Summary

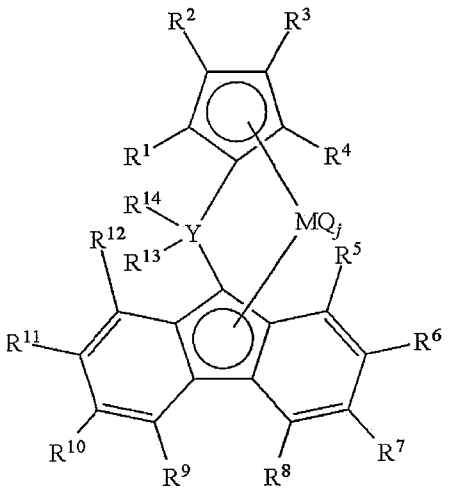
[0011a]

In one aspect of the present disclosure, there is provided a process for producing an ethylene/α-olefin copolymer, comprising copolymerizing ethylene and an α-olefin having 3 or more carbon atoms in the presence of an olefin polymerization catalyst comprising:

(A) a bridged metallocene compound represented by the following general formula [I], and

(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 bridged metallocene compound (A) to form an ion pair,

[I].



wherein Y is selected from a carbon atom, a silicon atom, a germanium atom and a tin atom,

M is a titanium atom, a zirconium atom or a hafnium atom,

R^1, R^2, R^3, R^4, R^5, R^6, R^7, R^8, R^9, R^{10}, R^{11} and R^{12} are each an atom or a substituent selected from a hydrogen atom, a hydrocarbon group having 1 to 20 carbon atoms, a silicon-containing group, a nitrogen-containing group, an oxygen-containing group, a halogen atom and a halogen-containing group, and are optionally the same as or different from each other,

adjacent substituents of R^1 to R^{12} are optionally bonded to

each other to form a ring,

R^{13} and R^{14} are each an aryl group or a substituted aryl group, and are optionally the same as or different from each other when being both substituted aryl groups,

at least one of R^{13} and R^{14} is a substituted aryl group, said substituted aryl group being a substituted aryl group in which one or more hydrogen atoms of an aryl group are each substituted with an electron-donating substituent having a Hammett substituent constant σ of not more than -0.2, wherein when the substituted aryl group has a plurality of the electron-donating substituents, these electron-donating substituents are the same as or different from each other, said substituted aryl group optionally having a substituent which is a substituent other than the electron-donating substituent and is selected from a hydrocarbon group having 1 to 20 carbon atoms, a silicon-containing group, a nitrogen-containing group, an oxygen-containing group, a halogen atom and a halogen-containing group, wherein when the substituted aryl group has a plurality of the substituents, these substituents are the same as or different from each other,

Q is selected from a halogen atom, a hydrocarbon group having 1 to 20 carbon atoms, an anionic ligand and a neutral ligand capable of coordination with a lone pair of electrons, in a combination of the same or different kinds, and

j is an integer of 1 to 4, and,

wherein polymerization is carried out in such a manner as to obtain an ethylene-based polymer in which the proportion of constituent units derived from ethylene is not less than 50% by mol when the total of constituent units derived from monomers in the polymer is 100% by mol.

Technical Problem

[0012]

In conventional processes for producing ethylene-based polymers such as the processes disclosed in the patent literatures 1 to 3, however, there is room for further improvement in points of molecular weight of the ethylene-based polymer produced in the high-temperature polymerization and responsiveness to hydrogen.

[0013]

A problem (1) to be solved by the present invention in view of such problems as associated with the prior art is to provide a process for producing an ethylene/α-olefin copolymer having a high molecular weight. As previously described, the high-temperature solution polymerization has advantages such as enhancement in productivity and reduction in production cost but simultaneously induces lowering of a molecular weight of the resulting olefin polymer, and in a process using a conventional polymerization catalyst, it was difficult to produce an ethylene/α-olefin copolymer having a high molecular weight under the conditions of a sufficiently high polymerization temperature. In order to solve this disadvantage and to acquire the advantages of the high-temperature solution polymerization, development of a process capable of producing an ethylene/α-olefin copolymer having a high molecular weight even under the conditions of a

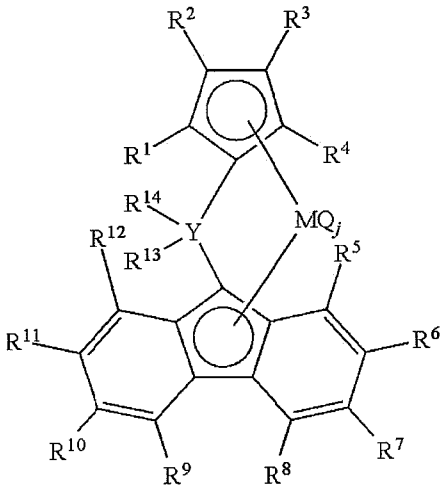
CLAIMS

1. A process for producing an ethylene/α-olefin copolymer, comprising copolymerizing ethylene and an α-olefin having 3 or more carbon atoms in the presence of an olefin polymerization catalyst comprising:

(A) a bridged metallocene compound represented by the following general formula [I], and

(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 bridged metallocene compound (A) to form an ion pair,

[I]



wherein Y is selected from a carbon atom, a silicon atom, a germanium atom and a tin atom,

M is a titanium atom, a zirconium atom or a hafnium atom,

R^1, R^2, R^3, R^4, R^5, R^6, R^7, R^8, R^9, R^{10}, R^{11} and R^{12} are each an atom or a substituent selected from a hydrogen atom, a hydrocarbon group having 1 to 20 carbon atoms, a silicon-containing group, a nitrogen-containing group, an oxygen-

containing group, a halogen atom and a halogen-containing group, and are optionally the same as or different from each other,

adjacent substituents of R^1 to R^{12} are optionally bonded to each other to form a ring,

R^{13} and R^{14} are each an aryl group or a substituted aryl group, and are optionally the same as or different from each other when being both substituted aryl groups,

at least one of R^{13} and R^{14} is a substituted aryl group, said substituted aryl group being a substituted aryl group in which one or more hydrogen atoms of an aryl group are each substituted with an electron-donating substituent having a Hammett substituent constant σ of not more than -0.2, wherein when the substituted aryl group has a plurality of the electron-donating substituents, these electron-donating substituents are the same as or different from each other, said substituted aryl group optionally having a substituent which is a substituent other than the electron-donating substituent and is selected from a hydrocarbon group having 1 to 20 carbon atoms, a silicon-containing group, a nitrogen-containing group, an oxygen-containing group, a halogen atom and a halogen-containing group, wherein when the substituted aryl group has a plurality of the substituents, these substituents are the same as or different from each other,

Q is selected from a halogen atom, a hydrocarbon group having 1 to 20 carbon atoms, an anionic ligand and a neutral ligand capable of coordination with a lone pair

of electrons, in a combination of the same or different kinds, and

j is an integer of 1 to 4, and,

wherein polymerization is carried out in such a manner as to obtain an ethylene-based polymer in which the proportion of constituent units derived from ethylene is not less than 50% by mol when the total of constituent units derived from monomers in the polymer is 100% by mol.

2. The process for producing an ethylene/α-olefin copolymer as claimed in claim 1, wherein R^1, R^2, R^3 and R^4 in the general formula [I] are all hydrogen atoms.

3. The process for producing an ethylene/α-olefin copolymer as claimed in claim 1, wherein Y in the general formula [I] is a carbon atom.

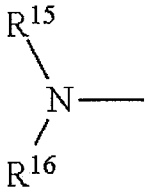
4. The process for producing an ethylene/α-olefin copolymer as claimed in claim 1, wherein the electron-donating substituent is a group selected from a nitrogen-containing group and an oxygen-containing group.

5. The process for producing an ethylene/α-olefin copolymer as claimed in claim 4, wherein R^{13} and R^{14} in the general formula [I] are the same substituted aryl group.

6. The process for producing an ethylene/α-olefin copolymer as claimed in claim 4, wherein R^{13} and R^{14} in the general formula [I] are each a substituted phenyl group containing, as the electron-donating substituent, a group selected from a nitrogen-containing group and an oxygen-containing group at the meta position and/or the para position with respect to bonding to Y.

7. The process for producing an ethylene/α-olefin copolymer as claimed in claim 4, wherein R^{13} and R^{14} in the general formula [I] are each a substituted phenyl group containing, as the electron-donating substituent, a nitrogen-containing group represented by the following general formula [II]:

[II]



wherein R^{15} and R^{16} are each an atom or a substituent selected from a hydrogen atom, a hydrocarbon group having 1 to 20 carbon atoms, a silicon-containing group, an oxygen-containing group and a halogen-containing group, are optionally the same as or different from each other, and are optionally bonded to each other to form a ring, and a line drawn on the right-hand side of N represents bonding to the phenyl group.

8. The process for producing an ethylene/α-olefin copolymer as claimed in claim 4, wherein R^{13} and R^{14} in the general formula [I] are each a substituted phenyl group containing, as the electron-donating substituent, an oxygen-containing group represented by the following general formula [III]:

R^{17}–O– [III]

wherein R^{17} is an atom or a substituent selected from a hydrogen atom, a hydrocarbon group having 1 to 20 carbon atoms, a silicon-containing group, a nitrogen-containing group and a halogen-containing group, and a line drawn on the right-hand side of O represents bonding to the phenyl group.

9. The process for producing an ethylene/α-olefin copolymer as claimed in claim 1, wherein R^5, R^8, R^9 and R^{12} in the general formula [I] are all hydrogen atoms.

10. The process for producing an ethylene/α-olefin copolymer as claimed in claim 1, wherein at least two of R^6, R^7, R^{10} and R^{11} in the general formula [I] are each a substituent selected from a hydrocarbon group, a silicon-containing group, a nitrogen-containing group, an oxygen-containing group, a halogen atom and a halogen-containing group.

11. The process for producing an ethylene/α-olefin copolymer as claimed in claim 10, wherein R^6 and R^7, and/or R^{10} and R^{11} in the general formula [I] are bonded to each other to form a ring.

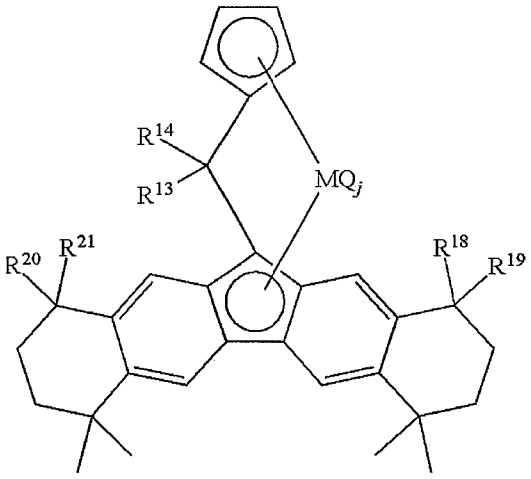
12. The process for producing an ethylene/α-olefin copolymer as claimed in claim 11, wherein R^6 and R^7, and R^{10} and R^{11} in the general formula [I] are both bonded to each other to form a ring.

13. The process for producing an ethylene/α-olefin copolymer as claimed in claim 12, wherein the rings formed by bonding of R^6 and R^7, and R^{10} and R^{11} in the general formula [I] to each other are each a five-membered to seven-membered ring.

14. The process for producing an ethylene/α-olefin copolymer as claimed in claim 13, wherein the rings formed by bonding of R^6 and R^7, and R^{10} and R^{11} in the general formula [I] to each other are each a six-membered ring.

15. The process for producing an ethylene/α-olefin copolymer as claimed in claim 14, wherein ethylene and an α-olefin are copolymerized in the presence of an olefin polymerization catalyst comprising a bridged metallocene compound represented by the following general formula [IV]:

[IV]



wherein M is a titanium atom, a zirconium atom or a hafnium atom,

R^{13} and R^{14} are each a substituted phenyl group containing, as the electron-donating substituent, a nitrogen-containing group represented by the general formula [II], or are each a substituted phenyl group containing, as the

electron-donating substituent, an oxygen-containing group represented by the general formula [III],

R^{18}, R^{19}, R^{20} and R^{21} are each a hydrogen atom or a methyl group,

Q is selected from a halogen atom, a hydrocarbon group having 1 to 20 carbon atoms, an anionic ligand and a neutral ligand capable of coordination with a lone pair of electrons, in a combination of the same or different kinds, and

j is an integer of 1 to 4.

16. The process for producing an ethylene/α-olefin copolymer as claimed in claim 1, wherein M in the general formula [I] is hafnium.

17. The process for producing an ethylene/α-olefin copolymer as claimed in claim 1, wherein the polymerization temperature is 100 to 300° C.

18. The process for producing an ethylene/α-olefin copolymer as claimed in claim 1, wherein polymerization is carried out in such a manner as to obtain an ethylene-based polymer in which the proportion of constituent units derived from ethylene is not less than 65% by mol when the total of constituent units derived from monomers in the polymer is 100% by mol.

19. The process for producing an ethylene/α-olefin copolymer as claimed in claim 18, wherein polymerization is carried out in such a manner as to obtain an ethylene-based polymer in which the proportion of constituent units derived from ethylene is not less than 70% by mol when the total of constituent units derived from monomers in the polymer is 100% by mol.