

TITLECOMPOSITIONS FOR PEELABLE PROTECTIVE FILMS AND
FILMS OF THE SAME

5

FIELD OF THE INVENTION

The present invention relates to compositions for peelable protective films and films of the compositions. More particularly, the invention relates to compositions capable of providing peelable protective polyolefin films which are used to temporarily cover metal articles, plastic articles, rubber articles, etc. and thereby protect those articles from scratches, contamination or corrosion occurring when they are worked, transported or stored and which can be peeled from those articles easily and at a stroke after the above object is attained. The present invention also relates to films of the compositions.

BACKGROUND OF THE INVENTION

Peelable protective films are used to protect metal articles, plastic articles, rubber articles, etc. from scratches, contamination or corrosion occurring when those articles are worked, transported or stored, so that these films are required to have such properties that they can temporarily cover surfaces of the above articles and can be peeled therefrom easily and at a stroke after the above object is attained.

For the conventional peelable protective films, resin compositions containing an ethylene/vinyl acetate copolymer

(hereinafter referred to as "EVA") or a low-crystalline ethylene/ α -olefin copolymer as their major component have been employed. These compositions have good initial adhesion, but they have a problem in that when they are
5 stored for a long period of time, particularly at high temperatures, their adhesive strength increases because of too low heat resistance, resulting in marked deterioration of the peelability.

In Japanese Patent Publication No. 45974/1983, a
10 peelable protective polyolefin film having been improved in heat resistance of the EVA film is proposed.

This film shows improved heat resistance, but it has a problem in that the adhesive strength of the film increases due to a change with time in the long time storage or a
15 bleed substance remains on a surface of an adherend after the film is peeled from the adherend.

In Japanese Patent Laid-Open Publications No. 66737/1991, No. 62838/1991 and No. 62839/1991, the present applicant has proposed methods to reduce a change in the
20 adhesive strength with time occurring in the long time storage or to reduce a bleed substance remaining on a surface of an adherend by using a propylene/1-butene random copolymer. For example, in Japanese Patent Laid-Open Publication No. 66737/1991, a composition comprising a
25 specific propylene/1-butene random copolymer and a specific ethylene/ α -olefin copolymer, a composition comprising a specific propylene/1-butene random copolymer, a specific ethylene/ α -olefin copolymer and a tackifier, and peelable

protective films formed from these compositions are proposed. These compositions can provide films showing excellent flexibility, scratch resistance and heat resistance, having moderate initial adhesive strength to an adherend and peelability, having good cold workability even in the bonded state, being almost free from a change in the adhesive strength with time when stored for a long period of time, and leaving no bleed substance on surfaces of adherends even when the films are peeled from the adherends.

However, since the propylene/1-butene random copolymer is prepared in the presence of a conventional Ziegler catalyst, it has a wide molecular weight distribution and a wide composition distribution. Therefore, the peelable protective film using this propylene/1-butene random copolymer is not always satisfactory in change of adhesive strength with time though the change is small. Besides, a bleed substance sometimes remains on a surface of an adherend though the amount is small. For these reasons, the film is desired to be further improved in the quality.

The present inventors have earnestly studied to solve such problems as mentioned above, and as a result they have found that a peelable protective film, which shows smaller change in the adhesive strength with time when stored for a long period of time and leaves no bleed substance on an adherend surface as compared with the peelable protective films of Japanese Patent Laid-Open Publication No. 66737/1991, can be obtained from a composition comprising

specific proportions of a specific propylene/1-butene random copolymer (A), that is obtained by copolymerizing propylene and 1-butene in the presence of an olefin polymerization catalyst containing a specific transition metal compound (metallocene compound) as a catalyst component, and a specific ethylene/ α -olefin copolymer (B) comprising ethylene and an α -olefin of 3 to 10 carbon atoms. Based on this finding, the present invention has been accomplished.

10

OBJECT OF THE INVENTION

The present invention is intended to solve such problems associated with the prior art as described above, and it is an object of the invention to provide a composition for a peelable protective film, said composition being capable of forming a peelable protective film showing excellent flexibility, scratch resistance and heat resistance, having moderate initial adhesive strength to an adherend and peelability, having good cold workability even in the bonded state, being almost free from a change in the adhesive strength with time when stored for a long period of time, and leaving no bleed substance on a surface of an adherend even when the film is peeled from the adherend. It is another object of the invention to provide a peelable protective film of the composition.

25

SUMMARY OF THE INVENTION

The composition for a peelable protective film according to the invention comprises:

a propylene/1-butene random copolymer (A) in an amount of 30 to 100 parts by weight,

5 an ethylene/ α -olefin copolymer (B) comprising ethylene and an α -olefin of 3 to 10 carbon atoms in an amount of 0 to 70 parts by weight, and

a tackifier (C) in an amount of 0 to 20 parts by weight,

10 the total amount of said components (A), (B) and (C) being 100 parts by weight;

wherein the propylene/1-butene random copolymer (A) has the following properties:

(i) the propylene content is 50 to 95 % by mol,
15 (ii) the melt flow rate (ASTM D 1238, 230 °C, 2.16 kg) is 0.1 to 100 g/10 min,

(iii) the molecular weight distribution (Mw/Mn, Mw: weight-average molecular weight, Mn: number-average molecular weight), as determined by gel permeation
20 chromatography (GPC), is not more than 3.0, and

(iv) the parameter B value, which indicates randomness of the copolymerized monomer sequence distribution, is 1.0 to 1.5;

and

25 the ethylene/ α -olefin copolymer (B) has the following properties:

(i) the melt flow rate (ASTM D 1238, 190 °C, 2.16 kg) is 0.1 to 100 g/10 min, and

(ii) the ethylene content is 75 to 95 % by mol.

The propylene/1-butene random copolymer (A) preferably has the following properties:

(iv) the parameter B value, which indicates randomness
5 of the copolymerized monomer sequence distribution, is 1.0 to 1.3,

(v) the melting point (T_m), as measured by a differential scanning calorimeter, is 60 to 140 °C,

(vi) the melting point (T_m) and the 1-butene content
10 (M [% by mol]) satisfy the following relation

$$-2.6M + 130 \leq T_m \leq -2.3M + 155$$

and

(vii) the crystallinity (α [%]), as measured by X-ray diffractometry, and the 1-butene content (M [% by mol])
15 satisfy the following relation

$$\alpha \geq -1.5M + 75.$$

The propylene/1-butene random copolymer (A) is obtained by copolymerizing propylene and 1-butene in the presence of an olefin polymerization catalyst comprising:

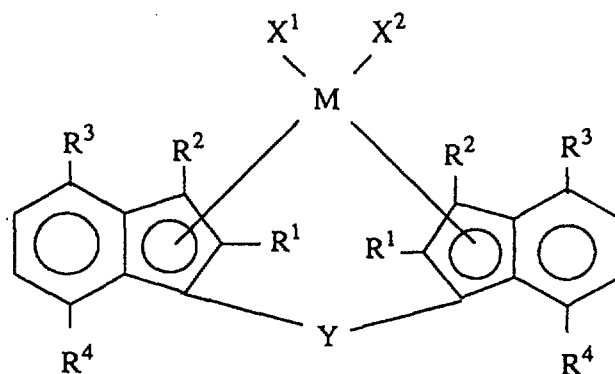
20 [A] a transition metal compound represented by the following formula [I],

[B] [B-1] an organoaluminum oxy-compound and/or [B-2] a compound which reacts with the transition metal compound [A] to form an ion pair,

25 and optionally

[C] an organoaluminum compound.

The transition metal compound [A] represented by the following formula [I]:



[I]

wherein M is a transition metal of Group IVa, Group Va or
 5 Group VIA of the periodic table,

R¹ and R² are each a hydrogen atom, a halogen atom, a
 hydrocarbon group of 1 to 20 carbon atoms, a halogenated
 hydrocarbon group of 1 to 20 carbon atoms, a silicon-
 containing group, an oxygen-containing group, a sulfur-
 10 containing group, a nitrogen-containing group or a
 phosphorus-containing group,

R³ is a secondary or tertiary alkyl group of 3 to 20
 carbon atoms or an aromatic group,

R⁴ is a hydrogen atom or an alkyl group of 1 to 20
 15 carbon atoms,

X¹ and X² are each a hydrogen atom, a halogen atom, a
 hydrocarbon group of 1 to 20 carbon atoms, a halogenated
 hydrocarbon group of 1 to 20 carbon atoms, an oxygen-
 containing group or a sulfur-containing group,

20 Y is a divalent hydrocarbon group of 1 to 20 carbon
 atoms, a divalent halogenated hydrocarbon group of 1 to 20
 carbon atoms, a divalent silicon-containing group, a

divalent germanium-containing group, a divalent tin-containing group, -O-, -CO-, -S-, SO-, -SO₂-, -NR⁵-, -P(R⁵)-, -P(O)(R⁵)-, -BR⁵- or -AlR⁵- (wherein R⁵ 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).

In the above formula [I], R¹ is preferably a methyl group.

The peelable protective film according to the invention is a single-layer or multi-layer film in which at least one of the outermost layers is formed from the above-mentioned composition.

DETAILED DESCRIPTION OF THE INVENTION

The composition for a peelable protective film and the peelable protective film according to the invention will be described in detail hereinafter.

The composition for a peelable protective film according to the invention comprises a propylene/1-butene random copolymer (A), and this composition further comprises an ethylene/ α -olefin copolymer (B) and/or a tackifier (C) in addition to the component (A).

Each component is described below.

Propylene/1-butene random copolymer (A)

(1) Composition

In the propylene/1-butene random copolymer (A) used in the invention, the content of constituent units derived from propylene (propylene content) is 50 to 95 % by mol,

preferably 50 to 90 % by mol, more preferably 55 to 90 % by mol, particularly preferably 60 to 85 % by mol; and the content of constituent units derived from 1-butene (1-butene content) is 5 to 50 % by mol, preferably 10 to 50 %
5 by mol, more preferably 10 to 45 % by mol, particularly preferably 15 to 40 % by mol.

The propylene/1-butene random copolymer (A) may contain constituent units derived from olefins other than propylene and 1-butene in small amounts, for example, not
10 more than 10 % by mol, preferably not more than 5 % by mol.

(2) Melt flow rate

The propylene/1-butene random copolymer (A) has a melt flow rate (MFR, ASTM D 1238, 230 °C, load of 2.16 kg) of 0.1 to 100 g/10 min, preferably 0.5 to 50 g/10 min, more
15 preferably 1 to 20 g/10 min.

(3) Molecular weight distribution (Mw/Mn)

The propylene/1-butene random copolymer (A) has a molecular weight distribution (Mw/Mn), as determined by gel permeation chromatography (GPC), of not more than 3.0,
20 preferably 1.5 to 3.0, more preferably 1.5 to 2.5.

(4) Randomness

The propylene/1-butene random copolymer (A) has a parameter B value, which indicates randomness of the copolymerized monomer sequence distribution, of 1.0 to 1.5,
25 preferably 1.0 to 1.3, more preferably 1.0 to 1.2.

The parameter B value was proposed by B.D. Coleman and T.G. Fox (J. Polym. Sci., A1, 3183 (1963)), and this parameter is defined as follows.

$$B = P_{12} / (2P_1 \cdot P_2)$$

wherein P_1 and P_2 are a first monomer content fraction and a second monomer content fraction, respectively, and P_{12} is a proportion of the first monomer-second monomer sequences to all the two molecule sequences.

In the case of $B = 1$, Bernoulli's statistics apply to the copolymer; in the case of $B < 1$, the copolymer tends to be block; and in the case of $B > 1$, the copolymer tends to be alternating.

In addition to the above properties, the propylene/1-butene random copolymer (A) preferably further has the following properties (5) and (6).

(5) The melting point (T_m), as measured by a differential scanning calorimeter, is 60 to 140 °C, preferably 80 to 130 °C, and besides the melting point (T_m) and the 1-butene content (M [% by mol]) satisfy the following relation:

$$-2.6M + 130 \leq T_m \leq -2.3M + 155.$$

(6) The crystallinity (α [%]), as measured by X-ray diffractometry, and the 1-butene content (M [% by mol]) satisfy the following relation:

$$\alpha \geq -1.5M + 75.$$

The propylene/1-butene random copolymer (A) is desired to have a crystallinity of 15 to 65 %, preferably 30 to 60 %.

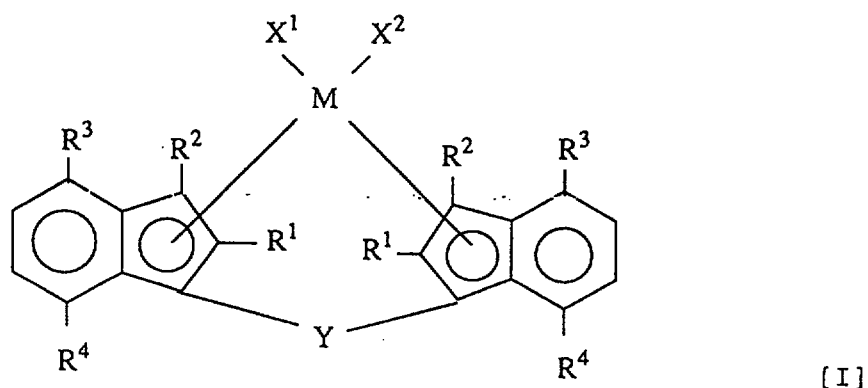
The propylene/1-butene random copolymer (A) can be prepared by copolymerizing propylene and 1-butene in the presence of an olefin polymerization catalyst comprising

[A] the later-described specific transition metal compound,
 [B] [B-1] an organoaluminum oxy-compound and/or [B-2] a
 compound which reacts with the transition metal compound
 [A] to form an ion pair, and optionally, [C] an
 5 organoaluminum compound.

The propylene/1-butene random copolymer prepared as
 above has high stereoregularity and shows excellent scratch
 resistance, heat resistance, transparency and blocking
 resistance.

10 The olefin polymerization catalyst used for the
 invention is described below.

The transition metal compound [A] (hereinafter
 sometimes referred to as "component [A]") forming the
 olefin polymerization catalyst used in the invention is
 15 represented by the following formula [I]:



wherein M is a transition metal of Group IVa, Group Va or
 20 Group VIa of the periodic table. Examples thereof include
 titanium, zirconium, hafnium, vanadium, niobium, tantalum,
 chromium, molybdenum and tungsten. Of these, preferred are

titanium, zirconium and hafnium. Particularly preferred is zirconium.

Substituents R^1 and R^2

R^1 and R^2 are each a hydrogen atom, a halogen atom, a hydrocarbon group of 1 to 20 carbon atoms, a halogenated hydrocarbon group of 1 to 20 carbon atoms, a silicon-containing group, an oxygen-containing group, a sulfur-containing group, a nitrogen-containing group or a phosphorus-containing group.

Examples of the halogens include fluorine, chlorine, bromine and iodine.

Examples of the hydrocarbon groups of 1 to 20 carbon atoms include alkyl groups, such as methyl, ethyl, propyl, butyl, hexyl, cyclohexyl, octyl, nonyl, dodecyl, icosyl, norbornyl and adamantyl; alkenyl groups, such as vinyl, propenyl and cyclohexenyl; arylalkyl groups, such as benzyl, phenylethyl and phenylpropyl; and aryl groups, such as phenyl, tolyl, dimethylphenyl, trimethylphenyl, ethylphenyl, propylphenyl, biphenyl, naphthyl, methylnaphthyl, anthracenyl and phenanthryl.

Examples of the halogenated hydrocarbon groups include those obtained by substituting the above-mentioned hydrocarbon groups with halogen atoms.

Examples of the silicon-containing substituents include monohydrocarbon-substituted silyls, such as methylsilyl and phenylsilyl; dihydrocarbon-substituted silyls, such as dimethylsilyl and diphenylsilyl; trihydrocarbon-substituted silyls, such as trimethylsilyl,

triethylsilyl, tripropylsilyl, tricyclohexylsilyl, triphenylsilyl, dimethylphenylsilyl, methyldiphenylsilyl, tritolylsilyl and trinaphthylsilyl; silyl ethers of hydrocarbon-substituted silyls, such as trimethylsilyl ether; silicon-substituted alkyl groups, such as trimethylsilylmethyl; and silicon-substituted aryl groups, such as trimethylphenyl.

Examples of the oxygen-containing substituents include hydroxyl groups; alkoxy groups, such as methoxy, ethoxy, propoxy and butoxy; aryloxy groups, such as phenoxy, methylphenoxy, dimethylphenoxy and naphthoxy; and arylalkoxy groups, such as phenylmethoxy and phenylethoxy.

Examples of the sulfur-containing groups include substituents obtained by replacing oxygen with sulfur in the above-exemplified oxygen-containing groups.

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

Examples of the phosphorus-containing groups include phosphino groups, such as dimethylphosphino and diphenylphosphino.

Of these, preferred as R^1 are hydrogen, methyl, hydrocarbon groups of 2 to 6 carbon atoms and aromatic groups, and particularly preferred are methyl and hydrocarbon groups of 2 to 6 carbon atoms.

As R^2 , preferred are hydrogen and hydrocarbon groups, and particularly preferred is hydrogen.

Substituent R^3

R^3 is a hydrocarbon group of 1 to 20 carbon atoms or a group obtained by substituting said hydrocarbon group with a halogen atom or a silicon-containing group, and is preferably a secondary or tertiary alkyl group of 3 to 20 carbon atoms or an aromatic group.

Examples of the secondary or tertiary alkyl groups include i-propyl, i-butyl, sec-butyl, tert-butyl, 1,2-dimethylpropyl, 2,3-dimethylbutyl, iso-pentyl, tert-pentyl, neopentyl, cyclopentyl, cyclohexyl, 4-methylcyclohexyl, iso-hexyl, norbornyl and adamantyl.

Examples of the aromatic groups include aryl groups, such as phenyl, tolyl, dimethylphenyl, trimethylphenyl, ethylphenyl, propylphenyl, biphenyl, α - or β -naphthyl, methylnaphthyl, anthracenyl, phenanthryl, benzylphenyl, pyrenyl, acenaphthyl, phenalenyl, aceanthrylenyl, tetrahydronaphthyl, indanyl and biphenylyl; and arylalkyl groups, such as benzyl, phenylethyl, phenylpropyl and tolylmethyl. These groups may contain double or triple bond.

Further, these groups may be substituted with such halogen atoms and silicon-containing groups as described for R^1 .

Substituent R^4

R^4 is a hydrogen atom or an alkyl group of 1 to 20 carbon atoms.

Examples of the alkyl groups include chain and cyclic alkyl groups, such as methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, sec-butyl, tert-butyl, pentyl, hexyl, cyclohexyl, heptyl, octyl, nonyl, dodecyl, icosyl,
 5 norbornyl and adamantyl.

These groups may be substituted with such halogen atoms and silicon-containing groups as described for R^1 .

X^1 and X^2

X^1 and X^2 are each a hydrogen atom, a halogen atom, a
 10 hydrocarbon group of 1 to 20 carbon atoms, a halogenated hydrocarbon group of 1 to 20 carbon atoms, an oxygen-containing group or a sulfur-containing group.

Examples of the halogen atoms, the oxygen-containing groups, the hydrocarbon groups of 1 to 20 carbon atoms and
 15 the halogenated hydrocarbon groups of 1 to 20 carbon atoms are the same as those described for R^1 .

Examples of the sulfur-containing groups include the same groups as described for R^1 ; sulfonato groups, such as methylsulfonato, trifluoromethanesulfonato
 20 phenylsulfonato, benzylsulfonato, p-toluenesulfonato, trimethylbenzenesulfonato, triisobutylbenzenesulfonato, p-chlorobenzenesulfonato and pentafluorobenzenesulfonato; and sulfinato groups, such as methylsulfinato, phenylsulfinato, benzenesulfinato, p-toluenesulfinato,
 25 trimethylbenzenesulfinato, pentafluorobenzenesulfinato and trifluoromethanesulfinato.

Y

Y is a divalent hydrocarbon group of 1 to 20 carbon atoms, a divalent halogenated hydrocarbon group of 1 to 20 carbon atoms, a divalent silicon-containing group, a divalent germanium-containing group, a divalent tin-containing group, -O-, -CO-, -S-, SO-, -SO₂-, -NR⁵-, -P(R⁵)-, -P(O)(R⁵)-, -BR⁵- or -AlR⁵- (R⁵ 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).

10 More specifically, there can be mentioned:

divalent hydrocarbon groups of 1 to 20 carbon atoms, such as alkylene groups (e.g., methylene, dimethylmethylene, 1,2-ethylene, dimethyl-1,2-ethylene, 1,3-trimethylene, 1,4-tetramethylene, 1,2-cyclohexylene and 1,4-cyclohexylene) and arylalkylene groups (e.g., diphenylmethylene and diphenyl-1,2-ethylene);

divalent halogenated hydrocarbon groups obtained by halogenating the above-exemplified divalent hydrocarbon groups of 1 to 20 carbon atoms, such as chloromethylene;

20 divalent silicon-containing groups, such as alkylsilylene, alkylarylsilylene and arylsilylene groups (e.g., methylsilylene, dimethylsilylene, diethylsilylene, di(n-propyl)silylene, di(i-propyl)silylene, di(cyclohexyl)silylene, methylphenylsilylene, 25 diphenylsilylene, di(p-tolyl)silylene and di(p-chlorophenyl)silylene) and alkyldisilyl, alkylaryldisilyl and aryldisilyl groups (e.g., tetramethyl-1,2-disilyl and tetraphenyl-1,2-disilyl);

divalent germanium-containing groups obtained by replacing silicon with germanium in the above-exemplified divalent silicon-containing groups; and

divalent tin-containing groups obtained by replacing
5 silicon with tin in the above-exemplified divalent silicon-containing groups.

R⁵ is the same halogen atom, hydrocarbon group of 1 to 20 carbon atoms or halogenated hydrocarbon group of 1 to 20 carbon atoms as described for R¹.

10 Of these, preferred are divalent silicon-containing groups, divalent germanium-containing groups and divalent tin-containing groups, and more preferred are divalent silicon-containing groups. Of these, particularly preferred are alkylsilylene, alkylarylsilylene and
15 arylsilylene.

Listed below are examples of the transition metal compounds represented by the above formula [I].

rac-Dimethylsilylene-bis(2,7-dimethyl-4-ethyl-1-indenyl)zirconium dichloride,

20 rac-Dimethylsilylene-bis(2,7-dimethyl-4-n-propyl-1-indenyl)zirconium dichloride,

rac-Dimethylsilylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium dichloride,

25 rac-Dimethylsilylene-bis(2,7-dimethyl-4-n-butyl-1-indenyl)zirconium dichloride,

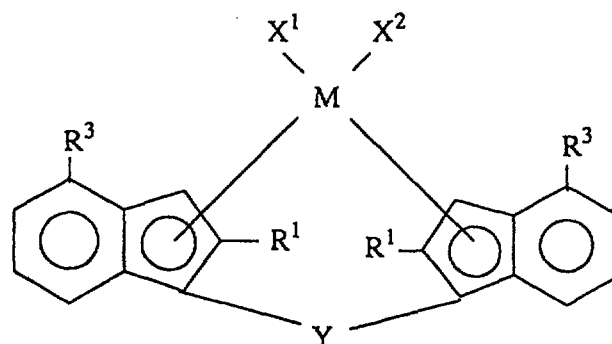
rac-Dimethylsilylene-bis(2,7-dimethyl-4-sec-butyl-1-indenyl)zirconium dichloride,

- rac-Dimethylsilylene-bis(2,7-dimethyl-4-t-butyl-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2,7-dimethyl-4-n-pentyl-1-indenyl)zirconium dichloride,
- 5 rac-Dimethylsilylene-bis(2,7-dimethyl-4-n-hexyl-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2,7-dimethyl-4-cyclohexyl-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2,7-dimethyl-4-
- 10 methylcyclohexyl-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2,7-dimethyl-4-phenylethyl-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2,7-dimethyl-4-phenyldichloromethyl-1-indenyl)zirconium dichloride,
- 15 rac-Dimethylsilylene-bis(2,7-dimethyl-4-chloromethyl-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2,7-dimethyl-4-trimethylsilylenemethyl-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2,7-dimethyl-4-
- 20 trimethylsiloxymethyl-1-indenyl)zirconium dichloride,
- rac-Diethylsilylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium dichloride,
- rac-Di(i-propyl)silylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium dichloride,
- 25 rac-Di(n-butyl)silylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium dichloride,
- rac-Di(cyclohexyl)silylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium dichloride,

- rac-Methylphenylsilylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium dichloride,
- rac-Methylphenylsilylene-bis(2,7-dimethyl-4-t-butyl-1-indenyl)zirconium dichloride,
- 5 rac-Diphenylsilylene-bis(2,7-dimethyl-4-t-butyl-1-indenyl)zirconium dichloride,
- rac-Diphenylsilylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium dichloride,
- rac-Diphenylsilylene-bis(2,7-dimethyl-4-ethyl-1-indenyl)zirconium dichloride,
- 10 rac-Di(p-tolyl)silylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium dichloride,
- rac-Di(p-chlorophenyl)silylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium dichloride,
- 15 rac-Dimethylsilylene-bis(2-methyl-4-i-propyl-7-ethyl-1-indenyl)zirconium dibromide,
- rac-Dimethylsilylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium dimethyl,
- rac-Dimethylsilylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium methylchloride,
- 20 rac-Dimethylsilylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium-bis(trifluoromethanesulfonato),
- rac-Dimethylsilylene-bis(2,7-dimethyl-4-i-propyl-1-indenyl)zirconium-bis(p-phenylsulfinato), and
- 25 rac-Dimethylsilylene-bis(2-phenyl-4-i-propyl-7-methyl-1-indenyl)zirconium dichloride.

Of the transition metal compounds represented by the above formula [I], a transition metal compound represented

by the following formula [I-a] is particularly preferably used in the invention.



[I-a]

5

wherein M, X¹, X², R¹, R³ and Y are the same as those in the formula [I], and further R¹ is preferably hydrogen atom, methyl or an aromatic group.

Listed below are preferred examples of the transition
10 metal compounds represented by the above formula [I-a].

rac-Dimethylsilylene-bis(4-phenyl-1-indenyl)zirconium
dichloride,

rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-
indenyl)zirconium dichloride,

15 rac-Dimethylsilylene-bis(2-methyl-4-(α -naphthyl)-1-
indenyl)zirconium dichloride,

rac-Dimethylsilylene-bis(2-methyl-4-(β -naphthyl)-1-
indenyl)zirconium dichloride,

rac-Dimethylsilylene-bis(2-methyl-4-(1-anthracenyl)-1-
20 indenyl)zirconium dichloride,

rac-Dimethylsilylene-bis(2-methyl-4-(2-anthracenyl)-1-
indenyl)zirconium dichloride,

- rac-Dimethylsilylene-bis(2-methyl-4-(9-anthracenyl)-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-(9-phenanthryl)-1-indenyl)zirconium dichloride,
- 5 rac-Dimethylsilylene-bis(2-methyl-4-(p-fluorophenyl)-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-(pentafluorophenyl)-1-indenyl)zirconium dichloride,
- 10 rac-Dimethylsilylene-bis(2-methyl-4-(p-chlorophenyl)-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-(m-chlorophenyl)-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-(o-chlorophenyl)-1-indenyl)zirconium dichloride,
- 15 rac-Dimethylsilylene-bis(2-methyl-4-(o,p-dichlorophenyl)phenyl-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-(p-bromophenyl)-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-(p-tolyl)-1-indenyl)zirconium dichloride,
- 20 rac-Dimethylsilylene-bis(2-methyl-4-(m-tolyl)-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-(o-tolyl)-1-indenyl)zirconium dichloride,
- 25 rac-Dimethylsilylene-bis(2-methyl-4-(o,o'-dimethylphenyl)-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-(p-ethylphenyl)-1-indenyl)zirconium dichloride,

- rac-Dimethylsilylene-bis(2-methyl-4-(p-i-propylphenyl)-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-(p-benzylphenyl)-1-indenyl)zirconium dichloride,
- 5 rac-Dimethylsilylene-bis(2-methyl-4-(p-biphenyl)-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-(m-biphenyl)-1-indenyl)zirconium dichloride,
- 10 rac-Dimethylsilylene-bis(2-methyl-4-(p-trimethylsilylphenyl)-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-(m-trimethylsilylphenyl)-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-phenyl-4-phenyl-1-indenyl)zirconium dichloride,
- 15 rac-Diethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,
- rac-Di(i-propyl)silylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,
- rac-Di(n-butyl)silylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,
- 20 rac-Dicyclohexylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,
- rac-Methylphenylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,
- 25 rac-Diphenylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,
- rac-Di(p-tolyl)silylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,

- rac-Di(p-chlorophenyl)silylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,
- rac-Methylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,
- 5 rac-Ethylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,
- rac-Dimethylgermylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,
- 10 rac-Dimethylstannylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dichloride,
- rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dibromide,
- rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium dimethyl,
- 15 rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium methylchloride,
- rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium chloride SO₂Me,
- 20 rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium chloride OSO₂Me,
- rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium monochloride mono(trifluoromethanesulfonate),
- 25 rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium di(trifluoromethanesulfonato),
- rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium di(p-toluenesulfonato),

rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium di(methylsulfonate),

rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium di(trifluoromethanesulfinato),

5 rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium di(trifluoroacetate),

rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium monochloride (n-butoxide),

10 rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium di(n-butoxide), and

rac-Dimethylsilylene-bis(2-methyl-4-phenyl-1-indenyl)zirconium monochloride(phenoxide).

Of the above compounds, particularly preferred are compounds of the formula [I-a] wherein R¹ is methyl.

15 Also preferably used are transition metal compounds of the formula [I-a] wherein R¹ is a hydrocarbon group of 2 to 6 carbon atoms and R³ is an aryl group of 6 to 16 carbon atoms.

Preferred examples of such compounds of the formula [I] are listed below.

rac-Dimethylsilylene-bis{1-(2-ethyl-4-phenylindenyl)}zirconium dichloride,

rac-Dimethylsilylene-bis{1-(2-ethyl-4-(α -naphthyl)indenyl)}zirconium dichloride,

25 rac-Dimethylsilylene-bis{1-(2-ethyl-4-(β -naphthyl)indenyl)}zirconium dichloride,

rac-Dimethylsilylene-bis{1-(2-ethyl-4-(2-methyl-1-naphthyl)indenyl)}zirconium dichloride,

- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(5-acenaphthyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(9-anthracenyl)indenyl)}zirconium dichloride,
- 5 rac-Dimethylsilylene-bis{1-(2-ethyl-4-(9-phenanthryl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(o-methylphenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(m-
- 10 methylphenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(p-methylphenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(2,3-dimethylphenyl)indenyl)}zirconium dichloride,
- 15 rac-Dimethylsilylene-bis{1-(2-ethyl-4-(2,4-dimethylphenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(2,5-dimethylphenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(2,4,6-
- 20 trimethylphenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(o-chlorophenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(m-chlorophenyl)indenyl)}zirconium dichloride,
- 25 rac-Dimethylsilylene-bis{1-(2-ethyl-4-(p-chlorophenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(2,3-dichlorophenyl)indenyl)}zirconium dichloride,

- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(2,6-dichlorophenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(3,5-dichlorophenyl)indenyl)}zirconium dichloride,
- 5 rac-Dimethylsilylene-bis{1-(2-ethyl-4-(2-bromophenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(3-bromophenyl)indenyl)}zirconium dichloride,
- 10 rac-Dimethylsilylene-bis{1-(2-ethyl-4-(4-bromophenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(4-biphenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-ethyl-4-(4-trimethylsilylphenyl)indenyl)}zirconium dichloride,
- 15 rac-Dimethylsilylene-bis{1-(2-n-propyl-4-phenylindenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-n-propyl-4-(α -naphthyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-n-propyl-4-(β -naphthyl)indenyl)}zirconium dichloride,
- 20 rac-Dimethylsilylene-bis{1-(2-n-propyl-4-(2-methyl-1-naphthyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-n-propyl-4-(5-acenaphthyl)indenyl)}zirconium dichloride,
- 25 rac-Dimethylsilylene-bis{1-(2-n-propyl-4-(9-anthracenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-n-propyl-4-(9-phenanthryl)indenyl)}zirconium dichloride,

- rac-Dimethylsilylene-bis{1-(2-i-propyl-4-phenylindenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-i-propyl-4-(α -naphthyl)indenyl)}zirconium dichloride,
- 5 rac-Dimethylsilylene-bis{1-(2-i-propyl-4-(β -naphthyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-i-propyl-4-(2-methyl-1-naphthyl)indenyl)}zirconium dichloride,
- 10 rac-Dimethylsilylene-bis{1-(2-i-propyl-4-(5-acenaphthyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-i-propyl-4-(9-anthracenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-i-propyl-4-(9-phenanthryl)indenyl)}zirconium dichloride,
- 15 rac-Dimethylsilylene-bis{1-(2-s-butyl-4-phenylindenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-s-butyl-4-(α -naphthyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-s-butyl-4-(β -naphthyl)indenyl)}zirconium dichloride,
- 20 rac-Dimethylsilylene-bis{1-(2-s-butyl-4-(8-methyl-9-naphthyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-s-butyl-4-(5-acenaphthyl)indenyl)}zirconium dichloride,
- 25 rac-Dimethylsilylene-bis{1-(2-s-butyl-4-(9-anthracenyl)indenyl)}zirconium dichloride,
- rac-Dimethylsilylene-bis{1-(2-s-butyl-4-(9-phenanthryl)indenyl)}zirconium dichloride,

- rac-Dimethylsilylene-bis(1-(2-n-pentyl-4-phenylindenyl))zirconium dichloride,
 rac-Dimethylsilylene-bis(1-(2-n-pentyl-4-(α -naphthyl)indenyl))zirconium dichloride,
- 5 rac-Dimethylsilylene-bis(1-(2-n-butyl-4-phenylindenyl))zirconium dichloride,
 rac-Dimethylsilylene-bis(1-(2-n-butyl-4-(α -naphthyl)indenyl))zirconium dichloride,
 rac-Dimethylsilylene-bis(1-(2-n-butyl-4-(β -
- 10 naphthyl)indenyl))zirconium dichloride,
 rac-Dimethylsilylene-bis(1-(2-n-butyl-4-(2-methyl-1-naphthyl)indenyl))zirconium dichloride,
 rac-Dimethylsilylene-bis(1-(2-n-butyl-4-(5-acenaphthyl)indenyl))zirconium dichloride,
- 15 rac-Dimethylsilylene-bis(1-(2-n-butyl-4-(9-anthracenyl)indenyl))zirconium dichloride,
 rac-Dimethylsilylene-bis(1-(2-n-butyl-4-(9-phenanthryl)indenyl))zirconium dichloride,
 rac-Dimethylsilylene-bis(1-(2-i-butyl-4-
- 20 phenylindenyl))zirconium dichloride,
 rac-Dimethylsilylene-bis(1-(2-i-butyl-4-(α -naphthyl)indenyl))zirconium dichloride,
 rac-Dimethylsilylene-bis(1-(2-i-butyl-4-(β -naphthyl)indenyl))zirconium dichloride,
- 25 rac-Dimethylsilylene-bis(1-(2-i-butyl-4-(2-methyl-1-naphthyl)indenyl))zirconium dichloride,
 rac-Dimethylsilylene-bis(1-(2-i-butyl-4-(5-acenaphthyl)indenyl))zirconium dichloride,

- rac-Dimethylsilylene-bis(1-(2-i-butyl-4-(9-anthracenyl)indenyl))zirconium dichloride,
- rac-Dimethylsilylene-bis(1-(2-i-butyl-4-(9-phenanthryl)indenyl))zirconium dichloride,
- 5 rac-Dimethylsilylene-bis(1-(2-neopentyl-4-phenylindenyl))zirconium dichloride,
- rac-Dimethylsilylene-bis(1-(2-neopentyl-4-(α -naphthyl)indenyl))zirconium dichloride,
- rac-Dimethylsilylene-bis(1-(2-n-hexyl-4-phenylindenyl))zirconium dichloride,
- 10 rac-Dimethylsilylene-bis(1-(2-n-hexyl-4-(α -naphthyl)indenyl))zirconium dichloride,
- rac-Methylphenylsilylene-bis(1-(2-ethyl-4-phenylindenyl))zirconium dichloride,
- 15 rac-Methylphenylsilylene-bis(1-(2-ethyl-4-(α -naphthyl)indenyl))zirconium dichloride,
- rac-Methylphenylsilylene-bis(1-(2-ethyl-4-(9-anthracenyl)indenyl))zirconium dichloride,
- rac-Methylphenylsilylene-bis(1-(2-ethyl-4-(9-phenanthryl)indenyl))zirconium dichloride,
- 20 rac-Diphenylsilylene-bis(1-(2-ethyl-4-phenylindenyl))zirconium dichloride,
- rac-Diphenylsilylene-bis(1-(2-ethyl-4-(α -naphthyl)indenyl))zirconium dichloride,
- 25 rac-Diphenylsilylene-bis(1-(2-ethyl-4-(9-anthracenyl)indenyl))zirconium dichloride,
- rac-Diphenylsilylene-bis(1-(2-ethyl-4-(9-phenanthryl)indenyl))zirconium dichloride,

- rac-Diphenylsilylene-bis{1-(2-ethyl-4-(4-biphenyl)indenyl)}zirconium dichloride,
- rac-Methylene-bis{1-(2-ethyl-4-phenylindenyl)}zirconium dichloride,
- 5 rac-Methylene-bis{1-(2-ethyl-4-(α -naphthyl)indenyl)}zirconium dichloride,
- rac-Ethylene-bis{1-(2-ethyl-4-phenylindenyl)}zirconium dichloride,
- rac-Ethylene-bis{1-(2-ethyl-4-(α -naphthyl)indenyl)}zirconium dichloride,
- 10 rac-Ethylene-bis{1-(2-n-propyl-4-(α -naphthyl)indenyl)}zirconium dichloride,
- rac-Dimethylgermylene-bis{1-(2-ethyl-4-phenylindenyl)}zirconium dichloride,
- 15 rac-Dimethylgermylene-bis{1-(2-ethyl-4-(α -naphthyl)indenyl)}zirconium dichloride,
- rac-Dimethylgermylene-bis{1-(2-n-propyl-4-phenylindenyl)}zirconium dichloride,
- rac-Dimethylstannylene-bis{1-(2-ethyl-4-phenylindenyl)}zirconium dichloride,
- 20 rac-Dimethylstannylene-bis{1-(2-ethyl-4-(α -naphthyl)indenyl)}zirconium dichloride,
- rac-Dimethylstannylene-bis{1-(2-n-ethyl-4-(9-phenanthryl)indenyl)}zirconium dichloride, and
- 25 rac-Dimethylstannylene-bis{1-(2-n-propyl-4-phenylindenyl)}zirconium dichloride.

In the present invention, also employable are transition metal compounds obtained by replacing zirconium

metal with metals of titanium, hafnium, vanadium, niobium, tantalum, chromium, molybdenum and tungsten in the above-exemplified compounds.

5 The transition metal compound is generally used as the olefin polymerization catalyst component in the form of racemic modification, and it can be used either in the form of R type or S type.

The transition metal compound used in the invention can be prepared in accordance with "Journal of
10 Organometallic Chem.", 288 (1985), pp. 63-67 and European Patent Publication No. 0,320,762.

The organoaluminum oxy-compound [B-1] (hereinafter sometimes referred to as "component [B-1]") forming the olefin polymerization catalyst used in the invention may be
15 either conventionally known aluminoxane or such a benzene-insoluble organoaluminum oxy-compound as exemplified in Japanese Patent Laid-Open Publication No. 78687/1990.

The conventionally known aluminoxane can be prepared by, for example, the following procedures.

20 (1) Adding an organoaluminum compound, e.g., trialkylaluminum, to a hydrocarbon medium suspension of compounds containing adsorbed water or salts containing water of crystallization, e.g., magnesium chloride hydrate, copper sulfate hydrate, aluminum sulfate hydrate, nickel
25 sulfate hydrate and cerous chloride hydrate, so as to allow the organoaluminum compound to react with the adsorbed water or the water of crystallization.

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

- 5 (3) Allowing organotin oxide such as dimethyltin oxide or dibutyltin oxide to react with an organoaluminum compound, e.g., 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 solution after recovery of aluminoxane and the residue is redissolved in a solvent or suspended in a poor solvent for aluminoxane.

- 15 Examples of the organoaluminum compounds used for preparing aluminoxane include:

trialkylaluminums, such as trimethylaluminum, triethylaluminum, tripropylaluminum, triisopropylaluminum, tri-n-butylaluminum, triisobutylaluminum, tri-sec-butylaluminum, tri-tert-butylaluminum, tripentylaluminum, trihexylaluminum, trioctylaluminum and tridecylaluminum;

20 tricycloalkylaluminums, such as tricyclohexylaluminum and tricyclooctylaluminum;

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

25 dialkylaluminum hydrides, such as diethylaluminum hydride and diisobutylaluminum hydride;

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

dialkylaluminum aryloxides, such as diethylaluminum phenoxide.

- 5 Of these, preferred are trialkylaluminums and tricycloalkylaluminums, and particularly preferred is trimethylaluminum.

Also employable as the organoaluminum compound used for preparing aluminoxane is isoprenylaluminum represented
10 by the following formula [II]:



wherein x, y and z are each a positive number, and $z \geq 2x$.

The organoaluminum compounds mentioned above are used singly or in combination.

- 15 Examples of the solvents used for preparing aluminoxane include aromatic hydrocarbons, such as benzene, toluene, xylene, cumene and cymene; aliphatic hydrocarbons, such as pentane, hexane, heptane, octane, decane, dodecane, hexadecane and octadecane; alicyclic hydrocarbons, such as
20 cyclopentane, cyclohexane, cyclooctane and methylcyclopentane; petroleum fractions, such as gasoline, kerosine and gas oil; and halides of these aromatic, aliphatic and alicyclic hydrocarbons, particularly chlorides and bromides thereof. Also employable are ethers
25 such as ethyl ether and tetrahydrofuran. Of the solvents, preferred are aromatic hydrocarbons and aliphatic hydrocarbons.

The compound [B-2] which reacts with the transition metal compound [A] to form an ion pair (hereinafter sometimes referred to as "component [B-2]"), that is used for forming the olefin polymerization catalyst used in the invention, includes Lewis acid, ionic compounds and carborane compounds, as 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. 5321106.

10 Examples of the Lewis acid include triphenylboron, tris(4-fluorophenyl)boron, tris(p-tolyl)boron, tris(o-tolyl)boron, tris(3,5-dimethylphenyl)boron, tris(pentafluorophenyl)boron, MgCl_2 , Al_2O_3 and $\text{SiO}_2\text{-Al}_2\text{O}_3$.

15 Examples of the ionic compounds include triphenylcarbeniumtetrakis(pentafluorophenyl)borate, tri-n-butylammoniumtetrakis(pentafluorophenyl)borate, N,N-dimethylaniliniumtetrakis(pentafluorophenyl)borate and ferroceniumtetrakis(pentafluorophenyl)borate.

20 Examples of the carborane compounds include dodecaborane, 1-carbaundecaborane, bis-n-butylammonium(1-carbedodeca)borate, tri-n-butylammonium(7,8-dicarbaundeca)borate and tri-n-butylammonium(tridecahydride-7-carbaundeca)borate.

25 The compounds [B-2] which react with the transition metal compound [A] to form an ion pair can be used in combination of two or more kinds.

The organoaluminum compound [C] (hereinafter sometimes referred to as "component [C]") for forming the olefin

polymerization catalyst used in the invention is, for example, an organoaluminum compound represented by the following formula [III]:



5 wherein R^9 is a hydrocarbon group of 1 to 12 carbon atoms, X is a halogen atom or a hydrogen atom, and n is 1 to 3.

In the above formula [III], R^9 is a hydrocarbon group of 1 to 12 carbon atoms, for example, an alkyl group, a cycloalkyl group or an aryl group. Particular examples
10 thereof include methyl, ethyl, n-propyl, isopropyl, isobutyl, pentyl, hexyl, octyl, cyclopentyl, cyclohexyl, phenyl and tolyl.

Examples of such organoaluminum compound [C] include:

trialkylaluminums, such as trimethylaluminum,
15 triethylaluminum, triisopropylaluminum, triisobutylaluminum, trioctylaluminum, tri(2-ethylhexyl)aluminum and tridecylaluminum;

alkenylaluminums, such as isoprenylaluminum;

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

alkylaluminum sesquihalides, such as methylaluminum
sesquichloride, ethylaluminum sesquichloride,
25 isopropylaluminum sesquichloride, butylaluminum sesquichloride and ethylaluminum sesquibromide;

alkylaluminum dihalides, such as methylaluminum dichloride, ethylaluminum dichloride, isopropylaluminum dichloride and ethylaluminum dibromide; and

alkylaluminum hydrides, such as diethylaluminum
5 hydride and diisobutylaluminum hydride.

Also employable as the organoaluminum compound [C] is a compound represented by the following formula [IV]:



wherein R^9 is the same as above; L is $-OR^{10}$ group, $-OSiR^{11}_3$
10 group, $-OAlR^{12}_2$ group, $-NR^{13}_3$ group, $-SiR^{14}_3$ group or
 $-N(R^{15})AlR^{16}_2$ group; n is 1 to 2; R^{10} , R^{11} , R^{12} and R^{16} are
each methyl, ethyl, isopropyl, isobutyl, cyclohexyl, phenyl
or the like, R^{13} is hydrogen, methyl, ethyl, isopropyl,
phenyl, trimethylsilylene or the like; and R^{14} and R^{15} are
15 each methyl, ethyl or the like.

Of such organoaluminum compounds, preferred are compounds of $R^9_n Al(OAlR^{10}_2)_{3-n}$, for example, $Et_2AlOAlEt_2$ and $(iso-Bu)_2AlOAl(iso-Bu)_2$.

Of the organoaluminum compounds represented by the
20 formulas [III] and [IV], preferred are compounds of the
formula R^9_3Al , and particularly preferred are compounds of
said formula wherein R^9 is an isoalkyl group.

The olefin polymerization catalyst used in the
invention comprises the component [A], the component [B-1]
25 and/or the component [B-2] (hereinafter referred to as the
component [B]), and if desired, the component [C]. The
olefin polymerization catalyst can be prepared by mixing

the components [A] and [B], and if desired, the component [C] in an inert hydrocarbon solvent or an olefin solvent.

Examples of the inert hydrocarbon solvents used for preparing the olefin polymerization catalyst include

- 5 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
- 10 as ethylene chloride, chlorobenzene and dichloromethane; and mixtures of these hydrocarbons.

The order of mixing the above components may be arbitrarily determined, but preferred is:

- mixing the component [B] with the component [A];
- 15 mixing first the component [B] with the component [C] and then with the component [A];
- mixing first the component [A] with the component [B] and then with the component [C]; or
- mixing first the component [A] with the component [C]
- 20 and then with the component [B].

- In the mixing of the above components, when the component [B-1] is used, the atomic ratio of aluminum in the component [B-1] to the transition metal in the transition metal compound [A], (Al/transition metal), is in
- 25 the range of usually 10 to 10,000, preferably 20 to 5,000, and the concentration of the component [A] is in the range of about 10^{-8} to 10^{-1} mol/liter, preferably 10^{-7} to 5×10^{-2} mol/liter.

When the component [B-2] is used, the molar ratio of the component [A] to the component [B-2], (component [A]/component [B-2]), is in the range of usually 0.01 to 10, preferably 0.1 to 5, and the concentration of the
5 component [A] is in the range of about 10^{-8} to 10^{-1} mol/liter, preferably 10^{-7} to 5×10^{-2} mol/liter.

When the component [C] is used, the atomic ratio of the aluminum atom (Al_c) in the component [C] to the aluminum atom (Al_{B-1}) in the component [B-1], (Al_c/Al_{B-1}), is
10 in the range of usually 0.02 to 20, preferably 0.2 to 10.

The above-mentioned catalyst components may be mixed in a polymerizer, or a mixture of the components preliminarily prepared may be added to a polymerizer.

If the components are preliminarily mixed, the mixing
15 temperature is in the range of usually -50 to 150 °C, preferably -20 to 120 °C; and the contact time is in the range of 1 to 1,000 minutes, preferably 5 to 600 minutes. The mixing temperature may be varied during the mixing procedure.

20 The olefin polymerization catalyst used in the invention may be a solid olefin polymerization catalyst in which at least one of the above components [A], [B] and [C] is supported on an inorganic or organic, granular or particulate solid carrier.

25 The inorganic carrier is preferably a porous oxide, and examples thereof include SiO_2 and Al_2O_3 .

Examples of the granular or particulate solid organic compounds include polymers or copolymers produced by using

α -olefins (e.g., ethylene, propylene and 1-butene) or styrene, as major component.

In the present invention, the above-described components can be used after prepolymerization of an olefin
5 with those components.

Examples of the olefins preferably used in the prepolymerization include propylene, ethylene and 1-butene. Mixtures of these olefins and other olefins are also employable.

10 The olefin polymerization catalyst used in the invention can contain other components than the above-mentioned ones, which are useful for the olefin polymerization, e.g., water as a catalyst component.

The propylene/1-butene random copolymer (A) used in
15 the invention can be prepared by copolymerizing propylene and 1-butene in the presence of the olefin polymerization catalyst in such a manner that the aforesaid composition ratio is finally obtained.

The copolymerization can be carried out by any of a
20 liquid phase polymerization process such as suspension polymerization or solution polymerization and a gas phase polymerization process.

In the solution polymerization, the same inert hydrocarbon solvent as used for preparing the catalyst is
25 employable as the solvent, or propylene is also employable as the solvent.

In the suspension polymerization, the polymerization temperature is usually -50 to 100 °C, preferably 0 to 90 °C; in the solution polymerization, it is usually 0 to 250 °C, preferably 20 to 200 °C; and in the gas phase
5 polymerization, it is usually 0 to 120 °C, preferably 20 to 100 °C.

The polymerization pressure is usually atmospheric pressure to 100 kg/cm², preferably atmospheric pressure to 50 kg/cm². The polymerization reaction can be carried out
10 batchwise, semi-continuously or continuously.

Further, the polymerization reaction can be carried out in two or more stages under different reaction conditions.

The molecular weight of the resulting propylene/1-
15 butene random copolymer can be adjusted by allowing hydrogen to exist in the polymerization system or by varying the polymerization temperature or the polymerization pressure.

To the propylene/1-butene random copolymer (A) may be
20 added additives, such as heat stabilizer, ultraviolet light absorber, anti-blocking agent, slip agent and antistatic agent.

Ethylene/ α -olefin copolymer (B)

The ethylene/ α -olefin copolymer (B) used in the
25 invention comprises ethylene and an α -olefin of 3 to 10 carbon atoms.

In the ethylene/ α -olefin copolymer (B), the content of constituent units derived from ethylene (ethylene content) is 75 to 95 % by mol, preferably 80 to 91 % by mol.

Examples of the α -olefins of 3 to 10 carbon atoms include propylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 3-methyl-1-pentene, 1-heptene, 1-octene, 1-nonene, 1-decene, 1-dodecene, 1-tetradecene and 1-octadecene.

The ethylene/ α -olefin copolymer (B) has a melt flow rate (MFR, ASTM D 1238, 190 °C, load of 2.16 kg) of 0.1 to 100 g/10 min, preferably 0.2 to 50 g/10 min, more preferably 0.4 to 20 g/10 min.

Use of the ethylene/ α -olefin copolymer (B) can impart tackiness to the composition of the propylene/1-butene random copolymer (A) and the copolymer (B).

The ethylene/ α -olefin copolymer (B) can be prepared by the use of a vanadium catalyst or a metallocene catalyst conventionally known.

Tackifier (C)

In the present invention, a tackifier (C) may be used to give moderate initial adhesive strength between a base resin comprising the propylene/1-butene random copolymer (A) and the ethylene/ α -olefin copolymer (B) and an adherend.

This tackifier (C) is a solid, amorphous polymer, and is a tackifying resin conventionally used in various fields such as fields of adhesive tapes, paints and hot melt

adhesives. As the tackifier, the below-described various resins, which are classified based on the difference between monomers polymerized, are employable.

For example, there can be mentioned:

- 5 aliphatic hydrocarbon resins prepared by using, as main starting materials, a C₄ fraction and a C₅ fraction obtained by cracking petroleum, naphtha or the like, mixtures thereof, and isoprene and 1,3-pentadiene in an optional fraction thereof such as a C₅ fraction;
- 10 aromatic hydrocarbon resins prepared by using, as main starting materials, styrene derivatives and indenenes in a C₉ fraction obtained by cracking petroleum, naphtha or the like;
- 15 aliphatic/aromatic copolymerization hydrocarbon resins prepared by copolymerizing an optional fraction of a C₄ fraction and a C₅ fraction with a C₉ fraction.
- 20 alicyclic hydrocarbon resins prepared by hydrogenating aromatic hydrocarbon resins;
- 20 synthetic terpene hydrocarbon resins having structures containing aliphatic, alicyclic and aromatic groups;
- 20 terpene hydrocarbon resins prepared by using, as starting materials, α,β -pinenes in turpentine oil;
- 25 coumarone-indene hydrocarbon resins prepared by using, as starting materials, indenenes and styrenes in coal tar naphtha;
- 25 low-molecular weight styrene resins; and
- 25 rosin hydrocarbon resins.

Of these tackifiers (C), the aliphatic hydrocarbon resins and the alicyclic hydrocarbon resins prepared by hydrogenating aromatic hydrocarbon resins are preferable because they have good compatibility with the component (A). Among them, more preferable are alicyclic hydrocarbon resins having a softening point (measured by a ring and ball method) of 70 to 150 °C, preferably 80 to 140 °C, and having a hydrogenation degree of aromatic groups of not less than 80 %, preferably not less than 85 %.

10

Composition

In the composition for a peelable protective film according to the invention, the propylene/1-butene random copolymer (A) is used in an amount of 30 to 100 parts by weight, preferably 30 to 99 parts by weight, more preferably 30 to 85 parts by weight, still more preferably 35 to 80 parts by weight, most preferably 35 to 75 parts by weight, the ethylene/ α -olefin copolymer (B) is used in an amount of 0 to 70 parts by weight, preferably 1 to 70 parts by weight, more preferably 15 to 70 parts by weight, still more preferably 20 to 65 parts by weight, most preferably 25 to 65 parts by weight, and the tackifier (C) is used in an amount of 0 to 20 parts by weight, preferably 2 to 15 parts by weight, more preferably 3 to 10 parts by weight, each based on 100 parts by weight of the total of the propylene/1-butene random copolymer (A), the ethylene/ α -olefin copolymer (B) and the tackifier (C).

20

25

The composition of the invention comprises the components (A), (B) and (C) in the above-mentioned

proportions, so that the film obtained from the composition has moderate initial adhesive strength, small change in the adhesive strength with time and good peelability even after bonded, and therefore the film is free from blocking or
5 bleeding after peeling.

The composition for a peelable protective film according to the invention can be blended with other resin components within limits not prejudicial to the objects of the present invention. Further, to the composition of the
10 invention can be added various additives, such as antioxidant, ultraviolet light absorber, lubricant, nucleating agent, antistatic agent, flame retarder, pigment, dye and inorganic or organic filler, within limits not prejudicial to the objects of the present invention.

15 The composition for a peelable protective film according to the invention can be prepared by an optional method conventionally known. For example, it can be prepared by mixing the components by means of mixing machines such as twin-cylinder mixer, ribbon blender,
20 tumbling blender and Henschel mixer, and/or by kneading the components by means of kneading machines such as extruder, mixing roll, Banbury mixer and kneader.

Peelable protective film

The peelable protective film of the invention uses a
25 film formed from the above-described composition. The peelable protective film of the invention may be a single-layer film formed from the composition, or may be a laminated film consisting of a layer of the composition and

other resin layer (film). When the peelable protective film of the invention is a laminated film having other resin layer, the layer of the composition is laminated in such a manner that this layer is positioned on at least one
5 of the outermost sides to form an adhesive layer bonded to an adherend.

As the other resin film serving as a base layer, a film of polyolefin having good adhesion to the composition of the invention, e.g., polyethylene, polypropylene or
10 poly-1-butene, is preferably employed. Further, a film of polyamide, polyester or the like may be laminated through other adhesive layer.

The peelable protective film of the invention can be produced by molding the composition into a single-layer
15 film or by co-extruding the composition and other resin to give a laminated film. In the production of the film, a film molding machine such as a T-die molding machine or an inflation molding machine is employed.

The peelable protective film produced as above is used
20 to cover the surfaces of various articles such as metal articles, plastic articles and rubber articles so that the layer of the composition of the invention serves as an adhesive layer, and thereby prevent scratches, contamination or corrosion of those articles when they are
25 worked, transported or stored.

In the peelable protective film of the invention, the adhesive layer formed from the composition of the invention has moderate adhesive strength, so that covering by

adhesion bonding or peeling is easily made. Besides, the film has a small change in the adhesive strength with time because of good heat resistance, so that the adhesion properties and peelability can last for a long period of time. Further, since the film of the invention has excellent scratch resistance and transparency and is free from blocking, the article covered with the film can be readily worked even in the bonded state and has good appearance.

10

EFFECT OF THE INVENTION

From the composition for a peelable protective film according to the invention, there can be obtained a peelable protective film showing excellent flexibility, scratch resistance, heat resistance and transparency, having moderate initial adhesive strength to an adherend and peelability from the adherend, having good cold workability even in the bonded state, being almost free from a change in the adhesive strength with time when stored for a long period of time, and leaving no bleed substance on a surface of an adherend even when the film is peeled from the adherend.

The peelable protective film of the invention, which exerts such effects as mentioned above, can be widely used. For example, the film can be suitably used to cover metallic, plastic or rubber surfaces of various articles such as electrical appliances, automobiles, glass articles, building materials and general merchandise.

EXAMPLE

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

Examples 1 and 2Preparation of propylene/1-butene random copolymer

A 2 liter autoclave thoroughly purged with nitrogen was charged with 900 ml of hexane and 50 g of 1-butene. To the autoclave was added 1 mmol of triisobutylaluminum, and the temperature of the system was elevated to 70 °C. Then, propylene was fed so that the total pressure of the system became 7 kg/cm²-G. To the system were added 0.30 mmol of methylaluminoxane and 0.001 mmol (in terms of Zr atom) of rac-dimethylsilylene-bis{1-(2-methyl-4-phenylindenyl)}zirconium dichloride. Further, propylene was continuously fed to perform polymerization for 30 minutes, while the total pressure of the system was maintained at 7 kg/cm²-G. After the polymerization, the system was degassed, and a polymer produced was recovered in a large amount of methanol. The polymer was vacuum dried at 110 °C for 12 hours.

The propylene/1-butene random copolymer thus obtained had a 1-butene content of 15 % by mol (i.e., propylene content of 85 % by mol), a melt flow rate (230 °C, 2.16 kg) of 8.0 g/10 min, a melting point (T_m) of 110 °C, a

molecular weight distribution (M_w/M_n) of 2.18, a B value of 1.03 and a crystallinity of 53 %.

Preparation of ethylene/1-butene random copolymer

5 A mixture of ethylene and 1-butene was polymerized in a hexane solvent and in the presence of hydrogen using vanadium oxytrichloride and ethylenealuminum sesquichloride as catalysts.

The ethylene/1-butene random copolymer obtained above had an ethylene content of 89 % by mol, a density of 0.885 g/cm³ and a melt flow rate (190 °C, 2.16 kg) of 3.5 g/10 min.

Preparation of composition and film

15 The propylene/1-butene random copolymer (hereinafter referred to as "PBC-1") and the ethylene/1-butene random copolymer (hereinafter referred to as "EBC") were blended in a blending ratio shown in Table 1 for 2 minutes by means of a tumbling blender. The mixture was melt kneaded by an extruder (diameter: 40 mm, preset temperature: 230 °C) to prepare a composition.

20 Then, the composition was fed to a T-die molding machine (preset temperature: 230 °C) to produce a single-layer film having a thickness of 50 μ m.

The properties of the composition and the film were evaluated in accordance with the following methods.

25 (1) Adhesive strength to adherend

Onto an acrylic resin substrate preheated to 55 °C, the film prepared above was bonded by means of pinch rolls under a line pressure of 3.6 kg/cm. The substrate with the

film was allowed to stand in an air oven at 50 °C for a given time. Then, the adhesive strength of the film was measured in accordance with a 180° peel test (rate of pulling: 300 mm/min).

5 (2) Surface hardness

From the composition, a pressed sheet having a thickness of 2 mm was prepared (200 °C, quenching method). The surface hardness of the pressed sheet was measured in accordance with ASTM D 2240.

10 (3) Surface appearance of adherend

After the film was peeled from the substrate (adherend) in the measurement of the adhesive strength, the surface appearance of the adherend was visually observed and evaluated based on the following criteria.

15 AA: No stain was observed.

BB: Slight stain was observed.

CC: Stain was observed.

The results are set forth in Table 1.

20 Example 3

The procedures of Example 1 were repeated except that a composition prepared by blending PBC-1 and EBC of Example 1 and an alicyclic hydrocarbon resin (trade name: ARKON P125, available from Arakawa Chemical Industries, Ltd.) in
25 a blending ratio shown in Table 1 was used.

The results are set forth in Table 1.

Example 4

The procedures of Example 1 were repeated except that an ethylene/1-octene copolymer (hereinafter referred to as "EOC") obtained by polymerizing a mixture of ethylene and 1-octene using a metallocene catalyst was used in place of
5 the ethylene/1-butene random copolymer of Example 1.

The EOC used herein had an ethylene content of 90 % by mol, a density of 0.885 g/cm³ and a melt flow rate (190 °C, 2.16 kg) of 4.7 g/10 min.

The results are set forth in Table 1.

10

Example 5

The procedures of Example 1 were repeated except that PBC-1 of Example 1 was used singly in place of the composition of Example 1.

15

The results are set forth in Table 1.

Comparative Example 1

A composition was prepared in the same manner as in Example 1 except that the blending ratio between PBC-1 and
20 EBC was varied to that shown in Table 1. The subsequent procedures were carried out in the same manner as in Example 1.

The results are set forth in Table 1.

25 Comparative Example 2

A composition was prepared in the same manner as in Example 3 except that a propylene/1-butene random copolymer (PBC-2) prepared by the below-described process was used in

place of PBC-1. The subsequent procedures were carried out in the same manner as in Example 3.

The results are set forth in Table 1.

Preparation of propylene/1-butene random copolymer (PBC-2)

- 5 In a 20 liter stainless steel polymerizer equipped with a stirring blade, 200 g of anhydrous magnesium chloride, 46 ml of ethylene benzoate and 30 ml of methylpolysiloxane as catalyst components were ball-milled in an atmosphere of nitrogen.
- 10 The catalyst components ball-milled as above were suspended in titanium tetrachloride and filtered. Then, the filtration product was fed so that the titanium concentration became 0.01 mmol/l, triethylaluminum was further fed so that the triethylaluminum concentration in
- 15 the polymerizer became 1.0 mmol/l, and methyl p-toluate was furthermore fed as an electron donor so that the methyl p-toluate concentration in the polymerizer became 0.33 mmol/l. Simultaneously, a mixed gas of propylene and 1-butene (propylene: 68 % by mol, 1-butene: 32 % by mol) was
- 20 fed at a rate of 4 kl/hr to perform copolymerization reaction at 70 °C using n-heptane as a polymerization solvent.

- The propylene/1-butene random copolymer obtained above had a propylene content, as measured by nuclear magnetic
- 25 resonance spectrum, of 71.0 % by mol, a melt flow rate (ASTM D 1238, 230 °C, 2.16 kg) of 7.0 g/10 min, a melting point (T_m) of 110 °C, a molecular weight distribution

(Mw/Mn) of 4.52, a B value of 0.92, and a crystallinity of 48 %.

Table 1

	Ex.1	Ex.2	Ex.3	Ex.4	Ex.5	Comp. Ex.1	Comp. Ex.2
PBC-1 (part by wt.)	70	40	50	70	100	20	--
PBC-2 (part by wt.)	--	--	--	--	--	--	50
EBC (part by wt.)	30	60	45	--	--	80	45
EOC (part by wt.)	--	--	--	30	--	--	--
ARKON P125 (part by wt.)	--	--	5	--	--	--	5
Adhesive strength to acrylic resin plate (g/25 mm)							
50°C, 1 day after	4.7	7.4	12.2	4.5	1.0>	9.7	15.1
50°C, 15 days after	5.3	9.0	14.3	5.1	1.0>	18.3	19.3
50°C, 30 days after	5.6	10.6	15.0	5.3	1.0>	25.1	24.8
Surface hardness (Shore D)	49	53	48	55	60	38	45
Surface appearance of adherend	AA	AA	AA	AA	AA	BB	AA



What is claimed is:

1. A composition for a peelable protective film, comprising:

5 a propylene/1-butene random copolymer (A) in an amount of 30 to 100 parts by weight,

an ethylene/ α -olefin copolymer (B) comprising ethylene and an α -olefin of 3 to 10 carbon atoms, in an amount of 0 to 70 parts by weight, and

10 a tackifier (C) in an amount of 0 to 20 parts by weight,

the total amount of said components (A), (B) and (C) being 100 parts by weight;

wherein the propylene/1-butene random copolymer (A) has the following properties:

15 (i) the propylene content is 50 to 95 % by mol,
(ii) the melt flow rate (ASTM D 1238, 230 °C, 2.16 kg) is 0.1 to 100 g/10 min,

(iii) the molecular weight distribution (M_w/M_n , M_w : weight-average molecular weight, M_n : number-average
20 molecular weight), as determined by gel permeation chromatography (GPC), is not more than 3.0, and

(iv) the parameter B value, which indicates randomness of the copolymerized monomer sequence distribution, is 1.0 to 1.5;

25 and

the ethylene/ α -olefin copolymer (B) has the following properties:



(i) the melt flow rate (ASTM D 1238, 190 °C, 2.16 kg) is 0.1 to 100 g/10 min, and

(ii) the ethylene content is 75 to 95 % by mol.

5 2. The composition for a peelable protective film as claimed in claim 1, wherein the propylene/1-butene random copolymer (A) has the following properties:

(iv) the parameter B value, which indicates randomness of the copolymerized monomer sequence distribution, is 1.0
10 to 1.3,

(v) the melting point (T_m), as measured by a differential scanning calorimeter, is 60 to 140 °C,

(vi) the melting point (T_m) and the 1-butene content (M [% by mol]) satisfy the following relation
15 $-2.6M + 130 \leq T_m \leq -2.3M + 155$

and

(vii) the crystallinity (α [%]), as measured by X-ray diffractometry, and the 1-butene content (M [% by mol]) satisfy the following relation
20 $\alpha \geq -1.5M + 75.$

3. The composition for a peelable protective film as claimed in claim 1, wherein the propylene/1-butene random copolymer (A) is obtained by copolymerizing propylene and
25 1-butene in the presence of an olefin polymerization catalyst comprising:

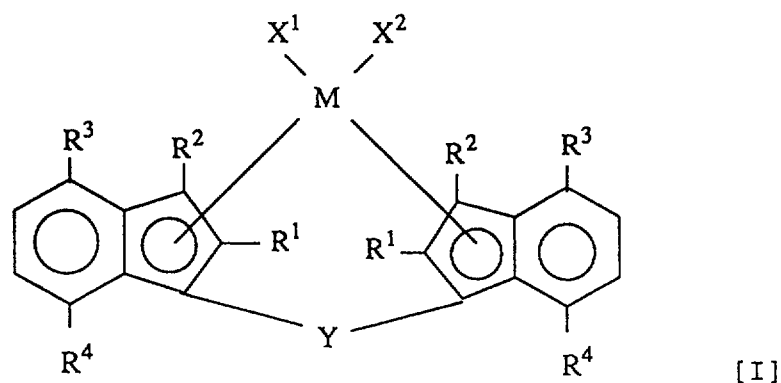
[A] a transition metal compound represented by the following formula [I],



[B] [B-1] an organoaluminum oxy-compound and/or [B-2]
a compound which reacts with the transition metal compound
[A] to form an ion pair,

and optionally

5 [C] an organoaluminum compound;



wherein M is a transition metal of Group IVa, Va or VIa of
10 the periodic table,

R¹ and R² are each hydrogen, a halogen atom, a
hydrocarbon group of 1 to 20 carbon atoms, a halogenated
hydrocarbon group of 1 to 20 carbon atoms, a silicon-
containing group, an oxygen-containing group, a sulfur-
15 containing group, a nitrogen-containing group or a
phosphorus-containing group,

R³ is a secondary or tertiary alkyl group of 3 to 20
carbon atoms or an aromatic group,

R⁴ is hydrogen or an alkyl group of 1 to 20 carbon
20 atoms,

X¹ and X² are each hydrogen, a halogen atom, a
hydrocarbon group of 1 to 20 carbon atoms, a halogenated



hydrocarbon group of 1 to 20 carbon atoms, an oxygen-containing group or a sulfur-containing group, and

Y is a divalent hydrocarbon group of 1 to 20 carbon atoms, a divalent halogenated hydrocarbon group of 1 to 20 carbon atoms, a divalent silicon-containing group, a divalent germanium-containing group, a divalent tin-containing group, -O-, -CO-, -S-, -SO-, -SO₂-, -NR⁵-, -P(R⁵)-, -P(O)(R⁵)-, -BR⁵- or -AlR⁵- (R⁵ is hydrogen, a halogen atom, a hydrocarbon group of 1 to 20 carbon atoms or a halogenated hydrocarbon group of 1 to 20 carbon atoms).

4. The composition for a peelable protective film as claimed in claim 3, wherein R¹ in the formula [I] representing the transition metal compound [A] is methyl.

5. A peelable protective film comprising a single-layer or multi-layer, which has at least one of the outermost layers formed from the composition as claimed in any one of claims 1 to 4.