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Aktenzeichen / Case Number / N^o du recours : T 575/89

Anmeldenummer / Filing No / N^o de la demande : 82 304 319.5

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Bezeichnung der Erfindung: Propylene-ethylene random copolymer, production
Title of invention: process thereof and film derived therefrom.
Titre de l'invention :

Klassifikation / Classification / Classement : C08F 210/06

ENTSCHEIDUNG / DECISION

vom / of / du 24 July 1990

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Showa Denko K.K.

Einsprechender / Opponent / Opposant :

01 BASF AG
02 Solvay & Cie
03 Hoechst AG

Stichwort / Headword / Référence :

EPO / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé :

"Inventive step (yes) - process features not following the prior art teaching - unexpected properties of the resulting product"

Leitsatz / Headnote / Sommaire



Case Number : T 575/89 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 24 July 1990

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Decision under appeal : Interlocutory decision of the Opposition Division of
the European Patent Office dated 25 July 1989
concerning maintenance of European Patent
No. 0 074 194 in amended form.

Composition of the Board :

Chairman : F. Antony

Members : C. Gérardin
M. Aúz Castro

Summary of Facts and Submissions

I. The mention of the grant of the patent No. 74 194 in respect of European patent application No. 82 304 319.5 filed on 16 August 1982 and claiming five priorities of earlier applications in Japan, was published on 26 November 1986 on the basis of 10 claims.

Claim 1 read as follows:

"A propylene-ethylene random copolymer having the following characteristics:

- (a) a melting point of 115°C to 140°C;
- (b) an ethylene content, determined by the C¹³-NMR method, of 6 to 20 mol %;
- (c) a block index, calculated according to the C¹³-NMR method, of 1.1 or less;
- (d) an MFI, determined at 230°C under a load of 2.16 kg, of 0.5 to 100 g/10 min;
- (e) a ratio of the MLMFI, determined at 230°C under a load of 10.0 kg, to the MFI, determined at 230°C under a load of 2.16 kg, of 10 to 16."

II. Notices of opposition were filed on 8 August 1987 by Opponent 1 (Appellant 1), on 12 August 1987 by Opponent 2 (Appellant 2) and on 25 August 1987 by Opponent 3 against the patent in suit on the grounds that its subject-matter was not novel and did not involve an inventive step.

These objections which were emphasised and elaborated in several later submissions were based on 15 documents.

III. By a decision issued on 25 July 1989 the Opposition Division held that there were no grounds of opposition to the maintenance of the patent in amended form on the basis

of a new Claim 1 drafted as a product-by-process claim filed on 23 March 1988, and Claims 2 to 10 as granted, the amendments consisting in introducing the new lower limit of 9.6 mol % for the ethylene content in the propylene-ethylene random copolymer (feature (b)) and in specifying that said copolymer was prepared "by (i) copolymerizing propylene and ethylene in the presence of a Ziegler-type catalyst and substantially in the absence of a solvent other than a monomer to form a random copolymer having an MFI, determined at 230°C under a load of 2.16 kg, of 0.01 to 0.3 g/10 min; and (ii) visbreaking the resultant copolymer in the presence of peroxide to form a random copolymer."

In that decision, wherein neither the closest state of the art was identified, nor the technical problem underlying the patent in suit was defined objectively, it was first stated that the requirement of novelty was met; then, each of the 15 documents was merely analysed by itself, as if the issue of novelty had been raised, to reach the unsubstantiated conclusion that from the numerous cited prior art, taken in isolation or in any combination, it would not have been obvious for a skilled man to arrive at the claimed subject-matter.

IV. The Appellants 1 and 2, thereafter, lodged notices of appeal against that decision on, respectively, 2 September 1989 and 13 September 1989, and paid the prescribed fee at the same time. The arguments presented in the Statements of Grounds of Appeal filed, respectively, on 1 December 1989 and 10 November 1989, as well as in later submissions, can be summarised as follows:

The lower limit of 9.6 mol%, by which novelty of the claimed subject-matter was established, contravened the requirements of Article 123(2) EPC. Although that limit did

correspond to the ethylene content of the copolymer according to Examples 16 and 26, these copolymers were in fact terpolymers containing, besides propylene as major component, 9.6 mol% ethylene and 1.3 mol% butene-1; the generalisation of the former feature to copolymers not containing the latter monomer was thus not permissible.

The substantive grounds of appeal relied essentially upon DE-A-2 827 764 (document (10)). The lower limit of 9.6% for the ethylene molar ratio in the claimed copolymers was totally arbitrary in the light of the original description and could not be regarded as a purposive selection contributing to an inventive step. In particular, the regular increase of film impact strength at -5°C parallel with increasing ethylene content according to Figure 2 of the patent in suit provided evidence that the above limit was not critical.

- V. Following an objection under Article 123(2) EPC raised in a communication of the Board on 12 June 1990, and after two auxiliary requests filed on 19 July 1990, the Appellant submitted during oral proceedings held on 24 July 1990 the following main claim:

"A propylene-ethylene random copolymer having the following characteristics:

- (a) a melting point of 115°C to 140°C;
- (b) an ethylene content, determined by the C¹³-NMR method, of 10 to 20 mol % with the proviso that when the copolymer further contains butene the said ethylene content is 9.6 to 20 mol %;
- (c) a block index, calculated according to the C¹³-NMR method, of 1.1 or less;
- (d) an MFI, determined at 230°C under a load of 2.16 kg of 0.5 to 100 g/10 min;
- (e) a ratio of the MLMFI, determined at 230°C under a

load of 10.0 kg, to the MFI, determined at 230°C under a load of 2.16 kg, of 10 to 16, said propylene-ethylene random copolymer being prepared by (i) copolymerizing propylene and ethylene in the presence of a Ziegler-type catalyst and in the absence of a solvent other than a monomer to form a random copolymer having an MFI, determined at 230°C under a load of 2.16 kg, of 0.01 to 0.3 g/10 min; and (ii) visbreaking the resultant copolymer in the presence of peroxide to form a random copolymer."

VI. To support the patentability of this claim, the Respondent put forward that inventiveness derived from both compositional and operative features. Firstly, the use of the monomer as the polymerisation solvent reduced the increase in viscosity which occurred at higher ethylene contents and, thereby, ensured an easy and smooth polymerisation process. The combination of a higher amount of ethylene and visbroken random copolymers conferred not only a high impact strength, but also excellent transparency and low-temperature heat seal strength to the random copolymers. This clearly appeared from the summary of Tables 2 and 4, wherein the copolymers were classified according to their ethylene content.

VII. The Appellants requested that the decision under appeal be set aside and that the patent be revoked.

The Respondent requested that the appeal be dismissed and that the patent be maintained on the basis of the documents submitted in the oral proceedings - Claims 1 to 10 and an adapted description - and of two sheets of drawings according to the patent specification.

Reasons for the Decision

1. The appeals comply with Articles 106 to 108 and Rule 64 EPC and are, therefore, admissible.

2. As mentioned in paragraph III above, the decision under appeal is seriously flawed in that it has not identified the closest prior art, nor has it correctly applied the problem/solution approach when investigating the existence of any inventive step. This could in itself have been a reason for it to be set aside and for the case to be remitted to the first instance in order to complete a proper examination of the oppositions (cf. Decision T 638/88 of 28 May 1990, particularly paragraph 6.2, last two sentences; unpublished). After thorough consideration of such possibility, the Board has reluctantly refrained from such remittal, mainly because this could not have led, in its opinion, to any different ultimate outcome of the case.

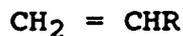
3. The current version of the claims does not give rise to objections under Article 123 EPC.

With regard to Claim 1 as originally filed, Claim 1 drafted as a product-by-process claim differs by the range of ethylene content in the copolymer specified under (b) and by the incorporation of various process features. As far as the amount of ethylene in the copolymer is concerned, two alternatives are now claimed. According to the first one, the copolymer contains 10 to 20 mol% ethylene; the lower limit of 10 mol% is disclosed on page 3, line 29 and page 7, line 60 of the patent specification corresponding to page 5, line 7 and page 17, line 12 of the application as originally filed. According to the second alternative, the copolymer contains 9.6 to 20 mol% ethylene and, additionally, butene; both the lower limit of 9.6 mol% and

the additional presence of butene correspond to the copolymer described in Examples 16 and 26 of the patent specification and the original application. Regarding the process features, the use of a Ziegler-type catalyst is mentioned on page 4, lines 43 to 45, respectively page 8, lines 21 to 24. The absence of any solvent other than the monomer subjected to the copolymerisation reaction finds basis in the patent specification on page 5, lines 15 to 21, respectively original page 10, lines 6 to 18. The formation of a random copolymer having an MFI, determined at 230°C under a load of 2.16 kg, of 0.01 to 0.3 g/10 min and, further, the visbreaking of the resultant copolymer in the presence of peroxide to form a random copolymer are both disclosed in Claim 3 as granted and originally filed.

As to Claims 2 to 10, they are identical to Claims 2 to 10 of the patent in suit, which in turn correspond to original Claims 2 to 10 after deletion of the features (a) to (e) from Claims 8 to 10 and use of the dependent form.

4. The patent in suit concerns a propylene-ethylene random copolymer, a production process thereof and a film derived therefrom. Such subject-matter is disclosed in document (10) which was cited originally in the three statements of opposition and which the Board regards as the closest state of the art. This document describes a two-step process for the preparation of copolymers of propylene with ethylene and, optionally, a higher α -olefin. More specifically, according to its Claim 1, the first step consists in copolymerising a monomer mixture of 93.5 to 99.5 weight% of propylene and 0.5 to 6.5 weight% (i.e. 0.75 to 9.44 mol%) of ethylene, or, alternatively, a monomer mixture of 85 to 97.5 weight% of propylene, 0.5 to 5 weight% of ethylene and 2 to 10 weight% of a higher α -olefin of formula



wherein R is an alkyl radical having 2 to 6 carbon atoms, in the presence of a Ziegler catalyst system; in the second step, the resulting copolymer is subjected to a visbreaking treatment in the absence of oxygen and radical initiators at a temperature between 200 and 300°C, whereby the molecular weight is reduced to RSV values of from 1.8 to 3.5 dl/g. Although the resulting propylene copolymers exhibit desirable heat sealability properties as well as a relatively low melting point which make them suitable for the production of films (page 4, paragraph 2; page 23, Table 3), their low temperature characteristics are regarded as unsatisfactory.

In the light of this prior art teaching the problem underlying the patent in suit may thus be seen in providing propylene polymers having improved impact resistance at low temperature, without impairing the heat sealability properties or raising the low melting point thereof.

According to the patent in suit this problem is solved by increasing the relative amount of ethylene in the propylene copolymer to at least 10 mol%, or to at least 9.6 mol% when butene is used additionally, excluding the use of an inert solvent for the polymerisation reaction and visbreaking the first stage copolymer in the presence of a peroxide, as specified in Claim 1.

In view of the undisputed advantages put forward in the description, especially on page 4, lines 23 to 38, page 6, lines 43 to 60 and page 8, lines 39 to 45, as well as in the examples of the patent in suit, the Board is satisfied that the above defined technical problem is effectively solved.

5. After examination of the cited documents, the Board has come to the conclusion that this technical teaching is not disclosed in any one of them and that the subject-matter of

the patent in suit is, therefore, novel. Since the issue of novelty is no longer raised by the Appellants, it is not necessary to consider this matter in further detail.

6. It still remains to be examined whether the subject-matter of the patent in suit as defined in Claim 1 involves an inventive step with regard to the teaching of the cited documents.

6.1 As noted above in point 3, the amount of ethylene in the propylene copolymers described in document (10) does not exceed 6.5 weight% or 9.44 mol%; preferably, however, as illustrated in the examples, the amount of ethylene copolymerised is significantly lower and varies between 0.9 weight% in Example 4 and 2.5 weight% in Example 1. These values are not arbitrary, but must be regarded as a compromise between opposite requirements.

Document (10) specifies that the incorporation of comonomers, especially ethylene and higher α -olefins, into the polypropylene is known to improve the low temperature characteristics of the polymer, provided a statistical distribution of the additional units along the polymer main chain can be achieved (page 2, paragraph 2). Although this general statement would appear to suggest increasing of the amount of comonomer in order to improve the low temperature properties of polypropylene, practical difficulties in carrying out the polymerisation reaction would in fact deter the skilled person from considering solutions along this line (page 2, paragraph 4). When this reaction is performed in suspension, the higher content of comonomer in the propylene copolymer results in increased solubility of the latter in the polymerisation solvent; as a consequence, the polymer particles in the slurry state become too viscous or sticky in the polymerisation system for an easy and smooth process to be possible due to

deposition on the reactor walls, which in turn has a detrimental effect on the overall yield. Similarly, agglomeration of the polymer particles occurs in the case of gas phase polymerisation when higher amounts of comonomer are used. In the Board's view, this explicit teaching must be regarded as a warning against any further increase of ethylene in the propylene copolymer described in document (10).

- 6.2 When discussing the practical difficulties in connection with the introduction of higher amounts of comonomers in polypropylene, document (10) basically refers to suspension and gas phase polymerisation and even quotes many inert solvents which would be suitable in the former case (page 2, paragraph 4 and page 8, paragraph 6 to page 9, paragraph 1). Although there is a general statement underlined by the Appellants, according to which the polymerisation reaction could be conducted in an appropriate monomer mixture as well, this cannot be regarded as more than a passing reference since all the examples are actually performed in an inert solvent.

In the Board's view, thus, the use of a liquid monomer solvent would at most be interpreted by the skilled person as a remote alternative within the context of the process according to document (10), thus based on rather low ethylene contents. For this reason, the advantages resulting from the use of a monomer as the solvent for the polymerisation reaction were not suggested by the prior art document. In particular, the higher polymerisation activity and the lower amount of the soluble fraction of the copolymers put forward by the Respondent (patent in suit, page 5, lines 19 to 21; statement filed on 19 July 1990, page 2, paragraph 3) must be regarded as unexpected in view of the restrictive teaching of document (10).

6.3 During oral proceedings the Appellants argued that the oxidative degradation used to achieve the visbreaking of the copolymer in step (ii) of Claim 1 according to the patent in suit was known to be a mere alternative to thermal degradation and that consequently the choice made by the Respondent could not be inventive. In the Board's view, however, neither document (10) nor US-A-4 061 694, already considered in opposition procedure, would support this conclusion.

Firstly, it is essential to observe that the authors of document (10) have considered the oxidative degradation method (page 3, paragraph 4 to page 4, paragraph 1), but found it less attractive than the thermal degradation, for it requires the use of special extruders as well as expensive radical initiators which have a detrimental effect on the purity of the final product. More specifically, the radical degradation would give rise to oxidation products, such as aldehydes or fatty acids, which would affect the properties of the final product and make the latter unsuitable for the fabrication of films to be used as packaging material (page 11, paragraph 4 to page 12, paragraph 1). No reason has been given by the Appellants or can otherwise be seen by the Board as to why the recommendations of this prior art teaching should be ignored.

Nor can the teaching of US-A-4 061 694 lead to the claimed subject-matter. This document describes the treatment of a polypropylene composition, wherein a block copolymer is contacted with air, oxygen or a peroxide in a high shear zone, to achieve oxidative degradation and thereby recover a polymer product having a melt flow rate which is higher than that of the initial block copolymer, the latter containing 60 to 95 percent by weight of a polypropylene block and 5 to 40 percent by weight of a block of a random

copolymer of ethylene and another α -olefin containing 30 to 60 percent by weight of ethylene (Claim 1). Although it is specified that this treatment results in a great improvement of impact strength, especially at low temperatures (column 5, line 64 to column 6, line 2), this statement cannot be regarded as a promising teaching for the solution of the above defined technical problem, for two reasons: Firstly, this effect is completely contrary to what could be expected from previous experiences, including propylene homopolymer, according to which the degradation is accompanied by a lowering in impact strength when a polymer of low melt flow rate is degraded to a higher melt flow rate (column 6, lines 2 to 9); secondly, the copolymer to be degraded differs significantly in its structure - block structure containing only 5 to 40 weight% of random portion - and in its composition - 30 to 60 weight% of ethylene in the random portion - from the random copolymers envisaged in the patent in suit. In view of these differences, there is no basis for saying that the improvement in low-temperature impact strength observed in the prior art would indicate a similar improvement in the case of the random copolymers presently claimed.

6.4 Likewise, the level of impact strength and heatseal strength exhibited by the propylene-ethylene random copolymers according to the patent in suit must be regarded as surprising. From Figure 2 of the patent in suit as well as from the Summaries of Tables 2 and 4 in the counterstatement filed on 7 April 1990, wherein both properties are indicated as functions of the ethylene content of the copolymer, it appears quite clearly that these properties are improved significantly when the ethylene content is at least 9.6, respectively 10 mol%, as specified in Claim 1.

The argument repeatedly put forward by the Appellants that these two properties vary in fact continuously with the ethylene content and, consequently, the limits of 9.6 and 10 mol% of ethylene in the random copolymer are arbitrary, cannot be accepted. Firstly, the alleged continuity takes ethylene amounts into account (Statement of Grounds of Appeal by Appellant 1, page 3, Table) which do not correspond to actual figures taken from the prior art, but from Comparative Examples in the patent in suit; such figures cannot be taken into account to object to a lack of inventive step. Secondly, impact strength is not at all considered in document (10), let alone the features which may influence this parameter at low temperature, so that no objective comparison is possible.

- 6.5 In addition to these qualitative advantages, the combination of process features as specified in Claim 1 makes it possible to operate at commercial scale at low cost.

According to the examples of document (10), only 4 kg of propylene and less than 0.1 kg of ethylene are introduced into the reactor per hour, and the polymerisation reaction is carried out during 7 hours to give rise, after various operations of purification, drying and stabilisation, to 25 kg of polymer, which is then degraded and pelletized.

By contrast, according to Example 1 of the patent specification, which must be regarded as a reference example, 91 kg of propylene and 4 kg of ethylene are fed into the reactor per hour and 43 kg of crude propylene-ethylene copolymer can be produced per hour; it is further indicated that 200 kg of this copolymer can be purified, dried, then visbroken by oxidative degradation and finally pelletized in one single batch. This can be carried out on a continuous basis and even after 24 hours of polymerisation, one observes neither any build up of

polymer in the reactor system nor any clogging, agglomeration or conglomeration of polymer in the subsequent purification, centrifugation and drying steps. In the Board's view, these advantages were totally unforeseeable and confirm the inventiveness of the claimed subject-matter.

- 6.6 In conclusion, in the light of the above defined technical problem, it was not obvious to select the combination of features in accordance with Claim 1, or Claim 3 which is based on the same inventive idea. These claims are thus to be maintained.
7. Claims 1 and 3 being allowable, the same applies to the dependent product Claim 2 and to the dependent process Claims 4 to 7, as well as to Claims 8 to 10, which are directed respectively to a water-cooling tubular film, a T-die cast film and a composite film from the copolymer according to Claim 1, and whose inventiveness is supported by that of the main claim.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the first instance with the order to maintain the patent on the basis of the documents submitted during oral proceedings - Claims 1 to 10 and an adapted description - and the two sheets of drawings according to the patent specification.

The Registrar:



M. Beer

03959

The Chairman:



F. Antony