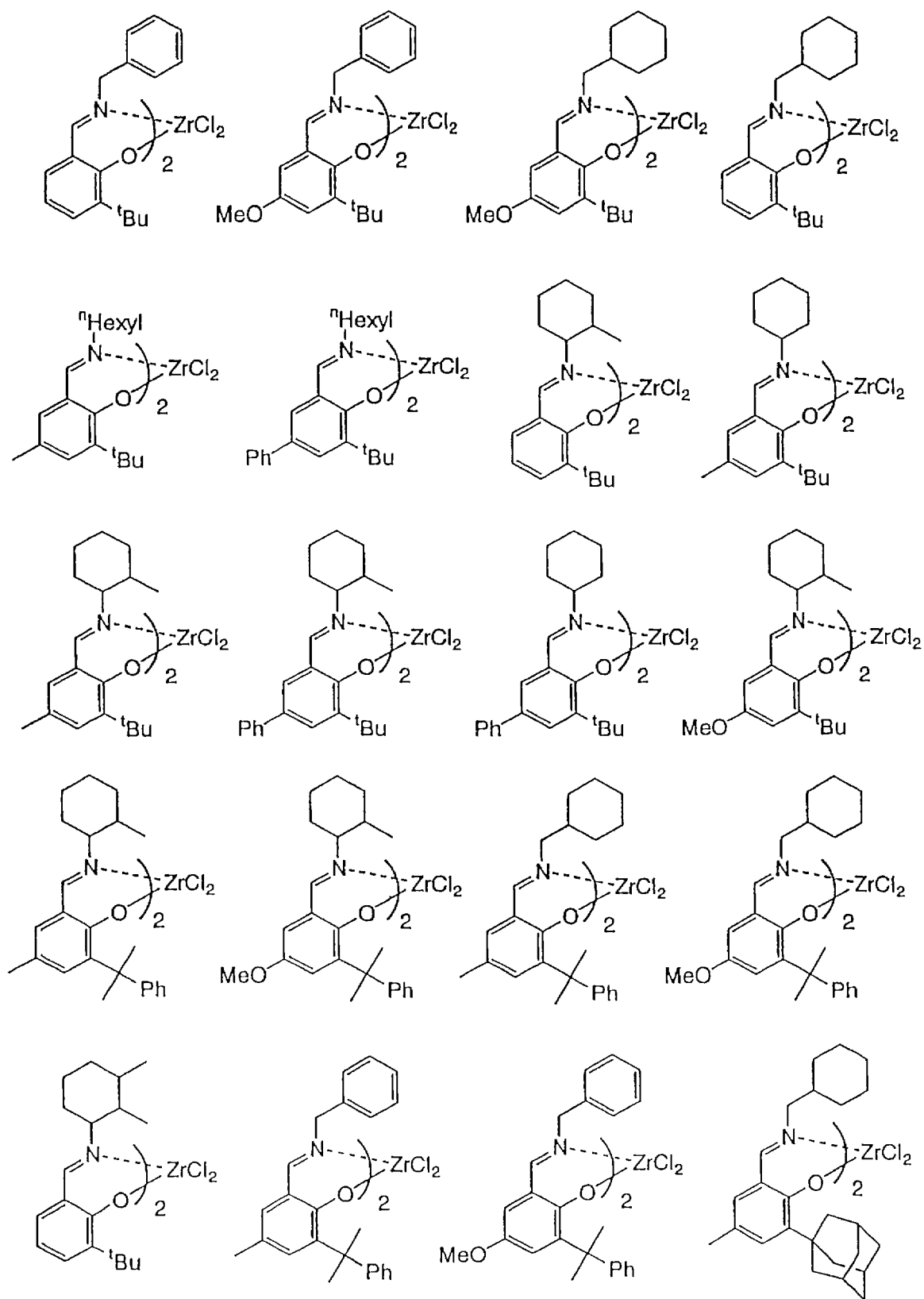
-9 APR 2003



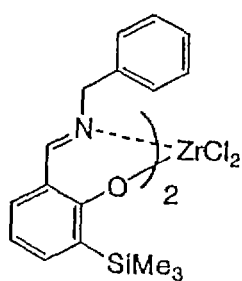
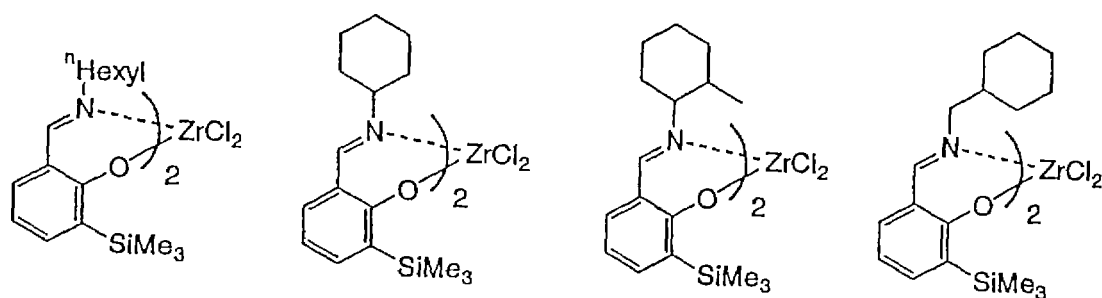
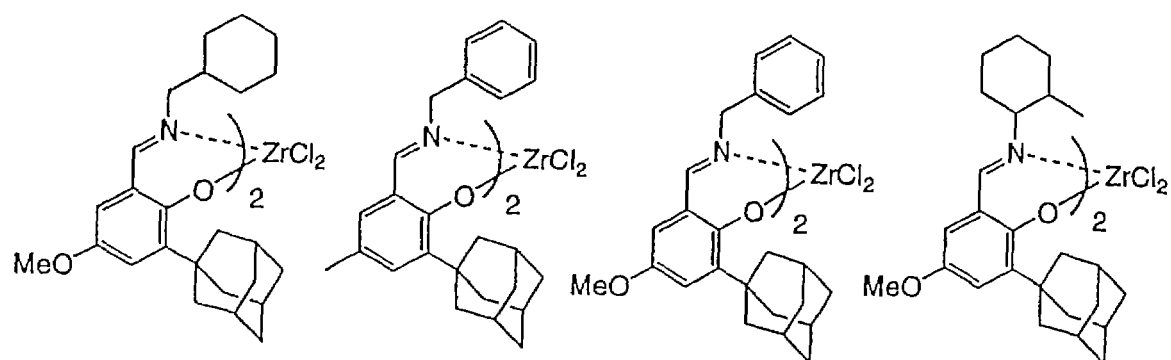
-9 APR 2003



-9 APR 2003



-9 APR 2003

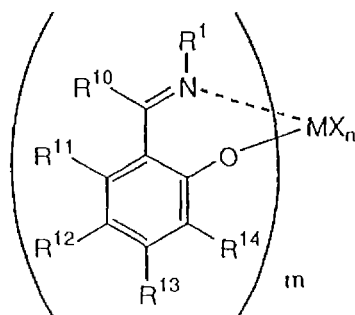


-9 APR 2003

In the above examples, Me denotes a methyl group, Et denotes an ethyl group, ⁿPr denotes a n-propyl group, ⁱPr denotes an isopropyl group, ⁿBu denotes a n-butyl group, ^tBu denotes a t-butyl group, ⁿPentyl denotes a n-pentyl group, ⁿHexyl denotes a n-hexyl group, ⁿHeptyl denotes a n-heptyl group, ⁿOctyl denotes a n-octyl group, ⁿNonyl denotes a n-nonyl group, ⁿDecyl denotes a n-decyl group, ⁿUndecyl denotes a n-undecyl group, ⁿDodecyl denotes a n-dodecyl group, ⁿOctadecyl denotes a n-octadecyl group, and Ph denotes a phenyl group.

In the present invention, transition metal compounds wherein the zirconium metal is replaced with a metal other than zirconium, such as titanium or hafnium, in the above-mentioned compounds are also employable.

Another embodiment of the olefin polymerization catalyst of the invention may comprise a transition metal compound represented by the following formula (II).



(II)

-9 APR 2003

In the above formula, M, R¹, n and X have the same meanings as those of M, R¹, n and X in the formula (I).

R¹ is the same aliphatic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group as R¹ in the above formula (I),
5 or the same alicyclic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group as R¹ in the above formula (I), and R¹ is desirably an aliphatic hydrocarbon group which
10 may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group and has preferably not less than total 5 carbon atoms, more preferably total 5 to 30 carbon atoms, or desirably an alicyclic hydrocarbon group which may be substituted with an aromatic
15 hydrocarbon group or an aliphatic hydrocarbon group and has not less than total 7 carbon atoms, more preferably total 7 to 30 carbon atoms.

The aliphatic hydrocarbon group is, for example, a group of 1 to 30 carbon atoms, preferably 4 to 30 carbon
20 atoms, more preferably 5 to 30 carbon atoms.

Examples of the aliphatic hydrocarbon groups include methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, n-pentyl, tert-amyl, 1,2-dimethylpropyl, 1-ethylpropyl, isoamyl, 1-methylbutyl, 2-methylbutyl, neopentyl, n-hexyl, 1,3-dimethylbutyl, 3,3-
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-9 APR 2003

dimethylbutyl, n-heptyl, 1-ethylpentyl, 1-methylhexyl, n-octyl, 1,5-dimethylhexyl, 2-ethylhexyl, 1-methylheptyl, tert-octyl, n-nonyl, n-decyl, n-undecyl, n-dodecyl, n-tridecyl, n-tetradecyl, n-pentadecyl, n-hexadecyl, n-heptadecyl and n-octadecyl.

Of the above groups, preferable are n-pentyl, tert-amyl, 1,2-dimethylpropyl, 1-ethylpropyl, isoamyl, 1-methylbutyl, 2-methylbutyl, neopentyl, n-hexyl, 1,3-dimethylbutyl, 3,3-dimethylbutyl, n-heptyl, 1-ethylpentyl, 1-methylhexyl, n-octyl, 1,5-dimethylhexyl, 2-ethylhexyl, 1-methylheptyl, tert-octyl, n-nonyl, n-decyl, n-undecyl, n-dodecyl, n-tridecyl, n-tetradecyl, n-pentadecyl, n-hexadecyl, n-heptadecyl and n-octadecyl.

The aliphatic hydrocarbon group may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group. Examples of the aliphatic hydrocarbon group substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group include groups of 4 to 30 carbon atoms, such as benzyl, methylene naphthyl, methylene fluorenyl, methylene anthranyl, methylene cyclopropyl, methylene cyclobutyl, methylene cyclopentyl, methylene cyclohexyl, methylene-2-methylcyclohexyl, methylene-2,3-dimethylcyclohexyl, methylene-2,2-dimethylcyclohexyl, methylene-2,6-dimethylcyclohexyl, methylene-2,2,6,6-tetramethylcyclohexyl, methylene

-9 APR 2003

adamantyl, methylene cyclopropylmethyl, methylene cyclobutylmethyl, methylene cyclopentylmethyl, or the like.

Of those groups, preferable are the groups of 5 to 30 carbon atoms, such as benzyl, methylene naphthyl, methylene fluorenyl, methylene anthranyl, methylene cyclobutyl, methylene cyclopentyl, methylene cyclohexyl, methylene-2-methylcyclohexyl, methylene-2,3-dimethylcyclohexyl, methylene-2,2-dimethylcyclohexyl, methylene-2,6-dimethylcyclohexyl, methylene-2,2,6,6-tetramethylcyclohexyl, methylene adamantyl, methylene cyclopropylmethyl, methylene cyclobutylmethyl, methylene cyclopentylmethyl, or the like.

Examples of the alicyclic hydrocarbon groups include groups of 3 to 30 carbon atoms, preferably 7 to 30 carbon atoms, such as cyclopropyl, cyclobuthyl, cyclopentyl, cyclohexyl, adamantyl, norbornyl, or the like.

The alicyclic hydrocarbon group may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group.

Examples of the alicyclic hydrocarbon groups which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group include groups of 7 to 30 carbon atoms, such as 2-methylcyclohexyl, 2,3-dimethylcyclohexyl, 2,2-dimethylcyclohexyl, 2,6-

-9 APR 2003

dimethylcyclohexyl, 2,2,6,6-tetramethylcyclohexyl, cyclopropylmethyl, cyclobuthylmethyl, cyclopentylmethyl, cyclopropylphenyl, cyclobutylphenyl, cyclopentylphenyl, cyclohexylphenyl, or the like.

5 m is 1 or 2, preferably 2.

R^{10} and R^{11} to R^{13} may be the same or different and are each a hydrogen atom, a hydrocarbon group, a hydrocarbon-substituted silyl group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group. Examples of such groups include the same hydrocarbon groups, the same hydrocarbon-substituted silyl groups, the same oxygen-containing groups, the same nitrogen-containing groups and the same sulfur-containing groups as previously described with respect to R^2 to R^5 in the formula (I).

10

15

R^{12} is preferably a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group, specifically preferably an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group.

20

 Examples of such groups include the same hydrocarbon groups, the same oxygen-containing groups, the same nitrogen-containing groups and the same sulfur-containing groups as previously described, and of these groups, preferable are an alkoxy group, an alkylthio group, an

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-9 APR 2003

aryloxy group, an arylthio group, an amino group, an imino group, an amido group, an imido group, a nitro group or a hydroxyl group, specifically preferable is an alkoxy group.

5 R^{14} is a hydrocarbon group or a hydrocarbon-substituted silyl group, and examples of such groups include the same hydrocarbon groups and the same hydrocarbon-substituted silyl groups as previously described with respect to R^2 to R^5 in the formula (I).

10 The above substituent groups are described below in more detail.

R^{12} is preferably an oxygen-containing group, such as an alkoxy group, an aryloxy group, a hydroxyl group or the like, a nitrogen-containing group, such as an amino
15 group, an imino group, an amido group, an imido group, a nitro group or the like, or a sulfur-containing group, such as an alkylthio group, an arylthio group or the like, more preferably an alkoxy group, an aryloxy group or an amino group, most preferably an alkoxy group.

20 R^{14} is a hydrocarbon group or a hydrocarbon-substituted silyl group. Examples of the hydrocarbon groups preferable as R^{14} include straight-chain or branched alkyl groups of 1 to 30 carbon atoms, preferably
1 to 20 carbon atoms, such as methyl, ethyl, n-propyl,
25 isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl,

-9 APR 2003

neopentyl and n-hexyl; cyclic saturated hydrocarbon groups of 3 to 30 carbon atoms, preferably 3 to 20 carbon atoms, such as cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl and adamantyl; aryl groups of 6 to 30 carbon atoms, preferably 6 to 20 carbon atoms, such as phenyl, benzyl, naphthyl, biphenyl and triphenyl; and groups wherein these groups are further substituted with substituent groups such as alkyl groups of 1 to 30 carbon atoms, preferably 1 to 20 carbon atoms, or aryl groups of 6 to 30 carbon atoms, preferably 6 to 20 carbon atoms.

Examples of the hydrocarbon-substituted silyl groups preferable as R^{14} include methylsilyl, dimethylsilyl, trimethylsilyl, ethylsilyl, diethylsilyl, triethylsilyl, diphenylmethylsilyl, triphenylsilyl, dimethylphenylsilyl, dimethyl-t-butylsilyl and dimethyl(pentafluorophenyl)silyl. Particularly preferable are trimethylsilyl, triethylphenylsilyl, diphenylmethylsilyl, isophenylsilyl, dimethylphenylsilyl, dimethyl-t-butylsilyl and dimethyl(pentafluorophenyl)silyl.

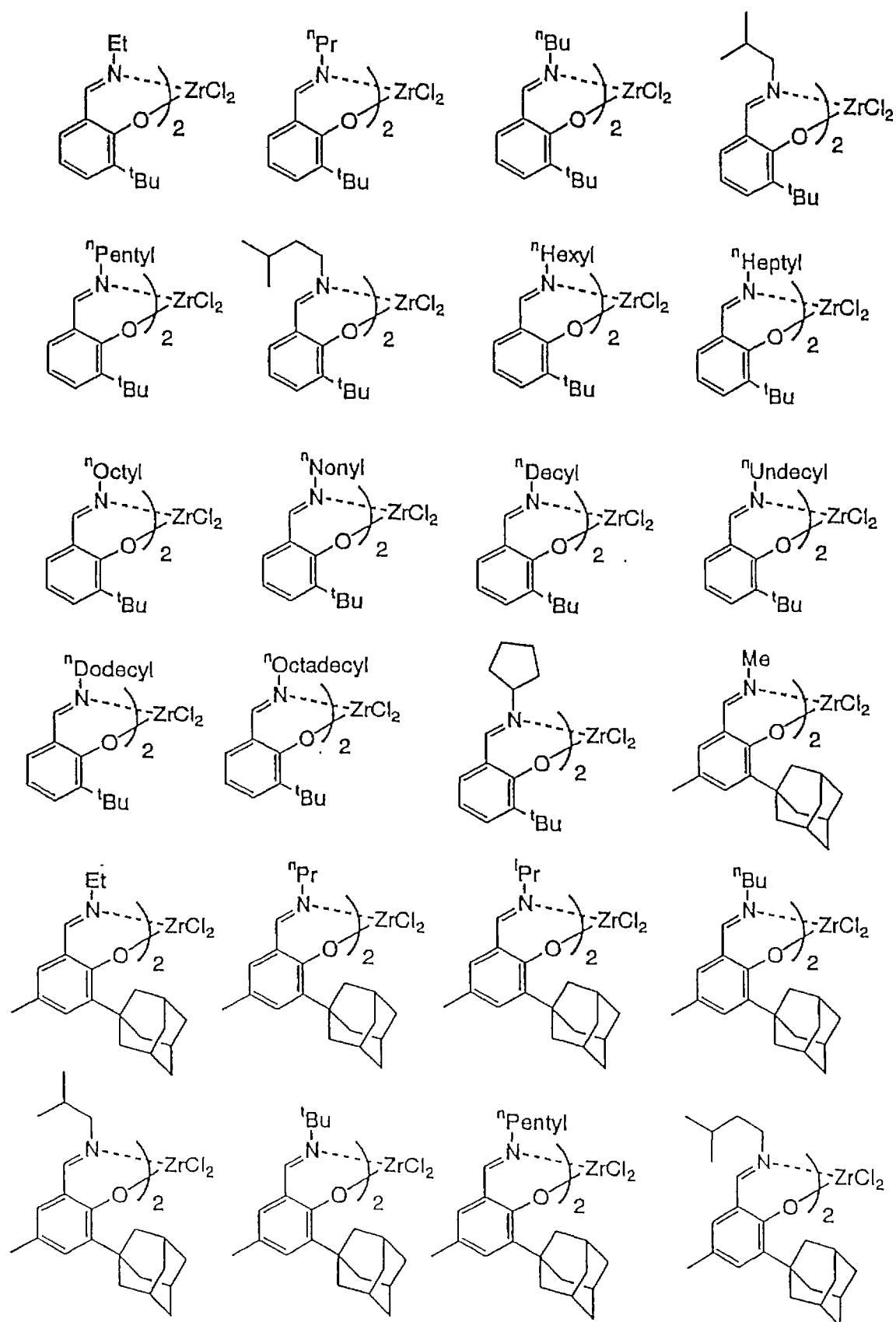
In the present invention, R^{14} is particularly preferably a group selected from branched alkyl groups of 3 to 30 carbon atoms, preferably 3 to 20 carbon atoms, e.g., isopropyl, isobutyl, sec-butyl, tert-butyl and neopentyl, groups wherein aryl groups of 6 to 30 carbon

9 APR 2003

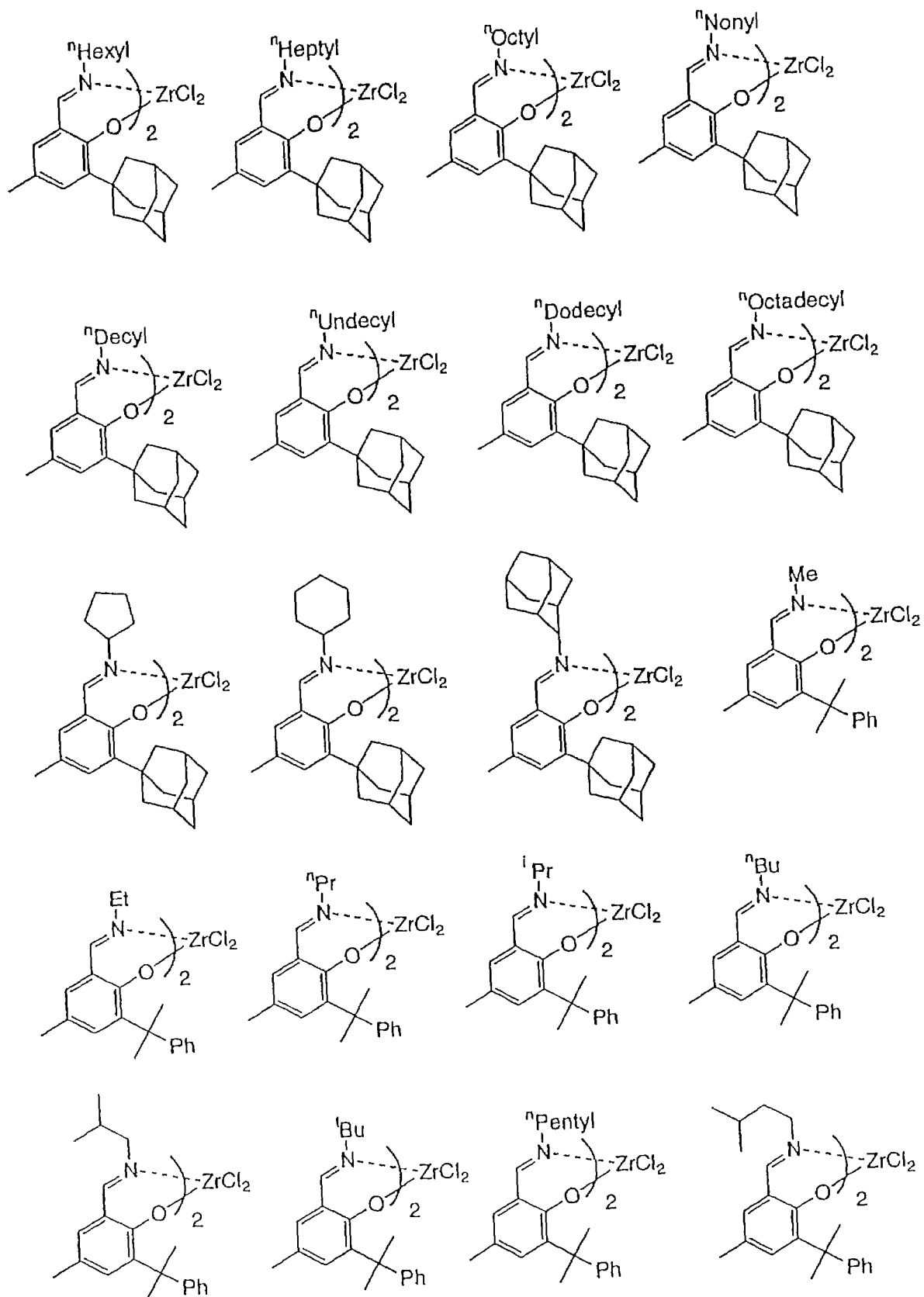
atoms, preferably 6 to 20 carbon atoms, are substituted
for the hydrogen atoms of these groups, e.g., cumyl, and
cyclic saturated hydrocarbon groups of 3 to 30 carbon
atoms, preferably 3 to 20 carbon atoms, e.g., adamantyl,
5 cyclopropyl, cyclobutyl, cyclopentyl and cyclohexyl. An
aryl group of 6 to 30 carbon atoms, preferably 6 to 20
carbon atoms, such as phenyl, naphthyl, fluorenyl,
anthranyl or phenanthryl, or a hydrocarbon-substituted
silyl group is also preferable.

10 Examples of the transition metal compounds
represented by the formula (II) are given below, but the
compounds of the formula (II) are not limited to those
examples. In the following examples, the compounds
represented by the formula (I) are included.

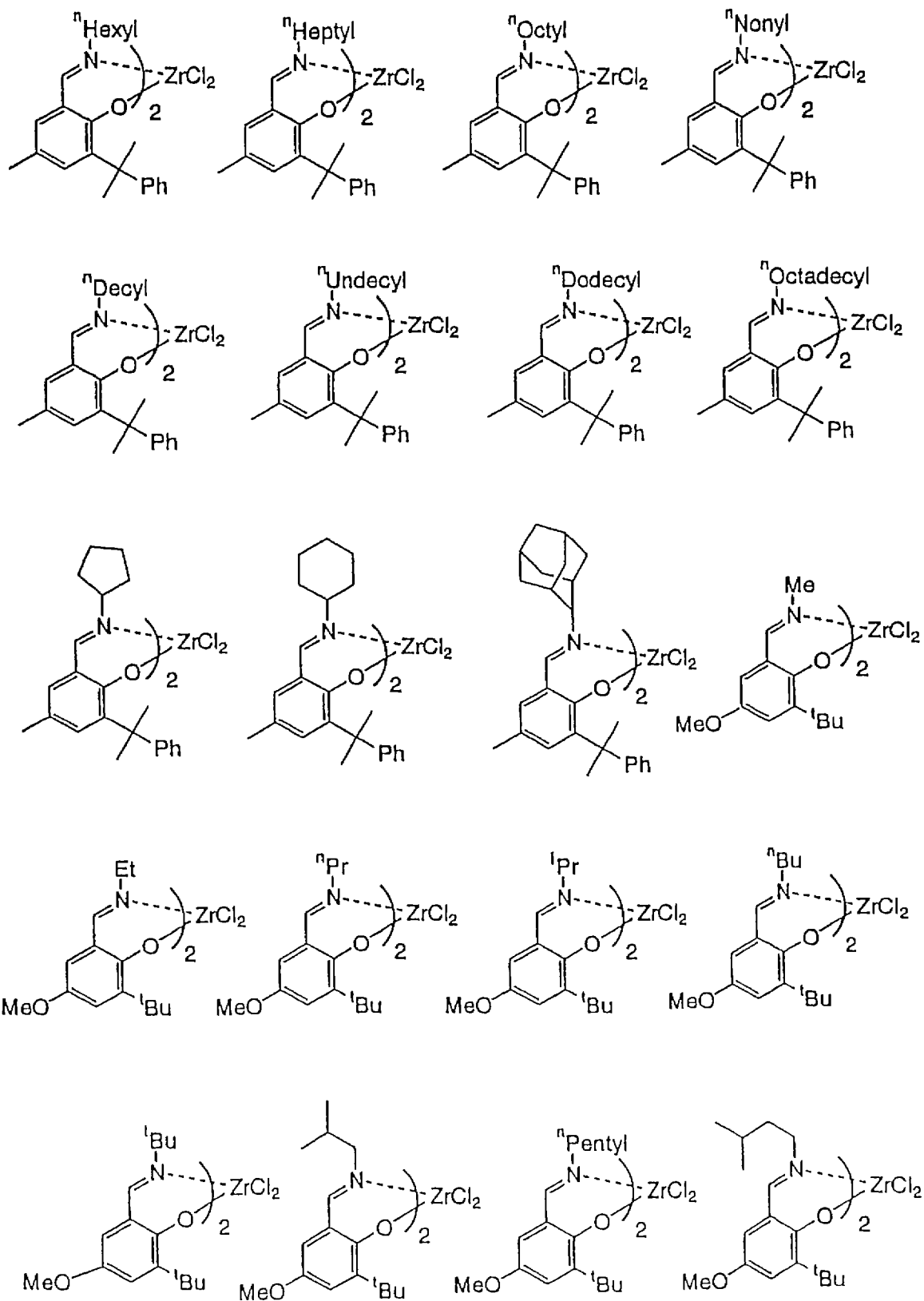
-9 APR 2003



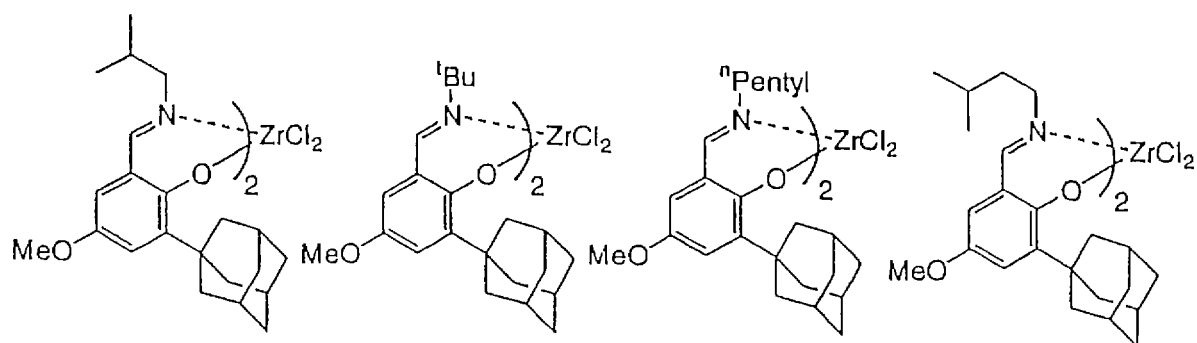
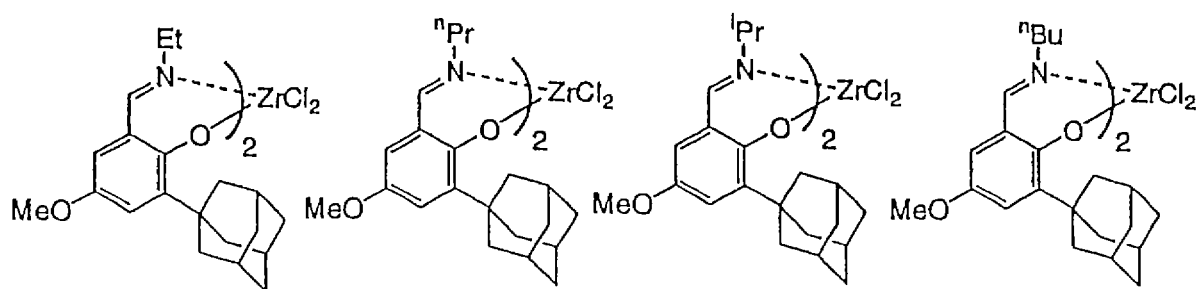
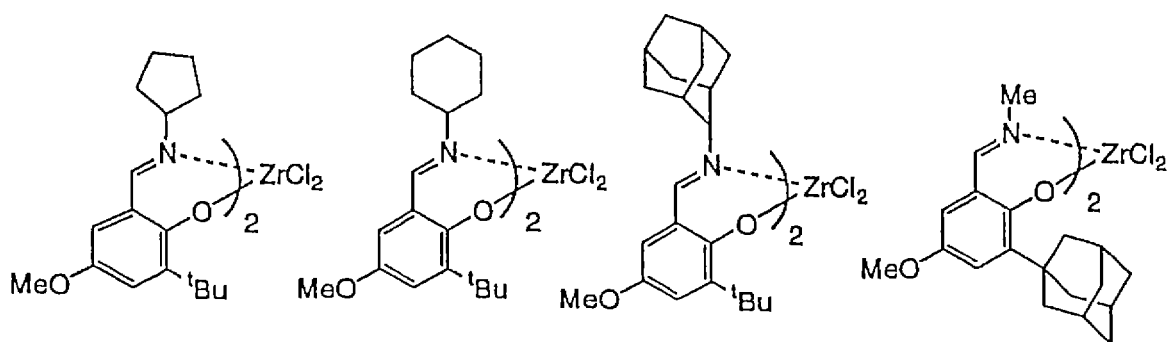
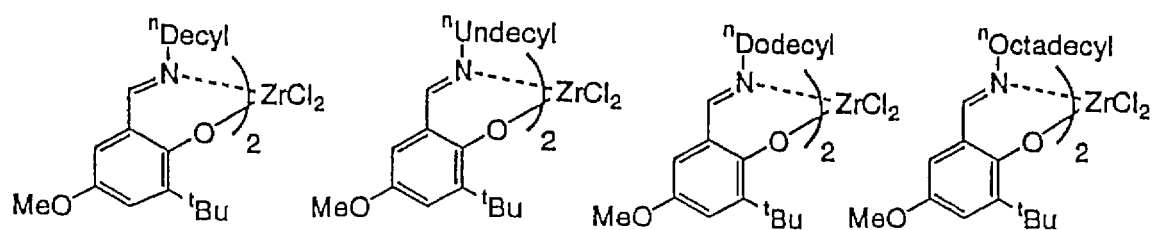
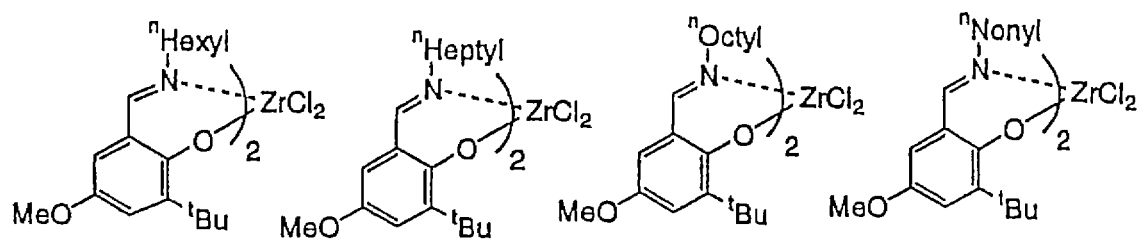
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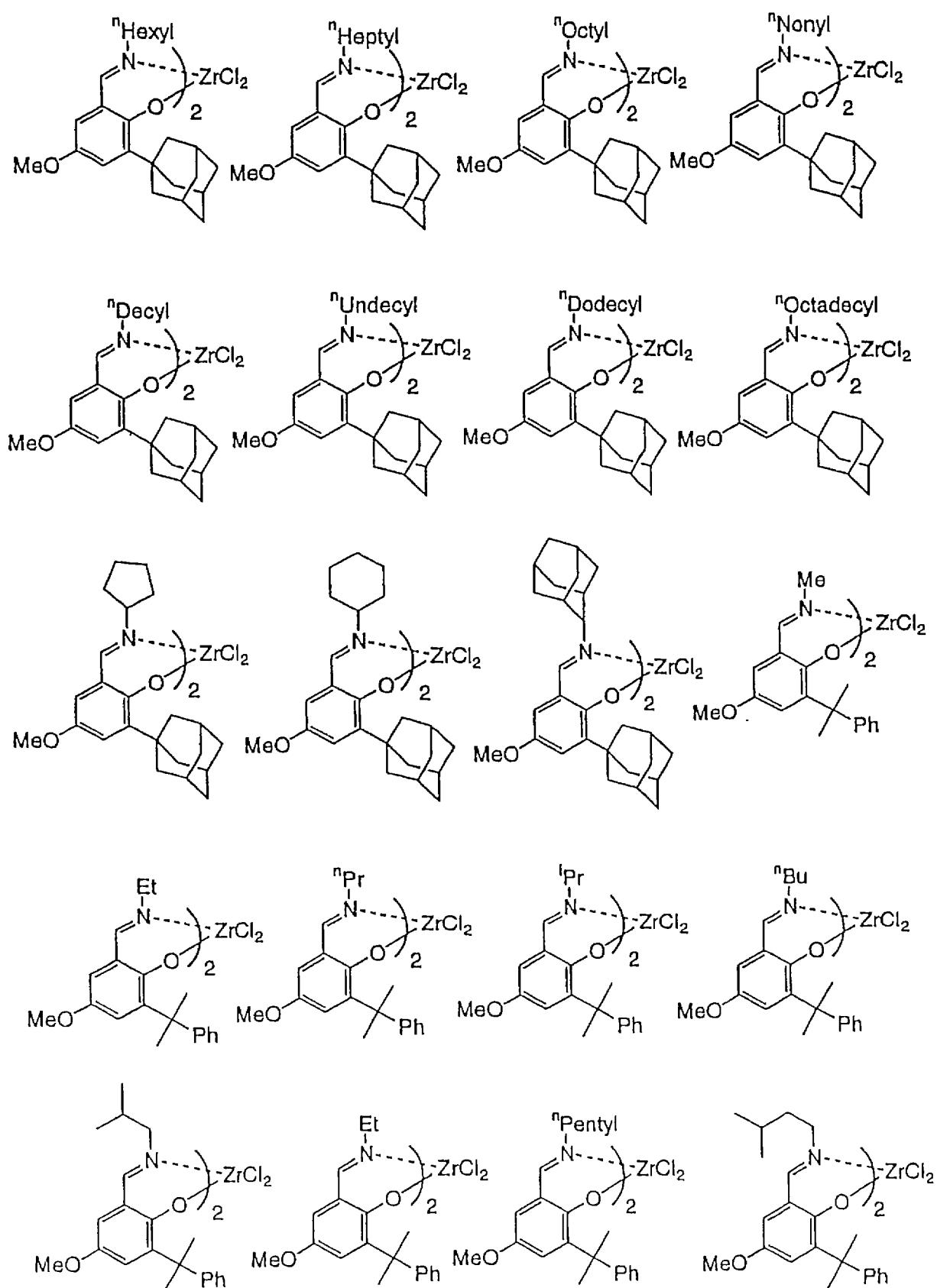
-9 APR 2003



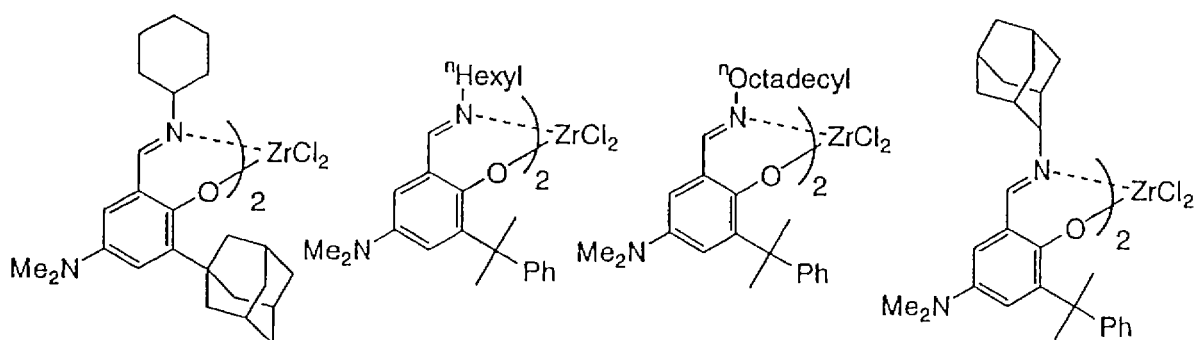
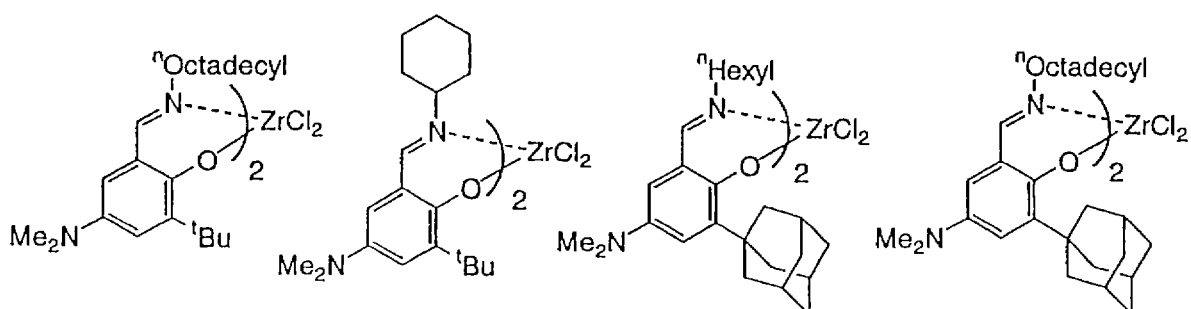
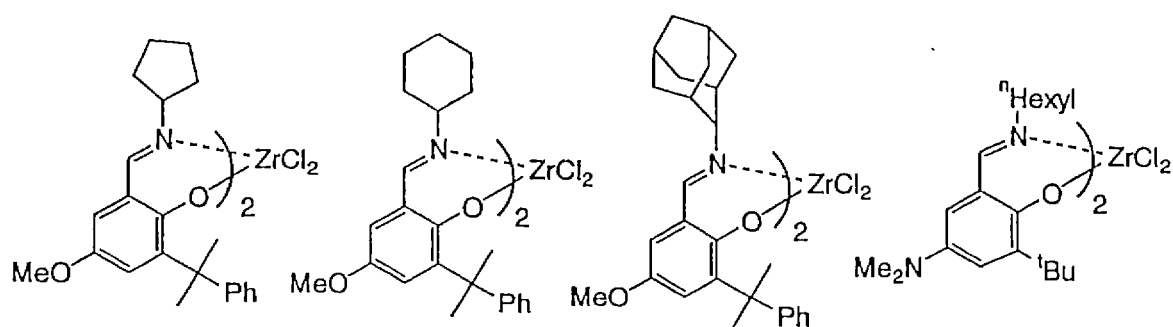
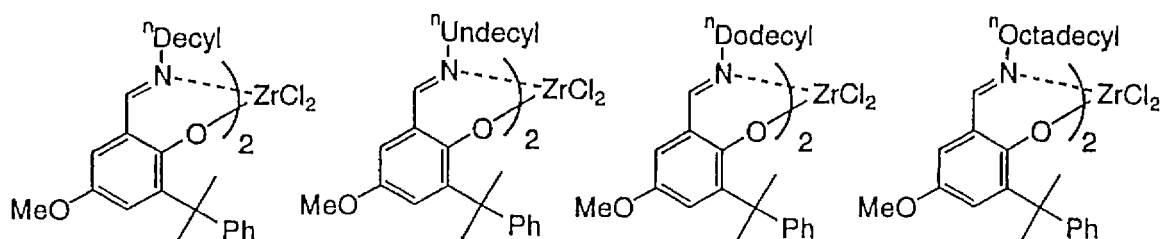
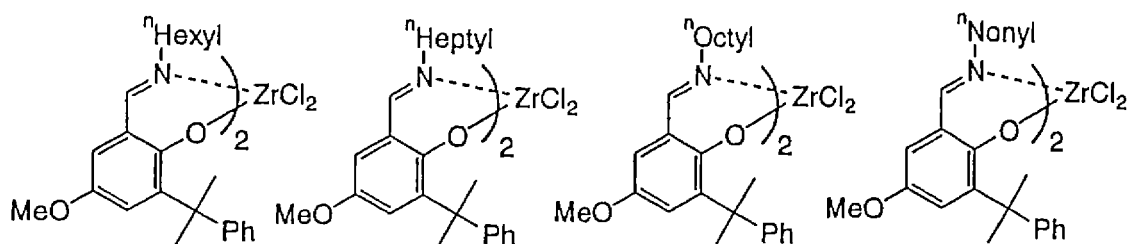
-9 APR 2003



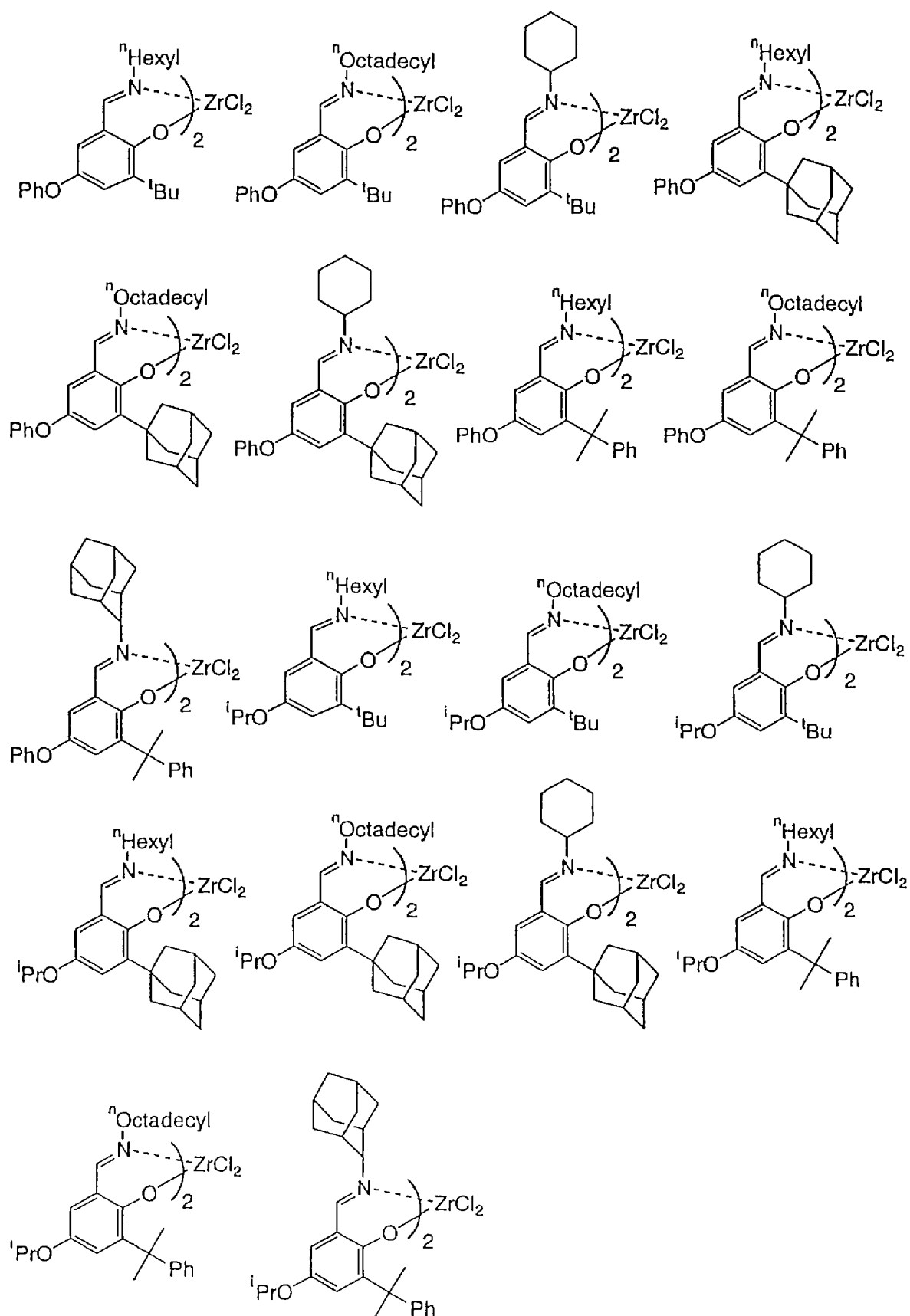
-9 APR 2003



-9 APR 2003



-9 APR 2003



-9 APR 2003

In the above examples, Me denotes a methyl group, Et denotes an ethyl group, ⁿPr denotes a n-propyl group, ⁱPr denotes an isopropyl group, ⁿBu denotes a n-butyl group, ^tBu denotes a t-butyl group, ⁿPentyl denotes a n-pentyl group, ⁿHexyl denotes a n-hexyl group, ⁿHeptyl denotes a n-heptyl group, ⁿOctyl denotes a n-octyl group, ⁿNonyl denotes a n-nonyl group, ⁿDecyl denotes a n-decyl group, ⁿUndecyl denotes a n-undecyl group, ⁿDodecyl denotes a n-dodecyl group, ⁿOctadecyl denotes a n-octadecyl group, and Ph denotes a phenyl group.

In the present invention, transition metal compounds wherein the zirconium metal is replaced with a metal other than zirconium, such as titanium or hafnium, in the above-mentioned compounds are also employable.

The process for preparing the transition metal compound represented by the formula (I) or (II) is not specifically limited, and for example, the following process is available.

The transition metal compound represented by the formula (I) can be synthesized by allowing a compound (ligand precursor) which will become a ligand in the synthesized transition metal compound to react with a transition metal M-containing compound represented by MX_k (M and X have the same meanings as those of M and X in

-9 APR 2003

the formula (I) and k is a number satisfying a valence of M).

The ligand precursor is obtained by allowing a β -diketone, a β -ketoester compound (including thioketone and thioketoester), an acetylacetone compound or the like to react with a primary amine compound represented by the formula R^1-NH_2 (R^1 has the same meaning as that of R^1 in the formula (I)) such as an aniline compound or an alkylamine compound. β -Diketones, β -ketoester compounds (including thioketones and thioketoesters), acetylacetone compounds, etc. are commercially available or obtainable by the processes known in literatures.

For synthesizing the ligand precursor, both of the starting compounds are first dissolved in a solvent. Then, the resulting solution is stirred at a temperature of room temperature to reflux temperature for about 1 to 48 hours, whereby the corresponding ligand precursor is obtained in a high yield.

A solvent usually used for such reaction is employable as the solvent. Above all, an alcohol solvent such as methanol or ethanol or a hydrocarbon solvent such as toluene is preferable.

In the synthesis of the ligand precursor, an acid catalyst such as formic acid, acetic acid or toluenesulfonic acid may be used as the catalyst. It is

-9 APR 2003

effective for the progress of the reaction to use a dehydrating agent such as molecular sieves, magnesium sulfate or sodium sulfate or to perform dehydration by the Dean and Stark method.

5 Preferred examples of the transition metal M-containing compounds include TiCl_3 , TiCl_4 , TiBr_3 , TiBr_4 , $\text{Ti}(\text{benzyl})_4$, $\text{Ti}(\text{NiMe}_2)_4$, ZrCl_4 , $\text{Zr}(\text{NiMe}_2)_4$, $\text{Zr}(\text{benzyl})_4$, ZrBr_4 , HfCl_4 , HfBr_4 , VCl_4 , VCl_5 , VBr_4 , VBr_5 , $\text{Ti}(\text{acac})_3$, and complexes of these compounds and tetrahydrofuran (THF),
10 acetonitrile or diethyl ether.

Then, the ligand precursor obtained above is allowed to react with the transition metal M-containing compound, whereby the corresponding transition metal compound can be synthesized.

15 More specifically, the synthesized ligand precursor is dissolved in a solvent. If necessary, the resulting solution may be contacted with a base to prepare a salt. Then, the solution or the solution with the salt is mixed with a metallic compound such as a metallic halide or a
20 metallic alkylate at a low temperature and stirred at a temperature of -78°C to room temperature or under reflux for about 1 to 48 hours. A solvent usually used for such reaction is employable as the solvent. Above all, a polar solvent such as ether or tetrahydrofuran or a
25 hydrocarbon solvent such as toluene is preferably used.

-9 APR 2003

Preferred examples of the bases used for preparing a phenoxide salt include metallic salts such as lithium salts (e.g., n-butyllithium) and sodium salts (e.g., sodium hydride) and organic bases such as triethylamine
5 and pyridine.

Depending upon properties of the transition metal compound, the ligand precursor is directly reacted with the metallic compound without producing a salt, whereby the corresponding transition metal compound can be
10 synthesized.

Further, it is possible to exchange the metal M in the synthesized transition metal compound with another transition metal by a conventional method. Furthermore, when any of R^1 to R^5 is H, a substituent group other than
15 H can be introduced in any stage of the synthesis process.

The transition metal compound represented by the formula (I-a) or (I-b) can be synthesized by allowing a compound (ligand precursor) for forming a ligand such as a thiosalicylidene ligand or an anilino ligand to react
20 with a transition metal M-containing compound.

The ligand precursor for forming the thiosalicylidene ligand is obtained by, for example, allowing a thiosalicylaldehyde compound to react with an aniline compound or an amine compound.

-9 APR 2003

To obtain the ligand precursor, o-acylbenzenethiol may be allowed to react with an aniline compound or an amine compound.

More specifically, a thiosalicylaldehyde compound or
5 o-acylbenzenethiol and an aniline compound wherein the nitrogen part has no substituent group or a primary amine compound are dissolved in a solvent, and the solution is stirred at a temperature of room temperature to reflux temperature for about 1 to 48 hours, whereby the ligand
10 precursor is obtained. As the solvent, an alcohol solvent such as methanol or ethanol or a hydrocarbon solvent such as toluene is preferably employed. As the catalyst, an acid catalyst such as formic acid, acetic acid or toluenesulfonic acid can be employed. It is
15 effective for the progress of the reaction to remove water from the reaction system by the Dean and Stark method during the reaction. Molecular sieves, magnesium sulfate, sodium sulfate or the like is employable as a dehydrating agent.

20 The o-acylbenzenethiol used herein can be obtained by, for example, allowing the OH group of o-acylphenol to react with dimethyl thiocarbamate to obtain thiocarbamate and then performing conversion reaction of oxygen atom and sulfur atom by means of heat or the like.

-9 APR 2003

The anilino ligand precursor can be obtained by allowing an o-formaniline compound to react with an aniline compound or an amine compound. To obtain the ligand precursor, o-acylaniline may be allowed to react
5 with aniline acid or an amine compound.

More specifically, the ligand precursor can be synthesized in the same manner as described above using, for example, an o-formaniline compound wherein the nitrogen part has no substituent group or o-acylaniline
10 wherein the nitrogen part has no substituent group and an aniline compound wherein the nitrogen part has no substituent group or a primary amine compound.

The o-acylaniline used herein can be obtained by, for example, reducing the carboxylic acid group of an o-aminobenzoic acid compound. A N-alkylation reaction of
15 an anthranil compound may be performed to obtain the corresponding N-alkyl-o-acylaniline compound.

The ligand precursor obtained as above is allowed to react with a transition metal M-containing compound,
20 whereby the corresponding transition metal compound can be synthesized. More specifically, the ligand precursor is dissolved in a solvent. If necessary, the resulting solution may be contacted with a base to prepare a thiophenoxide salt or an anilino salt. Then, the
25 solution or the solution with the salt is mixed with a

-9 APR 2003

transition metal M-containing compound such as a metallic halide or a metallic alkylate at a low temperature and stirred at a temperature of -78 °C to room temperature or under reflux for about 1 to 24 hours to obtain a
5 transition metal compound.

Examples of the solvents preferably used herein include polar solvents such as ether and tetrahydrofuran and hydrocarbon solvents such as toluene, without limiting thereto. Examples of the bases preferably used
10 herein include lithium salts such as n-butyllithium, sodium salts such as sodium hydride, and nitrogen-containing compounds such as pyridine and triethylamine, without limiting thereto.

Depending upon the type of the transition metal
15 compound, the ligand compound is directly reacted with the metallic compound, without preparing a thiophenoxide salt or an anilino salt, whereby the corresponding compound can be synthesized.

The transition metal compound represented by the
20 formula (II) can be synthesized in a manner similar to that described above.

The structure of the resulting transition metal compound is determined by 270 MHz ¹H-NMR (Japan Electron Optics Laboratory GSH-270), FT-IR (SHIMADZU FT-IR8200D),
25 FD-mass spectrometry (Japan Electron Optics Laboratory

-9 APR 2003

SX-102A), metal content analysis (analysis by ICP method after dry ashing and dissolution in dilute nitric acid, device: SHIMADZU ICPS-8000), and carbon, hydrogen and nitrogen content analysis (Halauss CHNO Model).

5 The transition metal compound obtained by the process described above is generally isolated by a conventional method and then used for polymerization. However, the reaction solution of the ligand precursor and the metallic compound can be used, as it is, for
10 polymerization, without isolating the transition metal compound.

 The transition metal compounds represented by the formula (I) are used singly or in combination of two or more kinds as an olefin polymerization catalyst.

15 It is a preferred embodiment of the present invention to use the transition metal compound (A-1) or (A-2) together with at least one compound (B) selected from (B-1) an organometallic compound, (B-2) an organoaluminum oxy-compound and (B-3) a compound which
20 reacts with the transition metal compound to form an ion pair.

(B-1) Organometallic compound

 Examples of the organometallic compounds (B-1) for
25 use in the invention may include the below-described

-9 APR 2003

compounds which contain a metal selected from Group 1, Group 2, Group 12 and Group 13 of the periodic table.

(B-1a) Organoaluminum compound represented by the following formula:



wherein R^a and R^b may be the same or different and are each a hydrocarbon group of 1 to 15 carbon atoms, preferably 1 to 4 carbon atoms; X is a halogen atom; and m, n, p and q are numbers satisfying the conditions of

10 $0 < m \leq 3, 0 \leq n < 3, 0 \leq p < 3, 0 \leq q < 3$ and $m+n+p+q = 3$.

(B-1b) Alkyl complex compound comprising a metal of Group 1 of the periodic table and aluminum, said compound being represented by the following formula:



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

(B-1c) Dialkyl compound containing a metal of Group 2 or Group 12 of the periodic table, said compound being represented by the following formula:



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

Examples of the organoaluminum compounds (B-1a)

25 include:

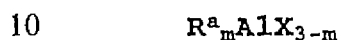
-9 APR 2003

an organoaluminum compound represented by the following formula:



wherein R^a and R^b may be the same or different and are
5 each a hydrocarbon group of 1 to 15 carbon atoms,
preferably 1 to 4 carbon atoms, and m is preferably a
number satisfying the condition of $1.5 \leq m \leq 3$;

an organoaluminum compound represented by the following formula:



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

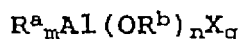
15 an organoaluminum compound represented by the following formula:



wherein R^a is a hydrocarbon group of 1 to 15 carbon atoms,
preferably 1 to 4 carbon atoms, and m is preferably a
20 number satisfying the condition of $2 \leq m < 3$;

and

an organoaluminum compound represented by the following formula:



-9 APR 2003

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

Particular examples of the organoaluminum compounds (B-1a) include:

tri-n-alkylaluminums, such as trimethylaluminum, triethylaluminum, tri-n-butylaluminum, tripropylaluminum, tripentylaluminum, trihexylaluminum, trioctylaluminum and tridecylaluminum;

branched-chain trialkylaluminums, such as triisopropylaluminum, triisobutylaluminum, tri-sec-butylaluminum, tri-tert-butylaluminum, tri-2-methylbutylaluminum, tri-3-methylbutylaluminum, tri-2-methylpentylaluminum, tri-3-methylpentylaluminum, tri-4-methylpentylaluminum, tri-2-methylhexylaluminum, tri-3-methylhexylaluminum and tri-2-ethylhexylaluminum;

tricycloalkylaluminums, such as tricyclohexylaluminum and tricyclooctylaluminum;

triarylaluminums, such as triphenylaluminum and tritolylaluminum;

dialkylaluminum hydrides, such as diisobutylaluminum hydride;

-9 APR 2003

trialkenylaluminums represented by (i-
 $C_4H_9)_xAl_y(C_5H_{10})_z$ (wherein x, y and z are each a positive
number, and $z \geq 2x$), such as triisoprenylaluminum;

alkylaluminum alkoxides, such as isobutylaluminum
5 methoxide, isobutylaluminum ethoxide and isobutylaluminum
isopropoxide;

dialkylaluminum alkoxides, such as dimethylaluminum
methoxide, diethylaluminum ethoxide and dibutylaluminum
butoxide;

10 alkylaluminum sesquialkoxides, such as ethylaluminum
sesquiethoxide and butylaluminum sesquibutoxide;

partially alkoxyated alkylaluminums having an
average composition represented by $R^a_{2.5}Al(OR^b)_{0.5}$;

dialkylaluminum aryloxides, such as diethylaluminum
15 phenoxide, diethylaluminum(2,6-di-t-butyl-4-
methylphenoxide), ethylaluminumbis(2,6-di-t-butyl-4-
methylphenoxide), diisobutylaluminum(2,6-di-t-butyl-4-
methylphenoxide) and isobutylaluminumbis(2,6-di-t-butyl-
4-methylphenoxide);

20 dialkylaluminum halides, such as dimethylaluminum
chloride, diethylaluminum chloride, dibutylaluminum
chloride, diethylaluminum bromide and diisobutylaluminum
chloride;

-9 APR 2003

alkylaluminum sesquihalides, such as ethylaluminum sesquichloride, butylaluminum sesquichloride and ethylaluminum sesquibromide,

partially halogenated alkylaluminums, such as
5 ethylaluminum dichloride, propylaluminum dichloride and butylaluminum dibromide;

dialkylaluminum hydrides, such as diethylaluminum hydride and dibutylaluminum hydride;

partially hydrogenated other alkylaluminums,
10 specifically, alkylaluminum dihydrides, such as ethylaluminum dihydride and propylaluminum dihydride; and

partially alkoxyated and halogenated alkylaluminums, such as ethylaluminum ethoxychloride, butylaluminum butoxychloride and ethylaluminum ethoxybromide.

15 Compounds analogous to the organoaluminum compound (B-1a) are also employable. For example, there can be mentioned organoaluminum compounds wherein two or more aluminum compounds are combined through a nitrogen atom, such as $(C_2H_5)_2AlN(C_2H_5)Al(C_2H_5)_2$.

20 Examples of the compounds (B-1b) include $LiAl(C_2H_5)_4$ and $LiAl(C_7H_{15})_4$.

Other compounds, such as methyllithium, ethyllithium, propyllithium, butyllithium, methylmagnesium bromide, methylmagnesium chloride, ethylmagnesium bromide,
25 ethylmagnesium chloride, propylmagnesium bromide,

-9 APR 2003

propylmagnesium chloride, butylmagnesium bromide,
butylmagnesium chloride, dimethylmagnesium,
diethylmagnesium, dibutylmagnesium and
butylethylmagnesium, are also employable as the
5 organometallic compounds (B-1).

Combinations of compounds capable of producing the
above-mentioned organoaluminum compounds in the
polymerization system, e.g., a combination of halogenated
aluminum and alkylolithium and a combination of
10 halogenated aluminum and alkylmagnesium, are also
employable.

Of the organometallic compounds (B-1), the
organoaluminum compounds are preferable.

The organometallic compounds (B-1) mentioned above
15 are used singly or in combination of two or more kinds.

(B-2) Organoaluminum oxy-compound

Examples of the organoaluminum oxy-compound (B-2)
for use in the invention may be conventional aluminoxane
or such a benzene-insoluble organoaluminum oxy-compound
20 as exemplified in Japanese Patent Laid-Open Publication
No. 78687/1990.

The conventional aluminoxane can be prepared by, for
example, the following processes, and is generally
obtained as a hydrocarbon solvent solution.

-9 APR 2003

(1) An organoaluminum compound such as trialkylaluminum is added to a hydrocarbon medium suspension of a compound containing adsorption water or a salt containing water of crystallization, e.g., magnesium chloride hydrate, copper sulfate hydrate, aluminum sulfate hydrate, nickel sulfate hydrate or cerous chloride hydrate, to allow the organoaluminum compound to react with the adsorption water or the water of crystallization.

(2) Water, ice or water vapor is allowed to directly act on an organoaluminum compound such as trialkylaluminum in a medium such as benzene, toluene, ethyl ether or tetrahydrofuran.

(3) An organotin oxide such as dimethyltin oxide or dibutyltin oxide is allowed to react with an organoaluminum compound such as trialkylaluminum in a medium such as decane, benzene or toluene.

The aluminoxane may contain a small amount of an organometallic component. Further, it is possible that the solvent or the unreacted organoaluminum compound is distilled off from the recovered solution of aluminoxane and the remainder is redissolved in a solvent or suspended in a poor solvent for aluminoxane.

Examples of the organoaluminum compounds used for preparing the aluminoxane include the same organoaluminum

-9 APR 2003

compounds as previously described with respect to the organoaluminum compound (B-1a). Of these, preferable are trialkylaluminums and tricycloalkylaluminums.

Particularly preferable is trimethylaluminum.

5 The organoaluminum compounds are used singly or in combination of two or more kinds.

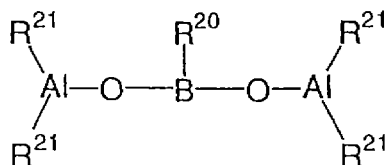
Examples of the solvents used for preparing the aluminoxane include aromatic hydrocarbons, such as benzene, toluene, xylene, cumene and cymene; aliphatic
10 hydrocarbons, such as pentane, hexane, heptane, octane, decane, dodecane, hexadecane and octadecane; alicyclic hydrocarbons, such as cyclopentane, cyclohexane, cyclooctane and methylcyclopentane; petroleum fractions, such as gasoline, kerosine and gas oil; and halogenated
15 products of these aromatic, aliphatic and alicyclic hydrocarbons, particularly chlorinated or brominated products thereof. Ethers such as ethyl ether and tetrahydrofuran are also employable. Of the solvents, particularly preferable are aromatic hydrocarbons and
20 aliphatic hydrocarbons.

The benzene-insoluble organoaluminum oxy-compound for use in the invention is preferably an organoaluminum oxy-compound containing an Al component that is soluble in benzene at 60 °C in an amount of usually not more than
25 10 %, preferably not more than 5 %, particularly

-9 APR 2003

preferably not more than 2 %, in terms of Al atom. That is, the benzene-insoluble organoaluminum oxy-compound is preferably insoluble or sparingly soluble in benzene.

The organoaluminum oxy-compound for use in the invention is, for example, an organoaluminum oxy-compound containing boron and represented by the following formula (III):

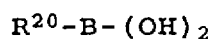


(III)

10

wherein R^{20} is a hydrocarbon group of 1 to 10 carbon atoms; and each R^{21} may be the same or different and is a hydrogen atom, a halogen atom or a hydrocarbon group of 1 to 10 carbon atoms.

15 The organoaluminum oxy-compound containing boron and represented by the formula (III) can be prepared by allowing an alkylboronic acid represented by the following formula (IV) to react with an organoaluminum compound in an inert solvent at a temperature of $-80\text{ }^{\circ}\text{C}$
20 to room temperature for a period of 1 minute to 24 hours under an inert gas atmosphere,



(IV)

-9 APR 2003

wherein R^{20} is the same group as described above.

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

Examples of the organoaluminum compounds to be reacted with the alkylboronic acid include the same organoaluminum compounds as previously described with respect to the organoaluminum compound (B-1a). Of these, preferable are trialkylaluminums and tricycloalkylaluminums. Particularly preferable are trimethylaluminum, triethylaluminum and triisobutylaluminum. These organoaluminum compounds are used singly or in combination of two or more kinds.

The organoaluminum oxy-compounds (B-2) mentioned above are used singly or in combination of two or more kinds.

-9 APR 2003

(B-3) Compound which reacts with the transition metal
compound to form ion pair

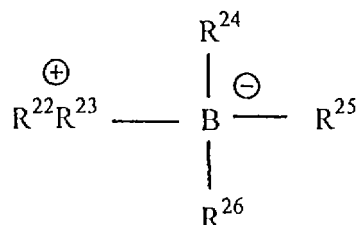
Examples of the compound (B-3) which reacts with the
transition metal compound (A) to form an ion pair

5 (referred to as "ionizing ionic compound" hereinafter)
may include a compound which reacts with the transition
metal compound represented by the above formula (I) or
(II) to form an ion pair, such as Lewis acid, an ionic
compound, a borane compound and a carborane compound
10 described in Japanese Patent Laid-Open Publications No.
501950/1989, No. 502036/1989, No. 179005/1991, No.
179006/1991, No. 207703/1991 and No. 207704/1991, and U.S.
Patent No. 5,321,106. A heteropoly compound and an
isopoly compound are also available.

15 The Lewis acid is, for example, a compound
represented by BR_3 (R is fluorine or a phenyl group which
may have a substituent group such as fluorine, methyl or
trifluoromethyl). Examples of such compounds include
trifluoroboron, triphenylboron, tris(4-fluorophenyl)boron,
20 tris(3,5-difluorophenyl)boron, tris(4-
fluoromethylphenyl)boron, tris(pentafluorophenyl)boron,
tris(p-tolyl)boron, tris(o-tolyl)boron and tris(3,5-
dimethylphenyl)boron.

The ionic compound is, for example, a compound
25 represented by the following formula (V).

-9 APR 2003



(v)

In the above formula, R²² is H⁺, carbonium cation,
 5 oxonium cation, ammonium cation, phosphonium cation,
 cycloheptyltrienyl cation, ferrocenium cation having a
 transition metal, or the like.

R²³ to R²⁶ may be the same or different and are each
 an organic group, preferably an aryl group or a
 10 substituted aryl group.

Examples of the carbonium cations include tri-
 substituted carbonium cations, such as triphenylcarbonium
 cation, tri(methylphenyl)carbonium cation and
 tri(dimethylphenyl)carbonium cation.

15 Examples of the ammonium cations include
 trialkylammonium cations, such as trimethylammonium
 cation, triethylammonium cation, tripropylammonium cation,
 tributylammonium cation and tri(n-butyl)ammonium cation;
 N,N-dialkylanilinium cations, such as N,N-
 20 dimethylanilinium cation, N,N-diethylanilinium cation and
 N,N-2,4,6-pentamethylanilinium cation; and

-9 APR 2003

dialkylammonium cations, such as di(isopropyl)ammonium cation and dicyclohexylammonium cation.

Examples of the phosphonium cations include triarylphosphonium cations, such as triphenylphosphonium cation, tri(methylphenyl)phosphonium cation and tri(dimethylphenyl)phosphonium cation.

R^{22} is preferably carbonium cation, ammonium cation or the like, particularly preferably triphenylcarbonium cation, N,N-dimethylanilinium cation or N,N-diethylanilinium cation.

Also employable as the ionic compound is a trialkyl-substituted ammonium salt, a N,N-dialkylanilinium salt, a dialkylammonium salt or a triarylphosphonium salt.

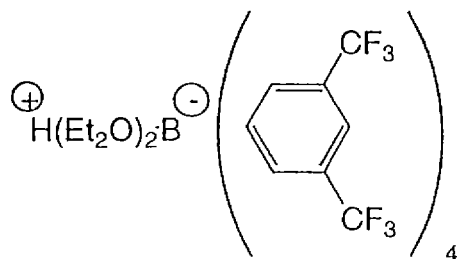
Examples of the trialkyl-substituted ammonium salts include triethylammoniumtetra(phenyl)boron, tripropylammoniumtetra(phenyl)boron, tri(n-butyl)ammoniumtetra(phenyl)boron, trimethylammoniumtetra(p-tolyl)boron, trimethylammoniumtetra(o-tolyl)boron, tri(n-butyl)ammoniumtetra(pentafluorophenyl)boron, tripropylammoniumtetra(o,p-dimethylphenyl)boron, tri(n-butyl)ammoniumtetra(m,m-dimethylphenyl)boron, tri(n-butyl)ammoniumtetra(p-trifluoromethylphenyl)boron, tri(n-butyl)ammoniumtetra(3,5-ditrifluoromethylphenyl)boron and tri(n-butyl)ammoniumtetra(o-tolyl)boron.

-9 APR 2003

Examples of the N,N-dialkylanilinium salts include N,N-dimethylaniliniumtetra(phenyl)boron, N,N-diethylaniliniumtetra(phenyl)boron and N,N-2,4,6-pentamethylaniliniumtetra(phenyl)boron.

5 Examples of the dialkylammonium salts include di(1-propyl)ammoniumtetra(pentafluorophenyl)boron and dicyclohexylammoniumtetra(phenyl)boron.

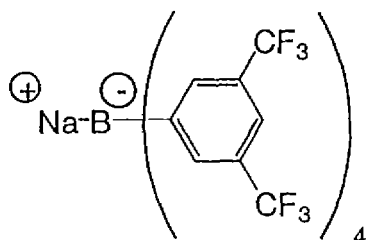
Further employable as the ionic compound is triphenylcarbeniumtetrakis(pentafluorophenyl)borate, N,N-
 10 dimethylaniliniumtetrakis(pentafluorophenyl)borate, ferroceniumtetra(pentafluorophenyl)borate, triphenylcarbeniumpentaphenylcyclopentadienyl complex, N,N-diethylaniliniumpentaphenylcyclopentadienyl complex or a boron compound represented by the following formula
 15 (VI) or (VII).



(VI)

wherein Et is an ethyl group.

-9 APR 2003



(VII)

Examples of the borane compounds include:

decaborane(14);

- 5 salts of anions, such as bis(tri(n-butyl)ammonium)nonaborate, bis(tri(n-butyl)ammonium)decaborate, bis(tri(n-butyl)ammonium)undecaborate, bis(tri(n-butyl)ammonium)dodecaborate, bis(tri(n-butyl)ammonium)decachlorodecaborate and bis(tri(n-butyl)ammonium)dodecachlorododecaborate; and
- 10 salts of metallic borane anions, such as tri(n-butyl)ammoniumbis(dodecahydridododecaborate)cobaltate(III) and bis(tri(n-butyl)ammonium)bis-
- 15 (dodecahydridododecaborate)nickelate(III).

Examples of the carborane compounds include:

- salts of anions, such as 4-carbanonaborane(14), 1,3-dicarbanonaborane(13), 6,9-dicarbadecaborane(14), dodecahydrido-1-phenyl-1,3-dicarbanonaborane,
- 20 dodecahydrido-1-methyl-1,3-dicarbanonaborane, undecahydrido-1,3-dimethyl-1,3-dicarbanonaborane, 7,8-

-9 APR 2003

dicarbaundecaborane(13), 2,7-dicarbaundecaborane(13),
undecahydrido-7,8-dimethyl-7,8-dicarbaundecaborane,
dodecahydrido-11-methyl-2,7-dicarbaundecaborane, tri(n-
butyl)ammonium-1-carbadecaborate, tri(n-butyl)ammonium-1-
5 carbaundecaborate, tri(n-butyl)ammonium-1-
carbadodecaborate, tri(n-butyl)ammonium-1-trimethylsilyl-
1-carbadecaborate, tri(n-butyl)ammoniumbromo-1-
carbadodecaborate, tri(n-butyl)ammonium-6-
carbadecaborate(14), tri(n-butyl)ammonium-6-
10 carbadecaborate(12), tri(n-butyl)ammonium-7-
carbaundecaborate(13), tri(n-butyl)ammonium-7,8-
dicarbaundecaborate(12), tri(n-butyl)ammonium-2,9-
dicarbaundecaborate(12), tri(n-
butyl)ammoniumdodecahydrido-8-methyl-7,9-
15 dicarbaundecaborate, tri(n-butyl)ammoniumundecahydrido-8-
ethyl-7,9-dicarbaundecaborate, tri(n-
butyl)ammoniumundecahydrido-8-butyl-7,9-
dicarbaundecaborate, tri(n-butyl)ammoniumundecahydrido-8-
allyl-7,9-dicarbaundecaborate, tri(n-
20 butyl)ammoniumundecahydrido-9-trimethylsilyl-7,8-
dicarbaundecaborate and tri(n-
butyl)ammoniumundecahydrido-4,6-dibromo-7-
carbaundecaborate; and
salts of metallic carborane anions, such as tri(n-
25 butyl)ammoniumbis(nonahydrido-1,3-

-9 APR 2003

- dicarbanonaborate)cobaltate(III), tri(n-butyl)ammoniumbis(undecahydrido-7,8-dicarbaundecaborate)ferrate(III), tri(n-butyl)ammoniumbis(undecahydrido-7,8-
- 5 dicarbaundecaborate)cobaltate(III), tri(n-butyl)ammoniumbis(undecahydrido-7,8-dicarbaundecaborate)nickelate(III), tri(n-butyl)ammoniumbis(undecahydrido-7,8-dicarbaundecaborate)cuprate(III), tri(n-
- 10 butyl)ammoniumbis(undecahydrido-7,8-dicarbaundecaborate)aurate(III), tri(n-butyl)ammoniumbis(nonahydrido-7,8-dimethyl-7,8-dicarbaundecaborate)ferrate(III), tri(n-butyl)ammoniumbis(nonahydrido-7,8-dimethyl-7,8-
- 15 dicarbaundecaborate)chromate(III), tri(n-butyl)ammoniumbis(tribromooctahydrido-7,8-dicarbaundecaborate)cobaltate(III), tris(tri(n-butyl)ammonium)bis(undecahydrido-7-carbaundecaborate)chromate(III), bis(tri(n-
- 20 butyl)ammonium)bis(undecahydrido-7-carbaundecaborate)manganate(IV), bis(tri(n-butyl)ammonium)bis(undecahydrido-7-carbaundecaborate)cobaltate(III) and bis(tri(n-butyl)ammonium)bis(undecahydrido-7-
- 25 carbaundecaborate)nickelate(IV).

The heteropoly compound comprises an atom selected from silicon, phosphorus, titanium, germanium, arsenic and tin and one or more atoms selected from vanadium, niobium, molybdenum and tungsten. Examples of such compounds include phosphovanadic acid, germanovanadic acid, arsenovanadic acid, phosphoniobic acid, germanoniobic acid, silicomolybdic acid, phosphomolybdic acid, titanomolybdic acid, germanomolybdic acid, arsenomolybdic acid, stannomolybdic acid, phosphotungstic acid, germanotungstic acid, stannotungstic acid, phosphomolybdovanadic acid, phosphotungstovanadic acid, germanotungstovanadic acid, phosphomolybdotungstovanadic acid, germanomolybdotungstovanadic acid, phosphomolybdotungstic acid, phosphomolybdoniobic acid and salts of these acids, for example, the salt of these acids and metals of Group 1 or Group 2 of the periodic table (e.g., lithium, sodium, potassium, rubidium, cesium, beryllium, magnesium, calcium, strontium, barium), and organic salts such as salts of these acids and triphenylethyl salt, but the compounds are not limited to these examples.

The ionizing ionic compounds (B-3) mentioned above are used singly or in combination of two or more kinds.

In the process for olefin polymerization according to the invention,

-9 APR 2003

the transition metal compound (A-1) or (A-2) may be used alone, or

the transition metal compound (A-1) or (A-2) may be used together with at least one compound (B) selected

5 from:

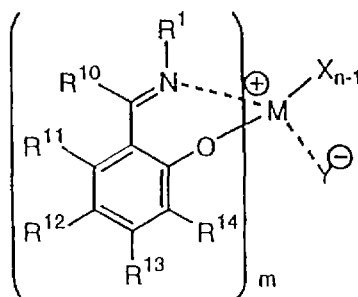
(B-1) an organometallic compound,

(B-2) an organoaluminum oxy-compound, and

(B-3) a compound which reacts with the transition metal compound to form an ion pair.

10 When the compound represented by the formula (II) is used as a transition metal compound and the component (B) is used in combination, the transition metal compound (A) forms a compound represented by the following formula (II-a) in the polymerization system.

15



(II-a)

-9 APR 2003

wherein R^1 , R^{10} to R^{14} , M, m, n and X have the same meanings as those of R^1 , R^{10} to R^{14} , M, m, n and X in the formula (II), and Y is a "weak coordination anion".

In the formula (II-a), the bond between the metal M
5 and Y may be a covalent bond or an ionic bond.

Examples of Y include weak coordination anions described in *Chemical Review*, Vol. 88, p. 1405 (1988), *Chemical Review*, Vol. 93, p. 927 (1993), and WO 98/30612, p. 6.

10 More specifically, there can be mentioned:



wherein each R may be the same or different and is an atom or a group selected from an oxygen atom, a nitrogen atom, a phosphorus atom, a hydrogen atom, a halogen atom,
15 a substituent group containing any of these atoms, an aliphatic hydrocarbon group, an aromatic hydrocarbon group, an alicyclic hydrocarbon group, an aliphatic, aromatic or alicyclic hydrocarbon group substituted with an oxygen atom, a nitrogen atom, a phosphorus atom or a
20 halogen atom, and an aliphatic, aromatic or alicyclic hydrocarbon group substituted with a substituent group having an oxygen atom, a nitrogen atom, a phosphorus atom or a halogen atom;



-9 APR 2003

wherein each R may be the same or different and is an atom or a group selected from an oxygen atom, a nitrogen atom, a phosphorus atom, a hydrogen atom, a halogen atom, a substituent group containing any of these atoms, an
5 aliphatic hydrocarbon group, an aromatic hydrocarbon group, an alicyclic hydrocarbon group, an aliphatic, aromatic or alicyclic hydrocarbon group substituted with an oxygen atom, a nitrogen atom, a phosphorus atom or a halogen atom, and an aliphatic, aromatic or alicyclic
10 hydrocarbon group substituted with a substituent group having an oxygen atom, a nitrogen atom, a phosphorus atom or a halogen atom;

PF₆⁻;

SbF₅⁻;

15 trifluoromethanesulfonato; and
p-toluenesulfonato.

In addition to the transition metal compound (A-1) or (A-2) (sometimes referred to as a "component (A)" hereinafter) and at least one compound (B) (sometimes
20 referred to as a "component (B)" hereinafter) selected from the organometallic compound (B-1), the organoaluminum oxy-compound (B-2) and the compound (B-3) which reacts with the transition metal compound to form an ion pair, said component (B) being optionally used,
25 the olefin polymerization catalyst of the invention can

-9 APR 2003

further contain the below-described carrier (C) and/or the later-described organic compound (D), if necessary.

(C) Carrier

5 The carrier (C) for optional use in the invention is an inorganic or organic compound in the form of granular or particulate solid.

As the inorganic compound, porous oxide, inorganic halogenide, clay, clay mineral or an ion-exchange layered
10 compound is preferable.

Examples of the porous oxides include SiO_2 , Al_2O_3 , MgO , ZrO , TiO_2 , B_2O_3 , CaO , ZnO , BaO , ThO_2 , and complex compounds or mixtures containing these oxides, such as natural or synthetic zeolite, $\text{SiO}_2\text{-MgO}$, $\text{SiO}_2\text{-Al}_2\text{O}_3$, $\text{SiO}_2\text{-TiO}_2$, $\text{SiO}_2\text{-V}_2\text{O}_5$, $\text{SiO}_2\text{-Cr}_2\text{O}_3$ and $\text{SiO}_2\text{-TiO}_2\text{-MgO}$. Of these,
15 preferable are compounds containing SiO_2 and/or Al_2O_3 as their major component.

The inorganic oxides may contain small amounts of carbonate, sulfate, nitrate and oxide components, such as
20 Na_2CO_3 , K_2CO_3 , CaCO_3 , MgCO_3 , Na_2SO_4 , $\text{Al}_2(\text{SO}_4)_3$, BaSO_4 , KNO_3 , $\text{Mg}(\text{NO}_3)_2$, $\text{Al}(\text{NO}_3)_3$, Na_2O , K_2O and Li_2O .

Although the porous oxides differ in their properties depending upon the type and the preparation process thereof, the carrier preferably used in the
25 invention has a particle diameter of 10 to 300 μm ,

-9 APR 2003

preferably 20 to 200 μm , a specific surface area of 50 to 1000 m^2/g , preferably 100 to 700 m^2/g , and a pore volume of 0.3 to 3.0 cm^3/g . If necessary, the carrier is calcined at 100 to 1000 $^{\circ}\text{C}$, preferably 150 to 700 $^{\circ}\text{C}$,
5 prior to use.

Examples of the inorganic halogenides employable in the invention include MgCl_2 , MgBr_2 , MnCl_2 and MnBr_2 . The inorganic halogenide may be used as it is, or may be used after pulverized by a ball mill or an oscillating mill.
10 The inorganic halogenide may be used as fine particles of a precipitate obtained by dissolving the inorganic halogenide in a solvent such as alcohol and then conducting precipitation using a precipitant.

The clay employable in the invention is generally
15 mainly composed of clay mineral. The ion-exchange layered compound employable is a compound having a crystal structure wherein layers structured by ionic bonding or the like are laminated in parallel to one another with a weak bond strength, and the ions contained
20 in the compound are exchangeable. Most of clay minerals are ion-exchange layered compounds. The clay, the clay minerals and the ion-exchange layered compounds employable in the invention are not limited to natural ones but include synthetic ones.

Examples of such clay, clay minerals and ion-exchange layered compounds include clay, clay minerals and ion crystalline compounds having layered crystal structures such as hexagonal closest packing type, antimony type, CdCl_2 type and CdI_2 type.

Particular examples of the clay and the clay minerals include kaolin, bentonite, kibushi clay, gairome clay, allophane, hisingerite, pyrophyllite, mica, montmorillonite, vermiculite, chlorite, palygorskite, kaolinite, nacrite, dickite and halloysite. Particular examples of the ion-exchange layered compounds include crystalline acid salts of polyvalent metals, such as $\alpha\text{-Zr}(\text{HAsO}_4)_2 \cdot \text{H}_2\text{O}$, $\alpha\text{-Zr}(\text{HPO}_4)_2$, $\alpha\text{-Zr}(\text{KPO}_4)_2 \cdot 3\text{H}_2\text{O}$, $\alpha\text{-Ti}(\text{HPO}_4)_2$, $\alpha\text{-Ti}(\text{HAsO}_4)_2 \cdot \text{H}_2\text{O}$, $\alpha\text{-Sn}(\text{HPO}_4)_2 \cdot \text{H}_2\text{O}$, $\gamma\text{-Zr}(\text{HPO}_4)_2$, $\gamma\text{-Ti}(\text{HPO}_4)_2$ and $\gamma\text{-Ti}(\text{NH}_4\text{PO}_4)_2 \cdot \text{H}_2\text{O}$.

The clay, the clay minerals and the ion-exchange layered compounds are preferably those having a pore volume, as measured on pores having a radius of not less than 20 Å by a mercury penetration method, of not less than 0.1 cc/g, and are particularly preferably those having a pore volume of 0.3 to 5 cc/g. The pore volume is measured on the pores having a radius of 20 to 3×10^4 Å by a mercury penetration method using a mercury porosimeter.

-9 APR 2003

If a compound having a pore volume, as measured on pores having a radius of not less than 20 Å, of less than 0.1 cc/g is used as the carrier, it tends to be difficult to obtain high polymerization activity.

5 It is preferable that the clay and the clay minerals for use in the invention are subjected to chemical treatments. Any of surface treatments to remove impurities attached to the surface and treatments having an influence on the crystal structure of the clay are
10 employable. Examples of such chemical treatments include acid treatment, alkali treatment, salt treatment and organic substance treatment. The acid treatment contributes to not only removing impurities from the surface but also eluting cations such as Al, Fe and Mg
15 present in the crystal structure to increase the surface area. The alkali treatment destroys crystal structure of clay to bring about change in the structure of the clay. The salt treatment and the organic substance treatment can produce ionic complex, molecular complex, organic
20 derivative or the like to change the surface area or the distance between layers.

 The ion-exchange layered compound for use in the invention may be a layered compound in which the exchangeable ions between layers have been exchanged with
25 other large and bulky ions utilizing ion exchange

-9 APR 2003

properties to enlarge the distance between the layers. The bulky ion plays a pillar-like roll to support the layer structure and is generally called a "pillar". Introduction of other substances between layers of a layered compound is called "intercalation". Examples of the guest compounds to be intercalated include cationic inorganic compounds, such as TiCl_4 and ZrCl_4 ; metallic alkoxides, such as $\text{Ti}(\text{OR})_4$, $\text{Zr}(\text{OR})_4$, $\text{PO}(\text{OR})_3$ and $\text{B}(\text{OR})_3$ (R is a hydrocarbon group or the like); and metallic hydroxide ions, such as $(\text{Al}_{13}\text{O}_4(\text{OH})_{24})^{7+}$, $(\text{Zr}_4(\text{OH})_{14})^{2+}$ and $(\text{Fe}_3\text{O}(\text{OCOCH}_3)_6)^+$. These compounds are used singly or in combination of two or more kinds. The intercalation of the compounds may be carried out in the presence of polymers obtained by hydrolysis of metallic alkoxides such as $\text{Si}(\text{OR})_4$, $\text{Al}(\text{OR})_3$ and $\text{Ge}(\text{OR})_4$ (R is a hydrocarbon group or the like) or in the presence of colloidal inorganic compounds such as SiO_2 . Examples of the pillars include oxides produced by intercalation of the above-mentioned metallic hydroxide ions between layers, followed by dehydration under heating.

The clay, clay minerals and ion-exchange layered compounds mentioned above may be used as they are, or may be used after they are subjected to a treatment of ball milling, sieving or the like. Moreover, they may be used after they are subjected to water adsorption or

-7 APR 2003

dehydration under heating. The clay, clay minerals and ion-exchange layered compounds may be used singly or in combination of two or more kinds.

Of the above-mentioned materials, preferable are
5 clay and clay minerals, and particularly preferable are montmorillonite, vermiculite, pectolite, taeniolite and synthetic mica.

The organic compound is, for example, a granular or particulate solid compound having a particle diameter of
10 10 to 300 μm . Examples of such compounds include (co)polymers produced using an α -olefin of 2 to 14 carbon atoms such as ethylene, propylene, 1-butene or 4-methyl-1-pentene as a chief ingredient, (co)polymers produced using vinylcyclohexane or styrene as a chief ingredient,
15 and modified products thereof.

(D) Organic compound component

In the present invention, the organic compound component (D) is optionally used for the purpose of
20 improving polymerizability and properties of the resulting polymer. Examples of the organic compounds include alcohols, phenolic compounds, carboxylic acids, phosphorus compounds and sulfonates, but the organic compounds are not limited to these examples.

-9 APR 2003

As the alcohols and the phenolic compounds, those represented by R^{20} -OH (R^{20} is a hydrocarbon group of 1 to 50 carbon atoms or a halogenated hydrocarbon group of 1 to 50 carbon atoms) are generally employed.

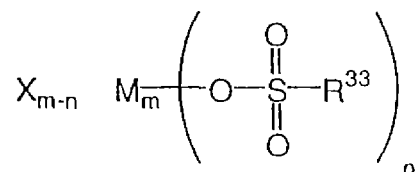
5 The alcohols are preferably those of the above formula wherein R^{20} is a halogenated hydrocarbon group. The phenolic compounds are preferably those wherein the α, α' -positions of the hydroxyl group are substituted with hydrocarbon groups of 1 to 20 carbon atoms.

10 As the carboxylic acids, those represented by R^{21} -COOH (R^{21} is a hydrocarbon group of 1 to 50 carbon atoms or a halogenated hydrocarbon group of 1 to 50 carbon atoms, preferably a halogenated hydrocarbon group of 1 to 50 carbon atoms) are generally employed.

15 As the phosphorus compounds, phosphoric acids having P-O-H bond, phosphates having P-OR bond or P=O bond and phosphine oxide compounds are preferably employed.

The sulfonates employable herein are those represented by the following formula (VIII):

20



(VIII)

-9 APR 2003

wherein M is an atom selected from Group 1 to Group 14 of the periodic table; R³³ is hydrogen, a hydrocarbon group of 1 to 20 carbon atoms or a halogenated hydrocarbon group of 1 to 20 carbon atoms; X is a hydrogen atom, a halogen atom, a hydrocarbon group of 1 to 20 carbon atoms or a halogenated hydrocarbon group of 1 to 20 carbon atoms; m is an integer of 1 to 7; and $1 \leq n \leq 7$.

In Fig. 1, steps for preparing the olefin polymerization catalyst of the invention are shown.

In the polymerization, the components can be used in any way and in any order. Some examples of the processes are given below.

(1) The component (A) and the compound (B) are fed to the polymerization reactor in an arbitrary order.

(2) A catalyst component wherein the component (A) is supported on the carrier (C), and the component (B) are fed to the polymerization reactor in an arbitrary order.

(3) A catalyst component wherein the component (B) is supported on the carrier (C), and the component (A) are fed to the polymerization reactor in an arbitrary order.

-9 APR 2003

(4) A catalyst component wherein the component (A) and the component (B) are supported on the carrier (C) is fed to the polymerization reactor.

In the above processes (1) to (4), at least two of
5 the catalyst components may be previously contacted.

In the processes (3) and (4) using a component (B) which is a supported type, a component (B) which is not a supported type may be added in an arbitrary order. In this case, the components (B) may be the same or
10 different.

Onto the solid catalyst component wherein the component (A) and the component (B) are supported on the carrier (C), an olefin may be prepolymerized. On the prepolymerized solid catalyst component, a catalyst
15 component may be further supported.

In the process for olefin polymerization according to the invention, an olefin is polymerized or copolymerized in the presence of the above-mentioned olefin polymerization catalyst to obtain an olefin
20 polymer.

In the present invention, the polymerization can be carried out as any of liquid phase polymerization, such as solution polymerization or suspension polymerization, and gas phase polymerization.

Examples of inert hydrocarbon media for use in the liquid phase polymerization include aliphatic hydrocarbons, such as propane, butane, pentane, hexane, heptane, octane, decane, dodecane and kerosine; alicyclic hydrocarbons, such as cyclopentane, cyclohexane and methylcyclopentane; aromatic hydrocarbons, such as benzene, toluene and xylene; halogenated hydrocarbons, such as ethylene chloride, chlorobenzene and dichloromethane; and mixtures of these hydrocarbons. The olefin itself can be used as the solvent.

In the polymerization of an olefin using the olefin polymerization catalyst, the component (A) is used in an amount of usually 10^{-12} to 10^{-2} mol, preferably 10^{-10} to 10^{-3} mol per 1 liter of reaction volume.

The component (B-1) is used in such an amount that the molar ratio $((B-1)/(M))$ of the component (B-1) to the transition metal atom (M) in the component (A) becomes usually 0.01 to 100000, preferably 0.05 to 50000. The component (B-2) is used in such an amount that the molar ratio $((B-2)/(M))$ of the aluminum atom in the component (B-2) to the transition metal atom (M) in the component (A) becomes usually 10 to 500000, preferably 20 to 100000. The component (B-3) is used in such an amount that the molar ratio $((B-3)/(M))$ of the component (B-3) to the

-9 APR 2003

transition metal atom (M) in the component (A) becomes usually 1 to 10, preferably 1 to 5.

When the component (B-1) is used as the component (B), the component (D) is optionally used in such an amount that the (D)/(B-1) ratio by mol becomes usually 0.01 to 10, preferably 0.1 to 5. When the component (B-2) is used as the component (B), the component (D) is optionally used in such an amount that the (D)/(B-2) ratio by mol becomes usually 0.001 to 2, preferably 0.005 to 1. When the component (B-3) is used as the component (B), the component (D) is optionally used in such an amount that the (D)/(B-3) ratio by mol becomes usually 0.01 to 10, preferably 0.1 to 5.

In the olefin polymerization using the olefin polymerization catalyst, the polymerization temperature is in the range of usually -50 to +250 °C, preferably 0 to 200 °C, more preferably 60 to 170 °C. The polymerization pressure is in the range of usually atmospheric pressure to 100 kg/cm², preferably atmospheric pressure to 50 kg/cm². The polymerization reaction can be carried out by any of batchwise, semi-continuous and continuous processes. The polymerization can be conducted in two or more stages under different reaction conditions.

-9 APR 2003

The molecular weight of the resulting olefin polymer can be controlled by allowing hydrogen to be present in the polymerization system or by changing the polymerization temperature. The molecular weight can be controlled also by changing the type of the component (B).

In the present invention, the olefin polymer with high molecular weight can be produced with excellent polymerization activity, especially at high temperatures by the use of the above olefin polymerization catalyst.

Examples of the olefins which can be polymerized by the use of the olefin polymerization catalyst include:

straight-chain or branched α -olefins of 2 to 30 carbon atoms, preferably 2 to 20 carbon atoms, such as ethylene, propylene, 1-butene, 2-butene, 1-pentene, 3-methyl-1-butene, 1-hexene, 4-methyl-1-pentene, 3-methyl-1-pentene, 1-octene, 1-decene, 1-dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene and 1-eicosene; and

cycloolefins of 3 to 30 carbon atoms, preferably 3 to 20 carbon atoms, such as cyclopentene, cycloheptene, norbornene, 5-methyl-2-norbornene, tetracyclododecene and 2-methyl-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene.

Polar monomers are also employable. Examples of the polar monomers include α,β -unsaturated carboxylic acids, such as acrylic acid, methacrylic acid, fumaric acid,

-9 APR 2003

maleic anhydride, itaconic acid, itaconic anhydride and
bicyclo[2,2,1]-5-heptene-2,3-dicarboxylic anhydride;
metallic salts, such as sodium salts, potassium salts,
lithium salts, zinc salts, magnesium salts and calcium
5 salts of the above acids; α,β -unsaturated carboxylic
esters, such as methyl acrylate, ethyl acrylate, n-propyl
acrylate, isopropyl acrylate, n-butyl acrylate, isobutyl
acrylate, tert-butyl acrylate, 2-ethylhexyl acrylate,
methyl methacrylate, ethyl methacrylate, n-propyl
10 methacrylate, isopropyl methacrylate, n-butyl
methacrylate and isobutyl methacrylate; vinyl esters,
such as vinyl acetate, vinyl propionate, vinyl caproate,
vinyl caprate, vinyl laurate, vinyl stearate and vinyl
trifluoroacetate; unsaturated glycidyl esters, such as
15 glycidyl acrylate, glycidyl methacrylate and monoglycidyl
itaconate; and halogen-containing olefins, such as vinyl
fluoride, vinyl chloride, vinyl boromide and vinyl iodide.

Vinylcyclohexane, dienes and polyenes are also
employable as olefins. The dienes and the polyenes
20 employable herein are cyclic or chain compounds having 4
to 30 carbon atoms, preferably 4 to 20 carbon atoms, and
having two or more double bonds. Examples of such
compounds include butadiene, isoprene, 4-methyl-1,3-
pentadiene, 1,3-pentadiene, 1,4-pentadiene, 1,5-hexadiene,
25 1,4-hexadiene, 1,3-hexadiene, 1,3-octadiene, 1,4-

-9 APR 2003

octadiene, 1,5-octadiene, 1,6-octadiene, 1,7-octadiene, ethylidene norbornene, vinyl norbornene, dicyclopentadiene, 7-methyl-1,6-octadiene, 4-ethylidene-8-methyl-1,7-nonadiene and 5,9-dimethyl-1,4,8-decatriene.

5 Aromatic vinyl compounds are also employable as olefins. Examples of such compounds include mono- or polyalkylstyrenes, such as styrene, o-methylstyrene, m-methylstyrene, p-methylstyrene, o,p-dimethylstyrene, o-ethylstyrene, m-ethylstyrene and p-ethylstyrene;
10 functional group-containing styrene derivatives, such as methoxystyrene, ethoxystyrene, vinylbenzoic acid, methyl vinylbenzoate, vinylbenzyl acetate, hydroxystyrene, o-chlorostyrene, p-chlorostyrene and divinylbenzene; and
15 other compounds, such as 3-phenylpropylene, 4-phenylpropylene and α -methylstyrene.

The olefins mentioned above can be used singly or in combination of two or more kinds.

EFFECT OF THE INVENTION

20 The olefin polymerization catalyst and the process for olefin polymerization according to the present invention can produce polyolefins with a high polymerization activity at high temperatures.

25

EXAMPLE

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

5 In the following examples, the intrinsic viscosity (η) was measured in decalin at 135 °C. The structures of the compounds obtained in the synthesis examples were determined by 270 MHz ¹H-NMR (Japan Electron Optics Laboratory GSH-270) and FD-mass spectrometry (Japan
10 Electron Optics Laboratory SX-102A), etc.

Synthesis Example 1

 Into a 500 ml reactor thoroughly dried and purged with argon, 7.51 g (50 mmol) of 2-tert-butylphenol and 54 ml of THF were introduced. To the reactor, 18.53 ml of
15 an ether solution containing 55.6 mmol of ethylmagnesium bromide was dropwise added at 0 °C, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 1 hour. Then, 180 ml of toluene was added, and the
20 system was heated to 100 °C to distill off about 40 ml of a mixed solution of ether and THF, whereby an opaque white slurry was obtained. After the slurry was cooled to room temperature, 3.75 g (125 mmol) of
25 paraformaldehyde and 10.45 ml (75 mmol) of triethylamine were added, followed by stirring at 88 °C for 1 hour.

-9 APR 2003

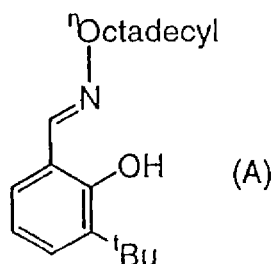
The reaction solution was allowed to cool to room temperature and then quenched with 10% hydrochloric acid. The organic layer was concentrated and purified by a silica gel column to obtain 6.22 g (yield: 70 %) of 3-t-butylsalicylaldehyde.

$^1\text{H-NMR}$ (CDCl_3): 1.42 (s, 9H), 6.94 (t, 1H), 7.25-7.54 (m, 2H), 9.86 (s, 1H), 11.79 (s, 1H)

Into a 200 ml reactor thoroughly purged with nitrogen, 80 ml of ethanol, 6.06 g (22.5 mmol) of n-octadecylamine and 2.84 g (15.0 mmol) of 3-t-butylsalicylaldehyde were introduced, then a small amount of acetic acid was added, and the mixture was stirred at room temperature for 24 hours. The reaction solution was vacuum concentrated to remove the solvent, whereby 4.56 g (yield: 70.7 %) of a compound (A) represented by the following formula (A) was obtained as a yellow crystal.

$^1\text{H-NMR}$ (CDCl_3): 0.89 (t, 3H), 1.26 (s, 30H), 1.44 (s, 9H), 1.53-1.75 (m, 2H), 3.57 (t, 2H), 6.79 (t, 1H), 7.08-7.11 (dd, 1H), 7.26-7.32 (dd, 1H), 8.32 (s, 1H), 14.22 (s, 1H)

- 9 APR 2003



Into a 100 ml reactor thoroughly dried and purged with argon, 0.86 g (2.0 mmol) of the compound (A) and 20 ml of ether were introduced, and they were cooled to -

5 78 °C and stirred. To the reactor, 1.43 ml of a n-hexane solution containing 2.2 mmol of n-butyllithium was dropwise added over a period of 10 minutes, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at

10 room temperature for 4 hours to prepare a lithium salt solution. The solution was dropwise added to 20 ml of a THF solution containing 0.38 g (1.0 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to -78 °C. After the dropwise addition was completed, stirring was

15 continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 15 hours, and the solvent was distilled off from the reaction solution. The resulting solid was recrystallized from a mixed solution of ether

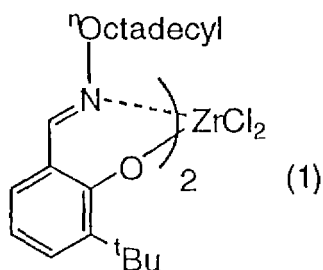
20 and hexane to obtain 0.220 g (yield: 21.6 %) of a

-9 APR 2003

compound (1) represented by the following formula (1) as a bright yellow crystal.

$^1\text{H-NMR}$ (CDCl_3): 0.88 (t, 6H), 1.25 (s, 78H), 1.57 (s, 4H), 3.59 (t, 4H), 6.92 (t, 2H), 7.20-7.26 (dd, 2H),
5 7.55-7.62 (dd, 2H), 8.15 (s, 1H)

FD-mass spectrometry: 1018

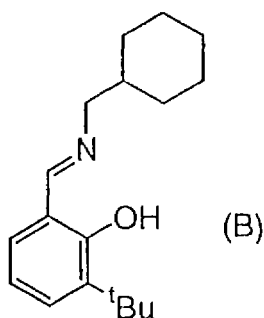


10 Synthesis Example 2

Into a 100 ml reactor thoroughly purged with nitrogen, 50 ml of ethanol, 2.28 g (20.14 mmol) of cyclohexanemethylamine and 2.98 g (16.73 mmol) of 3-*t*-butylsalicylaldehyde were introduced, and they were
15 stirred at room temperature for 20 hours. The reaction solution was vacuum concentrated to remove the solvent, and the resulting liquid was purified by silica gel column chromatography to obtain 3.38 g (yield: 74 %) of a compound (B) represented by the following formula (B) as
20 a yellow liquid.

-9 APR 2003

$^1\text{H-NMR}$ (CDCl_3): 0.95-1.82 (m, 11H), 1.44 (s, 9H), 3.42 (d, 2H), 6.76-7.32 (m, 3H), 8.28 (s, 1H), 14.19 (s, 1H)



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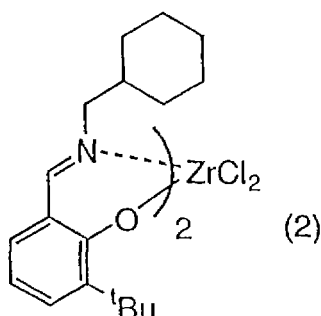
Into a 100 ml reactor thoroughly dried and purged with argon, 0.97 g (3.55 mmol) of the compound (B) and 20 ml of ether were introduced, and they were cooled to -78 °C and stirred. To the reactor, 2.40 ml of a n-hexane solution containing 3.84 mmol of n-butyllithium was dropwise added over a period of 10 minutes, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 4 hours to prepare a lithium salt solution. The solution was dropwise added to 30 ml of a THF solution containing 0.66 g (1.75 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to -78 °C. After the dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was

-9 APR 2003

further performed at room temperature for 15 hours, and the solvent was distilled off from the reaction solution. The resulting solid was washed with 120 ml of methylene chloride. The filtrate was concentrated and washed again
5 with ether to obtain 0.57 g (yield: 46 %) of a compound (2) represented by the following formula (2) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.50-1.74 (m, 22H), 1.58 (s, 18H),
3.32 (m, 4H), 6.90-7.60 (m, 6H), 8.01 (s, 2H)

10 $\text{FD-mass spectrometry: 706}$



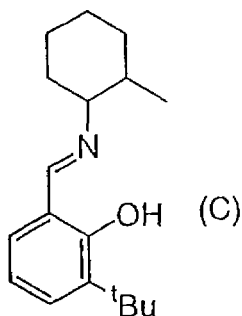
Synthesis Example 3

Into a 100 ml reactor thoroughly purged with
15 nitrogen, 50 ml of ethanol, 2.27 g (20.05 mmol) of 2-methylcyclohexylamine and 2.89 g (16.23 mmol) of 3-tert-butylsalicylaldehyde were introduced, then a small amount of acetic acid was added, and the mixture was stirred at room temperature for 24 hours. The reaction solution was
20 vacuum concentrated to remove the solvent, and the

-9 APR 2003

resulting liquid was purified by silica gel column chromatography to obtain 3.65 g (yield: 82 %) of a compound (C) represented by the following formula (C) as a yellow liquid.

5 ¹H-NMR (CDCl₃): 0.81-1.85 (m, 3H), 1.03-1.83 (m, 9H), 1.44 (s, 9H), 2.65-2.74 (td, 1H), 6.78-7.32 (m, 3H), 8.30, 8.33 (s, s, 1H), 14.19, 14.24 (s, s, 1H)



10

Into a 100 ml reactor thoroughly dried and purged with argon, 0.96 g (3.51 mmol) of the compound (C) and 20 ml of ether were introduced, and they were cooled to -78 °C and stirred. To the reactor, 2.41 ml of a n-hexane solution containing 3.86 mmol of n-butyllithium was dropwise added over a period of 10 minutes, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 2 hours to prepare a lithium salt solution. The solution was dropwise added to 20 ml of a

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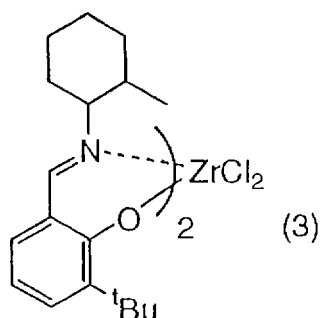
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-9 APR 2003

THF solution containing 0.66 g (1.75 mmol) of a
ZrCl₄(THF)₂ complex, said THF solution having been cooled
to -78 °C. After the dropwise addition was completed,
stirring was continued with slowly raising the
5 temperature to room temperature. Then, stirring was
further performed at room temperature for 15 hours, and
the solvent was distilled off from the reaction solution.
The resulting solid was washed with 50 ml of ether. The
filtrate was concentrated and recrystallized from a mixed
10 solution of methylene chloride and hexane to obtain 0.47
g (yield: 38 %) of a compound (3) represented by the
following formula (3) as a yellow powder.

¹H-NMR (CDCl₃): 0.69-1.90 (m, 24H), 1.62 (s, 18H),
3.65-3.90 (m, 2H), 6.86-7.64 (m, 6H), 8.14-8.27 (m, 2H)

15 FD-mass spectrometry: 706



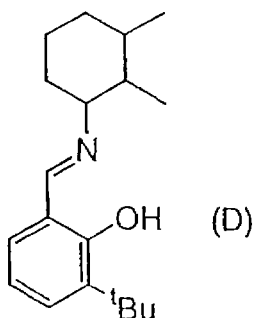
Synthesis Example 4

Into a 100 ml reactor thoroughly purged with
20 nitrogen, 50 ml of ethanol, 2.31 g (18.16 mmol) of 2,3-

-9 APR 2003

dimethylcyclohexylamine and 3.11 g (17.45 mmol) of 3-*t*-butylsalicylaldehyde were introduced, and they were stirred at room temperature for 24 hours. The reaction solution was vacuum concentrated to remove the solvent, whereby 4.97 g (yield: 99.1 %) of a compound (D) represented by the following formula (D) was obtained as a yellow liquid.

$^1\text{H-NMR}$ (CDCl_3): 0.75-1.00 (m, 6H), 1.00-1.95 (m, 8H), 1.44 (s, 9H), 2.73-3.30 (m, 1H), 6.77-7.32 (m, 3H), 8.30-8.36 (m, 1H), 14.20-14.33 (m, 1H)



Into a 100 ml reactor thoroughly dried and purged with argon, 1.20 g (4.18 mmol) of the compound (D) and 20 ml of ether were introduced, and they were cooled to -78 °C and stirred. To the reactor, 2.70 ml of a *n*-hexane solution containing 4.24 mmol of *n*-butyllithium was

-9 APR 2003

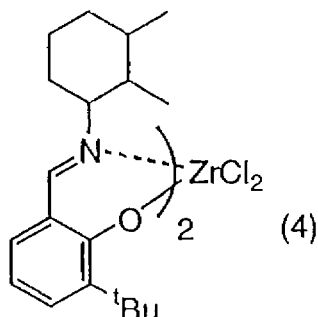
dropwise added over a period of 10 minutes, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 4 hours to prepare a lithium salt solution. The solution was dropwise added to 30 ml of a THF solution containing 0.79 g (2.09 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to $-78\text{ }^\circ\text{C}$. After the dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 15 hours, and the solvent was distilled off from the reaction solution. The resulting solid was washed with 30 ml of ether and 70 ml of methylene chloride. The filtrate was concentrated and washed again with a mixed solution of ether and hexane to obtain 0.37 g (yield: 24 %) of a compound (4) represented by the following formula (4) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.10-1.90 (m, 28H), 1.61 (s, 18H), 2.55-2.75 (m, 2H), 6.70-7.70 (m, 6H), 8.10-8.40 (m, 2H)

FD-mass spectrometry: 734

-9 APR 2003

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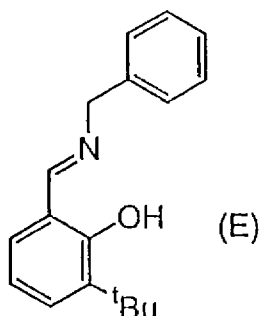
Synthesis Example 5

Into a 100 ml reactor thoroughly purged with
10 nitrogen, 40 ml of ethanol, 0.90 g (0.84 mmol) of
benzylamine and 1.50 g (8.42 mmol) of 3-*t*-
butylsalicylaldehyde were introduced, then a small amount
of molecular sieves was added with stirring at room
temperature, and the mixture was stirred at 65 °C for 4
15 hours. The reaction solution was filtered, and the
filtrate was vacuum concentrated. To the concentrate, 40
ml of cold MeOH was added, and the mixture was cooled at
-78 °C with stirring to obtain 1.80 g (yield: 80 %) of a
compound (E) represented by the following formula (E) as
20 a yellow solid.

¹H-NMR (CDCl₃): 1.45 (s, 9H), 4.82 (s, 2H), 6.78-7.45
(m, 8H), 8.47 (s, 1H), 13.89 (bs, 1H)

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-9 APR 2003



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Into a 100 ml reactor thoroughly dried and purged with argon, 0.50 g (1.87 mmol) of the compound (E) and 40 ml of ether were introduced, and they were cooled to -78 °C and stirred. To the reactor, 1.20 ml of a n-hexane solution containing 1.93 mmol of n-butyllithium was dropwise added over a period of 10 minutes, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 15 hours to prepare a lithium salt solution. The solution was dropwise added to 50 ml of a THF solution containing 0.352 g (1.87 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to -78 °C. After the dropwise addition was completed, the temperature of the system was naturally raised, and the solution was stirred all night at room temperature. After the solution was further stirred at 60 °C for 3 hours, the solvent was distilled off from the reaction solution. The resulting solid was reslurried in a mixed

25

-9 APR 2003

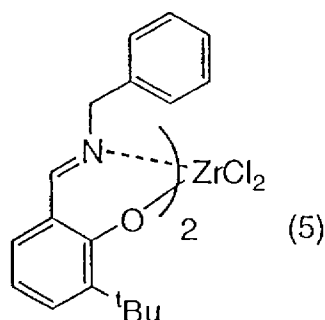
solvent of 100 ml of ether and 100 ml of hexane, and the slurry was filtered. The solid obtained by filtration was washed with 100 ml of hexane and vacuum concentrated to obtain 0.30 g (yield: 46 %) of a compound (5)

5 represented by the following formula (5) as a light yellowish white solid.

$^1\text{H-NMR}$ (CDCl_3): 1.60 (s, 18H), 4.65 (d, 2H), 4.95 (d, 2H), 6.70-7.70 (m, 16H), 7.85 (s, 2H)

FD-mass spectrometry: 694

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Synthesis Example 6

Into a 1 liter reactor thoroughly dried and purged with argon, 36.6 ml of an ether solution containing 110 mmol of ethylmagnesium bromide and 73.4 ml of ether were introduced. To the reactor, a solution containing 18.39 g (100 mmol) of 2-tert-butyl-4-methoxyphenol diluted with 90 ml of a THF was dropwise added at 0 °C over a period of 30 minutes, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 1 hour.

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-9 APR 2003

Then, 350 ml of toluene was added, and the system was heated to 100 °C to distill off about 180 ml of a mixed solution of ether and THF, whereby an opaque white slurry was obtained. After the slurry was cooled to 50 °C, 4.50 g (150 mmol) of paraformaldehyde and 21.0 ml (150 mmol) of triethylamine were added, followed by stirring at 80 to 90 °C for 1.5 hours. The reaction solution was allowed to cool to room temperature and then quenched with 10% hydrochloric acid. The organic layer was concentrated, and the solid precipitated was vacuum dried to obtain 11.66 g (yield: 56 %) of 3-t-butyl-5-methoxysalicylaldehyde.

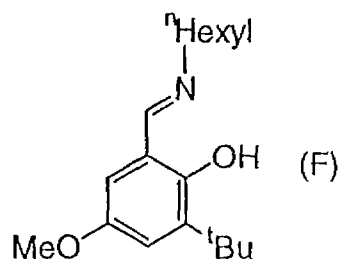
¹H-NMR (CDCl₃): 1.4 (s, 9H), 3.8 (s, 1H), 6.8 (s, 1H)-7.2 (d, 1H), 9.9 (s, 1H), 11.5 (s, 1H)

Into a 500 ml reactor thoroughly purged with nitrogen, 156 ml of ethanol, 2.53 g (25 mmol) of n-hexylamine and 5.20 g (25 mmol) of 3-t-butyl-5-methoxysalicylaldehyde were introduced, and they were stirred at room temperature for 24 hours. The reaction solution was vacuum concentrated to remove the solvent, whereby 6.96 g (yield: 97 %) of a compound (F) represented by the following formula (F) was obtained as a yellow crystal.

-9 APR 2003

$^1\text{H-NMR}$ (CDCl_3): 0.9 (t, 3H), 1.3 (s, 6H), 1.4 (s, 9H),
1.7-1.8 (m, 2H), 3.6 (t, 2H), 3.8 (s, 3H), 6.6 (s, 1H),
7.0 (s, 1H), 8.3 (s, 1H), 13.8 (s, 1H)

5



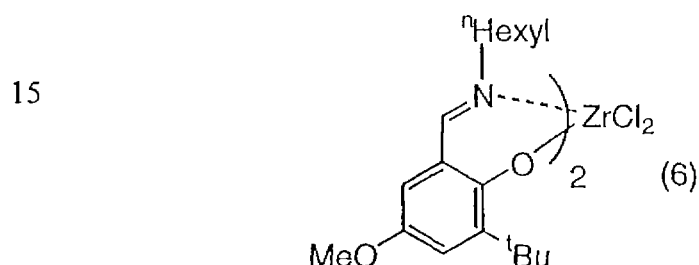
10 Into a 100 ml reactor thoroughly dried and purged
with argon, 1.17 g (4.0 mmol) of the compound (F) and 40
ml of ether were introduced, and they were cooled to -
78 °C and stirred. To the reactor, 2.61 ml of a n-hexane
solution containing 4.2 mmol of n-butyllithium was slowly
15 dropwise added, then the temperature of the system was
slowly raised to room temperature, and the reaction
solution was stirred at room temperature for 4 hours to
prepare a lithium salt solution. The solution was
dropwise added to 40 ml of a THF solution containing
20 0.755 g (2.0 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF
solution having been cooled to -78 °C. After the
dropwise addition was completed, stirring was continued
with slowly raising the temperature to room temperature.
Then, stirring was further performed at room temperature
25 for 15 hours, and the solvent was distilled off from the

-9 APR 2003

reaction solution. To the resulting solid, 20 ml of ether was added to give a slurry, and the slurry was filtered with washing with ether (10 ml $\times$ 2). The filtrate was concentrated until 10 ml of a concentrate was obtained, and the concentrate was allowed to stand one night at 0 °C to obtain 0.85 g (yield: 58 %) of a compound (6) represented by the following formula (6) as a yellowish orange crystal.

$^1\text{H-NMR}$ (CDCl_3): 0.9 (t, 6H), 1.0-1.4 (s, 12H), 1.6 (s, 18H), 3.6 (t, 4H), 3.8 (s, 6H), 6.7 (s, 2H), 7.2 (s, 2H), 8.2 (s, 1H)

FD-mass spectrometry: 742



Synthesis Example 7

20 Into a 200 ml reactor thoroughly purged with nitrogen, 50 ml of ethanol, 1.23 g (12.4 mmol) of cyclohexylamine and 2.55 g (12.2 mmol) of 3-*t*-butyl-5-methoxysalicylaldehyde were introduced, and they were stirred at room temperature for 24 hours. The solid
25 precipitated was filtered off, washed with ethanol and

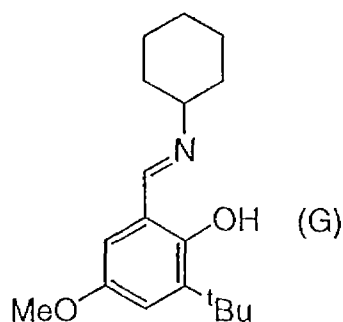
-9 APR 2003

vacuum dried to obtain 2.34 g (yield: 66 %) of a compound (G) represented by the following formula (G) as a yellowish orange crystal.

$^1\text{H-NMR}$ (CDCl_3): 1.20-1.85 (m, 19H), 3.18-3.23 (m, 1H),
5 3.77 (s, 3H), 6.59-6.60 (d, 1H), 6.95-6.96 (d, 1H), 8.33
(s, 1H), 13.75 (s, 1H)

FD-mass spectrometry: 289

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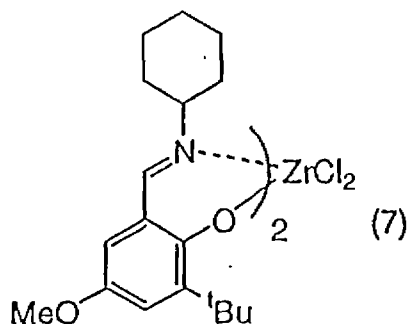
Into a 100 ml reactor thoroughly dried and purged
15 with argon, 0.86 g (2.99 mmol) of the compound (G) and 20
ml of ether were introduced, and they were cooled to -
78 °C and stirred. To the reactor, 2.15 ml of a n-hexane
solution containing 3.31 mmol of n-butyllithium was
slowly dropwise added, then the temperature of the system
20 was slowly raised to room temperature, and the reaction
solution was stirred at room temperature for 1 hour to
prepare a lithium salt solution. The solution was
dropwise added to 20 ml of a THF solution containing
0.566 g (1.50 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF
25 solution having been cooled to -78 °C. After the

-9 APR 2003

dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 15 hours, and the solvent was distilled off from the reaction solution. The resulting solid was washed with 100 ml of ether and 90 ml of methylene chloride. The filtrate was concentrated to obtain 0.27 g (yield: 25 %) of a compound (7) represented by the following formula (7) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.98-2.12 (m, 38H), 3.46-3.87 (m, 8H), 6.63-6.64 (d, 2H), 7.19-7.20 (d, 2H), 8.20 (s, 2H)

FD-mass spectrometry: 738

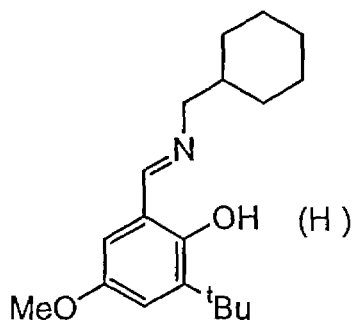


-9 APR 2003

Synthesis Example 8

Into a 100 ml reactor thoroughly purged with nitrogen, 30 ml of ethanol, 1.05 g (9.27 mmol) of cyclohexanemethylamine and 1.92 g (9.23 mmol) of 3-t-butyl-5-methoxysalicylaldehyde were introduced, and they were stirred at room temperature for 73 hours. The solid precipitated was filtered off, washed with ethanol and vacuum dried to obtain 2.70 g (yield: 96 %) of a compound (H) represented by the following formula (H) as an orange liquid.

$^1\text{H-NMR}$ (CDCl_3): 0.93-1.86 (m, 11H), 1.43 (s, 9H), 3.42 (d, 2H), 3.77 (s, 3H), 6.60 (d, 1H), 6.96 (d, 1H), 8.25 (s, 1H), 13.71 (bs, 1H)



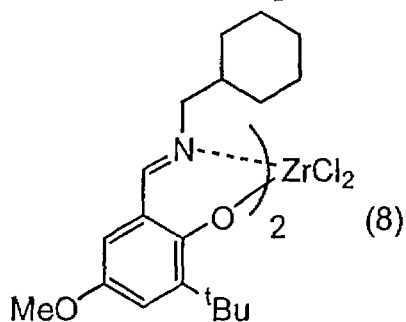
Into a 100 ml reactor thoroughly dried and purged with argon, 0.84 g (2.57 mmol) of the compound (H) and 20 ml of ether were introduced, and they were cooled to -78°C and stirred. To the reactor, 1.90 ml of a n-hexane solution containing 3.04 mmol of n-butyllithium was slowly dropwise added, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 1.5 hours to prepare a lithium salt solution. The solution was

-9 APR 2003

dropwise added to 20 ml of a THF solution containing 0.52 g (1.34 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to $-78\text{ }^\circ\text{C}$. After the dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 15 hours, and the solvent was distilled off from the reaction solution. The resulting solid was washed with 40 ml of ether and 100 ml of methylene chloride. The filtrate was concentrated and washed again with a mixture of ether and hexane to obtain 0.67 g (yield: 63 %) of a compound (8) represented by the following formula (8) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.53-1.85 (m, 22H), 1.55 (s, 18H), 3.22-3.40 (m, 4H), 3.80 (s, 6H), 6.64-6.67 (m, 2H), 7.10-7.26 (m, 2H), 7.96 (s, 2H)

FD-mass spectrometry: 766

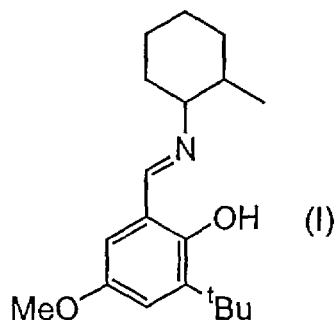


-9 APR 2003

Synthesis Example 9

Into a 100 ml reactor thoroughly purged with nitrogen, 30 ml of ethanol, 1.72 g (15.19 mmol) of 2-methylcyclohexylamine and 2.64 g (12.68 mmol) of 3-*t*-butyl-5-methoxysalicylaldehyde were introduced, and they were stirred at room temperature for 24 hours. The solid precipitated was filtered off, washed with ethanol and vacuum dried to obtain 2.82 g (yield: 73 %) of a compound (I) represented by the following formula (I) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.83 (d, 3H), 1.00-1.89 (m, 9H), 1.43 (s, 9H), 2.64-2.73 (td, 1H), 3.78 (s, 3H), 6.60 (d, 1H), 6.96 (d, 1H), 8.27 (s, 1H), 13.7 (s, 1H)



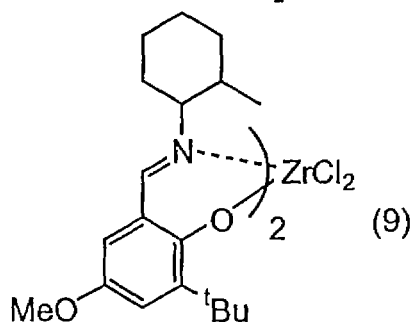
Into a 100 ml reactor thoroughly dried and purged with argon, 0.91 g (3.00 mmol) of the compound (I) and 20 ml of ether were introduced, and they were cooled to -78 °C and stirred. To the reactor, 2.10 ml of a *n*-hexane solution containing 3.36 mmol of *n*-butyllithium was slowly dropwise added, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 2 hours to prepare a lithium salt solution. The solution was

-9 APR 2003

dropwise added to 20 ml of a THF solution containing 0.57 g (1.51 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to $-78\text{ }^\circ\text{C}$. After the dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 15 hours, and the solvent was distilled off from the reaction solution. The resulting solid was washed with 10 ml of ether and 60 ml of methylene chloride. The filtrate was concentrated and washed again with a mixture of ether and hexane to obtain 0.54 g (yield: 46 %) of a compound (9) represented by the following formula (9) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.60-2.90 (m, 24H), 1.60 (s, 18H), 2.50-2.70 (m, 2H), 3.47 (m, 6H), 6.61-6.64 (m, 2H), 7.21, 7.22 (s, s, 2H), 8.15-8.23 (m, 2H)

FD-mass spectrometry: 766



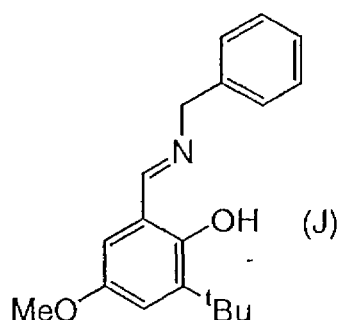
Synthesis Example 10

-9 APR 2003

Into a 100 ml reactor thoroughly purged with nitrogen, 40 ml of ethanol, 1.61 g (15.0 mmol) of benzylamine and 3.12 g (15.0 mmol) of 3-*t*-butyl-5-methoxysalicylaldehyde were introduced, and they were stirred at room temperature for 1.5 hours. The solid precipitated was filtered off, washed with methanol and vacuum dried to obtain 3.25 g (yield: 73 %) of a compound (J) represented by the following formula (J) as a yellowish orange crystal.

¹H-NMR (CDCl₃): 1.42 (s, 9H), 3.78 (s, 3H), 4.80 (s, 2H), 6.62-7.43 (m, 7H), 8.41 (s, 1H), 13.52 (s, 1H)

15.



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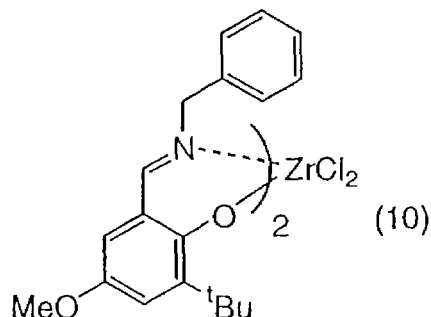
Into a 50 ml reactor thoroughly dried and purged with argon, 0.89 g (3.00 mmol) of the compound (J) and 30 ml of ether were introduced, and they were cooled to -78 °C and stirred. To the reactor, 2.01 ml of a *n*-hexane solution containing 3.15 mmol of *n*-butyllithium was slowly dropwise added, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 3 hours to

-9 APR 2003

prepare a lithium salt solution. The solution was dropwise added to 30 ml of a THF solution containing 0.57 g (1.50 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to $-78\text{ }^\circ\text{C}$. After the dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 15 hours, and the solvent was distilled off from the reaction solution. The resulting solid was washed with 30 ml of ether and 20 ml of methylene chloride. The filtrate was concentrated and washed again with 20 ml of ether to obtain 0.58 g (yield: 51 %) of a compound (10) represented by the following formula (10) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 1.51 (s, 18H), 3.75 (s, 6H), 4.60-5.10 (m, 4H), 6.35-7.25 (m, 14H), 7.82 (s, 2H)

FD-mass spectrometry: 754



Synthesis Example 11

-9 APR 2003

Into a 1 liter reactor thoroughly dried and purged with nitrogen, 9.68 g (58.93 mmol) of 2-tert-butyl-4-methylphenol and 100 ml of THF were introduced. To the reactor, 23.00 ml of an ether solution containing 69.00 mmol of ethylmagnesium bromide was dropwise added at 0 °C over a period of 30 minutes, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 1 hour. Then, 100 ml of toluene was added, and the system was heated to 95 °C to distill off a mixed solution of ether and THF, whereby an opaque white slurry was obtained. After the slurry was cooled to room temperature, 100 ml of toluene, 4.50 g (149.90 mmol) of paraformaldehyde and 12.50 ml (89.93 mmol) of triethylamine were added, followed by stirring at 95 °C for 2 hours. The reaction solution was allowed to cool to room temperature and then quenched with 300 ml of 1N hydrochloric acid. The organic layer was concentrated and purified by a silica gel column to obtain 7.36 g (yield: 65 %) of 3-t-butyl-5-methylsalicylaldehyde.

¹H-NMR (CDCl₃): 1.41 (s, 9H), 2.32 (s, 3H), 7.19 (d, 1H), 7.33 (d, 1H), 9.83 (s, 1H), 11.60 (s, 1H)

Into a 200 ml reactor thoroughly purged with nitrogen, 50 ml of ethanol, 1.52 g (15.02 mmol) of n-hexylamine and 2.86 g (14.90 mmol) of 3-t-butyl-5-

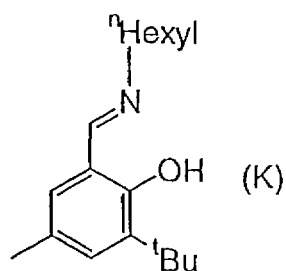
-9 APR 2003

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methylsalicylaldehyde were introduced, and they were stirred at room temperature for 24 hours. The reaction solution was vacuum concentrated to remove the solvent, whereby 4.14 g (yield: 100 %) of a compound (K)

5 represented by the following formula (K) was obtained as a yellow liquid.

$^1\text{H-NMR}$ (CDCl_3): 0.89 (t, 3H), 1.25-1.43 (m, 4H), 1.43 (s, 9H), 1.60-1.77 (m, 2H), 2.28 (s, 3H), 3.56 (t, 2H), 6.89 (s, 1H), 7.11 (d, 1H), 8.27 (s, 1H), 13.94 (s, 1H)



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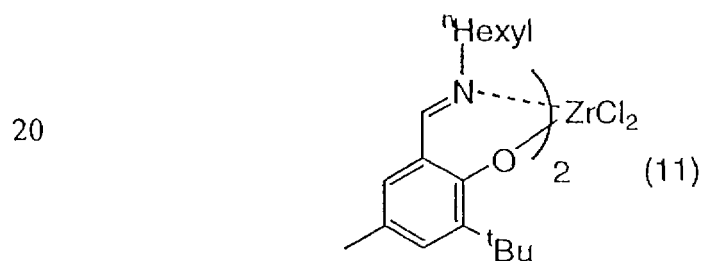
Into a 300 ml reactor thoroughly dried and purged with argon, 4.16 g (15.10 mmol) of the compound (K) and 70 ml of ether were introduced, and they were cooled to -78 °C and stirred. To the reactor, 9.40 ml of a n-hexane solution containing 15.04 mmol of n-butyllithium was dropwise added over a period of 30 minutes, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 4 hours to prepare a lithium salt solution. The solution was dropwise added to 80 ml of a

-9 APR 2003

THF solution containing 2.85 g (7.56 mmol) of a
ZrCl₄(THF)₂ complex, said THF solution having been cooled
to -78 °C. After the dropwise addition was completed,
stirring was continued with slowly raising the
5 temperature to room temperature. Then, stirring was
further performed at room temperature for 15 hours, and
the solvent was distilled off from the reaction solution.
The resulting solid was washed with 50 ml of ether and
200 ml of methylene chloride. The filtrate was
10 concentrated and washed again with 20 ml of ether to
obtain 4.30 g (yield: 80 %) of a compound (11)
represented by the following formula (11) as a yellow
powder.

¹H-NMR (CDCl₃): 0.74-1.54 (m, 18H), 1.55 (s, 18H),
15 2.31 (s, 6H), 3.37-3.68 (m, 4H), 6.99 (s, 2H), 7.36 (s,
2H), 8.09 (s, 2H)

FD-mass spectrometry: 710

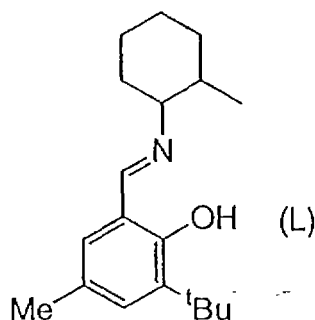


25 Synthesis Example 12

-9 APR 2003

Into a 200 ml reactor thoroughly purged with nitrogen, 70 ml of ethanol, 1.83 g (16.17 mmol) of 2-methylcyclohexylamine and 3.06 g (15.92 mmol) of 3-*t*-butyl-5-methylsalicylaldehyde were introduced, and they
5 were stirred at room temperature for 24 hours. The solid precipitated was filtered off and vacuum dried to obtain 3.00 g (yield: 66 %) of a compound (L) represented by the following formula (L) as a yellow crystal.

¹H-NMR (CDCl₃): 0.79-0.83 (m, 3H), 0.96-1.88 (m, 9H),
10 1.43 (s, 9H), 2.28 (s, 3H), 2.63-2.72 (td, 1H), 6.90 (d, 1H), 7.11 (d, 1H), 8.26 (s, 1H), 13.93 (s, 1H)



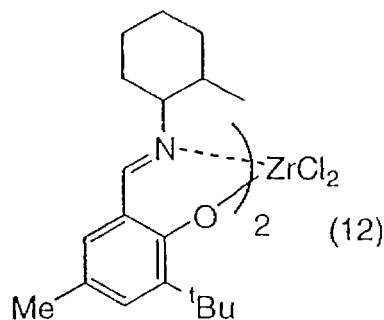
Into a 100 ml reactor thoroughly dried and purged with argon, 0.87 g (3.03 mmol) of the compound (L) and 20
15 ml of ether were introduced, and they were cooled to -78 °C and stirred. To the reactor, 1.90 ml of a *n*-hexane solution containing 2.98 mmol of *n*-butyllithium was dropwise added over a period of 10 minutes, then the temperature of the system was slowly raised to room
20 temperature, and the reaction solution was stirred at

-9 APR 2003

room temperature for 2 hours to prepare a lithium salt solution. The solution was dropwise added to 20 ml of a THF solution containing 0.57 g (1.50 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to -78 °C. After the dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 15 hours, and the solvent was distilled off from the reaction solution. The resulting solid was washed with 80 ml of ether. The filtrate was concentrated and recrystallized from a mixed solution of ether and hexane to obtain 0.44 g (yield: 40 %) of a compound (12) represented by the following formula (12) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.65-1.90 (m, 24H), 1.60 (s, 18H), 2.32 (s, 6H), 3.68-3.90 (m, 2H), 6.98 (s, 2H), 7.37 (s, 2H), 8.14-8.22 (m, 2H)

FD-mass spectrometry: 734



-9 APR 2003

Synthesis Example 13

Into a 200 ml reactor thoroughly dried and purged with nitrogen, 34.15 g (316.0 mmol) of paracresol, 2.50 g of Umberist-15E (trade name, available from Organo Co.) and 20 ml of toluene were introduced. To the reactor, a solution containing 14.40 g (105.0 mmol) of 4-cumylphenol diluted with 30 ml of a toluene was dropwise added at 80 °C, followed by stirring for 17 hours. After the system was cooled to room temperature, the reaction solution was filtered with washing with hexane. The resulting liquid was purified by silica gel column chromatography to obtain 10.52 g (yield: 44 %) of 2-cumyl-4-methylphenol.

¹H-NMR (CDCl₃): 1.75 (s, 6H), 2.40 (s, 3H), 7.10-7.30 (m, 8H), 11.17 (s, 1H)

Into a 500 ml reactor thoroughly purged with nitrogen, 21.0 ml of an ether solution containing 63.1 mmol of ethylmagnesium bromide and 40 ml of THF were introduced. To the reactor, a solution containing 13.61 g (60.1 mmol) of 2-cumyl-4-methylphenol diluted with 20 ml of a THF was dropwise added at 0 °C over a period of 1 hour, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 30 minutes. Then, 220 ml of toluene was added, and the system was heated to 100 °C

-9 APR 2003

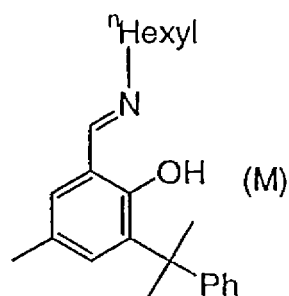
to distill off about 50 ml of a mixed solution of ether and THF, whereby an opaque white slurry was obtained. After the slurry was cooled to 24 °C, 4.37 g (145.5 mmol) of paraformaldehyde and 12.0 ml (86.0 mmol) of triethylamine were added, followed by stirring at 90 °C for 1 hour. The reaction solution was allowed to cool to room temperature and then quenched with 42 ml of 18% hydrochloric acid. The organic layer was concentrated, and the resulting liquid was purified by silica gel column chromatography to obtain 14.13 g (yield: 92 %) of 3-cumyl-5-methylsalicylaldehyde.

¹H-NMR (CDCl₃): 1.75 (s, 6H), 2.40 (s, 3H), 7.10-7.42 (m, 7H), 8.55 (s, 1H), 13.18 (s, 1H)

Into a 100 ml reactor thoroughly purged with nitrogen, 40 ml of ethanol, 1.82 g (18.0 mmol) of n-hexylamine and 3.81 g (15.0 mmol) of 3-cumyl-5-methylsalicylaldehyde were introduced, and they were stirred at room temperature for 3 hours. The reaction solution was vacuum concentrated to remove the solvent, and the resulting liquid was purified by silica gel column chromatography to obtain 3.97 g (yield: 78 %) of a compound (M) represented by the following formula (M).

¹H-NMR (CDCl₃): 0.85 (t, 3H), 1.27 (s, 6H), 1.52-1.70 (m, 2H), 1.71 (s, 6H), 2.34 (s, 3H), 3.45 (t, 2H), 6.92-7.35 (m, 7H), 8.22 (s, 1H), 13.49 (s, 1H)

-9 APR 2003



5

Into a 50 ml reactor thoroughly dried and purged with argon, 1.01 g (3.00 mmol) of the compound (M) and 30 ml of ether were introduced, and they were cooled to - 78 °C and stirred. To the reactor, 2.20 ml of a n-hexane solution containing 3.45 mmol of n-butyllithium was slowly dropwise added, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 4 hours to prepare a lithium salt solution. The solution was dropwise added to 30 ml of a THF solution containing 0.622 g (1.65 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to -78 °C. After the dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 15 hours, and the solvent was distilled off from the reaction solution. To the resulting solid, 20 ml of ether was added to give a slurry, and the slurry was filtered with washing with 10 ml of ether. The filtrate

25

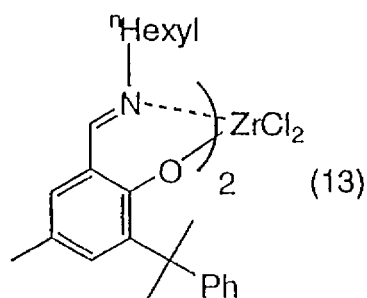
-9 APR 2003

was concentrated and washed again with a mixed solution of hexane and ether to obtain 0.53 g (yield: 42 %) of a compound (13) represented by the following formula (13) as a light yellow powder.

5 $^1\text{H-NMR}$ (CDCl_3): 0.50-2.00 (m, 34H), 2.25-2.45 (m, 6H), 2.50-2.75 (m, 4H), 6.90-7.55 (m, 14H), 8.85 (s, 2H)

FD-mass spectrometry: 832

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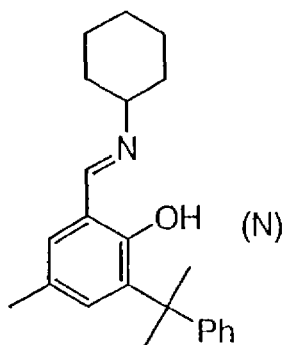


Synthesis Example 14

15 Into a 100 ml reactor thoroughly purged with nitrogen, 40 ml of ethanol, 0.99 g (10.0 mmol) of cyclohexylamine and 2.54 g (10.0 mmol) of 3-cumyl-5-methylsalicylaldehyde were introduced, and they were stirred at room temperature for 20 hours. The solid
20 precipitated was filtered off, washed with cold methanol and vacuum dried to obtain 2.85 g (yield: 85 %) of a compound (N) represented by the following formula (N) as a light yellow crystal.

-9 APR 2003

$^1\text{H-NMR}$ (CDCl_3): 1.10-1.65 (m, 6H), 1.65-1.82 (m, 4H),
1.75 (s, 6H), 2.35 (s, 3H), 3.12 (m, 1H), 6.95-7.33 (m,
7H), 8.28 (s, 1H), 13.42 (s, 1H)



10

Into a 50 ml reactor thoroughly dried and purged
with argon, 1.01 g (3.00 mmol) of the compound (N) and 30
ml of ether were introduced, and they were cooled to -
78 °C and stirred. To the reactor, 2.01 ml of a n-hexane
15 solution containing 3.15 mmol of n-butyllithium was
slowly dropwise added, then the temperature of the system
was slowly raised to room temperature, and the reaction
solution was stirred at room temperature for 4 hours to
prepare a lithium salt solution. The solution was
20 dropwise added to 30 ml of a THF solution containing
0.566 g (1.50 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF
solution having been cooled to -78 °C. After the
dropwise addition was completed, stirring was continued
with slowly raising the temperature to room temperature.
25 Then, stirring was further performed at room temperature

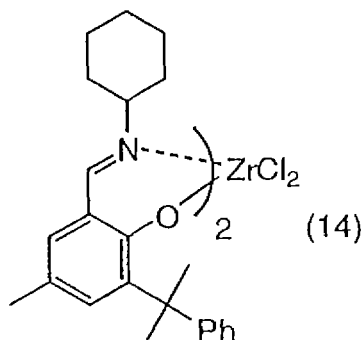
-9 APR 2003

for 15 hours, and the solvent was distilled off from the reaction solution. To the resulting solid, 30 ml of ether was added to give a slurry, and the slurry was filtered with washing with ether (10 ml $\times$ 2). The filtrate was concentrated and washed again with a mixed solution of hexane and ether to obtain 0.91 g (yield: 73 %) of a compound (14) represented by the following formula (14) as a light yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.85-2.10 (m, 32H), 2.28, 2.38 (s, s, 6H), 3.62 (m, 2H), 6.95-7.50 (m, 14H), 8.10 (s, 2H)

FD-mass spectrometry: 830

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Synthesis Example 15

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Into a 100 ml reactor thoroughly purged with nitrogen, 30 ml of ethanol, 0.91 g (8.04 mmol) of 2-methylcyclohexylamine and 1.96 g (7.71 mmol) of 3-cumyl-5-methylsalicylaldehyde were introduced, and they were stirred at room temperature for 15 hours. The solid precipitated was filtered off and vacuum dried to obtain

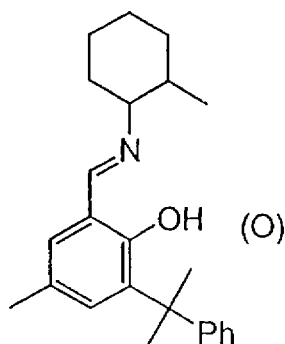
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-9 APR 2003

2.65 g (yield: 98 %) of a compound (O) represented by the following formula (O) as a yellow crystal.

$^1\text{H-NMR}$ (CDCl_3): 0.70-0.80 (m, 3H), 0.85-1.90 (m, 15H),
2.33, 2.34 (s, s, 3H), 2.60, 3.23 (m, 1H), 6.95-7.30 (m,
5 7H), 8.20-8.30 (m, 1H), 13.35 (s, 1H)

10



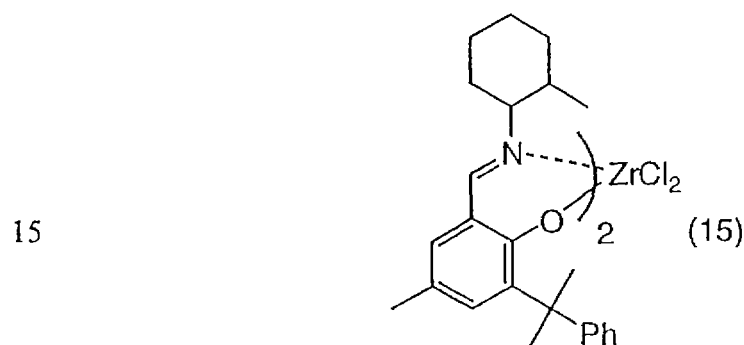
Into a 100 ml reactor thoroughly dried and purged with argon, 1.08 g (3.08 mmol) of the compound (O) and 20 ml of ether were introduced, and they were cooled to -
15 78 °C and stirred. To the reactor, 2.00 ml of a n-hexane solution containing 3.14 mmol of n-butyllithium was slowly dropwise added, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 1 hour to
20 prepare a lithium salt solution. The solution was dropwise added to 20 ml of a THF solution containing 0.58 g (1.54 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to -78 °C. After the dropwise addition was completed, stirring was continued with
25 slowly raising the temperature to room temperature. Then,

-9 APR 2003

stirring was further performed at room temperature for 20 hours, and the solvent was distilled off from the reaction solution. To the resulting solid, 30 ml of ether and 50 ml of methylene chloride were added, and the mixture was filtered. The filtrate was concentrated and recrystallized from ether to obtain 0.39 g (yield: 30 %) of a compound (15) represented by the following formula (15) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.20-2.50 (m, 42H), 3.38-3.65 (m, 2H), 6.90-7.70 (m, 14H), 8.05-8.25 (m, 2H)

FD-mass spectrometry: 858



Synthesis Example 16

Into a 100 ml reactor thoroughly dried and purged with nitrogen, 11.22 g (46.30 mmol) of 2-(1-adamantyl)-4-methylphenol and 100 ml of THF were introduced. To the reactor, 16.20 ml of an ether solution containing 48.60 mmol of ethylmagnesium bromide was dropwise added at 0 °C over a period of 30 minutes, then the temperature of the system was slowly raised to room temperature, and the

-9 APR 2003

reaction solution was stirred at room temperature for 2.5 hours. Then, 300 ml of toluene was added, and the system was heated to 100 °C to give an opaque white slurry.

After the slurry was cooled to room temperature, 3.80 g

5 (126.54 mmol) of paraformaldehyde and 7.10 ml (70.17

mmol) of triethylamine were added, followed by stirring

at 85 °C for 30 minutes. The reaction solution was

allowed to cool to room temperature and then quenched

with 10% hydrochloric acid. The organic layer was

10 concentrated, and the solid precipitated was vacuum dried

to obtain 10.51 g (yield: 84 %) of 3-(1-adamantyl)-5-

methylnsalicylaldehyde.

¹H-NMR (CDCl₃): 2.2-1.8 (s, 16H), 2.3 (s, 3H), 7.5-
7.0 (m, 2H), 9.8 (s, 1H), 11.6 (s, 1H)

15 Into a 100 ml reactor thoroughly purged with

nitrogen, 30 ml of ethanol, 0.55 g (5.55 mmol) of

cyclohexylamine and 1.51 g (5.57 mmol) of 3-(1-

adamantyl)-5-methylnsalicylaldehyde were introduced, and

they were stirred at room temperature for 18 hours. The

20 solid precipitated was filtered off and vacuum dried to

obtain 1.73 g (yield: 89 %) of a compound (P) represented

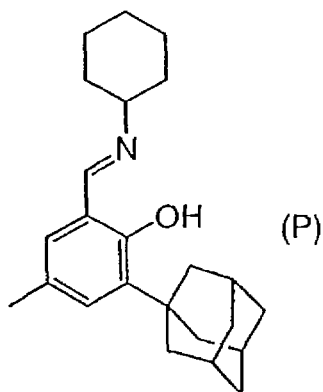
by the following formula (P) as a yellow crystal.

¹H-NMR (CDCl₃): 1.18-1.70 (m, 10H), 1.79 (t, 6H),
2.11 (s, 3H), 2.30 (s, 6H), 2.31 (s, 3H), 3.12-3.28 (m,

-9 APR 2003

1H), 6.97 (d, 1H), 7.02 (s, 1H), 8.17 (s, 1H), 13.98 (s, 1H)

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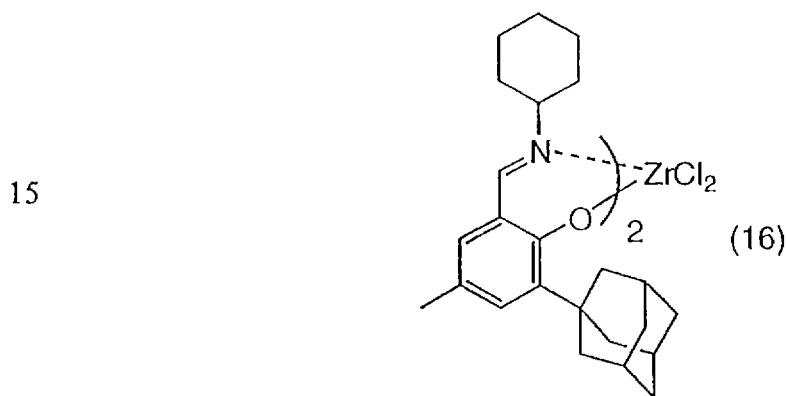
10 Into a 100 ml reactor thoroughly dried and purged
with argon, 1.01 g (2.88 mmol) of the compound (P) and 20
ml of THF were introduced, and they were cooled to -78 °C
and stirred. To the reactor, 1.83 ml of a n-hexane
solution containing 2.87 mmol of n-butyllithium was
15 dropwise added, then the temperature of the system was
slowly raised to room temperature, and the reaction
solution was stirred at room temperature for 1 hour to
prepare a lithium salt solution. The solution was
dropwise added to 20 ml of a THF solution containing 0.54
20 g (1.44 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution
having been cooled to -78 °C. After the dropwise
addition was completed, stirring was continued with
slowly raising the temperature to room temperature. Then,
stirring was further performed at room temperature for 15
25 hours, and the solvent was distilled off from the

-9 APR 2003

reaction solution. To the resulting solid, 30 ml of ether and 70 ml of methylene chloride were added to give a slurry, and the slurry was filtered. The filtrate was concentrated and recrystallized from a mixed solution of hexane and ether to obtain 0.99 g (yield: 79 %) of a compound (16) represented by the following formula (16) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.70-2.20 (m, 50H), 2.27 (s, 6H), 3.80-3.95 (m, 2H), 6.87 (d, 2H), 7.04 (d, 2H), 8.31 (s, 2H)

FD-mass spectrometry: 862



20 Synthesis Example 17

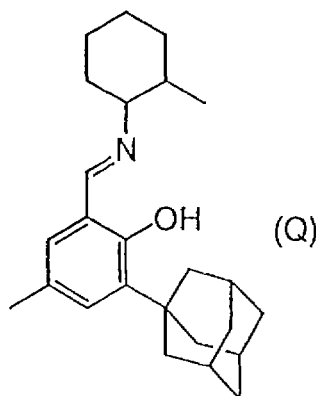
Into a 100 ml reactor thoroughly purged with nitrogen, 30 ml of ethanol, 0.67 g (5.92 mmol) of 2-methylcyclohexylamine and 1.50 g (5.55 mmol) of 3-(1-adamantyl)-5-methylsalicylaldehyde were introduced, and they were stirred at room temperature for 18 hours. The

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-9 APR 2003

solid precipitated was filtered off and vacuum dried to obtain 1.67 g (yield: 82 %) of a compound (Q) represented by the following formula (Q) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.81 (d, 3H), 0.95-1.75 (m, 9H), 1.79 (d, 6H), 2.08 (s, 3H), 2.18 (d, 6H), 2.28 (s, 3H), 2.60-2.75 (td, 1H), 6.89 (s, 1H), 7.05 (d, 1H), 8.25, 8.28 (s, s, 1H), 13.88, 13.90 (s, s, 1H)



Into a 100 ml reactor thoroughly dried and purged with argon, 1.00 g (2.74 mmol) of the compound (Q) and 20 ml of THF were introduced, and they were cooled to $-78\text{ }^{\circ}\text{C}$ and stirred. To the reactor, 1.75 ml of a n-hexane solution containing 2.75 mmol of n-butyllithium was slowly dropwise added, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 1.5 hours to prepare a lithium salt solution. The solution was dropwise added to 20 ml of a THF solution containing 0.52 g (1.37 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution

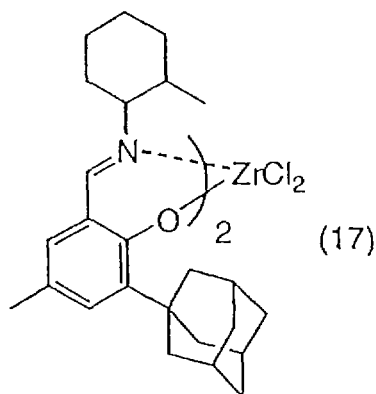
-9 APR 2003

having been cooled to $-78\text{ }^{\circ}\text{C}$. After the dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 20
5 hours, and the solvent was distilled off from the reaction solution. To the resulting solid, 30 ml of ether and 70 ml of methylene chloride were added to give a slurry, and the slurry was filtered. The filtrate was concentrated and recrystallized from a mixed solution of
10 hexane and ether to obtain 0.50 g (yield: 41 %) of a compound (17) represented by the following formula (17) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.60-2.20 (m, 54H), 2.32 (s, 6H),
3.70-3.92 (m, 2H), 6.90-7.00 (m, 2H), 7.26, 7.31 (s, s,
15 2H), 8.00-8.30 (m, 2H)

FD-mass spectrometry: 890

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-9 APR 2003

Synthesis Example 18

Into a 200 ml reactor thoroughly dried and purged with nitrogen, 25.31 g (203.90 mmol) of anisole, 2.00 g of Umberist-15 (trade name, available from Organo Co.) and 30 ml of toluene were introduced. To the reactor, a solution containing 15.52 g (101.9 mmol) of 1-adamantyl alcohol diluted with 20 ml of a toluene was dropwise added at 95 °C, followed by stirring for 5.5 hours.

After the system was cooled to room temperature, the crystal precipitated was washed with hexane and vacuum dried to obtain 12.20 g (yield: 46 %) of 2-(1-adamantyl)-4-methoxyphenol.

¹H-NMR (CDCl₃): 1.78 (s, 6H), 2.09 (s, 3H), 2.12 (s, 6H), 3.76 (s, 3H), 4.30 (bs, 1H), 6.58 (s, 2H), 6.83 (s, 1H)

Into a 500 ml reactor thoroughly purged with nitrogen, 16.50 ml of an ether solution containing 49.60 mmol of ethylmagnesium bromide and 10 ml of THF were introduced. To the reactor, a solution containing 12.31 g (47.20 mmol) of 2-(1-adamantyl)-4-methoxyphenol diluted with 50 ml of a THF was dropwise added at 0 °C over a period of 15 minutes. Then, 150 ml of toluene was added, and the system was heated to 95 °C to distill off about 40 ml of a mixed solution of ether and THF, whereby an

-9 APR 2003

opaque white slurry was obtained. After the slurry was cooled to 20 °C, 3.43 g (114.30 mmol) of paraformaldehyde and 9.40 ml (67.50 mmol) of triethylamine were added, followed by stirring at 90 °C for 30 minutes. The

5 reaction solution was allowed to cool to room temperature and then quenched with 32 ml of 18% hydrochloric acid.

The organic layer was washed with 100 ml of water, 50 ml of a sodium hydrogencarbonate aqueous solution and 100 ml of a sodium chloride aqueous solution, and then

10 concentrated. The resulting solid was purified by silica gel column chromatography to obtain 4.95 g (yield: 37 %) of 3-(1-adamantyl)-5-methoxysalicylaldehyde.

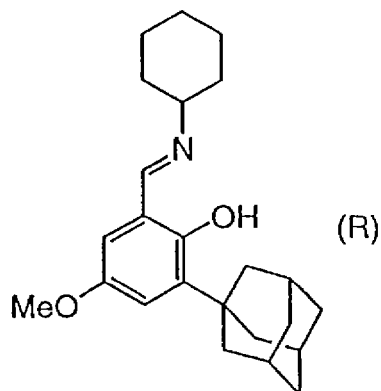
¹H-NMR (CDCl₃): 1.78 (s, 6H), 2.09 (s, 3H), 2.12 (s, 6H), 3.82 (s, 3H), 6.80 (s, 1H), 7.13 (s, 1H), 9.87 (s, 15 1H), 11.57 (s, 1H)

Into a 100 ml reactor thoroughly purged with nitrogen, 30 ml of ethanol, 0.37 g (3.73 mmol) of cyclohexylamine and 1.03 g (3.59 mmol) of 3-(1-adamantyl)-5-methoxysalicylaldehyde were introduced, and 20 they were stirred at room temperature for 5.5 hours. The solid precipitated was filtered off and vacuum dried to obtain 1.24 g (yield: 94 %) of a compound (R) represented by the following formula (R).

¹H-NMR (CDCl₃): 1.20-1.80 (m, 10H), 1.80 (s, 6H), 25 2.08 (s, 3H), 2.15 (s, 6H), 3.15-3.30 (m, 1H), 3.77 (s,

-9 APR 2003

3H), 6.58 (d, 1H), 6.89 (d, 1H), 8.32 (s, 1H), 13.76 (s, 1H)



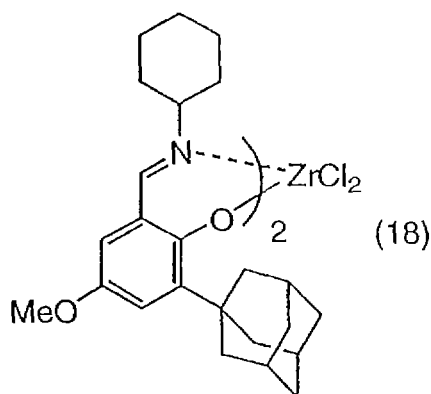
10 Into a 50 ml reactor thoroughly dried and purged
with argon, 1.00 g (2.72 mmol) of the compound (R) and 10
ml of THF were introduced, and they were cooled to -78 °C
and stirred. To the reactor, 1.80 ml of a n-hexane
solution containing 2.83 mmol of n-butyllithium was
15 slowly dropwise added, then the temperature of the system
was slowly raised to room temperature, and the reaction
solution was stirred at room temperature for 1.5 hours to
prepare a lithium salt solution. The solution was
dropwise added to 20 ml of a THF solution containing 0.51
20 g (1.36 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution
having been cooled to -78 °C. After the dropwise
addition was completed, stirring was continued with
slowly raising the temperature to room temperature. Then,
stirring was further performed at room temperature for 20
25 hours, and the solvent was distilled off from the

-9 APR 2003

reaction solution. To the resulting solid, 30 ml of ether and 70 ml of methylene chloride were added, and the mixture was filtered. The filtrate was concentrated and washed again with a mixed solution of ether and hexane to obtain 0.16 g (yield: 13 %) of a compound (18) represented by the following formula (18) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.50-2.40 (m, 50H), 3.80 (s, 6H), 3.80-4.00 (m, 2H), 6.62 (d, 2H), 7.14 (d, 2H), 8.18 (s, 2H)

FD-mass spectrometry: 894



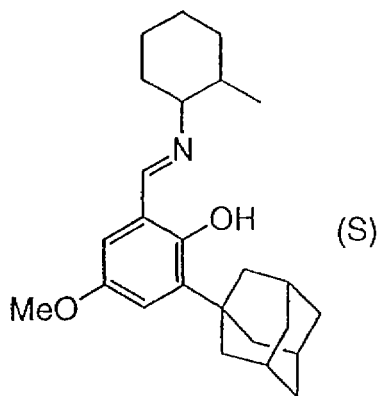
Synthesis Example 19

Into a 100 ml reactor thoroughly purged with nitrogen, 30 ml of ethanol, 0.44 g (3.89 mmol) of 2-methylcyclohexylamine and 1.06 g (3.70 mmol) of 3-(1-adamantyl)-5-methoxysalicylaldehyde were introduced, and they were stirred at room temperature for 5 hours. The

-9 APR 2003

solid precipitated was filtered off and vacuum dried to obtain 1.27 g (yield: 90 %) of a compound (S) represented by the following formula (S).

$^1\text{H-NMR}$ (CDCl_3): 0.82 (d, 3H), 1.00-1.80 (m, 9H), 1.79 (d, 6H), 2.07 (s, 3H), 2.17 (d, H), 2.65-2.75 (m, 1H), 3.77 (s, 3H), 6.59 (d, 1H), 6.90 (d, 1H), 8.27, 8.30 (s, s, 1H), 13.66, 13.88 (s, s, 1H)



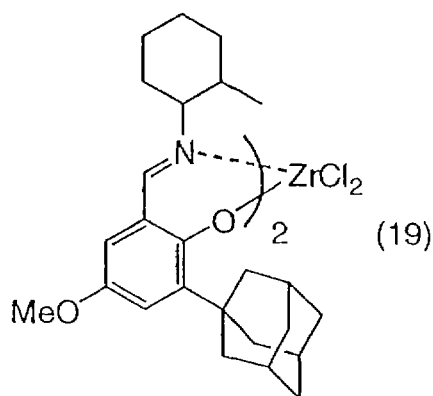
Into a 50 ml reactor thoroughly dried and purged with argon, 1.00 g (2.63 mmol) of the compound (S) and 20 ml of THF were introduced, and they were cooled to $-78\text{ }^{\circ}\text{C}$ and stirred. To the reactor, 1.70 ml of a n-hexane solution containing 2.67 mmol of n-butyllithium was slowly dropwise added, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 1.5 hours to prepare a lithium salt solution. The solution was dropwise added to 20 ml of a THF solution containing 0.50 g (1.32 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution

-9 APR 2003

having been cooled to $-78\text{ }^{\circ}\text{C}$. After the dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 20 hours, and the solvent was distilled off from the reaction solution. To the resulting solid, 30 ml of ether and 70 ml of methylene chloride were added, and the mixture was filtered. The filtrate was concentrated and washed again with ether to obtain 0.65 g (yield: 54 %) of a compound (19) represented by the following formula (19) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.20-2.40 (m, 54H), 3.65-3.95 (m, 2H), 3.80 (s, 6H), 6.59-6.65 (m, 2H), 7.13-7.16 (m, 2H), 8.14-8.23 (m, 2H)

FD-mass spectrometry: 923



-9 APR 2003

Synthesis Example 20

Into a 200 ml reactor thoroughly dried and purged with nitrogen, 43.00 g (346.4 mmol) of anisole, 4.30 g of
5 Umberist-15 (trade name, available from Organo Co.) and 50 ml of toluene were introduced. To the reactor, a solution containing 81.88 g (692.8 mmol) of α -methylstyrene diluted with 50 ml of a toluene was dropwise added at 105 °C, followed by stirring for 7.5
10 hours. After the system was cooled to room temperature, the reaction solution was filtered with washing with toluene, and the resulting liquid was purified by silica gel column chromatography to obtain 24.28 g (yield: 28 %) of 2-cumyl-4-methoxyphenol.

15 ¹H-NMR (CDCl₃): 1.67 (s, 6H), 3.82 (s, 3H), 3.96 (s, 1H), 6.72-7.38 (m, 8H)

Into a 1 liter reactor thoroughly purged with nitrogen, 43.5 ml of an ether solution containing 130.7 mmol of ethylmagnesium bromide and 70 ml of THF were
20 introduced. To the reactor, a solution containing 30.16 g (124.5 mmol) of 2-cumyl-4-methoxyphenol diluted with 80 ml of a THF was dropwise added at 0 °C over a period of 30 minutes. Then, 500 ml of toluene was added, and the system was heated to 100 °C to distill off about 40 ml of
25 a mixed solution of ether and THF, whereby an opaque

-9 APR 2003

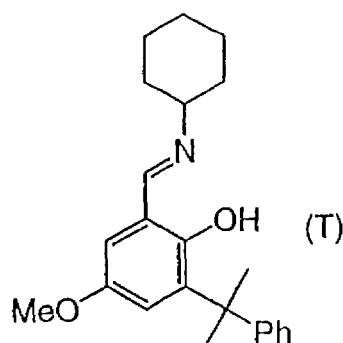
white slurry was obtained. After the slurry was cooled to 15 °C, 9.04 g (301.3 mmol) of paraformaldehyde and 24.8 ml (178.0 mmol) of triethylamine were added, followed by stirring at 90 °C for 15 minutes. The reaction solution was allowed to cool to room temperature and then quenched with 84 ml of 18% hydrochloric acid. The organic layer was washed with 400 ml of water, 200 ml of a sodium hydrogencarbonate aqueous solution and a sodium chloride aqueous solution (200 ml × 2), and then concentrated. The resulting liquid was purified by silica gel column chromatography to obtain 27.27 g (yield: 81 %) of 3-cumyl-5-methoxysalicylaldehyde.

¹H-NMR (CDCl₃): 1.82 (s, 6H), 3.85 (s, 3H), 7.15-7.38 (m, 7H), 9.80 (s, 1H), 11.3 (s, 1H)

Into a 100 ml reactor thoroughly purged with nitrogen, 40 ml of ethanol, 1.09 g (11.0 mmol) of cyclohexylamine and 2.85 g (10.0 mmol) of 3-cumyl-5-methoxysalicylaldehyde were introduced, and they were stirred at room temperature for 2 hours. The solid precipitated was filtered off and vacuum dried to obtain 2.54 g (yield: 72 %) of a compound (T) represented by the following formula (T).

¹H-NMR (CDCl₃): 1.11-1.55 (m, 6H), 1.55-1.85 (m, 4H), 1.75 (s, 6H), 3.11 (m, 1H), 3.82 (s, 3H), 6.63-7.29 (m, 7H), 8.28 (s, 1H), 13.20 (s, 1H)

-9 APR 2003



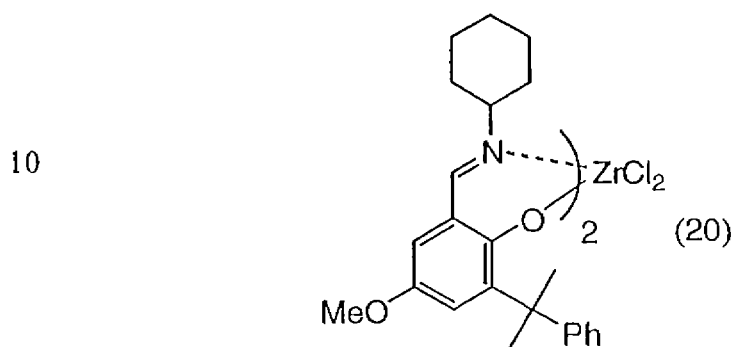
Into a 50 ml reactor thoroughly dried and purged
with argon, 1.05 g (3.00 mmol) of the compound (T) and 30
ml of ether were introduced, and they were cooled to -
10 78 °C and stirred. To the reactor, 2.01 ml of a n-hexane
solution containing 3.15 mmol of n-butyllithium was
slowly dropwise added, then the temperature of the system
was slowly raised to room temperature, and the reaction
solution was stirred at room temperature for 3 hours to
15 prepare a lithium salt solution. The solution was
dropwise added to 30 ml of a THF solution containing 0.57
g (1.50 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution
having been cooled to -78 °C. After the dropwise
addition was completed, stirring was continued with
20 slowly raising the temperature to room temperature. Then,
stirring was further performed at room temperature for 20
hours, and the solvent was distilled off from the
reaction solution. To the resulting solid, 30 ml of
ether was added to give a slurry, and the slurry was
25 filtered with washing with 50 ml of methylene chloride.

-9 APR 2003

The filtrate was concentrated and washed again with a mixed solution of methylene chloride and ether to obtain 0.67 g (yield: 52 %) of a compound (20) represented by the following formula (20) as a yellow powder.

5 $^1\text{H-NMR}$ (CDCl_3): 0.90-2.10 (m, 32H), 3.67 (m, 2H), 3.77, 3.85 (s, s, 6H), 6.60-7.41 (m, 14H), 8.11 (s, 2H)

FD-mass spectrometry: 863

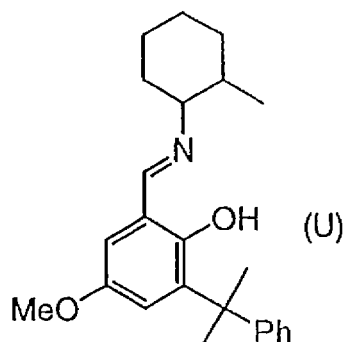


Synthesis Example 21

15 Into a 100 ml reactor thoroughly purged with nitrogen, 40 ml of ethanol, 1.25 g (11.0 mmol) of 2-methylcyclohexylamine and 2.85 g (10.0 mmol) of 3-cumyl-5-methoxysalicylaldehyde were introduced, and they were stirred at room temperature for 3 hours. The solid
20 precipitated was filtered off and vacuum dried to obtain 2.73 g (yield: 75 %) of a compound (U) represented by the following formula (U).

25 $^1\text{H-NMR}$ (CDCl_3): 0.70-0.80 (d, 3H), 0.9-1.83 (m, 9H), 1.75 (s, 6H), 2.62 (m, 1H), 3.82 (s, 3H), 6.68-7.31 (m, 7H), 8.23 (s, 1H), 13.13 (s, 1H)

-9 APR 2003



○ Into a 50 ml reactor thoroughly dried and purged with argon, 1.10 g (3.00 mmol) of the compound (U) and 30 ml of ether were introduced, and they were cooled to -78 °C and stirred. To the reactor, 2.01 ml of a n-hexane solution containing 3.15 mmol of n-butyllithium was slowly dropwise added, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 2 hours to prepare a lithium salt solution. The solution was dropwise added to 30 ml of a THF solution containing 0.57 g (1.50 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to -78 °C. After the dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 20 hours, and the solvent was distilled off from the reaction solution. To the resulting solid, 10 ml of ether and 30 ml of methylene chloride were added to give

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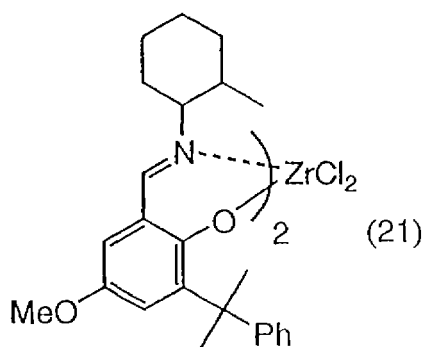
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-9 APR 2003

a slurry, and the slurry was filtered with washing with 20 ml of methylene chloride. The filtrate was concentrated and washed again with a mixed solution of methylene chloride and ether to obtain 0.78 g (yield: 58 %) of a compound (21) represented by the following formula (21) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.60-0.80 (m, 36H), 3.52 (m, 2H), 3.77 (s, 6H), 6.50-7.60 (m, 14H), 8.12 (s, 2H)

FD-mass spectrometry: 891



Synthesis Example 22

Into a 300 ml reactor thoroughly dried and purged with nitrogen, 34.04 g (200.0 mmol) of 4-phenylphenol, 3.40 g of Umberist-15E (trade name, available from Organo Co.) and 150 ml of toluene were introduced. To the reactor, a solution containing 48.20 g (200.0 mmol) of t-butyl alcohol diluted with 10 ml of a toluene was dropwise added at 105 °C, followed by stirring for 16 hours. After the system was cooled to room temperature,

-9 APR 2003

the solution was introduced into 500 ml of hexane and filtered with washing with hexane. After concentration, the resulting liquid was purified by silica gel column chromatography to obtain 31.15 g (yield: 69 %) of 2-t-butyl-4-phenylphenol.

$^1\text{H-NMR}$ (CDCl_3): 1.48 (s, 9H), 4.78 (s, 1H), 6.70-6.58 (m, 8H)

Into a 1 liter reactor thoroughly purged with nitrogen, 66.80 ml of an ether solution containing 200.4 mmol of ethylmagnesium bromide and 100 ml of THF were introduced. To the reactor, a solution containing 43.16 g (190.7 mmol) of 2-t-butyl-4-phenylphenol diluted with 90 ml of a THF was dropwise added at 0 °C over a period of 40 minutes, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 30 minutes. Then, 700 ml of toluene was added, and the system was heated to 100 °C to distill off 52 ml of a mixed solution of ether and THF, whereby an opaque white slurry was obtained. After the slurry was cooled to -20 °C, 13.84 g (460.9 mmol) of paraformaldehyde and 38.1 ml (273.4 mmol) of triethylamine were added, followed by stirring at 90 °C for 30 minutes. After ice cooling, the reaction solution was quenched with 133 ml of 18% hydrochloric acid. The organic layer was washed with 300 ml of water,

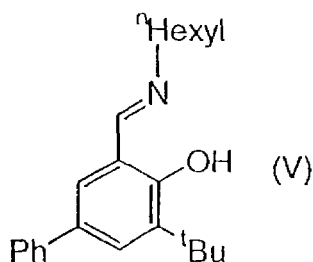
-9 APR 2003

300 ml of a sodium hydrogencarbonate aqueous solution and
300 ml of a sodium chloride aqueous solution, and then
concentrated. The resulting liquid was purified by
silica gel column chromatography to obtain 37.73 g
5 (yield: 78 %) of 3-*t*-butyl-5-phenylsalicylaldehyde.

$^1\text{H-NMR}$ (CDCl_3): 1.47 (s, 9H), 7.29-7.79 (m, 7H), 9.70
(s, 1H), 11.78 (s, 1H)

Into a 100 ml reactor thoroughly purged with
nitrogen, 50 ml of ethanol, 1.21 g (12.0 mmol) of *n*-
10 hexylamine and 2.54 g (10.0 mmol) of 3-*t*-butyl-5-
phenylsalicylaldehyde were introduced, and they were
stirred at room temperature for 1 hour. The reaction
solution was vacuum concentrated to remove the solvent,
and the resulting liquid was purified by silica gel
15 column chromatography to obtain 3.38 g (yield: 94 %) of a
compound (V) represented by the following formula (V).

$^1\text{H-NMR}$ (CDCl_3): 0.90 (t, 3H), 1.20-1.50 (m, 6H), 1.48
(s, 9H), 1.72 (m, 2H), 3.61 (t, 2H), 7.27-7.56 (m, 7H),
8.40 (s, 1H), 14.29 (s, 1H)



-9 APR 2003

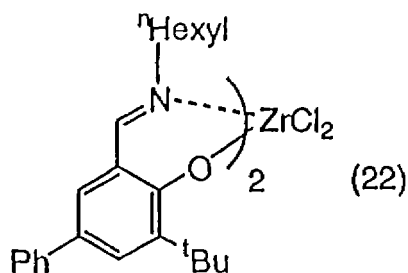
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Into a 50 ml reactor thoroughly dried and purged with argon, 1.01 g (3.00 mmol) of the compound (V) and 30 ml of ether were introduced, and they were cooled to -78 °C and stirred. To the reactor, 1.97 ml of a n-hexane solution containing 3.15 mmol of n-butyllithium was slowly dropwise added, then the temperature of the system was slowly raised to room temperature, and the reaction solution was stirred at room temperature for 4 hours to prepare a lithium salt solution. The solution was dropwise added to 30 ml of a THF solution containing 0.57 g (1.50 mmol) of a $\text{ZrCl}_4(\text{THF})_2$ complex, said THF solution having been cooled to -78 °C. After the dropwise addition was completed, stirring was continued with slowly raising the temperature to room temperature. Then, stirring was further performed at room temperature for 20 hours, and the solvent was distilled off from the reaction solution. To the resulting solid, 20 ml of ether was added to give a slurry, and the slurry was filtered with washing with 10 ml of methylene chloride. The filtrate was concentrated and washed again with 30 ml of ether to obtain 0.49 g (yield: 39 %) of a compound (22) represented by the following formula (22) as a yellow powder.

$^1\text{H-NMR}$ (CDCl_3): 0.7-0.90 (m, 6H), 1.1-2.0 (s, 34H), 3.8-4.1 (m, 4H), 7.2-8.1 (m, 14H), 8.3, 8.8 (brs, s, 2H)

-9 APR 2003

FD-mass spectrometry: 834



The NC values of the compounds obtained are set forth in Table 1.

Table 1

Compound No.	NC	Compound No.	NC
1	1.915	12	1.801
2	1.908	13	1.905
3	1.856	14	1.799
4	1.803	15	1.790
5	1.913	16	1.799
6	1.909	17	1.792
7	1.852	18	1.780
8	1.900	19	1.768
9	1.870	20	1.799
10	1.905	21	1.791
11	1.910	22	1.910

Examples 1 -17

Into a 1 liter stainless steel autoclave thoroughly purged with nitrogen, 500 ml of heptane was introduced. The temperature of the autoclave was set at 75 °C, and

-9 APR 2003

the liquid phase and the gas phase were saturated with ethylene. To the autoclave, 1.25 mmol (in terms of aluminum atom) of methylaluminoxane and the compound shown in Table 2 in the amount shown in Table 2 were added, and the polymerization was conducted for 15 minutes under the ethylene pressure of 8 kg/cm²-G.

The resulting polymer suspension was introduced into a large amount of a mixture of acetone and methanol (acetone:methanol = 1:1) containing a small amount of hydrochloric acid to precipitate a polymer. The polymer was filtered through a glass filter. After the solvent was removed, the polymer was washed with methanol. the polymer was vacuum dried at 80 °C for 10 hours to obtain polyethylene. The results are set forth in Table 2.

Comparative Example 1

Polymerization was carried out in the same manner as in Examples 1 to 22, except that the following zirconium compound (23) (NC = 2.01) was used as the zirconium compound in an amount of 0.00005 mmol. As a result, polyethylene was obtained in an amount of 2.61 g. The polymerization activity was 209 kg/mmol-Zr·hr, and the intrinsic viscosity (η) of the polyethylene was 1.95 dl/g. The results are set forth in Table 2.

-9 APR 2003

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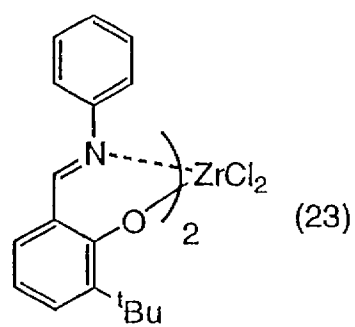


Table 2

	Catalyst		Yield (g)	Activity (kg/mmol -Zr·hr)	Intrinsic viscosity (η) (dl/g)
	Compound	Amount			
	No.	(mmol)			
Ex. 1	1	0.0002	15.06	301	0.55
Ex. 2	2	0.00005	7.17	573	0.56
Ex. 3	3	0.00005	17.54	1403	6.37
Ex. 4	4	0.00005	4.29	343	4.72
Ex. 5	5	0.0001	14.24	569	2.52
Ex. 6	6	0.00005	8.58	686	1.11
Ex. 7	7	0.00005	10.62	850	1.52
Ex. 8	8	0.00005	9.68	774	1.99
Ex. 9	9	0.00005	9.91	793	13.8
Ex. 10	10	0.00005	6.67	534	6.88
Ex. 11	11	0.0001	10.83	433	0.49
Ex. 12	12	0.00005	6.93	554	4.70
Ex. 13	13	0.00002	14.40	2881	0.34
Ex. 14	14	0.00002	8.89	1778	0.46
Ex. 15	16	0.0001	17.24	690	0.82
Ex. 16	17	0.00005	10.84	867	5.74
Ex. 17	22	0.00005	6.01	481	0.32
Comp. Ex.1	23	0.00005	2.61	209	1.95

Examples 18 - 29

Polymerization was carried out in the same manner as in Examples 1 to 17, except that the polymerization in Examples 1 to 17, except that the polymerization temperature was set at 90 °C and the amount of the catalyst was varied to that shown in Table 3. The results are set forth in Table 3.

-9 APR 2003

Comparative Example 2

Polymerization was carried out in the same manner as in Examples 18 to 29, except that the zirconium compound (23) was used as the zirconium compound in an amount of 0.00025 mmol. As a result, polyethylene was obtained in an amount of 3.87 g. The polymerization activity was 62 kg/mmol-Zr.hr, and the intrinsic viscosity (η) of the polyethylene was 4.19 dl/g. The results are set forth in Table 3.

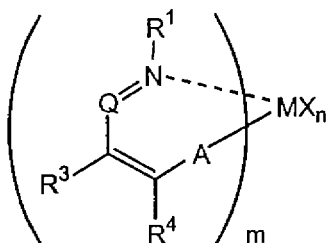
Table 3

	Catalyst		Yield (g)	Activity (kg/mmol -Zr·hr)	Intrinsic viscosity (η) (dl/g)
	Compound	Amount			
	No.	(mmol)			
Ex. 18	1	0.0002	12.26	245	0.38
Ex. 19	2	0.0001	6.69	267	0.55
Ex. 20	3	0.00005	12.00	956	4.84
Ex. 21	5	0.0001	5.03	201	1.69
Ex. 22	6	0.00005	10.29	823	0.78
Ex. 23	7	0.00005	14.98	1198	1.09
Ex. 24	8	0.00005	10.38	831	0.90
Ex. 25	9	0.00005	11.71	937	8.63
Ex. 26	11	0.0001	6.52	261	0.47
Ex. 27	12	0.00005	5.94	475	3.61
Ex. 28	13	0.00005	10.12	810	0.36
Ex. 29	22	0.0001	6.79	272	0.32
Comp. Ex. 2	23	0.00025	3.87	62	4.19

-9 APR 2003

What is claimed is:

1. An olefin polymerization catalyst comprising a transition metal compound which is represented by the following formula (I) and in which the net charge parameter of the central metal M is in the range of 1.70 to 2.00:



(I)

wherein M is a transition metal atom of Group 4 to Group 5 of the periodic table,

m is an integer of 1 to 5,

Q is a nitrogen atom or a carbon atom having a substituent group R²,

A is an oxygen atom, a sulfur atom, a selenium atom or a nitrogen atom having a substituent group R⁵,

R¹ is an aliphatic hydrocarbon group which may not be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group wherein the aliphatic hydrocarbon group has 18 to 30 carbon atoms, or may be substituted with an alicyclic hydrocarbon group,

R² to R⁵ may be the same or different, they are each a hydrogen atom, a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group, a sulfur-containing group, a boron-containing group, an aluminum-

-9 APR 2003

containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, two or more of them may be bonded to each other to form a ring, and when m is 2 or greater, at least one of the groups indicated by R^2 to R^5 contained in one ligand and at least one of the groups indicated by R^2 to R^5 contained in another ligand may be bonded to each other,

when m is 2 or greater, R^1s , R^2s , R^3s , R^4s , or R^5s may be the same or different,

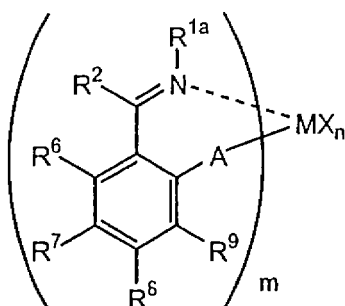
n is a number satisfying a valence of M , and

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

2. An olefin polymerization catalyst comprising a transition metal compound which is represented by the following formula (I-a) and in which the net charge

-9 APR 2003

parameter of the central metal M is in the range of 1.70 to 2.00:



(I-a)

wherein M is a transition metal atom of Group 4 to Group 5 of the periodic table,

m is an integer of 1 to 5,

A is an oxygen atom, a sulfur atom, a selenium atom or a nitrogen atom having a substituent group R⁵,

R^{1a} is an aliphatic hydrocarbon group which may not be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group wherein the aliphatic hydrocarbon group has 18 to 30 carbon atoms, or may be substituted with an alicyclic hydrocarbon group,

R² and R⁵ to R⁹ may be the same or different, they are each a hydrogen atom, a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group, a sulfur-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-

-9 APR 2003

containing group or a tin-containing group, two or more of them may be bonded to each other to form a ring, and when m is 2 or greater, at least one of the groups indicated by R^2 and R^5 to R^9 contained in one ligand and at least one of groups indicated by R^2 and R^5 to R^9 contained in another ligand may be bonded to each other,

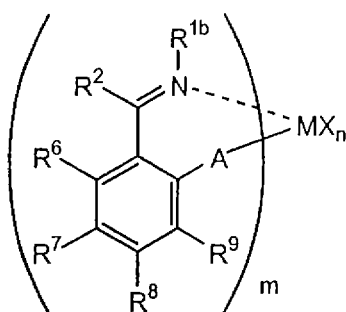
when m is 2 or greater, $R^{1a}s$, R^2s , R^5s , R^6s , R^7s , R^8s , or R^9s may be the same or different,

n is a number satisfying a valence of M , and

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

3. An olefin polymerization catalyst comprising a transition metal compound which is represented by the following formula (I-b) and in which the net charge parameter of the central metal M is in the range of 1.80 to 1.89:

-9 APR 2003



(I-b)

wherein M is a transition metal atom of Group 4 to Group 5 of the periodic table,

m is an integer of 1 to 5,

A is an oxygen atom, a sulfur atom, a selenium atom or a nitrogen atom having a substituent group R⁵,

R^{1b} is an alicyclic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group,

R², R⁵, R⁶, R⁸ and R⁹ may be the same or different, they are each a hydrogen atom, a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group, a sulfur-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, two or more of them may be bonded to each other to form a ring, and when *m* is 2 or greater, at least one of groups indicated by R², R⁵, R⁶, R⁸ and R⁹ contained in one ligand and at

-9 APR 2003

least one of groups indicated by R^2 , R^5 , R^6 , R^8 and R^9 contained in another ligand may be bonded to each other,

R^7 is a hydrogen atom,

when m is 2 or greater, $R^{1b}s$, R^2s , R^5s , R^6s , R^7s , R^8s , or R^9s may be the same or different,

n is a number satisfying a valence of M , and

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

4. The olefin polymerization catalyst of any one of claims 1 to 3, wherein A in each formula (I), (I-a) and (I-b) is an oxygen atom.

5. An olefin polymerization catalyst comprising:
(A-1) the transition metal compound of any one of claims 1 to 4, and

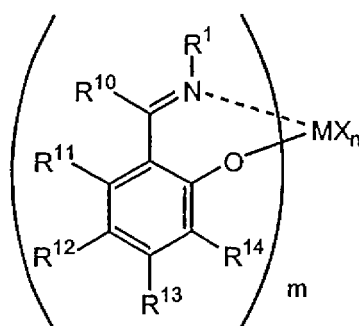
(B) at least one compound selected from:

(B-1) an organometallic compound,

-9 APR 2003

(B-2) an organoaluminum oxy-compound, and
 (B-3) a compound which reacts with the
 transition metal compound to form an ion pair.

6. An olefin polymerization catalyst comprising a
 transition metal compound represented by the following
 formula (II):



(II)

wherein M is a transition metal atom of Group 4 to Group
 5 of the periodic table,

m is 1 or 2,

R¹ is an aliphatic hydrocarbon group which may not
 be substituted with an aromatic hydrocarbon group or an
 alicyclic hydrocarbon group wherein the aliphatic
 hydrocarbon group has 18 to 30 carbon atoms, or may be
 substituted with an alicyclic hydrocarbon group,

R¹⁰ and R¹¹ to R¹³ may be the same or different, they
 are each a hydrogen atom, a hydrocarbon group, a
 hydrocarbon-substituted silyl group, an oxygen-containing
 group, a nitrogen-containing group or a sulfur-containing

-9 APR 2003

group, and two or more of them may be bonded to each other to form a ring,

R^{14} is a hydrocarbon group or a hydrocarbon-substituted silyl group,

when m is 2, at least one of groups indicated by R^{10} to R^{14} contained in one ligand and at least one of groups indicated by R^{10} to R^{14} contained in another ligand may be bonded to each other,

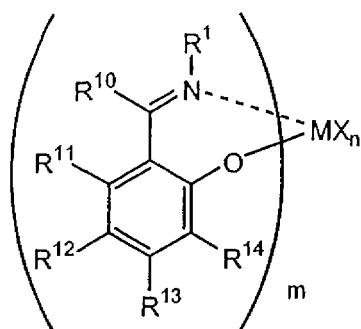
when m is 2, R^1s , $R^{10}s$, $R^{11}s$, $R^{12}s$, $R^{13}s$, or $R^{14}s$ may be the same or different,

n is a number satisfying a valence of M , and

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

7. An olefin polymerization catalyst comprising a transition metal compound represented by the following formula (II):

-9 APR 2003



(II)

wherein M is a transition metal atom of Group 4 to Group 5 of the periodic table,

m is 1 or 2,

R¹ is an aliphatic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group wherein the aliphatic hydrocarbon group has 5 to 30 carbon atoms,

R¹⁰, R¹¹ and R¹³ may be the same or different, they are each a hydrogen atom, a hydrocarbon group, a hydrocarbon-substituted silyl group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group, and two or more of them may be bonded to each other to form a ring,

R¹² is a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group,

R¹⁴ is a hydrocarbon group or a hydrocarbon-substituted silyl group,

-9 APR 2003

when m is 2, at least one of groups indicated by R^{10} to R^{14} contained in one ligand and at least one of groups indicated by R^{10} to R^{14} contained in another ligand may be bonded to each other,

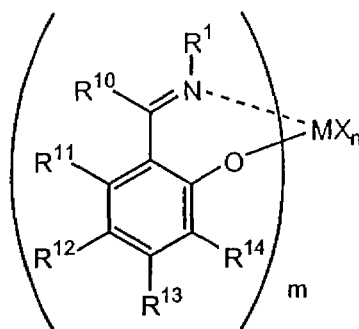
when m is 2, R^1s , $R^{10}s$, $R^{11}s$, $R^{12}s$, $R^{13}s$, or $R^{14}s$ may be the same or different,

n is a number satisfying a valence of M , and

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

8. An olefin polymerization catalyst comprising a transition metal compound represented by the following formula (II):

-9 APR 2003



(II)

wherein M is a transition metal atom of Group 4 to Group 5 of the periodic table,

m is 1 or 2,

R¹ is an alicyclic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group and has not less than total 7 carbon atoms,

R¹⁰, R¹¹ and R¹³ may be the same or different, they are each a hydrogen atom, a hydrocarbon group, a hydrocarbon-substituted silyl group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group, and two or more of them may be bonded to each other to form a ring,

R¹² is a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group,

R¹⁴ is a hydrocarbon group or a hydrocarbon-substituted silyl group,

-9 APR 2003

when m is 2, at least one of groups indicated by R^{10} to R^{14} contained in one ligand and at least one of groups indicated by R^{10} to R^{14} contained in another ligand may be bonded to each other,

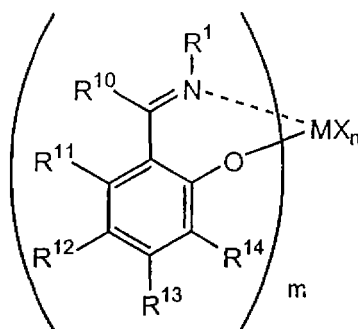
when m is 2, R^1s , $R^{10}s$, $R^{11}s$, $R^{12}s$, $R^{13}s$, or $R^{14}s$ may be the same or different,

n is a number satisfying a valence of M , and

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

9. An olefin polymerization catalyst comprising a transition metal compound represented by the following formula (II):

-9 APR 2003



(II)

wherein M is a transition metal atom of Group 4 to Group 5 of the periodic table,

m is 1 or 2,

R¹ is an alicyclic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group,

R¹⁰, R¹¹ and R¹³ may be the same or different, they are each a hydrogen atom, a hydrocarbon group, a hydrocarbon-substituted silyl group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group, and two or more of them may be bonded to each other to form a ring,

R¹² is a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group,

R¹⁴ is a group selected from a branched alkyl group of 3 to 30 carbon atoms wherein an aryl group is substituted for a hydrogen atom of the branched alkyl

-9 APR 2003

group, a cyclic saturated hydrocarbon group, an aryl group, and a hydrocarbon-substituted silyl group,

when m is 2, at least one of groups indicated by R^{10} to R^{14} contained in one ligand and at least one of groups indicated by R^{10} to R^{14} contained in another ligand may be bonded to each other,

when m is 2, R^1s , $R^{10}s$, $R^{11}s$, $R^{12}s$, $R^{13}s$, or $R^{14}s$ may be the same or different,

n is a number satisfying a valence of M , and

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

10. An olefin polymerization catalyst comprising:

(A-2) the transition metal compound of any one of claims 6 to 9, and

(B) at least one compound selected from:

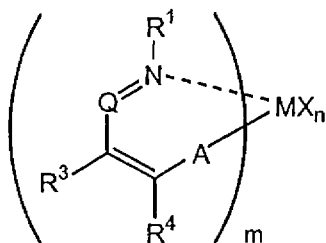
(B-1) an organometallic compound,

(B-2) an organoaluminum oxy-compound, and

-9 APR 2003

(B-3) a compound which reacts with the transition metal compound to form an ion pair.

11. A process for olefin polymerization, comprising polymerizing an olefin at polymerization temperature in the range of 60 to 250 °C in the presence of a olefin polymerization catalyst including a transition metal compound which is represented by the following formula (I) and in which the net charge parameter of the central metal M is not more than 2.00:



(I)

wherein M is a transition metal atom of Group 4 to Group 5 of the periodic table,

m is an integer of 1 to 5,

Q is a nitrogen atom or a carbon atom having a substituent group R²,

A is an oxygen atom, a sulfur atom, a selenium atom or a nitrogen atom having a substituent group R⁵,

R¹ is an aliphatic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group wherein the aliphatic hydrocarbon group has 5 to 30 carbon atoms,

R^2 to R^5 may be the same or different, they are each a hydrogen atom, a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group, a sulfur-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, two or more of them may be bonded to each other to form a ring, and when m is 2 or greater, at least one of groups indicated by R^2 to R^5 contained in one ligand and at least one of groups indicated by R^2 to R^5 contained in another ligand may be bonded to each other,

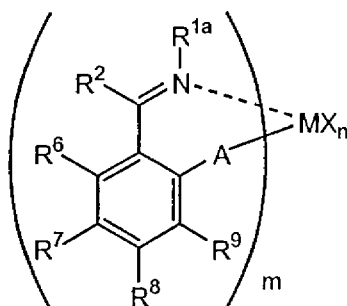
when m is 2 or greater, R^1s , R^2s , R^3s , R^4s , or R^5s may be the same or different,

n is a number satisfying a valence of M , and

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

-9 APR 2003

12. A process for olefin polymerization, comprising polymerizing an olefin at polymerization temperature in the range of 60 to 250 °C in the presence of an olefin polymerization catalyst including a transition metal compound which is represented by the following formula (I-a) and in which the net charge parameter of the central metal M is in the range of 1.70 to 2.00:



(I-a)

wherein M is a transition metal atom of Group 4 to Group 5 of the periodic table,

m is an integer of 1 to 5,

A is an oxygen atom, a sulfur atom, a selenium atom or a nitrogen atom having a substituent group R⁵,

R^{1a} is an aliphatic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group wherein the aliphatic hydrocarbon group has 5 to 30 carbon atoms,

R² and R⁵ to R⁹ may be the same or different, they are each a hydrogen atom, a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group, a

sulfur-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, two or more of them may be bonded to each other to form a ring, and when m is 2 or greater, at least one of groups indicated by R^2 and R^5 to R^9 contained in one ligand and at least one of groups indicated by R^2 and R^5 to R^9 contained in another ligand may be bonded to each other,

when m is 2 or greater, $R^{1a}s$, R^2s , R^5s , R^6s , R^7s , R^8s , or R^9s may be the same or different,

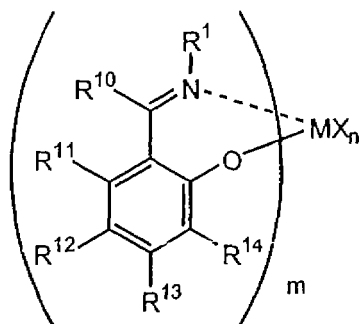
n is a number satisfying a valence of M , and

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

13. A process for olefin polymerization, comprising polymerizing an olefin at polymerization temperature in the range of 60 to 250 °C in the presence of an olefin

-9 APR 2003

polymerization catalyst including a transition metal compound which is represented by the following formula (II):



(II)

wherein M is a transition metal atom of Group 4 to Group 5 of the periodic table,

m is 1 or 2,

R^1 is an aliphatic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an alicyclic hydrocarbon group wherein the aliphatic hydrocarbon group has not less than total 5 carbon atoms,

R^{10} and R^{11} to R^{13} may be the same or different, they are each a hydrogen atom, a hydrocarbon group, a hydrocarbon-substituted silyl group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group, and two or more of them may be bonded to each other to form a ring,

R^{14} is a hydrocarbon group or a hydrocarbon-substituted silyl group,

when m is 2, at least one of groups indicated by R^{10} to R^{14} contained in one ligand and at least one of groups

-9 APR 2003

indicated by R^{10} to R^{14} contained in another ligand may be bonded to each other,

when m is 2, R^1s , $R^{10}s$, $R^{11}s$, $R^{12}s$, $R^{13}s$, or $R^{14}s$ may be the same or different,

n is a number satisfying a valence of M , and

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

14. A process for olefin polymerization of any one of claims 11 to 13, wherein the olefin polymerization catalyst further includes

(B) at least one compound selected from:

(B-1) an organometallic compound,

(B-2) an organoaluminum oxy-compound, and

(B-3) a compound which reacts with the transition metal compound to form an ion pair.

-9 APR 2003

15. A process for olefin polymerization, comprising polymerizing an olefin at polymerization temperature in the range of 60 to 250 °C in the presence of a olefin polymerization catalyst of claim 3.

16. A process for olefin polymerization, comprising polymerizing an olefin at polymerization temperature in the range of 60 to 250 °C in the presence of an olefin polymerization catalyst of claim 4.

17. A process for olefin polymerization of claims 15 or 16, wherein the olefin polymerization catalyst further includes

(B) at least one compound selected from:

(B-1) an organometallic compound,

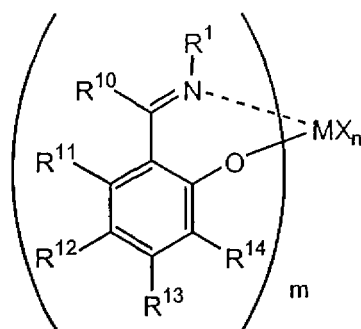
(B-2) an organoaluminum oxy-compound, and

(B-3) a compound which reacts with the

transition metal compound to form an ion pair.

18. A process for olefin polymerization, comprising polymerizing an olefin at polymerization temperature in the range of 60 to 250 °C in the presence of a olefin polymerization catalyst including a transition metal compound which is represented by the following formula (II):

-9 APR 2003



(II)

wherein M is a transition metal atom of Group 4 to Group 5 of the periodic table,

m is 1 or 2,

R¹ is an alicyclic hydrocarbon group which may be substituted with an aromatic hydrocarbon group or an aliphatic hydrocarbon group wherein the aliphatic hydrocarbon group has not less than total 5 carbon atoms,

R¹⁰, R¹¹ and R¹³ may be the same or different, they are each a hydrogen atom, a hydrocarbon group, a hydrocarbon-substituted silyl group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group, and two or more of them may be bonded to each other to form a ring,

R¹² is a hydrocarbon group, an oxygen-containing group, a nitrogen-containing group or a sulfur-containing group,

R¹⁴ is a hydrocarbon group or a hydrocarbon-substituted silyl group,

when m is 2, at least one of groups indicated by R¹⁰ to R¹⁴ contained in one ligand and at least one of groups

indicated by R^{10} to R^{14} contained in another ligand may be bonded to each other,

when m is 2, R^1s , $R^{10}s$, $R^{11}s$, $R^{12}s$, $R^{13}s$, or $R^{14}s$ may be the same or different,

n is a number satisfying a valence of M , and

X is a hydrogen atom, a halogen atom, a hydrocarbon group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group, a boron-containing group, an aluminum-containing group, a phosphorus-containing group, a halogen-containing group, a heterocyclic compound residual group, a silicon-containing group, a germanium-containing group or a tin-containing group, and when n is 2 or greater, plural groups indicated by X may be the same or different and plural groups indicated by X may be bonded to each other to form a ring.

19. A process for olefin polymerization of claim 18, wherein the olefin polymerization catalyst further includes

(B) at least one compound selected from:

(B-1) an organometallic compound,

(B-2) an organoaluminum oxy-compound, and

(B-3) a compound which reacts with the

transition metal compound to form an ion pair.