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D E C I S I O N
of 12 November 2003

Case Number: T 0771/00 - 3.3.3

Application Number: 91311764.4

Publication Number: 0491566

IPC: C08F 10/00

Language of the proceedings: EN

Title of invention:

A method for the modification of catalysts intended for the polymerization of olefins

Patentee:

BOREALIS A/S

Opponent:

Basell Technology Company B.V.

Headword:

-

Relevant legal provisions:

EPC Art. 54, 56, 83, 84, 100(a), 100(b), 123(2), 123(3)

Keyword:

"Late submitted material - admitted (yes)"

"Amendments - broadening of claim (main request: yes - auxiliary request: no)"

"Inventive step - problem and solution (auxiliary request: yes)"

Decisions cited:

T 0153/85

Catchword:

-



Case Number: T 0771/00 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 12 November 2003

Appellant: Basell Technology Company B.V.
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Decision under appeal: Interlocutory decision of the Opposition
Division of the European Patent Office dated
17 May 2000 and issued in writing on
31 May 2000 concerning maintenance of European
patent No. 0491566 in amended form.

Composition of the Board:

Chairman: R. Young
Members: A. Däweritz
J. Van Moer

Summary of Facts and Submissions

- I. The grant of European patent No. 0 491 566 in respect of European patent application No. 91 311 764.4, filed on 18 December 1991 and claiming priority of 19 December 1990 of an earlier application in Finland (906282), was announced on 2 October 1996 (Bulletin 1996/40) on the basis of 10 claims.

Independent Claims 1 and 8 to 10 read as follows:

- "1. Method for the preparation of a Ziegler-Natta catalyst composition for the polymerization of olefins, said method comprising contacting an alcohol of the formula R_1OH wherein R_1 is a first alkyl group and a first carboxylic acid ester, comprising an ester group of the formula $-COOR_2$, wherein R_2 is a second alkyl group, to form a first product, characterised in that said first product is subjected to conditions such that said alcohol is transesterified with said first ester to form a transesterification product having a second carboxylic acid ester, comprising an ester group of the formula $-COOR_1$, wherein R_1 is said first alkyl group, and recovering said transesterification product as said Ziegler-Natta catalyst composition."
- "8. A catalyst prepared by a method according to any one of the preceding claims, characterised in that the ratio of the signals at points 32.5° and 30° in the X-ray diffraction spectrum is 1."

"9. A catalyst prepared by a method according to any of claims 1 to 7, characterised in that in the X-ray diffraction spectrum of the catalyst the signal is divided into two at point 15° so that a new signal is created at point 13°."

"10. Process for the polymerization of an olefin characterised in that a catalyst prepared by the method of any one of claims 1 to 7 or as claimed in claim 8 or 9 is used."

The remaining Claims 2 to 7 were method claims dependent on Claim 1.

II. On 26 June 1997, a Notice of Opposition was filed in which revocation of the patent in its entirety was requested on the grounds of lack of novelty, lack of inventive step and insufficiency of disclosure according to Article 100(b) EPC.

(a) The objections were supported by five documents (D1 to D5), including

D2: EP-A-0 045 977.

(b) New sets of claims were filed on four occasions during the opposition proceedings to replace the respective previous request(s), viz. with letters dated

- 23 February 1998: a single request;

- 4 January 1999: a Main Request and an Auxiliary Request;

- 17 March 2000: a Main Request and two Auxiliary Requests;
- 15 May 2000: three additional requests identified as Requests "A", "B" and "C".

The requests of 23 February 1998 and 4 January 1999 had been objected to for non-compliance with the requirements of Articles 84 and 123(2) EPC already during the written proceedings (communication dated 29 May 1998; annex to summons to oral proceedings, dated 17 June 1999; and Opponent's letter dated 17 March 2000).

III. In an interlocutory decision orally announced at the end of the oral proceedings on 17 May 2000 and issued in writing on 31 May 2000, the patent in suit was maintained in the form of the first Auxiliary request which had been submitted with the letter dated 17 March 2000.

- (a) Independent Claims 1 and 7 to 10 according to this Auxiliary Request 1 as maintained read as follows:

"1. Method for the preparation of a titanium-containing Ziegler-Natta catalyst composition for the polymerization of olefins, said method comprising contacting an alcohol of the formula R_1OH wherein R_1 is a first alkyl group and a first carboxylic acid ester, comprising an ester group of the formula $-COOR_2$, wherein R_2 is a second alkyl group, to form a first product, characterised in that said first product is subjected to conditions

of at least 125°C such that said alcohol is transesterified with said first ester to form a transesterification product having a second carboxylic acid ester, comprising an ester group of the formula $-\text{COOR}_1$, wherein R_1 is said first alkyl group, and recovering said transesterification product as said Ziegler-Natta catalyst composition."

"7. A catalyst prepared by a method according to any one of the preceding claims, characterised in that the ratio of the signals at points 32.5° and 30° in the X-ray diffraction spectrum is 1.

8. A Ziegler-Natta catalyst composition for the polymerization of olefins prepared by a method comprising contacting an alcohol of the formula $R_1\text{OH}$ wherein R_1 is a first alkyl group and a first carboxylic acid ester, comprising an ester group of the formula $-\text{COOR}_2$, wherein R_2 is a second alkyl group, to form a first product, wherein said first product is subjected to conditions such that said alcohol is transesterified with said first ester to form a transesterification product having a second carboxylic acid ester, comprising an ester group of the formula $-\text{COOR}_1$, wherein R_1 is said first alkyl group, and recovering said transesterification product as said Ziegler-Natta catalyst composition characterised in that in the X-ray diffraction spectrum of the catalyst the signal is divided into two at point 15° so that a new signal is created at point 13°.

9. A catalyst as prepared by a method according to any of claims 1 to 6, characterised in that in the X-ray diffraction spectrum of the catalyst the signal is divided into two at point 15° so that a new signal is created at point 13°.

10. Process for the polymerization of an olefin characterised in that a catalyst prepared by the method of any one of claims 1 to 8 or as claimed in claim 7 to 9 is used."

Claims 2 to 6 were method claims dependent on Claim 1.

- (b) In particular, the decision held that the claims of this request met the requirements of Articles 83, 84, and 123(2) and (3) EPC. Moreover, whilst four of the five documents cited in the Notice of Opposition were characterised as background art, irrelevant for novelty and inventive step, novelty was acknowledged over D2, which was also considered to represent the closest state of the art for the assessment of inventive step.

- (c) According to a test filed with the Notice of Opposition, Example 20 of D2 gave all the features of Claim 1 except for the temperature limitation of at least 125°C. Thus, in that example of D2, 100°C and 120°C were the temperatures used for the preparation of the catalyst.

- (d) The technical problem to be overcome with regard to D2 was to provide an alternative catalyst system which, when used in the polymerisation of propylene, had a high catalytic activity and led to polypropylene having a high isotacticity from which it was very easy to remove the catalyst by washing (item 6.2 of the reasons).

- (e) The document did not suggest the use of the transesterification process in order to obtain a catalyst which would overcome the above technical problem, but rather suggested that the transesterification that took place in Example 20 of D2 should have been avoided. In order to achieve this, the skilled person would have lowered the reaction temperature. Therefore, D2 taught away from increasing the temperature used to at least 125°C.

- (f) Consequently, novelty and inventive step were acknowledged.

IV. On 25 July 2000, a Notice of Appeal was filed by the Opponent (Appellant) against this decision with simultaneous payment of the prescribed fee.

In the Statement of Grounds of Appeal, received on 6 October 2000, the initial objections of lack of novelty, lack of inventive step and insufficient disclosure, raised in the Notice of Opposition, were maintained and further elaborated.

Additionally, the claims as maintained were objected to under Articles 84 and 123(2) EPC, and a new document was cited as being relevant to the questions of novelty and inventive step:

D6: US-A-4 857 613,

which was accompanied by an associated experimental report referring to the content of phthalate esters containing ethyl and/or n-butyl groups in the catalyst component obtained in a repetition of Example 1 of the document. Specific reference was made to a temperature of 125°C in step 1.3 of the example.

- V. In reply to the appeal, in a letter dated 12 November 2001, the Respondent disputed these objections and the arguments presented and requested that the appeal be dismissed and that the patent be maintained according to the contested decision (Main request) or, in the alternative, according to one of three Auxiliary requests filed therewith.
- VI. On 14 March 2003, in a communication of the Rapporteur, doubts were expressed that the requirements of Article 123(2) EPC were fulfilled by the Claims 1 according to any one of the requests then on file. Moreover, objections were raised under Article 84 EPC, because it appeared doubtful that all the features essential to define the claimed subject-matter had been specified in the independent claims.

In a letter dated 22 July 2003, the Appellant supported these objections and further commented, in particular, on the auxiliary requests.

On 31 July 2003, the parties were summoned to oral proceedings arranged for 12 November 2003.

By letter dated 10 October 2003, all the requests then on file were replaced by a new Main Request and eight Auxiliary Requests (I) to (VIII), and comments on the issues raised by the Appellant and in the communication were given by the Respondent.

By letter dated 13 October 2003, a further new document was cited by the Appellant with respect to novelty and inventive step:

D7: EP-A-0 350 170,

which was also accompanied by an associated experimental report showing the content of phthalate esters containing ethyl, isobutyl and both of these groups, respectively, in the catalyst component obtained in a repetition of Example 3 of that document. Specific reference was made to two titanization steps in the example carried out at 120°C and 130°C, respectively.

VII. The Oral proceedings were held on 12 November 2003.

(a) Upon verification of the requests of the parties, the Respondent announced that it maintained its main request, but did not further pursue Auxiliary requests (I) to (VI) submitted by letter dated 10 October 2003. They should be replaced by new Auxiliary requests (I) and (II), both containing six claims. Auxiliary requests (VII) and (VIII)

should follow as renumbered Auxiliary requests (III) and (IV), respectively.

- (b) Whilst Claim 1 of the Main Request, filed with the letter dated 10 October 2003, had the same wording as Claim 1 as maintained (section III(a), above), Claims 7 and 8 of this request were worded as follows:

"7. Method as claimed in any one of claims 1 to 6 wherein said first product is subjected to conditions of between 125°C and 135°C such that said alcohol is transesterified with said first ester.

8. Process for the polymerization of an olefin characterised in that a catalyst prepared by the method of any one of claims 1 to 6 is used."

Claims 2 to 6 were method claims appendant to Claim 1, which corresponded to Claims 2 to 6, respectively, of the request as maintained by the Opposition Division.

- (c) Auxiliary request (I), as initially filed at the beginning of the oral proceedings, contained five method claims and a process claim (Claim 6), the latter being directed to the polymerisation of an olefin by means of a catalyst as obtained by the claimed method. After deletion of Claim 6 during the oral proceedings, the claims of this request read as follows:

- "1. Method for the preparation of a titanium-containing Ziegler-Natta catalyst composition for the polymerization of olefins, said method comprising contacting a spray crystallised or emulsion solidified adduct of the formula $MgCl_2 \cdot nR_1OH$ (wherein R_1OH is an alcohol and in which n is 1 to 6) with titanium tetrachloride to form a titanised carrier, adding to said titanised carrier an alkyl ester of a phthalic acid comprising a first ester $COOR_2$, wherein R_2 is an alkyl group, to form a first product, characterised in that said first product is subjected to conditions of higher than $136^\circ C$ such that said alcohol is transesterified with said first ester to form a transesterification product having a second carboxylic acid ester, comprising an ester group of the formula $COOR_1$, wherein R_1 is a first alkyl group, and recovering said transesterification product as a Ziegler-Natta catalyst composition.
2. Method according to claim 1 or 2, characterised in that the transesterification is effected in a medium having a high boiling point.
3. Method according to claim 2 characterised in that the said medium is a long-chained hydrocarbon, preferably nonane or decane.
4. Method according to any one of claims 1 to 3, characterised in that the alkyl ester is diisobutyl phthalate.

5. Method according to claim 4, in which the alcohol is EtOH."

- (d) Claim 1 of Auxiliary request (II) differed from Claim 1 of Auxiliary request (I) in that the feature ", and recovering said transesterification product as a Ziegler-Natta catalyst composition" was replaced by "and contacting the same with a trialkyl-Al-cocatalyst and an outer donor to form said Ziegler-Natta catalyst composition."
- (e) In the discussion, the Appellant raised objections under Article 123(2) EPC against the pending requests, in particular with regard to the feature of "at least 125°C" in the Main Request and the reaction details in both Claims 1 of Auxiliary Requests (I) and (II).

In view of the limitation of the temperature to "higher than 136°C", the objection of lack of novelty to Auxiliary Request (I) was withdrawn.

Despite the fact that it was used as a starting point for argumentation, D2 was not necessarily seen to represent the closest state of the art.

In line with the acknowledged state of the art in the patent in suit (page 2, lines 20 to 43), Document D2 also made use of different phthalates in order to attain good activities and high stereospecificity. The results exemplified in D2 were either similar to those in the patent in suit or even better, so that no effect could be derived from the application of the higher temperature.

Even the measured values of the examples outside the scope of Claim 1 presented in the tables of the patent in suit were in part better than those given for the experiments within the scope of the claim.

The lower temperatures in the preferred ranges disclosed in D6 did not invalidate the argument that temperatures above 136°C were mentioned in the document and that the results according to Claim 1 were in part worse than those in the prior art (D2 + D6/D7).

The question of lifetime (ie storage time) would be of minor importance. Furthermore, the result in Table 6 of the patent in suit at 135°C was poorer than that obtained at 110°C. No comments were given to the argument of the Respondent that storage time was different from the lifetime of a catalyst.

From D7, it was known that, in particular, the phthalate esters had an effect on activity and stereospecificity and that it was not necessary to use them as starting compounds. Rather, the trivial reaction of transesterification was within the common general knowledge, although not explicitly mentioned in D7.

- (f) The Respondent disputed these arguments and relied essentially on the previous arguments as accepted in the decision under appeal that the minimum temperature of 125°C was a valid limitation for the Main request. With regard to Auxiliary Request

(I), it was further pointed out that only D2 mentioned transesterification as one option for the preparation of the ester donor, whilst the other documents were completely silent in this respect, nor provided an incentive to carry out such a reaction. Rather, even D2 would rather discourage transesterification as held in the contested decision (section III(e), above). In D7, preparation of the electron donor by conversion of other starting compounds was suggested which, however, did not amount to suggest transesterification between donors. Moreover, none of the documents suggested to heat the reaction mixture to more than 136°C. Nor did any cited document refer to the use of a spray crystallised or emulsion solidified magnesium chloride carrier. Lifetime of the catalyst, which was different from storage time, was an important feature of the catalyst to be prepared by the claimed method, whilst comparable properties as to activity and stereospecificity were obtained. No experimental results had been produced by the Opponent (Appellant), which were directly comparable to the claimed subject-matter.

VIII. The Appellant requested that the decision under appeal be set aside and the patent be revoked in its entirety.

The Respondent requested that the appeal be dismissed and that the patent be maintained on the basis of the Main request (Claims 1 to 8) filed with the letter dated 10 October 2003 or, in the alternative, on the basis of Auxiliary Request (I) (Claims 1 to 5) filed at the oral proceedings or on the basis of Auxiliary

Request (II) (Claims 1 to 6) also filed at the oral proceedings or on the basis of Auxiliary Request (III) or (IV) corresponding to renumbered Auxiliary Requests (VII) and (VIII), respectively, filed with the letter dated 10 October 2003.

Reasons for the Decision

1. The appeal is admissible.
2. *Procedural Matters*
 - 2.1 At the beginning of the oral proceedings, the admissibility of the late-filed documents D6 and D7 and of the new Auxiliary Requests (I) and (II) to the proceedings was discussed (sections IV, VI and VII (a) to (d), above).
 - 2.2 According to the Respondent, these new auxiliary requests were intended to overcome the objections raised by the Appellant on the basis of document D7, which had only been cited in the letter dated 13 October 2003, so that the oral proceedings were the first opportunity for the Respondent to comment thereon.
 - 2.3 With respect to the two documents D6 and D7, cited for the first time in the appeal proceedings, the Respondent stated its strong interest in the patent in suit to be maintained in a valid form, and, therefore, confirmed *expressis verbis* that it did not raise any objections against their admission to the proceedings.

- 2.4 In view of this statement, the Board admitted both documents, D6 and D7, into the proceedings.
- 2.5 It is, however, evident that the late submission of D7, in particular, resulted in a factual change of the situation for the Respondent at a time of less than one month before the oral proceedings, and that it had had no opportunity, therefore, to comment and react thereon before the oral proceedings.
- 2.5.1 According to Rule 57a EPC, "the description, claims and drawings of a European patent may be amended, provided that the amendments are occasioned by grounds for opposition specified in Article 100 EPC, even if the respective ground has not been invoked by the opponent." The comparison of the two new Auxiliary Requests (I) and (II) (sections VII (c) and (d), above) with the finding in the decision under appeal as to novelty (section III(c), above) and the two late-filed documents D6 and, in particular, D7 (cf. sections IV and VI, above) demonstrate that this requirement is fulfilled by both requests (by limitation to a temperature of higher than 136°C in the transesterification step).
- 2.5.2 In view of the citation of D7 at this late stage of the proceedings, the requirement for admissibility of alternative claims to appeal proceedings is also fulfilled, that such claims should normally be filed as soon as possible in these proceedings (cf. T 153/85; OJ EPO 1988, 001).
- 2.5.3 With respect to new Auxiliary Request (II), the question arises whether the further additional

modification at the end of Claim 1 could be a bar to the admissibility of the request to the proceedings.

At first sight, the additional features in this modification ("contacting ... with a trialkyl-Al-cocatalyst and an outer donor") appear to be disclosed only within the context of a combination of specific constituents in the example (patent in suit, page 4, lines 8 and 9: "... by means of this procatalyst obtained and trialkyl-Al-cocatalyst as well as an outer donor").

However, this is not the case, since the use of an outer donor is also referred to in the description in connection with Ziegler-Natta catalysts, in general (page 2, lines 15, 16 and 20 to 43), and trialkyl-aluminium is also mentioned as a preferred cocatalyst to be used together with the transesterified catalyst composition prepared by the method as defined in Claim 1 (page 3, lines 11 to 45, in particular line 44). Hence, these features in the claim are not based on an unallowable generalisation of specifics of an example.

Furthermore, this modification, aiming at the compliance of Claim 1 with Article 84 EPC (section VI, above), had already been present in Claim 1 according to the previous Auxiliary Request (I) (filed with the letter dated and received on of 10 October 2003), ie within the time limit set in the communication dated 14 March 2003 and extended in the summons until one month before the oral proceedings (Rule 71a EPC).

Hence, this modification at the end of Claim 1 of Auxiliary request (II) does not contravene Article 123(2) EPC, has been filed in good time in

reply to an objection and is, thus, no obstacle to the admission of this request in the proceedings either (cf. T 153/85, above, sections 2.1 to 3).

2.6 Under the specific circumstances of the present case, the Board has, therefore, come to the conclusion that the new Auxiliary Requests (I) and (II) should be admitted to the discussion.

3. *Insufficiency of Disclosure (Article 100(b) EPC)*

In the Statement of Grounds of Appeal, two objections were raised under the heading of "Articles 83 and 84 EPC" (page 6).

In the first objection, the reason, arising from the limitation "at least 125°C", for an exclusion of a titanization temperature of 110°C whilst 143°C remained included, was considered as completely obscure in view of the fact that better results were obtained in terms of activity and isotacticity at 110°C than at 143°C (patent in suit: Table 3). This objection is clearly related to Article 84 EPC, but not to Article 83 EPC.

According to the other objection, former independent Claim 8 (section III(a), above) lacked enabling disclosure. The subject-matter of this claim is, however, no longer claimed in any one of the valid requests.

Since no further arguments have been provided by the Appellant with respect to Article 83 and 100(b) EPC, respectively, the Board is, therefore, satisfied that

the patent in suit and the subject-matter to which it relates meet the requirements of Article 83 EPC.

Consequently, the objection under Article 100(b) EPC fails.

4. *Main request*

4.1 Article 123(2) EPC

4.1.1 In the letter dated 10 October 2003, the Respondent appreciated that the "at least 125°C" limit in the Main Request (which limit, according to the decision under appeal, was the only distinguishing feature with regard to D2, section III(c), above) derived from a specific example, and expressed the opinion that "this temperature disclosure must be read in the context of the entire specification and in particular the requirement for a transesterification reaction to take place at elevated temperature." With full consideration of the application text, it was believed that the skilled person would have realised the criticality of the 125°C limit and its broad applicability (page 1, paragraph under the heading "Added Matter").

4.1.2 This opinion contrasts with the view expressed in the communication dated 14 March 2003, items 2 and 3 (cf. section VI, above), that the above minimum temperature had only been disclosed in connection with experiments, which had been carried out with catalysts prepared by reacting a particular combination of compounds under specific conditions. However, none of these particulars had been recited in Claim 1 of that time (which did not differ from the present Main Request in this respect),

although there had been no disclosure in the application as filed that any one of these particulars did not affect the results in the tables in the specification. Consequently, the wording of this claim was considered to be based on a generalisation which did not comply with Article 123(2) EPC, because a person skilled in the art had apparently been presented with information which had not been directly and unambiguously presented by the application as originally filed.

- 4.1.3 The Board does not resile from this view, since, on the one hand, it is evident from the description that the temperature necessary for this reaction depends on the nature of the acid - alcohol pair and, when titanization is involved, no transesterification takes place at temperatures of below 100°C (page 3, lines 5 to 10 and 36 to 38), but temperatures close to the boiling point of $TiCl_4$ (136°C) or above are required for such a reaction (page 3, lines 21, 22, 40 and 41).
- 4.1.4 In the exemplified experiments on page 4 *et seq.*, on the other hand, a temperature of 125°C is only mentioned in a list of five different titanization temperatures within a range of from 110°C to 143°C in close relation to "a certain catalyst composition" (patent in suit: page 4, lines 14/15), as already pointed out in the above communication.

Furthermore, it is *expressis verbis* stated in this context that a transesterification takes place between the *ethoxy groups originating from the carrier adduct and the iso-butyl groups of the donor* and that diethyl phthalate is created as a donor, if a titanization

temperature high enough is used (page 4, lines 6 to 12, in particular, lines 10 to 12; emphasis added).

However, it is not derivable from any results shown in Tables 1 to 6 in the patent in suit (and obtained in the preparation of the above "certain catalyst composition") that a titanization temperature of at least 125°C would have been a critical limitation in this respect, especially in view of the fact that the patent in suit is silent with respect to any minimum degree of transesterification necessary to achieve the desired results in the polymerisation of the olefin by means of such a catalyst. The only indication to a critical minimum temperature can be taken from the experiment in Table 1, the only one indicated to be a comparison, wherein heptane (which has a boiling point of about 98°C, ie below 100°C, cf. section 4.1.3, above) was used as the solvent.

Rather, as also demonstrated by the experimental data associated with D2 and D6, respectively, which had been presented in the Notice of Opposition and in the Statement of Grounds of Appeal, transesterification between ethanol in the carrier adduct and di-n-butyl and di-isobutyl phthalate, respectively, as the inner electron donor took place in the preparation of the respective catalyst compositions at temperatures even below this limit of 125°C, such as 100°C and 120°C, respectively. The occurrence of transesterification *per se* under these conditions, although allegedly with low degrees of conversion, was not denied by the Respondent in the oral proceedings.

Nor does the sentence that "it is possible to use this transesterification reaction also for the modification of other ester components of a catalyst" provide the information which would allow the generalisation of the specific temperature value as suggested in Claim 1 (page 4, lines 13 to 17).

4.1.5 Hence, in contrast with the situation concerning the basis of the additional features in Claim 1 of Auxiliary request (II) in the application as originally filed (section 2.5.3, above), it is evident that the question of support of the Main Request by the original description must, therefore, be answered in the negative.

4.1.6 Claim 7 further limits the reaction conditions to a temperature range of between 125°C and 135°C. However, temperatures above 135°C are clearly important for achieving the transesterification in parallel to the titanization (cf. section 4.1.3, above). It follows that the above arguments concerning the 125°C limit are also valid for both temperatures in this claim.

4.2 Consequently, neither Claim 1 nor Claim 7 complies with Article 123(2) EPC, and, therefore, the Main Request cannot prevail.

5. *Auxiliary request (I)*

5.1 Article 123(2) EPC

No objections under Article 100(c) EPC had been raised by the Opponent against the patent in suit as granted.

Nor were such objections raised by the Opposition Division (contested decision: item 2 of the reasons).

The amendments carried out in Claim 1 (in comparison to Claim 1 as granted) have their basis on page 3, lines 11 to 34 of the specification (page 4, line 32 to page 5, line 31 of the application as originally filed).

Claims 2 to 5 have their basis in Claims 3 to 5 and 7 as granted.

Consequently, the Board is satisfied that the claims of this request comply with Article 123(2) EPC.

5.2 Article 123(3) EPC

The above amendments further limit the scope of Claim 1. It follows therefrom that the requirements of Article 123(3) EPC are also met.

5.3 Article 84 EPC

The Board is satisfied that the claims meet the requirements of Article 84 EPC.

5.4 Problem and solution

The patent in suit relates to a method for the preparation of a Ziegler-Natta catalyst intended for the polymerisation of olefins, whereby the polymerisation is carried out by means of a procatalyst, a cocatalyst and an outer donor, to achieve a homo- or copolymerisation product of high stereospecificity at high productivity yield in terms of mass of polymer

produced per gram of catalyst composition employed (Claim 1; page 2, lines 50 to 52).

- 5.4.1 Such a method is known from D2 which was considered by the Respondent to represent the closest state of the art, a view also shared by the Opposition Division. The Appellant, while indicating reservations in this respect at the oral proceedings, did not elaborate on the reasons for this, nor did it propose an alternative state of the art as an appropriate starting point.

Consequently, the Board sees no reason to depart from the choice of the closest state of the art in the decision under appeal.

- 5.4.2 Document D2 concerns a solid component (c) to be used in combination with an Al-alkyl compound (a) and a certain type of silicon compounds (b) to form catalysts for the polymerisation of α -olefins. The solid component (c) comprises an anhydrous magnesium dihalide in active form, as the essential support for a titanium halide or halo-alcoholate and an ester (as an inner donor) selected from seven different types, inclusive of (in class 3) mono- and diesters of aromatic ortho-dicarboxylic acids, wherein at least one of the hydrocarbyl radicals bound in the ester groups contains from 3 to 20 carbon atoms (Claim 1 and page 4, line 4 to page 5, line 2). Examples of such esters are listed on page 6, including *inter alia* the diisobutyl and ethyl-isobutyl phthalates. However, the document does not refer to spray crystallisation or emulsion solidification of the carrier.

In the preparation of this solid component (c), the esters are contacted with the active Mg dihalide or precursors thereof or "the esters can be formed in situ by means of known reactions as for instance by esterification between an alcohol or an alcoholate and an aryl halide or between an anhydride or a[n] [h]emiester of a polycarboxylic acid with an alcohol or by transesterification. The esters can also be used in mixture with other known inside donors" (page 7, lines 14 to 22).

The activity and stereospecificity of the supported catalysts comprising as outside donor a silicon compound containing Si-O-C bonds can be further increased "by using as inside donor an ester having a particular structure" (page 2, lines 7 to 14; page 3, lines 12 to 16).

Document D2 aims at increasing the activity and stereospecificity of the prior art supported catalysts (page 3, lines 12 to 14). Thus, in Table III (page 29), the yields in gram polypropylene per gram of catalyst component and the isotacticity indices ("I.I.") of the polypropylene products in a number of examples and comparative examples are listed.

Example 20, specifically referred to by the Appellant and in the decision under appeal, describes a supported catalyst made of spherical $MgCl_2 \cdot 2.5C_2H_5OH$ particles and a suspension of a titanium tetrachloride-diisobutyl phthalate adduct with heating to $100^\circ C$, filtering, further treating the resulting solid product with additional $TiCl_4$ at $120^\circ C$, subsequent filtration and washing with n-heptane. The solid component was then

used together with aluminium triethyl and phenyltriethoxysilane in the manufacture of polypropylene having an "I.I." of 96.9%, in a yield of 13,900 g per gram of the solid titanium catalyst component.

- 5.4.3 In line with the description in the patent in suit, in particular page 2, lines 51/52 and page 6, lines 16 to 31, the technical problem underlying the patent in suit with respect to D2 may be seen as the provision of a method for the preparation of a Ziegler-Natta catalyst composition having (i) both a high activity and capability of producing a polymer with a high stereospecificity and (ii) the catalyst system in use exhibiting an improved lifetime.
- 5.4.4 The solution to this problem proposed according to Claim 1 is to subject a first product, formed from a spray crystallised or emulsion solidified adduct of the formula $MgCl_2 \cdot nR_1OH$ (wherein R_1OH is an alcohol and in which n is 1 to 6), titanium tetrachloride and an alkyl ester of a phthalic acid, to elevated temperature conditions of higher than $136^\circ C$ such that the alcohol and the phthalic acid ester undergo transesterification, in parallel to the titanization (Claim 1 and page 3, lines 35 to 41).
- 5.4.5 Whilst the Appellant contested aspect (i) of the technical problem on the basis that it had already been solved by D2, so that the catalyst system according to the patent in suit would have to be regarded as a mere alternative, this view cannot be shared by the Board for the following reasons:

- (a) The formulation of the technical problem under 5.4.3, above, in any case does not call for an improvement over that of D2 in terms of catalyst activity and stereospecificity of the polymer products thus produced. Rather, it calls for these capabilities to be present at acceptably high levels.
- (b) In this connection, whilst it might appear at first sight, in view of (i) the reported activity in Example 20 of D2 (13.9 kg polypropylene/g catalyst) in comparison with the corresponding value of the only example (at 143°C) within the scope of Claim 1 of the patent in suit (413 kg polypropylene/g Ti, equivalent to 9.9 kg/g catalyst; recalculation undisputed by the Respondent), (ii) nearly the same isotacticity of both polymers (D2: 96.9%; patent in suit: 97.0%) and (iii) an inferior isotacticity at higher titanization temperatures as shown for the different temperatures within Table 3 of the patent in suit, that the state of the art was, if anything, superior to the patent in suit in terms of catalyst activity and practically equal to it in terms of stereospecificity of the polymer product, this superficial view takes no account of the different reaction conditions, eg the use of a different outer donor, a different propylene pressure and a different reaction time (D2: phenyltriethoxysilane, 7 atmospheres, 4 hours; patent in suit: cyclohexylmethoxymethylsilane, 10 bar, 3 hours, respectively), so that the experimental data cannot directly be compared with each other.

- (c) The fact that the experimental results in D2 and the patent in suit cannot directly be compared with each other was, in the end, also conceded by the Appellant.

Consequently, the interpretation of the experimental data in D2 (Table III) and in the patent in suit (Table 3) and the conclusion drawn by the Appellant, that the technical problem underlying the patent in suit was not solved, is not convincing, because the arguments provided are neither based on comparable data and facts, nor do they concern the precise requirements of aspect (i) of the relevant technical problem.

- 5.4.6 As regards aspect (ii) of the technical problem, there is no mention in D2 of the lifetime of the catalyst.

In the latter connection and as demonstrated by Table 6 in the patent in suit, the use of the spray crystallised or emulsion solidified $MgCl_2$ -adduct and the reaction at a temperature of more than $136^\circ C$ have a significant effect on the lifetime of the catalyst.

Thus, the available data concerning the usability time (lifetime) of the catalysts subjected to different titanization temperatures are based on measurements of the decrease of their respective activities in percentages within one hour from the preparation of the catalyst (ie according to the undisputed explanation of the Respondent, the combination of procatalyst, cocatalyst and outer donor directly before the feed of the catalyst to the polymerisation reaction, which is

different from the storage time of the catalyst prior to its use; cf. page 4, lines 29 to 32 and section VII(f), above). From the data provided, it can be concluded that the percentage of the residual activity of catalysts treated at temperatures according to Claim 1 is better, ie the lifetime becomes longer (Table 6; page 6, lines 30/31).

These results have not been disproved by the Appellant, on whom the onus of proof had been.

5.4.7 Hence, the Board is satisfied that both aspects (i) and (ii) of the relevant technical problem have been effectively solved by the claimed measures.

5.5 Novelty

In the oral proceedings, the Appellant withdrew its novelty objections to Claim 1 with regard to the cited documents D2, D6 and D7. The Board has no reason to take a different position.

Hence, the subject-matter of Claim 1 is novel.

5.6 Inventive step

It remains to be decided whether the solution found was obvious to a person skilled in the art.

5.6.1 Although the use of inner and outer donors and of more than one donor at a time has been known (page 2, lines 20 to 43 of the patent in suit) and transesterification is mentioned in D2 as one method for the preparation of the ester donor (page 7), the

latter document neither specifically recommends this reaction, nor does it provide any information as to any effects on the polymerisation reaction which may be caused by this specific preparation route to the catalyst. Moreover, D2 and also the experimental report associated with this document, which was provided by the Appellant, are completely silent about any influence the temperature might have on the preparation of the solid catalyst component, on its use or on the polymer finally produced therewith. Nor does D2 provide an incentive to prepare the solid procatalyst from the specific MgCl_2 -adduct and, in particular, to apply temperatures of more than 136°C , ie above the boiling point of TiCl_4 , as required by Claim 1.

- 5.6.2 In agreement with these findings, emphasis was repeatedly put by the Respondent on the arguments that D2 did not provide any incentive to increase the titanization and transesterification temperature, and in particular, that D2 was completely silent with respect to the lifetime of the catalyst.

These arguments have not been refuted by the Appellant, whose arguments rather concentrated on the definition of the technical problem (section 5.4.5 *et seq.*, above).

Consequently, having regard to D2 itself, there is no guidance to take the measures constituting the solution of the technical problem.

- 5.6.3 Therefore, it remains to be decided whether any one of the other two documents relied upon by the Appellant would provide the missing information which would make the solution of the technical problem obvious.

5.6.4 Document D6 discloses a process for the preparation of homopolymers and copolymers of propylene by means of a Ziegler-Natta catalyst consisting of (1) a titanium component, (2) an aluminium trialkyl component and (3) a silane component. The titanium component (1) is obtained in a multi-stage procedure, wherein (1.1) a carrier (I) is prepared from finely divided silica (Ia), an organo magnesium compound (Ib) and a gaseous chlorinating agent (Ic).

The carrier (I) is then, in a second stage (1.2), reacted with a C₁- to C₈-alkanol (II), TiCl₄ (III) and a phthalic acid derivative (IV), such as an C₁- to C₁₀-alkyl ester. After the addition of the TiCl₄ (III) at room temperature, the substances combined are kept at from 10° to 150°C for 0.5 to 5 hours and the resulting solid-phase intermediate is isolated with removal of the liquid phase.

In a third stage (1.3), the solid-phase intermediate obtained in the second stage is subjected, at from 100 to 150°C for from 0.2 to 5 hours, to a single-stage or multi-stage or continuous extraction with TiCl₄ or its mixture with an alkylbenzene. Finally, in a fourth stage (1.4), the solid thus obtained is washed with an inert liquid hydrocarbon until the eluate contains less than 2% of the titanium tetrachloride and results in catalyst component (1) (Claim 1).

In the procedure of Example 1, the carrier (I) is combined with ethanol (II) in n-heptane at room temperature and the mixture is then kept at 80°C for 1.5 hours. Thereafter, TiCl₄ (III) is at first

introduced with stirring at room temperature. Then, di-n-butyl phthalate is added, and the resulting mixture is kept at 120°C for two hours with stirring. The solid-phase intermediate thereby formed is isolated by filtration under suction, with removal of the liquid phase. In the third stage (1.3), the solid-phase intermediate is subjected to a continuous extraction with a mixture of $TiCl_4$ and ethylbenzene at 125°C for 2 hours. Finally, after filtration, the resulting solid-phase product is washed in stage (1.4) with n-heptane.

The catalyst component, prepared in this way, contained 2.6% by weight of Ti, 9.7% by weight of Mg and 32.2% by weight of Cl.

The polymerisation of propylene with this catalyst component, triethylaluminium and triethoxytoluylsilane for 2 hours at 70°C and 28 bar resulted in 17,000 g polypropylene per g of catalyst, and the product contained 1.6% heptane-solubles (ie an I.I. of 98.4%).

Whilst it is true that, in D6, temperatures of up to 140° or even 150°C are mentioned in the description of the catalyst preparation (Claim 1; column 4, lines 24 and 35), the document only states that the temperature should be "kept" within ranges limited by the above values, it does not teach to heat the reaction mixture to a temperature of above 136°C, and it is completely silent about any possible effects correlated with such heating above 136°C or about a transesterification. Moreover, D6 neither teaches to use the specific $MgCl_2$ -adduct, nor does it refer to the relevant technical problem of the lifetime of the catalyst.

5.6.5 Consequently, this document provides no incentive to modify the preparation of the catalyst of D2 so as to arrive at something within the ambit of Claim 1, or, therefore, to solve the relevant technical problem.

5.6.6 The polymerisation process of D7 is carried out by means of an olefin polymerisation catalyst formed from (A) a solid Ti catalyst component containing Mg, Ti and halogen as essential ingredients, (B) an organo-aluminium compound and (C) an organosilicon compound (Claim 1).

The document aims at a catalyst having high polymerisation activity and being capable of giving a homopolyolefin having excellent stereoregularity (page 3, lines 24 and 25).

In the preparation of component (A), an electron donor is used referring amongst various different classes of compounds also to alcohols such as ethanol and organic acid esters having 2 to 30 C-atoms including a number of phthalates. They may be used as single compounds or as mixtures. The electron donors need not be used as starting compounds, but compounds convertible to the electron donors in the course of preparing the titanium catalyst component may also be used as starting materials (page 4, line 48 to page 7, line 21; in particular, page 5, lines 1 to 9; page 6, lines 31 to 45 and 55 to 57 and page 7, lines 19 to 21).

Specific reference was made by the Appellant to Example 3, wherein component (A) was prepared by the following steps: MgCl₂, ethanol and sorbitan distearate were charged to purified kerosene, heated to 120°C with

stirring and added to further purified kerosene at -10°C . The solid thus obtained was thoroughly washed with hexane to produce a carrier. The carrier was suspended in TiCl_4 and diisobutyl phthalate was added. The temperature was raised to 120°C and the mixture was stirred at this temperature for 2 hours. Then the solid portion was collected by filtration, again suspended in TiCl_4 and stirred for another 2 hours at 130°C . After filtration, the solid product was washed with purified hexane to give the solid catalyst component (A) which contained 63% by weight of Cl, 20% by weight of Mg and 5.0% by weight of diisobutyl phthalate.

After prepolymerisation for 1 hour with the use of triethyl aluminium, a silane and 5.9 Nl/h propylene, the solid, thus obtained, was isolated by filtration and dispersed in decane. The main polymerisation using this prepolymerised component (A), triethyl aluminium (B) and a silane (C) was carried out, in the presence of hydrogen, with 500 g of propylene for 40 minutes at 70°C to prepare polypropylene having a boiling n-heptane extraction residue of 98.9% (isotacticity index). The polymerisation activity was 45,800 g polypropylene/mmol Ti, corresponding to 954 g polypropylene/g Ti (this recalculation was not in dispute between the parties).

However, the document does not provide any data which would show whether the object to provide a catalyst, which does not easily decrease in activity with time, has indeed been achieved (page 3, lines 27/28).

Nor does D7 mention the importance of the starting material for the carrier and of the temperature used in

the preparation of the catalyst for aspect (ii) of the relevant technical problem, the lifetime of the catalyst. Moreover, a transesterification reaction has neither been contemplated in the document, nor has any hint to such a reaction been given therein (cf. the analysis data of component (A) in Example 3, mentioned above). The reference to the *in situ* preparation of the donor from different starting compounds does not amount to a reference to a transesterification in accordance with Claim 1 of the patent in suit. Whilst reference is made in D7 to the temperatures in the preliminary polymerisation (-20° to +100°C) and in the main polymerisation (20° to 200°C) (page 11, lines 22 to 24; page 12, line 27), the temperatures used during the preparation of the catalyst are only mentioned in Examples 1 and 3, all clearly below 136°C.

Hence, this document provides no incentive either to solve the relevant technical problem by modification of the preparation of the catalyst of D2 so as to arrive at something within the ambit of Claim 1.

5.6.7 The finding as to the missing meaningfulness of the experimental report associated to D2 is also valid for the corresponding data provided by the Appellant with respect to D6 and D7 (sections IV and VI and 5.6.1, above). Therefore, these data cannot change the above conclusions about the relevance of D6 and/or D7 for inventive step.

5.7 Consequently, the Board is satisfied that the method of Claim 1 is not derivable from the state of the art in a manner which would be obvious to a person skilled in the art. Hence, the subject-matter of Claim 1 involves

an inventive step (Article 56 EPC). By the same token, this is also valid for the remaining dependent claims.

6. Since Auxiliary Request 1 is successful, there is no need further to consider the remaining auxiliary requests.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The Main Request is refused.
3. The case is remitted to the First Instance with the order to maintain the patent on the basis of Claims 1 to 5 of Auxiliary Request (I) filed at the oral proceedings and after any necessary consequential amendment of the description.

The Registrar:

The Chairman:

E. Görgmaier

R. Young