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File Number: T 732/91 - 3.3.3

Application No.: 83 302 741.0

Publication No.: 0 094 818

Title of invention: Propylene-ethylene copolymers for high-rigidity moulded products and process for producing the copolymers

Classification: C08F 297/08

DECISION  
of 18 December 1992

Applicant: Chisso Corporation

Opponent: Solvay & Cie., S.A., Bruxelles

Headword:

EPC Articles 54, 56

Keyword: "Novelty (yes) - selection invention"  
"Inventive step (confirmed)"



Case Number : T 732/91 - 3.3.3

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.3  
of 18 December 1992

**Appellant :**  
(Opponent) Solvay & Cie. S.A., Bruxelles  
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**Representative :**  
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Solvay  
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**Respondent :**  
(Proprietor of the patent) Chisso Corporation  
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Kita-ku  
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**Representative :**  
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**Decision under appeal :**  
Decision of the Opposition Division of the  
European Patent Office dated 10 June 1991, posted  
on 19 July 1991 rejecting the opposition filed  
against European patent No. 0 094 818 pursuant to  
Article 102(2) EPC.

**Composition of the Board :**

**Chairman :** F. Antony  
**Members :** C. Gérardin  
G. Davies

## Summary of Facts and Submissions

- I. The mention of the grant of the patent No. 94 818 in respect of European patent application No. 83 302 741.0 filed on 16 May 1983 and claiming the priority of 19 May 1982 from an earlier application in Japan, was published on 27 July 1988 on the basis of 10 claims, Claim 1 reading as follows:

"A propylene-ethylene block copolymer for high-rigidity moulded products, obtained by

(i) polymerizing propylene in an amount of 70 to 95% by weight based on the total polymerized material, in the presence of a catalyst obtained by reacting with  $TiCl_4$  (C) an organoaluminium compound (I) or a reaction product of an organoaluminium compound (I) with an electron donor (A), further reacting the resulting solid product (II) with an electron donor (A) and an electron acceptor (B), and combining the resulting solid product (III) with an organoaluminium compound (IV) and an aromatic carboxylic acid ester (V), the gramme ratio of the organoaluminium compound (IV) to the solid product (III) being in the range 0.1:1 to 500:1 and, the molar ratio of said aromatic carboxylic acid ester (V) to said solid product (III) being in the range of 0.1:1 to 10.0:1, and then

(ii) polymerizing ethylene or ethylene and propylene in an amount of 30 to 5% by weight based on the total polymerized amount, at one or more stages, in the presence of the same catalyst as above, the ethylene content being in the range of 3 to 20% by weight based on the total polymerized material."

Claims 2 to 4 are dependent claims directed to preferred copolymers according to Claim 1. Claim 5 is an independent process claim concerning the preparation of a propylene-ethylene block copolymer for high-rigidity moulded products, which recites all the features of the main

product claim; further, Claims 6 to 9 are related to particular embodiments of the process as defined in Claim 5. As to Claim 10, it deals with a moulded product made from a copolymer as claimed in any of Claims 1 to 4.

II. On 20 April 1989 the Opponent filed a Notice of Opposition against the grant of the patent and requested revocation thereof in its entirety for lack of inventive step under Article 100(a) EPC. This objection, which was emphasised and elaborated in several later submissions as well as during oral proceedings, was based essentially on the following document

(2) US-A-4 259 461

as well as on

(4) GB-A-1 128 724

cited after the opposition period.

III. By a decision delivered orally on 10 June 1991, with written reasons posted on 19 July 1991, the Opposition Division rejected the opposition. It was first stated in that decision that novelty, which had been questioned by the Opponent for the first time during oral proceedings, was given since the claimed subject-matter differed from the teaching of document (2) in two respects, i.e. in the preparation of the first component of the catalyst and in the choice of an ester derived from an aromatic carboxylic acid for the final treatment of the catalyst system. The block copolymers prepared by using such catalyst systems exhibited a surprisingly high bending modulus, so that an inventive step could be acknowledged as well.

IV. The Appellant (Opponent) thereafter filed a Notice of Appeal against this decision on 19 September 1991 and paid

the prescribed fee at the same time. In the Statement of Grounds of Appeal filed on 18 November 1991 the Appellant raised again the objection of lack of novelty, maintaining that document (2) not only described the preparation of a catalyst system from initial and intermediate products corresponding to the compounds mentioned in the patent in suit, but also mentioned the use of such catalyst system to prepare propylene-ethylene block copolymers which consequently were identical with the polymers claimed in the patent in suit. Regarding the issue of inventive step, the final treatment of the catalyst system with an aromatic ester was known to enhance the crystallinity of the polymers prepared therewith; since crystallinity and stiffness were closely related, as mentioned in "Polyolefins: Structure and Properties" by H.V. Boenig published by Elsevier Publishing Company, 1966, page 119, there was a direct incentive to choose an aromatic ester for the final treatment of the catalyst system in order to improve the stiffness of the block copolymers according to the patent in suit.

- V. In the Counterstatement of Appeal filed on 20 May 1992 the Respondent (Patentee) argued that document (2) was in fact a broad teaching regarding both the titanium trichloride catalyst and the electron donor; the specific choices made in the patent in suit should be regarded as selections and novelty should be acknowledged on that basis. The relationship postulated by the Appellant between crystallinity and stiffness was not accepted for copolymers containing very small crystalline segments, such as the block copolymers according to the patent in suit. Further, the relatively small improvement of properties observed in document (4) by using an aromatic carboxylic acid ester could not have been an incentive to adopt the same solution in order to enhance the stiffness of block copolymers known from document (2).

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

The Respondent requested that the appeal be dismissed and, by way of auxiliary request, that oral proceedings be appointed.

#### Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is admissible.
2. As mentioned in point IV above, the Appellant has sought to introduce a new document in support of its objection of lack of inventive step. This citation, which specifies in particular that among other properties stiffness increases with high crystallinity, can be regarded as the missing link between the teachings of documents (2) and (4) (see decision, page 5, paragraphs 4 and 5). However, as will appear hereinafter, the choice of an aromatic carboxylic acid ester for the final treatment of the catalyst system is not the only feature of the solution proposed by the Respondent to overcome the shortcomings of the known block copolymers. The other feature being equally important for the issue of inventive step, and the Respondent having disputed the validity of the relationship between stiffness and crystallinity in the case of copolymers containing very small crystalline segments, an argument left unanswered by the Appellant, the Board has decided to disregard the late-filed document under Article 114(2) EPC.
3. Novelty of the claimed subject-matter has been questioned by the Appellant on the ground that document (2) mentions the same starting compounds for the preparation of the catalyst system as Claim 1 of the patent in suit.

3.1 The process described in this citation is directed to the preparation of propylene-ethylene block copolymers comprising, first, polymerising propylene, and, subsequently, randomly copolymerising propylene and ethylene. According to the general definition of the catalyst system the latter comprises (A) a titanium trichloride solid catalyst obtained by reducing titanium tetrachloride with an organoaluminium compound and activating, (B) an organoaluminium compound, and (C) an electron donor (column 3, lines 47 to 67).

In the passage dealing with the preparation of the catalyst component (A) (column 4, lines 6 to 52), it is indicated that the titanium trichloride solid catalyst may be prepared by four different methods, three of them encompassing several alternatives; according to the alternative (2)(i), the titanium trichloride solid catalyst (A) is obtained by first reducing titanium tetrachloride with a conventional organoaluminium compound, then treating the reaction product with an ether and subsequently with titanium tetrachloride. This particular embodiment corresponds thus to the preparation of the solid product III as defined in Claim 1 of the patent in suit.

In the final step of the preparation of the catalyst system the solid catalyst component (A) is combined with an organoaluminium catalyst component (B) and optionally an electron donor (C), the latter being selected among a large group of organic and inorganic compounds known to improve the stereoregularity of the copolymer. The list of compounds includes aliphatic as well as aromatic esters, in particular ethyl benzoate, ethyl p-anisate and methyl p-methylbenzoate (column 4, lines 53 to 65).

In the catalyst system the molar ratio of (C)/(A) is suitably 0.1:10 and the molar ratio of (B)/(A) is 1:500 (column 7, lines 1 to 7).

3.2 The teaching of document (2) is thus such that one of the classes of titanium trichloride solid catalyst (A) and some of the compounds suitable as electron donors (C) correspond respectively to the solid product III and to the aromatic carboxylic acid ester as defined in the patent in suit. In other words, document (2) must be regarded as a broad disclosure within which one specific solid catalyst component and one specific type of electron donor have been selected.

This situation corresponds to the one dealt with in the decision T 12/81 "Diastereoisomers" published in OJ EPO 1982, 296, wherein similarly the prior art document was a broad disclosure and the claimed subject-matter involved the selection of specific starting substances within two classes of products. In that case the Board took the view that "if ... two classes of starting substances are required to prepare the end products and examples of individual entities in each class are given in two lists of some length, then a substance resulting from the reaction of a specific pair from the two lists can nevertheless be regarded for patent purposes as a selection and hence as new" (Reasons for the Decision, point 13). It is further mentioned that "substances obtained in this way by selecting a specific pair of starting substances from the immense range of possibilities offered are in normal practice rightly regarded - in the absence of any additional information - as not having been anticipated by prior art description but as being a new selection. The new element - indispensable if a substance selection is to be recognised as new for patent law purposes - is not attributable to the absence of a reference to the end product but to the fact that the combination actually selected from the wide range of possibilities has not been disclosed to the public" (Reasons for the Decision, point 14.2).



By analogy, the catalyst system used in the patent in suit must thus be regarded as novel.

- 3.3 The comparison of the properties of the block copolymers obtained by using this novel catalyst system with those of the polymers prepared with a catalyst system within the terms of document (2) shows that the claimed products exhibit superior bending modulus and are thus themselves different from the prior art block copolymers.

From Example 2 and Comparative Example 8 of the patent in suit it appears that the addition of methyl p-toluate to the catalyst system increases the bending modulus of the resulting polymer by about 39%. This advantage has not been disputed by the Appellant, so that the Board regards this superior property as a distinguishing feature of the claimed block copolymers.

- 3.4 For these reasons, the subject-matter as defined in Claim 1 of the patent in suit is novel.

4. It remains to be decided whether the combination of features according to Claim 1, which involves the selection of a specific method of preparation of the titanium trichloride solid catalyst as well as the selection of a specific category of electron donors, can be regarded as inventive in the framework of the technical problem underlying the patent in suit, i.e. the improvement of the mechanical properties of the block copolymers disclosed in document (2).

- 4.1 The various embodiments illustrated in the examples of document (2), which correspond to particular choices within the broad teaching of this citation, do not provide an incentive for the skilled man to operate along the lines of the solution proposed in the patent in suit.

The comparison between Examples 1 and 2 shows that the final treatment of the catalyst system with an electron donor results in a limited improvement of flexural rigidity of the block copolymer. In Example 1, wherein the titanium trichloride solid catalyst component (A) has been prepared according to the procedure referred to above as (2)(i) and wherein no electron donor (C) has been used, the block copolymer has a flexural rigidity of 10,300 kg/cm<sup>2</sup>; in Example 2, wherein the catalyst system has been treated with methyl methacrylate as an electron donor, the flexural rigidity is 10,900 kg/cm<sup>2</sup> (columns 15/16, Table 2), which corresponds to an increase of 5.8%.

Example 3 shows that an improvement of the same order of magnitude can be achieved by using another method for the preparation of the catalyst component (A). In this example the reduced reaction product of titanium tetrachloride with the organoaluminium compound is first subjected to a heat treatment, then reacted with iodine and an ether, according to the procedure identified as (1)(ii) (column 4, lines 8 to 11 and 19 to 22). The flexural rigidity of the resulting block copolymer is 10,800 kg/cm<sup>2</sup> which corresponds to an increase of 4.9% with regard to Example 1.

These data show that in order to improve the mechanical properties of the block copolymers obtained in Example 1, which must be regarded as the closest state of the art in that it uses the method of preparation of the titanium trichloride catalyst component selected in the patent in suit, the skilled man could either look for alternative methods of preparation of that component or subject the catalyst system to a final treatment with an electron donor, but that in both cases the improvement would be limited. Clear information inviting the skilled man to

combine the selections required in the patent in suit is thus not to be found in document (2).

4.2 Although the teaching of document (4) would appear to give preference to the use of aromatic esters as electron donors in order to prepare crystalline polyolefins having good mechanical properties, this information could not by itself lead to the solution as claimed in the patent in suit.

4.2.1 Document (4) describes a process for producing olefin polymers which comprises bringing at least one olefin monomer into contact with a catalyst system comprising (a) a halide of transition metals of Groups IV, V and VI of the Periodic Table of Elements, (b) an organometallic compound of metals of Groups I, II and III of the Periodic Table, and (c) an aromatic carbonyl compound (page 2, lines 110 to 123). The catalyst component (a) is typically titanium trichloride obtained by reduction of titanium tetrachloride with an organoaluminium compound followed by an activating treatment in a ball mill or vibrating mill (page 3, lines 35 to 56; page 6, lines 22 to 32). Organoaluminium compounds are said to be particularly preferred as component (b) (page 3, lines 19 to 34). Further, ethyl p-toluate, ethyl anisate and isopropyl benzoate have been found to be particularly effective as component (c), as apparent from the isotactic indices in the examples (page 3, lines 86 to 89). When these catalysts are used for the copolymerisation of propylene with ethylene, the content of ethylene should not exceed 30% by weight based on the total weight of the resultant copolymer in order to ensure a satisfactory degree of crystallinity (page 4, lines 102 to 127).

4.2.2 In the Board's view, however, this preference for aromatic esters as electron donors should not be isolated from the whole technical content of the document, but on the

contrary considered in connection with the various features of the catalyst system. In that respect, as pointed out by the Respondent in its statement filed in the opposition procedure on 16 January 1991, an essential feature in the preparation of the catalyst component (a) is the grinding step following the reduction step; this clearly appears in the light of the experimental data in the patent in suit. A block copolymer obtained from a catalyst system comprising a first component activated by a grinding treatment has a bending modulus of 1.06 kg/cm<sup>2</sup> (average figure from Comparative Examples 3, 5 and 6); similarly, when the catalyst system derives from a first component within the terms of the patent in suit, the block copolymer has a bending modulus of 1.10 kg/cm<sup>2</sup> (Comparative Example 8). By contrast, when these catalyst systems are subsequently subjected to a final treatment with an aromatic ester, the bending moduli are increased up to 1.24 kg/cm<sup>2</sup> (Comparative Example 4) respectively 1.75 kg/cm<sup>2</sup> (average figure from Examples 1 to 20). This shows that the catalyst systems described in document (4) do not make it possible to prepare block copolymers having the desired mechanical properties and that the skilled man would not look for a general solution along that line.

4.3 Even, for the sake of argument, the selection of aromatic esters in document (2) on the basis of the preference taught in document (4) would not lead to the subject-matter as claimed in the patent in suit, for the skilled man would still be faced with the problem of finding the optimal method of preparation of the titanium trichloride solid catalyst (document (2), column 4, lines 6 to 52). For that purpose, the skilled man could only rely on the figures of Table 2, which show that the catalyst system used in Example 3 is intrinsically superior to that used in Example 1 as far as the flexural rigidity of the block copolymer is concerned. On that basis, in the Board's view, the skilled man would rather consider a combination

of the catalyst system used in Example 3 of document (2) with the aromatic esters recommended as electron donors by document (4) than any other catalyst system, in particular the one selected in the patent in suit. For this reason, the Board regards the combination of features in Claim 1 of the patent in suit as non-obvious and, therefore, the selection as inventive.

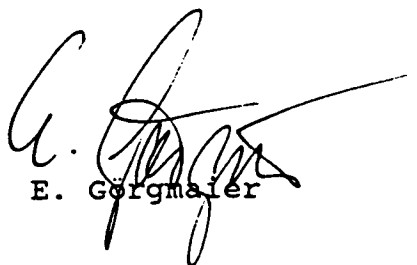
- 4.4 For these reasons, the subject-matter of the patent in suit as defined in Claim 1 involves an inventive step.
5. Claim 1 being allowable, the same applies to dependent Claims 2 to 4, which are directed to preferred block copolymers according to Claim 1, as well as to dependent Claim 10, which concerns a moulded product made from a copolymer as defined in these product claims, and whose inventiveness is supported by that of the main claim. Similar considerations apply to Claim 5, which is drafted as an independent process claim reciting all the features of the main product claim, and further to dependent process Claims 6 to 9, which deal with preferred embodiments of that claim.

#### Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar:



E. Gorgmaier

The Chairman:



F. Antony