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Aktenzeichen / Case Number / N° du recours : T 209/85

Anmeldenummer / Filing No / N° de la demande : 80 301 160.0

Veröffentlichungs-Nr. / Publication No / N° de la publication : 18 738

Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Solid titanium catalyst component and use thereof in the production of olefin polymers or copolymers

Klassifikation / Classification / Classement : C 08 F 10/00

ENTSCHEIDUNG / DECISION
vom / of / du 17 November 1986

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /
Titulaire du brevet : Mitsui Petrochemical Industries (respondent)

Einsprechender / Opponent / Opposant : N.V. DSM (appellant)

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Articles 54 and 56

Kennwort / Keyword / Mot clé :

"Novelty - Onus of proof"
"Inventive step - Simplicity of the solution"

Leitsatz / Headnote / Sommaire

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Case Number : T 209 /85

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 17 November 1986

Appellant : (Opponent)
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Decision under appeal : **Decision of Opposition Division of the European Patent Office dated 1 March 1985* rejecting the opposition filed against European patent No. 18 738 pursuant to Article 102(2) EPC.**

* posted on 28 June 1985

Composition of the Board :

Chairman : K. Jahn

Member : C. Gérardin

Member : G. D. Paterson

Summary of Facts and Submissions

I. The European patent application No. 80 301 160.0 which was filed on 10 April 1980 and for which priority is claimed from a previous application in Japan on 10 April 1979, was granted on the basis of 6 claims on 11 May 1983 and published under the publication No. 18738.

Claim 1 reads as follows:

"A solid titanium catalyst component for use in the production of olefin polymers or copolymers, comprising titanium, magnesium, halogen and an electron donor as essential ingredients, characterised by further comprising an inert liquid hydrocarbon in an amount of, based on the weight of said component, 1 to 10% by weight when said component has a uniformity coefficient of at least 4, and 1 to 25% by weight when said component has a uniformity coefficient of less than 4."

II. The opponents filed opposition to the grant on 9 February 1984 on the basis of following citations

(1) DE-A-2 700 774
(2) US-A-4 069 169
(3) US-A-4 342 856
(4) US-A-4 149 990 = (4') DE-A-2 735 672
(5) GB-A-1 525 693

and requested that the patent be revoked in its entirety on the grounds of lack of novelty and inventive step.

III. The opposition division rejected the opposition in its decision of 1 March 1985, posted on 28 June 1985 on the basis of the following grounds:

(i) with regard to novelty, the claimed catalyst is not inherently disclosed in example 1 of citation (1) or example 1 of citation (2) since the exact structure of a catalyst depends not only on its components, but as well on the sequence of reactions used to prepare it,

(ii) with regard to inventive step, the patentee has clearly demonstrated that the claimed ranges of the amount of inert liquid hydrocarbon are critical and essential to solve the problem underlying the disputed patent.

IV. The appellant (opponent) filed an appeal against the decision of the Opposition Division on 1 August 1985, paying the prescribed fee. The grounds for appeal were filed on 24 October 1985 and were based on citations (1) and (2), and further referred to arguments concerning (5) previously presented in opposition procedure; these grounds were approximately as follows:

(i) it is usual in the art to dry the catalyst component after having washed with an inert hydrocarbon; this drying step which is carried out as far as strictly necessary to obtain a flowing powder, but not further, must lead in the case of the catalysts described in citations (1) and (2) to a product corresponding to the catalyst component presently claimed. This is supported by a reaction scheme showing the three steps which lead to the catalyst; in the absence of side reactions there is no structural difference between the known compounds and the claimed compounds;

(ii) a pre-dried catalyst prepared according to example 1 of citation (1) has a hexane content of 10 to 11% by weight; if the drying step is carried out one more minute the hexane content is 6.2% by weight. The slight difference with regard to the values required according to Claim 1 of the patent only reflects differences in the drying procedure, not in the overall structure of the catalyst;

(iii) as far as the inventive step is concerned, the advantages resulting from a more regular size of the polymer particles mentioned in the patent have already been observed in citation (5) (page 1, lines 46 to 74; page 2, lines 11 to 15 and 123 to 125; page 4, lines 7 to 19).

Besides these substantive objections, the appellant stressed that the decision of the Opposition Division did not contain a reasoned argumentation concerning the mechanism of preparation of the catalyst component put forward by him during the oral proceedings, but only a mere repetition of the arguments presented by the patentee. This was considered to be a procedural violation.

V. The respondent's arguments can be summarized as follows:

(i) no prior document discloses a process which inherently has the result of yielding a solid titanium containing catalyst component which inevitably contains the specified amount of inert hydrocarbon liquid;

(ii) no document suggests that the drying operation can be controlled to provide titanium catalyst components containing varying amounts of inert hydrocarbon liquid, and no evidence has been provided as to the procedures normally followed by those skilled in the art;

(iii) the reaction scheme put forward to illustrate the changes which take place during the preparation of the catalyst is not supported by any documentary evidence; the reactions are in fact much more complex and the exact chemical structure of the catalyst is unknown;

(iv) the procedure used by the appellant to repeat example 1 of citation (1) in order to determine the amount of hydrocarbon differs in a number of significant respects from the method actually described in (1); any comparison of the amounts of hexane according to this prior art and according to the disputed patent is thus meaningless.

VI. After detailed discussion of these views at oral proceedings on 17 November 1986 the respondent asked that the appeal be dismissed and the patent be maintained unamended. The appellant requested the revocation of the patent as well as the reimbursement of the appeal fee.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is therefore admissible.

2. Solid titanium catalyst components containing titanium, magnesium, halogen and electron donor as essential ingredients, when combined with an organometallic compound

of a metal of groups I to III of the Mendeleeff's periodic table, provide catalyst systems which are useful in the homopolymerization or copolymerization of olefins. Solid titanium catalyst components containing the same elements as in the disputed patent are described in (1), which will thus be considered as representing the closest state of the art.

The use of such solid titanium catalyst components leads to olefin polymers and copolymers which exhibit high stereospecificity, high bulk density and low content of fine powdery polymer.

The final stage of preparation of the catalyst component is usually a washing treatment with an inert liquid hydrocarbon; the catalyst component is then stored either as a slurry in the inert hydrocarbon or as a dried product until it is used for polymerization (see citation (5), page 1, line 86 to page 2, line 5).

The prior art catalyst components, although prepared from the same ingredients and obtained by the same means of preparation, frequently differ considerably from batch to batch; this affects some properties of the polyolefins, such as the bulk density or stereospecificity, which cannot be reproduced consistently. This shortcoming has never been challenged or refuted by the appellant, so that the Board relies on this statement.

The problem underlying the disputed patent is thus to suggest a catalyst component of the above specified composition which safeguards a consistent and reproducible result when used in polymerising olefins. The solution thereof consists in providing a catalyst whose amount of inert liquid hydrocarbon has been carefully controlled during the drying stage; more specifically the solid

component comprises 1 to 10% by weight when said component has a uniformity coefficient of at least 4, and 1 to 25% by weight when said component has a uniformity coefficient of less than 4.

The patentee has demonstrated with appropriate comparative examples that such controlled amounts of hydrocarbon enable the reproducibility of the polymerization, so that it is entirely credible that the said problem has effectively been solved.

3. The question of novelty will be discussed by considering the preparation of the catalyst component, i.e. the drying step, the relevance of the comparative filed on 24 October 1985 and some structural features of the catalyst component.

3.1 The patentee found that the critical stage in the preparation of the catalyst component is the drying step. Each of the citations (1) and (2) dealing explicitly with catalyst components based on the same elements as the catalysts according to the disputed patent is silent on how to perform the drying step; moreover, the purpose of drying is not mentioned at all in these documents. From an objective reading of (1) and (2) the skilled man would only conclude that drying is carried out as completely as possible and that complete drying is the normal procedure. These two documents thus cannot be regarded as implicitly disclosing a controlled drying procedure.

3.2 A close comparison between example 1 of (1) and the comparative example filed by the appellant on 24 October 1985 reveals some differences:

- according to example 1 of (1), the product resulting from the pulverization of 1 kg of magnesium chloride and

0.23 l of ethyl benzoate is suspended in titanium tetrachloride and contacted at 80°C for two hours, with stirring. The solid portion is collected by filtration and washed with titanium tetrachloride first, then with hexane and eventually dried. There are thus two treatments with titanium tetrachloride. The resulting titanium catalyst component contains 2.1% by weight of titanium and 64.5% by weight of chlorine. On the basis of these figures the appellant has determined by difference that the hexane content should be 4.2% by weight which would demonstrate that drying in (1) is not complete.

- according to the appellant's comparative example, the product resulting from the pulverization of 5 g of magnesium chloride and 1.15 ml of ethyl benzoate is suspended in 100 ml of titanium tetrachloride and contacted at 80°C for two hours while stirring. The solid portion is collected by filtration, washed with hexane and eventually dried. There is here only one treatment with titanium tetrachloride. The resulting titanium catalyst component contains 2.1% by weight of titanium, 62.5% by weight of chlorine and 6.2% by weight of hexane.

The appellant does not dispute the fact that one treatment with titanium tetrachloride has been deleted in his comparative example; in fact he considers that elimination of titanium chloride followed by a washing step with the same compound would be pointless. The only distinguishing feature would thus be the drying procedure, more complete in (1) but without effect on the catalyst.

In reality the additional washing with titanium tetrachloride followed by the elimination of titanium tetrachloride is not a sequence of operations without technical effect. Even if one assumes that no proper chemical reaction occurs during this additional step, the appellant's approach overlooks the specific solubility of the various compounds in titanium tetrachloride which will modify the relative amounts of these compounds. This means that the amounts of titanium and chlorine used to determine the amount of hexane by difference are not a reliable basis, and that the comparison is meaningless.

3.3 The appellant's argumentation is further based on the assumption that the preparation of the catalyst occurs according to a very simple reaction scheme. As explicitly specified in the disputed patent, the exact structure of the solid titanium catalyst component is in fact quite complex (column 3, lines 33 to 38). The additional reaction with titanium tetrachloride results in a complex structure wherein presumably the magnesium atom and the titanium atom are firmly bonded to each other, having halogen in common, whereas the catalyst component referred to for comparative purposes is a simple coordination compound.

The structural complexity of the catalyst component is in fact foreshadowed in the prior art as well:

- (2) refers (column 1, lines 8 to 14) to catalyst components obtained by chemically bonding and supporting a Ziegler-type catalyst component to and on particles of magnesium dihalide, which means both chemical bonding and supporting;

- various methods of preparation of the catalyst component are disclosed in (1). The first one (page 12, paragraph 2) involves basically reacting the same

compounds as in the disputed patent; further treatments and/or additional steps can optionally be combined with the basic reactions. It is further specified (page 12, last paragraph) that the titanium atom in the final compound is not necessarily tetravalent; this statement, which is not related to a particular embodiment for the preparation of the catalyst, is evidence that this final compound does not contain the initial reactants in unmodified form.

3.4 The calculation made by the appellant concerning the hexane content is thus based on an idealized structure which ignores the above possibilities and assumes that

- all the titanium atoms are tetravalent
- all the available valencies of titanium and magnesium are satisfied with chlorine.
- chlorine is bonded only to titanium and magnesium.

Besides, the determination by simple difference of the inert liquid hydrocarbon does not take into account the washing effect of titanium tetrachloride on the other components of the catalyst component. For these reasons the hydrocarbon content cannot be regarded as implicitly disclosed in (1), i.e. drying in the prior art has the meaning of completely drying, which demonstrates novelty.

3.5 Even in the absence of detailed technical information from the prior art, the general principle of the onus of proof in opposition proceedings should apply here. According to the decision of this Board T 219/83 (see O.J. 1986, 211), if the parties to opposition proceedings make contrary assertions which they cannot substantiate, and the European Patent Office is unable to establish the facts of its own motion, the patent proprietor is given the benefit of the doubt (Headnote I). This decision further specifies quite

clearly (point 12, paragraph 4) that it is not sufficient in opposition proceedings for the opponent to impugn a granted patent with an assertion which cannot be substantiated.

3.6 This question of the onus of proof is particularly pertinent in relation to the dispute between the parties in the present case as to how the drying step would normally have been carried out, at the priority date of the disputed patent. As stated in paragraph IV(i) above, the appellant asserts that the skilled man would only have dried for long enough to obtain a flowing powder, and no further. The respondent stated, in his written observations in response to the statement of grounds of appeal and during the oral hearing, that this contention was not accepted; and he pointed out that no evidence had been provided by the appellant to support such contention. During the oral hearing the appellant was asked whether there was any relevant evidence before the Board to support such contention. It was stated in reply on behalf of the appellant that in his comparative example the drying step had been carried out by a skilled chemist only for long enough to obtain a flowing powder.

However, the fact that a skilled chemist, after the priority date and with full knowledge of the claimed invention, carried out the drying step in this way, cannot be relevant evidence as to how a skilled man would have carried out the drying step at the priority date and without knowledge of the claimed invention. There being no other relevant evidence to support the appellant's contention, in the Board's view the appellant has clearly failed to prove his case in this respect.

4. The inventive step will be discussed with reference to the teaching of (5). This citation describes the preparation of supported polymerization catalyst components which comprises mixing a solvent-soluble transition metal compound with a particulate support material to cause physical or chemical adsorption thereon, washing the particulate product with solvent to remove excess transition metal compound therefrom, adding to the washed product an inert organic liquid and evaporating at least 50% by weight of the said liquid (page 1, lines 54 to 65). It is further specified that at least 50%, preferably at least 90%, and most preferably substantially all of the inert liquid, is evaporated (page 1, lines 86 to 93). In practice the catalyst component which always requires the presence of a specific electron donor, isobutanol, can be used either in the slurry form (examples 1 and 3) or as a free-flowing solid produced by evaporating the solvent (examples 2 and 4).

As a result of the use of a particulate supported polymerization catalyst the polyolefin is formed directly as solid particles (page 1, lines 26 to 32); the size of these particles can be increased by using a catalyst which has been dried completely (page 4, table and lines 7 to 15). What (5) basically teaches is thus a method of increasing the size of polymer particles by evaporating as much inert liquid as possible; nothing is said in this citation about the quantities present initially, so that the actual quantities after evaporation are unclear. In fact (5) is only concerned with an improvement of one of the three properties the disputed patent deals with, whereas the disputed patent aims at a good balance of three properties to be achieved consistently. As such (5)

provides no teaching about a possible connection between the amount of inert liquid hydrocarbon and the reproducibility of the process and therefore cannot lead to the solution of the problem.

It is remarkable that in spite of the extensive studies carried out in the field of polymerization catalysts and orientated to the choice of specific components in order to improve a particular property, the solution to the problem of reproducibility was in the amount of inert liquid hydrocarbon. The simplicity of this solution speaks in favour of an inventive step.

5. Claims 2 to 5 are concerned with preferred embodiments of the catalyst component according to Claim 1. Their patentability is supported by that of Claim 1.

The use of the catalyst component according to Claims 1 to 5 resulting in beneficial properties for the polymers, Claim 6 is allowable as well.

6. The mere fact that the Board deems the appeal not allowable excludes the reimbursement of the appeal fee.

Order

For these reasons

it is decided that:

the appeal is rejected.

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