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D E C I S I O N
of 8 March 2000

Case Number: T 0770/97 - 3.3.3

Application Number: 87906209.9

Publication Number: 0287666

IPC: C08F 10/00

Language of the proceedings: EN

Title of invention:
Process for polymerizing olefins

Patentee:
MITSUI CHEMICALS, INC.

Opponent:
TARGOR GmbH

Headword:
-

Relevant legal provisions:
EPC Art. 123(2)

Keyword:
"Amendments - added subject-matter (yes)"

Decisions cited:
-

Catchword:
-



Case Number: T 0770/97 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 8 March 2000

Appellant: MITSUI CHEMICALS, INC.
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 13 May 1997
revoking European patent No. 0 287 666 pursuant
to Article 102(1) EPC.

Composition of the Board:

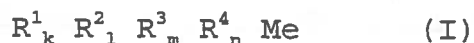
Chairman: C. Gérardin
Members: B. ter Laan
V. Di Cerbo

Summary of Facts and Submissions

I. Mention of the grant of European patent No. 0 287 666 in respect of European patent application No. 87 906 209.9, filed on 24 September 1987 as the International patent application No. PCT/JP87/00696, claiming priority from four earlier applications in Japan (223781/86 of 24 September 1986; 231242/86 of 1 October 1986; 231243/86 of 1 October 1986 and 293446/86 of 11 December 1986), was published on 26 May 1993 (Bulletin 93/21) on the basis of five claims, Claim 1 reading:

"A process for polymerizing olefins, which comprises polymerizing or copolymerizing olefins in the presence of a catalyst composed of

(A) a solid catalyst component composed of a compound of a transition metal which compound is represented by formula (I)



wherein R^1 represented a cycloalkadienyl group, R^2 , R^3 and R^4 are identical or different and each represents a cycloalkadienyl group, an aryl group, an alkyl group, an aralkyl group, a halogen atom or a hydrogen atom, Me represents zirconium, titanium or hafnium, k is 1, 2, 3 or 4, l, m and n are each 0, 1, 2 or 3, and $k+l+m+n=4$, supported on an inorganic carrier,

(B) an aluminoxane, and

(C) an organoaluminum compound having a hydrocarbon group other than n-alkyl groups."

Claims 2 to 5 referred to preferred embodiments of the process according to Claim 1.

II. On 25 February 1994 a Notice of Opposition against the granted patent was filed, in which the revocation of the patent in its entirety was requested on the grounds set out in Article 100(a) EPC.

The opposition was, *inter alia*, supported by

D2 EP-A-279 863.

III. By a decision issued in writing on 13 May 1997 the Opposition Division revoked the patent. That decision was based on two sets of claims as the main request and one auxiliary request, which were both filed during the oral proceedings. The Opposition Division held that the main request did not comply with the requirements of Article 123(2) EPC, whereas the auxiliary request was found not to be novel over D2, a prior art document according to Article 54(3)(4) EPC.

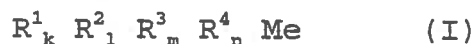
IV. On 11 July 1997 the Appellant (Proprietor) lodged an appeal against the above decision and paid the prescribed fee on 14 July 1997. With the Statement of Grounds of the Appeal, which was filed on 23 September 1997, the Appellant filed as its new main request an amended set of claims and submitted detailed arguments regarding their allowability pursuant to under Articles 123(2), 123(3) and 54 EPC.

In a letter filed on 21 January 2000, that sole set of claims was replaced by four new sets of claims as the main and three auxiliary requests. For those new claims no indication of their disclosure in the original application was given nor were any arguments brought forward as to why the new claims would overcome the previous objections and comply with the requirements of the EPC. Instead it was proposed to discuss the basis for the amendments and the significance thereof at the oral proceedings.

Claim 1 of the main request reads:

"A process for polymerizing olefins, which comprises:

- (a) prepolymerizing an olefin in the presence of
 - (A) a solid catalyst component composed of a compound of a transition metal which compound is represented by formula (I)



wherein R^1 represents a cycloalkadienyl group, R^2 , R^3 and R^4 are identical or different and each represents a cycloalkadienyl group, an aryl group, an alkyl [group], an aralkyl group, a halogen atom or a hydrogen atom, Me represents zirconium, titanium or hafnium, k is 1, 2, 3 or 4, l, m and n are each 0, 1, 2 or 3, and $k+l+m+n=4$, supported on an inorganic carrier, and

- (B) an aluminoxane,

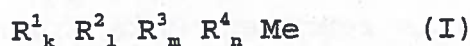
to form a prepolymerized olefin polymerization catalyst; and

- (b) charging (i) an organoaluminium compound (C) having a hydrocarbon group other than n-alkyl groups, and (ii) the prepolymerized olefin polymerization catalyst to a reaction vessel and polymerizing or copolymerizing olefins in the reaction vessel."

Claim 1 of the first auxiliary request, as further amended during the oral proceeding before the Board, reads:

"A process for polymerizing olefins, which comprises:

(a) prepolymerizing an olefin in the presence of a solid catalyst component composed of a compound of (A) a transition metal which compound is represented by formula (I)



wherein R^1 represents a cycloalkadienyl group, R^2 , R^3 and R^4 are identical or different and each represents a cycloalkadienyl group, an aryl [group], an alkyl group, an aralkyl group, a halogen atom or a hydrogen atom, Me represents zirconium, titanium or hafnium, k is 1, 2, 3 or 4, l, m and n are each 0, 1, 2 or 3, and $k+l+m+n=4$, and

(B) an aluminoxane,

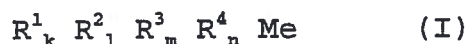
wherein (A) and (B) are supported on the same inorganic carrier so as to form a prepolymerized olefin polymerization catalyst; and

(b) charging (i) an organoaluminium compound (C) having a hydrocarbon group other than n-alkyl groups, and (ii) the prepolymerized olefin polymerization catalyst to a reaction vessel and polymerizing or copolymerizing olefins in the reaction vessel."

Claim 1 of the second auxiliary request reads:

"A process for polymerizing olefins, which comprises:

(a) prepolymerizing an olefin in the presence of (A) a solid catalyst component composed of a compound of a transition metal which compound is represented by formula (I)



wherein R¹ represents a cycloalkadienyl group, R², R³ and R⁴ are identical or different and each represents a cycloalkadienyl group, an aryl group, an alkyl [group], an aralkyl group, a halogen atom or a hydrogen atom, Me represents zirconium, titanium or hafnium, k is 1, 2, 3 or 4, l, m and n are each 0, 1, 2 or 3, and k+l+m+n=4, supported on an inorganic carrier, and

(B) an aluminoxane,

wherein (A) and (B) have been mixed in an inert hydrocarbon medium and the solvent removed using an evaporator at room temperature or an elevated temperature under atmospheric or reduced pressure so as to form a prepolymer having an intrinsic viscosity $[\eta]$, measured in decalin at 135°C of at least 0.2 dl/g, the mole ratio of the transition metal atom in catalyst component (A) to aluminium atom in catalyst component (B) in the prepolymerization being from 20 to 5000 and the prepolymerization being carried out (1) in the absence of solvent or (2) in an inert hydrocarbon medium; and

(b) charging (i) an organoaluminium compound (C) having a hydrocarbon group other than n-alkyl groups, and (ii) the prepolymer to a reaction vessel and polymerizing or copolymerizing olefins in the reaction vessel."

Claim 1 of the third auxiliary request differs from that of the second auxiliary request in that the organo-aluminium compound (C) is further specified as having a hydrocarbon group which is a branched alkyl group.

V. The Respondent (Opponent), in a letter dated 20 July 1998, observed that the Appellant in the Statement of Grounds of the Appeal had not criticised the Opposition Division's decision, but instead filed new

claims. Those claims did not comply with Articles 123(2) and 123(3) EPC, since there was no disclosure in the original application for the present combination of features and the claimed subject-matter now extended beyond the protection conferred by the patent as granted.

VI. The arguments of the Appellant brought forward during oral proceedings held on 23 February 2000 can be summarized as follows:

- (a) The support for the amendments in all requests was to be found in Examples 1 to 9 as granted (Examples 19 to 27 as originally filed), in which prepolymerization processes were described. The prepolymerization step was introduced into the claim in order to better distinguish between component (C) and a further organoaluminium compound which could also be present and which was different from component (C). The basis for that was indicated in the original description, which, to that end, was analysed and discussed in detail.
- (b) Since the claims were more limited than the ones as granted, Article 123(3) EPC was complied with.
- (c) Regarding novelty, D2 did not disclose the presence of component (C) during polymerization, which rendered the claimed subject-matter of all requests novel.

VII. The Respondent, during the oral proceedings, raised only objections under Article 123(2) to the newly filed claims.

According to the Respondent, the processes as defined in all said requests comprised the steps of conducting a prepolymerization in the presence of the transition metal compound on a carrier and the aluminoxane, whereafter catalyst component (C) was added and polymerization was carried out. The latter feature was essential since novelty was based upon it. The combination of features now required was not clearly and unambiguously derivable from the original application, in particular not from Examples 19 to 27, which described very specific process steps. Reference was also made to the original claims and description, the interpretation of which by the Appellant could not be accepted.

VIII. The Appellant requested that the decision under appeal be set aside and a patent be granted based upon the claims of the main request or, alternatively, the claims of one of the auxiliary requests.

The Respondent requested that the appeal be dismissed.

IX. The Board did not to come to a final decision at the end of the oral proceedings but reserved its decision till a later time.

The decision of the Board was taken on 8 March 2000 and the parties were informed by the Registrar of the results thereof.

Reasons for the Decision

1. The appeal is admissible.

Procedural matters

2. The Appellant filed four sets of new claims just one month before the oral proceedings without any comments as regards their allowability, in particular pursuant to Articles 123(2)(3) and 54 EPC. Moreover, during the oral proceedings an amended version of the first auxiliary request was filed.

However, given that the Respondent did not protest against the Appellant's behaviour and was capable of supplying its arguments in detail, the Board, in the exercise of its discretion, considered said sets of claims as admissible and invited the parties to discuss them.

Nevertheless, in view of the new arguments submitted during the oral proceedings on the one hand and the complexity of the claimed subject-matter on the other hand, the Board was not in the position to announce its decision at the end of the oral proceedings. The final deliberation took place on 8 March 2000.

Amendments

The wording of the claims

3. Original Claim 1 read:

"A process for polymerizing olefins, which comprises polymerizing or copolymerizing olefins in the presence of a catalyst composed of

(A) a solid catalyst component composed of a compound of a transition metal of group IVB of the periodic table supported on an inorganic carrier,
(B) an aluminoxane, and
(C) an organoaluminum compound having a hydrocarbon group other than n-alkyl groups."

From the wording of the claim as well as from the original description, where it is stated that catalyst component (A) is a solid catalyst component composed of a carrier and a transition metal compound (page 9, lines 5 to 7), it is clear that the support by an inorganic carrier of the transition metal compound is an obligatory feature; this supported transition metal compound is labelled (A). Regarding the other components, original Claim 1 contains no limitation as to whether they should also be supported or in which order they should be added. A prepolymerization step is not mentioned. From the expression "composed of", which is normally interpreted as "consisting of", it might be concluded that, apart from the specified compounds, no other ones should be present as catalyst components. However, an analysis of the original description (point 4 below) shows that this is not *per se* the case.

3.1 In the latest version of the main request a possible support of component (B) is not mentioned, so that its nonsupported presence is within the scope of the claim. However, although the wording "a solid catalyst component composed of ..." regarding component (A) would exclude the presence of any other compound on the support than the ones specified to begin with, it would still encompass the possibility of adding other compounds later, like e.g. in the second auxiliary request (see point 3.3 below).

3.2 According to the latest version of the first auxiliary request, the solid catalyst component is "composed of" the transition metal compound (contrary to the original disclosure here labelled "(A)") and component (B), so that only these compounds should be present on the inorganic carrier. However, the wording "prepolymerizing ... in the presence of..." does not exclude the presence of any further compounds during prepolymerization.

3.3 In the latest versions of the second and third auxiliary requests the transition metal compound should be deposited on the carrier first, resulting in Component (A), which is then mixed with the aluminoxane in a specific way resulting in both transition metal compound and aluminoxane being present on the support. Like in the main request, Component (A) is "a solid catalyst composed of ...", thus excluding the presence of other compounds than the ones specified before mixing with component (B).

3.4 Hence, Claim 1 of all new requests differs from Claim 1 as originally filed in that first a prepolymerization step takes place in the presence of compounds (A) and (B), and then a component (C) is required to be charged separately to a reaction vessel, after which the main polymerization is carried out in the presence of the prepolymerized components (A) and (B) as well as component (C). In other words, the process steps as well as the presence and order of addition of the various catalyst components are defined much more specifically than in original Claim 1.

- 3.5 Therefore, it should be investigated whether the information present in the application as originally filed provides adequate support for such more specified definition of the polymerization process and the catalyst system.

The application as originally filed

4. The original description can be divided in a number of sections:
- 4.1 After a summary of the background of the claimed subject-matter and the state of the art (page 1, line 1 to 8, line 3), the object of the invention is explained on page 8, lines 4 to 24, after which the definition of the invention is given (page 8, lines 26 to 36) as well as the definition of the catalyst (page 9, lines 1 to 4).
- 4.2 From page 9, line 8 onward, a detailed specification of the catalyst components is given:
- Component (A): page 9, line 8 to page 20, line 13;
 - Component (B): page 20, line 14 to page 23, line 5;
 - Component (C): page 23, lines 6 to 33.
- 4.3 The polymerization process is described on page 23, line 34 to page 25, line 21, prepolymerization being mentioned in the passage on page 24, lines 2 to 27, after which the amounts of catalyst components present during the process are given (page 24, lines 28 to page 25, line 18). Further information about suitable embodiments to carry out the polymerization is provided on page 29, line 30 to page 31, line 12.

- 4.4 The description also refers to a second and a third invention, according to which the use of a specific transition metal compound (A') or a limitation of the amount of component (B) render the use of an inorganic carrier superfluous (page 25, line 22 to page 29, line 29).
- 4.5 Then the worked examples are reported. In the majority of the examples no support for the catalyst components is used. Only Examples 19 to 27 refer to the use of an inorganic carrier for the support of some of the catalyst components.
5. The passages in the original description that refer to catalyst systems without a carrier are in clear contradiction with the requirement of the subject-matter as claimed that a support should be used. Since that particular part of the description has no bearing whatsoever on the use of supported catalyst components, it cannot provide any information relevant to the subject-matter now being claimed.
6. The first part of the description (page 1, line 1 to page 25, line 21) does refer to the claimed subject-matter, so that its disclosure may be of relevance to the present question.
- 6.1 Regarding the prepolymerization step, according to the original description (page 24, lines 7 to 12), the catalyst components (A) and (B) are desirably mixed in a hydrocarbon medium and the solvent removed, which results in a support containing the transition metal compound and the aluminoxane. Further details about mole ratio, prepolymerization and main polymerization step are given (page 24, lines 12 to 34), but nothing

is said about the presence or absence of catalyst component (C), either during the prepolymerization step or during the subsequent main polymerization step.

6.2 The only other information concerning polymerization in two steps can be found in the Examples 19 to 27.

6.2.1 In Example 19, first an aluminoxane is brought onto a support, upon which (bis(cyclopentadienyl)zirconium dichloride) is also deposited in a second step. Then, prepolymerization of ethylene is carried out. In the presence of both this prepolymerized solid catalyst and triisobutyl aluminium, ethylene is polymerized.

Examples 20 to 26 are basically a repetition of Example 19 under different polymerization conditions.

6.2.2 In Example 27, first dimethyl aluminium monochloride and then bis(cyclopentadienyl)zirconium dichloride are brought onto a support in two separate steps, after which the aluminoxane of Example 19 is deposited in a further step. Then, prepolymerization of ethylene is carried out in the same way as in Example 22, that is, in the presence of both this solid catalyst as well as triisobutyl aluminium.

6.2.3 Therefore, in the original description, only the Examples 19 to 27 disclose the use of a supported transition metal compound (A), together with an aluminoxane component (B), for prepolymerizing an olefin, followed by a main polymerization in the presence both of the prepolymerized catalyst components (A) and (B) and of an added component (C).

6.2.4 However, in view of the analyses of the various versions of Claim 1 on file, according to which, in the interpretation of the Board ("composed of" =

"consisting of"), no other compounds should be deposited on the carrier at all (first auxiliary request) or before the transition metal compound (main, second and third auxiliary requests), it is doubtful that the Examples 19 to 27 could provide the necessary support for the amendments.

6.2.5 Even if the wording of the claims would be read more broadly, the conditions of these examples are much more specific than those required in the claims. In particular, only one specific component (C) (triisobutyl aluminium), only one specific component (B) (the aluminoxane prepared in Example 19, page 44, lines 6 to 19), and only one specific transition metal compound (bis(cyclopentadienyl)-zirconium dichloride) are used in all examples and only ethylene is (co)polymerized. Also, in those examples, the aluminoxane component (B) is supported by the inorganic carrier, like the bis(cyclopentadienyl)zirconium dichloride. Therefore, the question arises whether the information contained in the other parts of the description would allow for a generalisation of the specific combination of features disclosed in the examples.

7. As pointed out above (point 4), the original description contains details about the various catalyst components within the terms of the claims as well as catalysts that have not been claimed at any stage of the proceedings. Numerous possible variations of these catalyst components are portrayed on a great number of pages and various possible embodiments of the polymerization process, including the possibility of prepolymerization, are also mentioned, thus including a huge amount of combinations of catalyst components and polymerization conditions that may be used in the claimed process, without however giving any specific disclosure. In particular, the

combination of prepolymerization in the presence of catalyst components (A) and (B), followed by charging catalyst component (C) and the prepolymerized catalyst components (A) and (B) to a reaction vessel and carrying out by the main polymerization, is not specifically indicated. Therefore, it remains to be seen whether the skilled person would recognize that the specific combination of features of Examples 19 to 27 could be generalized on the basis of the disclosure regarding those features.

7.1 Information about the prepolymerization in the presence of catalyst components (A) and (B) can, apart from the Examples 19 to 27, only be found in the passage on page 24, lines 5 to 27. According to the latter information, component (A), which is the supported transition metal, is mixed with component (B) in an inert hydrocarbon medium, after which the solvent is removed, as illustrated in Example 27. Contrary to this, in Example 19 component (B) is deposited before the transition metal. In both cases, this leads to a supported solid catalyst component containing both the transition metal compound as well as component (B). Therefore, there is no basis for an embodiment according to which component (B) could also be unsupported, as encompassed by the subject-matter of the main request. Hence, already for this reason, there is no basis in the original disclosure for those claims.

7.2 Information regarding catalyst component (C) can be found in the original description on page 23, lines 6 to 33. However, that information only describes the compounds that may be used as catalyst component (C). No details regarding the presence during polymerization and/or prepolymerization, or the way in which component (C) is added to the reaction mixture are given.

A further statement about a preferred component (C) can be found on page 16, lines 9 to 10, which follows the detailed description of catalyst component (A).

7.3 That detailed information concerning catalyst component (A) entails the statement that it is a solid catalyst component composed of a carrier and a transition metal compound (page 9, lines 5 to 7).

7.4 The description then identifies specific transition metal compounds (page 9, line 8 to page 15, line 7) and mentions the possibility of treating them with an organic metal compound or a halogen-containing silicon compound prior to deposition. The organic metal compound may be, for example, an organoaluminum compound, e.g. isoprenyl aluminium, which falls under the definition of catalyst component (C) (page 15, lines 8 to 31).

7.4.1 The Appellant stated that the passage on page 15, lines 8 to 11 about the treatment of the transition metal compound with an organic metal compound or a halogen-containing silicon compound before deposition was erroneous. It was the carrier that was meant to be treated, not the transition metal compound. The Appellant supported that statement by referring to EP-A-0 232 595, which was cited during the examination of the application which led to the present patent, as well as to page 17, line 2 to page 18, line 2 of the patent specification.

7.4.1.1 Regarding EP-A-0 232 595, this document was cited in the search report. It is not referred to in the original description and hence cannot serve to interpret the contents thereof in order to determine its original disclosure.

7.4.1.2 Regarding the information on page 17, line 2, to page 18, line 2, even if it could be accepted that the reference to the pretreatment of the transition metal compound (page 15, lines 8 to 11) was incorrect, the Appellant's argument is not to the point. Whether the carrier or the transition metal compound could be treated with the organometal compound, either way the above cited passage relates to the presence of an organometal compound, in particular an organoaluminium compound, on the carrier, whether or not premixed with the transition metal compound. In the light of page 16, lines 9 to 10, which state that "An organoaluminum compound is preferred as the organic metal compound (C).", there can be no doubt that a preferred such compound may be the same as the catalyst component (C). Therefore, this information teaches that component (C) may be present on the carrier, which, in view of the wording of all Claims 1, on the one hand, is contradictory to the requirements of the first auxiliary request and on the other hand, would mean that component (C) may be present during the prepolymerization step. Even if the Appellant's argument, that this particular sentence, too, was incorrect and did not refer to the pretreatment of catalyst component (A) would be accepted, the possibility of catalyst component (C), in supported or unsupported form, being present during prepolymerization is not excluded by the present wording of Claim 1 of all requests.

7.4.2 Whatever the interpretation, the information given on page 17, line 2 to page 18, line 2, contains no hint of adding catalyst component (C) to the reaction vessel after the prepolymerization and only then carrying out the main polymerization step, so that it cannot be considered as a teaching in that sense.

7.5 For the basis of the addition of component (C) during the polymerization step the Appellant also relied on the disclosure on page 24, line 2 to page 25, line 18.

7.5.1 That passage can be divided into two parts: page 24, lines 2 to 27 refers to the actual prepolymerization step, whereas on page 24, line 28 to page 25, line 18, the polymerization process in general is discussed.

7.5.2 As stated above (point 6.1), the passage about prepolymerization does not consider catalyst component (C) at all, so that no information regarding its addition, presence or absence during polymerization in whichever step can be obtained from this passage.

7.5.3 Regarding the polymerization in general, amounts of the catalyst components to be used are given. The amount of component (C) is related to the amount of component (B).

7.5.4 The Appellant argued that the latter particular passage supported the obligatory addition of catalyst component (C) during the main polymerization after a prepolymerization step had been carried out. However, the Board cannot follow that argument.

7.5.4.1 First, the discussion about the amounts of catalyst components to be used refers to "the process of this invention" (page 24, lines 29 and 35; page 25, lines 12/13). No particular relationship is indicated or even suggested between this passage and the one before it, describing prepolymerization. Therefore, there is no reason to assume that the indicated amounts of catalyst components would refer exclusively to the main polymerization step after the prepolymerization step has ended. On the contrary, the wording used would rather appear to refer to the polymerization as a whole, since no distinction is

made between the catalyst components present during a possible prepolymerization step and those present during the subsequent main polymerization step. Hence, it is not clear that the contents of this part of the description would refer exclusively to the main polymerization step.

7.5.4.2 Secondly, even if that particular passage would be interpreted as describing the main polymerization step only, the presence of catalyst component (C) does not mean that it was actually added during that step. Nothing excludes the possibility that, like component (B), it was added during the prepolymerization step as well or even was present deposited on the inorganic carrier.

7.6 For the above reasons, the Board cannot see any basis in the description as originally filed for the obligatory addition of catalyst component (C) during the main polymerization step of a two-step polymerization process. Hence, the subject-matter as now claimed cannot be derived clearly and unambiguously from the original disclosure, so that it has to be considered as extending beyond the content of the application as originally filed.

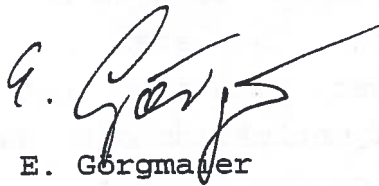
8. Since the main request as well as the first, second and third auxiliary requests share that common undisclosed feature of the obligatory addition of catalyst component (C) during the main polymerization step, the claimed subject-matter of all requests contravenes the requirements of Article 123(2) EPC.

Order

For these reasons it is decided that:

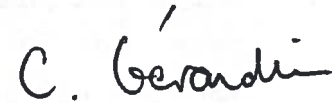
The appeal is dismissed.

The Registrar:



E. Gorgmayer

The Chairman:



C. Gérardin