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DECISION of 16 March 2005

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 Copolymers of ethylene with α-olefins

 Applicant:
 BASELL POLYOLEFINE GMBH

 Opponent:
 V

Headword:

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Relevant legal provisions: PCT Art. 17(3)(a) PCT R. 13.1, 13.2, 40.2(c),(d),(e)

Keyword: "Lack of unity - no"

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Boards of Appeal

Chambres de recours

Case Number: W 0029/04 - 3.3.3 International Application No. PCT/EP 03/014437

D E C I S I O N of the Technical Board of Appeal 3.3.3 of 16 March 2005

Applicant:	BASELL POLYOLEFINE	GMBH
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Representative:

Decision under appeal: Protest according to Rule 40.2(c) of the Patent Cooperation Treaty made by the applicants against the invitation (payment of additional fees) of the European Patent Office (International Searching Authority) dated 2 April 2004 .

Composition of the Board:

Chairman:	R. Young
Members:	C. Idez
	B. Günzel

Summary of Facts and Submissions

- I. International application PCT/EP 03/14437 entitled "Copolymers of ethylene with α -olefins" comprising 14 claims was filed on 18 December 2003.
- II. Independent Claims 1, 6, 7, 10, 11, 13, and 14 of the application as filed read as follows:

"1. A copolymer of ethylene with α -olefins which has a molar mass distribution M_w/M_n of from 1 to 8, a density of from 0.85 to 0.94 g/cm³, a molar mass M_n of from 10 000 g/mol to 4 000 000 g/mol and a CBDI of less than 50% and in which the side chain branching of the maxima of the individual peaks of the side chain branching distribution is in each case greater than 5 CH₃/1000 carbon atoms.

6. A process for preparing ethylene copolymers as claimed in any of claims 1 to 5, which comprises polymerizing ethylene with α -olefins in the presence of the following components:

A) at least one monocyclopentadienyl complex comprising the structural feature of the formula (Cp-Z-A)Cr (I), where the variables have the following meanings:

Cp-Z-A is a ligand of the formula (II)



where

 $R^{1A}-R^{4A}$ are each, independently of one another, hydrogen, C_1-C_{20} -alkyl, C_2-C_{20} -alkenyl, C_6-C_{20} -aryl, alkylaryl having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part, NR^{11A}_2 , $N(SiR^{11A}_3)_2$, OR^{11A} , $OSiR^{11A}_3$, BR^{11A}_2 , where the organic radicals $R^{1A}-R^{4A}$ may also be substituted by halogens and where at least two of the vicinal radicals $R^{1A}-R^{4A}$ are joined to form a five- or six-membered ring, and/or two vicinal radicals $R^{1A}-R^{4A}$ are joined to form a heterocycle which contains at least one atom from the group consisting of N, P, O and S,

Z is a bridge between A and Cp having the formula



where

L is carbon or silicon, preferably carbon, R^{5A} , R^{6A} are each hydrogen, C_1-C_{20} -alkyl C_2-C_{20} -alkenyl, C_6-C_{20} -aryl, alkylaryl having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part, or SiR^{11A}₃, where the organic radicals R^{5A} and R^{6A} may also be substituted by halogens and R^{5A} and R^{6A} may also be joined to form a five- or six-membered ring,

A is

 $R_{p}^{7A} \stackrel{|^{p}}{\underset{E}{\overset{2A}}} R_{p}^{9A} R_{p}^{9A}$ (III)

where $E^{1A}-E^{4A}$ are each carbon or nitrogen,

 $R^{7A}-R^{10A}$ are each, independently of one another, hydrogen, C₁-C₂₀-alkyl, C₂-C₂₀-alkenyl, C₆-C₂₀-aryl, alkylaryl having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part or SiR^{11A}₃, where the organic radicals $R7^{A}$ -R10^A may also bear halogens or nitrogen or further C₁-C₂₀-alkyl groups, C₂-C₂₀-alkenyl groups, C₆-C₂₀-aryl groups, alkylaryl groups having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part or SiR^{11A}₃ as substituents and two vicinal radicals R^{7A} -R^{10A} or R^{7A} and Z may also be joined to form a five- or six-membered ring,

 R^{11A} are each, independently of one another hydrogen, C_1 - C_{20} -alkyl, C_2 - C_{20} -alkenyl, C_6 - C_{20} -aryl, alkylaryl having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part and two geminal radicals R^{11A} may also be joined to form a five- or six -membered ring and

p is 0 when $E^{1A}-E^{4A}$ is nitrogen and is 1 when $E^{1A}-E^{4A}$ is carbon,

B) optionally an organic or inorganic support,

C) optionally one or more activating compounds and

D) optionally one or more metal compounds containing a metal of group 1, 2, or 13 of the Periodic Table.

7. A catalyst system for olefin polymerization comprising

A') at least one monocyclopentadienyl complex A') comprising the structural feature of the formula $(Cp-CR^{5B}R^{6B}-A)Cr$ (IV) where the variables have the following meanings:

Cp-CR^{5B}R^{6B}-A is



where

 $R^{1B}-R^{4B}$ are each independently of one another, hydrogen, C_1-C_{20} -alkyl, C_2-C_{20} -alkenyl, C_6-C_{20} -aryl, alkylaryl having 1 to 10 carbon atoms in the alkyl radical and 6-20 carbon atoms in the aryl radical, NR^{5A}_2 , $N(SiR^{11B}_3)_2$, OR^{11B} , $OSiR^{11B}_3$, BR^{11B}_2 , where the organic radicals $R^{1B}-R^{4B}$ may also be substituted by halogens and at least two vicinal radicals $R^{1B}-R^{4B}$ may also be joined to form a five- or six-membered ring,

 $R^{^{5B}},\ R^{^{6B}}$ are each hydrogen or methyl,

A is



where

 $E^{1B}-E^{4B}$ are each carbon or nitrogen,

 $R^{7B}-R^{10B}$ are each, independently of one another, hydrogen, C₁-C₂₀-alkyl, C₂-C₂₀-alkenyl, C₆-C₂₀-aryl, alkylaryl having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part or SiR^{11B}₃, where the organic radicals $R^{7B}-R^{10B}$ may also bear halogens or nitrogen or further C₁-C₂₀-alkyl groups, C₂-C₂₀-alkenyl groups, C₆-C₂₀-aryl groups, alkylaryl groups having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part or SiR^{11B}₃ as substituents and two vicinal radicals $R^{7B}-R^{10B}$ may also be joined to form a five- or six-membered ring,

 R^{11B} are each, independently of one another hydrogen, C_1 - C_{20} -alkyl, C_2 - C_{20} -alkenyl, C_6 - C_{20} -aryl, alkylaryl having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part and two radicals R^{11B} may also be joined to form a five- or six -membered ring,

p is 0 when $E^{1B} - E^{4B}$ is nitrogen and is 1 when $E^{1B} - E^{4B}$ is carbon,

where at least one radical $R^{7B}-R^{10B}$ is C_1-C_{20} -alkyl, C_2-C_{20} -alkenyl, C_6-C_{20} -aryl, alkylaryl having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part or SiR^{11B}_3 and the organic radicals $R^{7B}-R^{10B}$ may also bear halogens or nitrogen or further C_1-C_{20} -alkyl groups, C_2-C_{20} -alkenyl groups, C_6-C_{20} -aryl groups, alkylaryl groups having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part or SiR^{5C}_3 as substituents and two vicinal radicals $R^{7B}-R^{10B}$ may also be joined to form a five- or six-membered ring or at least one $E^{1B}-E^{4B}$ is nitrogen,

B) optionally an organic or inorganic support,

C) optionally one or more activating compounds and

D) optionally one or more metal compounds containing a metal of group 1, 2, or 13 of the Periodic Table.

9. A prepolymerized catalyst system comprising a catalyst system as claimed in claim 7 or 8 and linear C_2-C_{10} -1-alkenes polymerized onto it in a mass ratio of from 1:0.1 to 1:200.

10. The use of a catalyst system as claimed in any of claims 7 to 9 for the polymerization or copolymerization of ethylene with α -olefins.

11. A process for preparing ethylene copolymers as claimed in any of claims 1 to 4, which comprises polymerizing ethylene with α -olefins in the presence of a catalyst system as claimed in any of claims 7 to 9.

13. A polymer mixture comprising

(E) from 1 to 99% by weight of one or more ethylene copolymers as claimed in any of claims 1 to 5 and

(F) from 1 to 99% by weight of a polymer which is different from (E),

where the percentages by weight are based on the total polymer mixture.

14. A fiber, film or molding comprising an ethylene copolymer as claimed in any of the claims 1 to 5."

Claims 2 to 5 were dependent on Claim 1. Claims 8 to 9 were dependent on Claim 7, and Claim 12 was dependent on Claim 11.

- III. On 2 April 2004 the European Patent Office (EPO), acting as International Searching Authority (ISA), in compliance with Article 17(3)(a) PCT issued an "Invitation to pay Additional Fees" (hereinafter "Invitation") stating that the application contravened the requirements of unity of invention according to Rule 13 PCT and inviting the Applicant to pay, within a time limit of 30 days, 3 additional search fees.
- IV. This "Invitation" resulted from the EPO/ISA's conclusion that the general concept underlying the claimed subject-matter, i.e. the production of ethylene copolymers according to Claim 1 was known from the document D1: WO-A1-01/12641 (cf page 7; line 12 to page 8, line 32; Examples 28-51, 56, Table 1-3; Claims 1, 8 and 11). According to the "Invitation" the problem relating to the production of these copolymers could be solved in various ways, which were linked by the ethylene copolymer mentioned above as same or corresponding feature. In the light of D1, there was, however, no single general inventive concept (Rule 13.1 PCT) and no demonstrated same or corresponding special technical feature (Rule 13.2 PCT) linking the following groups of claims: Group 1: The subject-matter of Claims 1-5; 13,14 Group 2: the subject-matter of Claim 6; Group 3: the subject-mater of Claims 7 and 8, and the subject-matter of Claim 9, excluding the subject-matter of Claim 6; and

Group 4 The subject-matter of Claims 10-12 excluding the subject-matter of Claim 6.

- V. On 22 April 2004 the Applicant paid under protest these three additional search fees.
- VI. In its letter dated 21 April 2004 announcing the aforementioned payment the Applicant argued essentially as follows:

(a) Document D1 did not disclose the copolymers according to Claim 1, since the copolymers disclosed therein showed neither a bimodal branching distribution nor a CDBI of less than 50%.

(b) The catalysts system used in the Examples of D1 did not fall under the catalyst used in the process of Claim 6, which did not include chinolyl ligands attached directly to an indenyl group as described in D1.

(c) Furthermore, document WO-A-01/12687 (referred to as D2) disclosed in its Example 1 that copolymers obtained with an indenylchinolyl chromium complex had a CBDI of more than 90%. Thus, the catalyst used in the present application gave rise to different polymers.

VII. On 23 June 2004 the Review Panel of EPO/ISA issued a "Notification regarding Review of Justification for Invitation to pay Additional Search Fees" (hereinafter "Review Notification"), in which the Applicant was invited to pay a protest fee within a time limit of one month. In paragraph 1 of the "Review Notification", the Applicant was told that after review of the protest the three additional search fees should not be reimbursed.

The position of the Review Board (cf. paragraph 2.3 of the Review Notification) can be summarized as follows:

(i) D1 described a catalyst system comprising a chromium complex, i.e. indenyl- (C_6C_3N) CrCl₂ used in the production of ethylene-hexene copolymers (cf. Examples 28-51, 56).

(ii) This chromium complex fell under those disclosed in Claim 6 of the application, since in the formula II mentioned therein the group Z might be linked to the radical R^{7A} to form a six-membered ring.

(iii) The copolymers produced in D1 using this catalyst had the same parameters as those of the present application except for the CBDI parameter about which D1 was silent.

(iv) The bimodal branching distribution was not disclosed in D1 but it was not incorporated in Claim 1.

(v) The fact that this catalyst might lead to different polymers from those of the present application (cf. D2) was not pertinent, since the cocatalyst system and the operating conditions were totally different in D1 and D2.

(vi) Since the catalyst system and the operating conditions were the same in D1 as in the present application, the copolymers produced must be the same. (vii) Thus, the copolymers of the present application were anticipated by those of D1.

- VIII. On 29 June 2004 the Applicant paid the protest fee requested in the "Review Notification".
- IX. The Applicant requested the reimbursement of the additional search fees and of the protest fee which had been paid.

Reasons for the Decision

- 1. The protest is admissible.
- 2. As can be deduced from the description, the aim of the present application is the preparation of ethylene copolymers with α -olefins having a composition breadth distribution index (CBDI) of less than 50%, an at least bimodal short chain branching distribution, and a narrow molar mass distribution. According to the present application these copolymers can be prepared by copolymerizing ethylene with α -olefins in the presence of specific chromium catalysts.
- 3. Although, as indicated in Section II above, the present application comprises 7 independent claims, the claims of the present application should be grouped in the following manner:

Group I: Claims 1 to 5 which refer to the ethylene copolymers with α -olefins;

Group II: Claim 6 which refers to the preparation of ethylene- α -olefins copolymers in the presence of specific chromium catalysts;

Group III: Claims 7 to 9, and 10 which relate, respectively, to specific chromium catalysts *per se* and use thereof;

Group IV: Claims 11 to 12 which relate to the preparation of ethylene- α -olefins copolymers in the presence of specific chromium catalysts of Claims 7 to 9;

Group V: Claim 13 which refers to compositions comprising the ethylene- α -olefins copolymers; and

Group VI: Claim 14 which refers to products comprising the ethylene- α -olefins copolymers.

- 4. In the Board's view the subject-matter of the Group I can be considered as conceptually linked to the other groups by the specific ethylene copolymers specified in Claim 1, either directly with that of Groups II, IV, and V or indirectly, due to the relationship between the properties of the copolymers and the use of a specific chromium catalyst system in their preparation, with that of Group III. Thus, these ethylene copolymers would qualify as common unifying "special technical feature" within the meaning of Rule 13.2. PCT, provided this common concept is novel and has an inventive character.
- 5. In this connection, it has, however, been considered in the "Review Notification" (Section VII above) that

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document D1 disclosed a catalyst system comprising a chromium complex employed in the copolymerization of ethylene with hexene (Examples 28-51, 56), which fell under the scope of Claim 6 of the present application. According to the Review Panel, the ethylene copolymers of D1 would hence exhibit the same properties as those of the present application, and, consequently, the copolymers of D1 anticipated those of the present application.

6. Document D1 relates to a process for the polymerization or copolymerization of olefins in presence of a catalyst composition comprising:

> A) a substituted monocyclopentadienyl, monoindenyl, monofluorenyl or heterocyclopentadienyl of the formula I:

$$[Y-M-X_n]_m$$
 I,

where: M is chromium, molybdenum or tungsten, Y is described by the formula II:



II

where: E^1-E^5 are carbon or a maximum of one E^1 to E^5 is phosphorus or nitrogen, Z is NR^5R^6 , PR^5R^6 , OR^5 , SR^5 or an unsubstituted, substituted or fused, partially unsaturated heterocyclic or heteroaromatic ring system,

B is one of the following groups:



and in addition, if Z is an unsubstituted, substituted or fused, partially unsaturated heterocyclic or heteroaromatic ring system, may alternatively be



where L^1 and L^2 are silicon or carbon, k is 1, and is alternatively 0 if Z is an unsubstituted, substituted or fused, partially unsaturated heterocyclic or heteroaromatic ring system,

X, independently of one another, are fluorine, chlorine, bromine, iodine, hydrogen, C_1-C_{10} -alkyl, C_2-C_{10} -alkenyl, C_6-C_{20} -aryl, alkylaryl having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part, NR¹⁵R¹⁶, OR¹⁵, SR¹⁵, SO₃R¹⁵, OC(O)R¹⁵, CN, SCN, β -diketone, CO, BF₄, PF₆ or a bulky non-coordinating anion,

 $R^{1}-R^{16}$, independently of one another, are hydrogen, $C_{1}-C_{20}$ -alkyl, $C_{2}-C_{20}$ -alkenyl, $C_{6}-C_{20}$ -aryl, alkylaryl having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part or SiR^{17}_{3} , whereby the organic radicals $R^{1}-R^{16}$ may also be substituted by halogens, and in each case two geminal or vicinal radicals $R^{1}-R^{16}$ may also be linked to form a five- or six-membered ring,

 R^{17} , independently of one another, are hydrogen, C_1-C_{20} alkyl, C_2-C_{20} -alkenyl, C_6-C_{20} -aryl, alkylaryl having 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part, and in each case two geminal radicals R^{17} may also be linked to form a five- or six-membered ring,

and

n is 1, 2 or 3, and m is 1, 2 or 3;

B) if desired, one or more activator compounds, and

C) if desired, one or more additional conventional olefin-polymerization catalysts (D1, Claims 2, 8, 10, 11).

In a preferred embodiment the group Z is a substituted or unsubstituted 8-quinolyl group and k is 0 (Claim 7).

7. More precisely D1 discloses in its Examples 30 to 54, and 56 the copolymerization of ethylene with hexene in the presence of the following compounds:

1-(8-quinolyl)-2,3,4,5-tetramethylcyclopentadienyl)
chromium(III) dichloride (Examples 30 to 34;
Examples 52 to 54);
1-(8-quinolyl)indenylchromium(III) dichloride
(Examples 35 to 45, 56);
1-(8-quinolyl)-2-methylindenylchromium(III) dichloride)
(Examples 46 to 49), and

1-(2-methyl-8-quinolyl)-2,3,4,5-tetramethyl
cyclopentadienyl chromium(III) dichloride (Examples 50
to 51),
in all of which the quinolyl group is directly bound to
a cyclopentadienyl group.

- 8. In that respect, the Review Panel (cf. Section VII (ii) above) has considered that such catalysts would fall under the scope of Claim 6 of the present application for the sole reason that this claim includes the feature that the radical R^{7A} and the group Z in the formula II may be joined to form a six-membered ring.
- 9. In this connection, the Board notes that this feature would either result in a "pentavalent" carbon or a "pentavalent" silicon atom in group Z, or imply that a group R^{5A} or R^{6A} must be absent in group Z, which is, however, in contradiction with the definition given for Z in the same claim. Thus, for these reasons alone, the present case should have been considered as amounting to a border-line case as referred to in the decision G 1/89 (OJ EPO 1991, 155; point 8.1 of the reasons).
- 10. In any case, when considering a claim, one should rule out interpretations which are illogical or which do not make technical sense, so that, in the Board's view, Claim 6 of the present application should be interpreted as indeed requiring that the cyclopentadienyl radical in formula II be bound to the nitrogen containing aromatic ring of formula (III) over a carbon or a silicon containing bridge Z.
- 11. In that context, it is evident from paragraphs 6 and 7 above that D1 does not clearly and unambiguously

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disclose the use of a chromium catalyst exhibiting this structural feature. Furthermore, since the chromium catalysts disclosed in Examples 30 to 54, and 56 of D1 neither correspond to those of Claim 6 nor to those of Claims 7 to 9 of the present application, it cannot, correspondingly, be concluded that the copolymers prepared in these examples would fall under the scope of Claim 1 of the present application, let alone that the composition distribution index (CBDI) and the distribution of the short chain branching of the obtained copolymers, upon which D1 is totally silent, would automatically fulfil the requirements of Claim 1 of the patent application.

- 12. Consequently, the common concept as defined in paragraph 3 above is considered as novel over D1.
- 13. Since, in the Board's view, this common concept cannot be rendered obvious by the disclosure of D1, because this document gives no hint to the preparation of copolymers having a CBDI of less than 50% and a multimodal distribution short chain branching distribution in combination with a narrow molecular weight distribution, the subject-matter of Claims 1 to 5, of Claim 6, of Claims 7 to 10, of Claims 11 to 12, of Claim 13, and of Claim 14 is considered as so linked as to form a single general inventive concept within the meaning of Rule 13.1 PCT.
- 14. It thus follows from the above that the reasons given in the "Invitation" do not warrant the proposed lack of unity objection and the Applicant's protest against the payment of three additional search fees is therefore justified.

Order

For these reasons it is decided that:

The refund of the additional search fees and the protest fee is ordered.

The Registrar:

The Chairman:

C. Eickhoff

R. Young