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Anmeldenummer / Filing No / N° de la demande : 79 302 239.3

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Bezeichnung der Erfindung: Process for producing a random ethylene terpolymer
Title of invention: and melt-shaped articles of such terpolymer
Titre de l'invention :

Klassifikation / Classification / Classement : C08F 210/16

ENTSCHEIDUNG / DECISION

vom / of / du 3 October 1989

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Mitsui Petrochemical Industries Ltd.

Einsprechender / Opponent / Opposant :

OI Dow Benelux N.V.
OII Naamloze Vennootschap DSM

Stichwort / Headword / Référence : Terpolymers/Mitsui

EPÜ / EPC / CBE Article 56 EPC

Schlagwort / Keyword / Mot clé : "Inventive step (confirmed) - improper
combination of disclosure of prior art
documents"

Leitsatz / Headnote / Sommaire



Case Number : T 175 /87 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 3 October 1989

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Decision under appeal : **Decision of Opposition Division of the European Patent Office of 6 November 1986, posted on 10 March 1987, revoking European patent No. 0 010 428 pursuant to Article 102(1) EPC.**

Composition of the Board :

Chairman : K.J.A. Jahn

Members : R.W. Andrews

R. Schulte

Summary of Facts and Submissions

- I. The mention of the grant of the European patent No. 0 010 428 in respect of European patent application No. 79 302 239.3, filed on 17 October 1979 and claiming priority of 18 October 1978 from a prior application filed in Japan, was announced on 12 October 1983 (cf. Bulletin 83/41) on the basis of eight claims. The independent Claims 1 and 8 read as follows:

"1. A process for producing a random terpolymer by polymerizing ethylene and two alpha-olefins, a first alpha-olefin of 3 or 4 carbon atoms and a second alpha-olefin of more carbon atoms than the first, in the presence of a catalyst composed of (1) a titanium catalyst component containing at least magnesium and titanium and (2) an organoaluminum catalyst component in a hydrocarbon solvent at a temperature above the melting point of the terpolymer formed to 240°C and a pressure of 196 kPa to 9807 kPa (2 to 100 kg/cm²), under such conditions that the resulting terpolymer dissolves in the hydrocarbon solvent, characterized in that said terpolymer consists of

- (A) more than 90 mole% to 99.5 mole% of ethylene,
- (B) 0.2 mole% to 9.8 mole% of an alpha-olefin of 3 or 4 carbon atoms, and
- (C) 0.2 mole% to 9.8 mole% of an alpha-olefin of 5 to 18 carbon atoms,

the total of the proportions of the monomers (A), (B) and (C) being 100 mole%.

8. Melt-shaped articles, especially packaging films, of a random terpolymer produced by a process as claimed in any one of Claims 1 to 7."

- II. Notices of opposition were filed on 11 July 1984 and 12 July 1984 in which the revocation of the patent was requested on the grounds that its subject-matter lacked novelty and did not involve an inventive step. It was further alleged that the patent did not disclose the invention in a manner sufficiently clear and complete for it to be carried out by the skilled person. The oppositions were supported, inter alia, by the following documents:
- (1) DE-A-2 803 598, and
 - (2') US-A-3 645 992.
- III. By a decision delivered orally on 6 November 1986, with written reasons posted on 10 March 1987, the Opposition Division revoked the European patent. The Opposition Division concluded that the claimed subject-matter was novel and that the disclosure of the patent was sufficient. However, the Opposition Division decided that the claimed subject-matter did not involve an inventive step in the light of the teaching of document (1) combined with that of document (2').
- IV. An appeal was lodged against this decision on 2 May 1987 and the prescribed fee duly paid. A Statement of Grounds of Appeal was filed on 20 July 1987.

In his statement and during the oral proceedings held on 3 October 1989, the Appellant submitted that the problem underlying the disputed patent was to provide terpolymers having improved transparency in terms of reduced haze values compared with the copolymers disclosed in document

(1) without a substantial deterioration in their mechanical properties. Although an improvement in haze values is generally obtained at the expense of favourable mechanical properties, nevertheless the strength properties of the films are sufficient and acceptable for use as a packaging material. The Appellant claimed that the data in the disputed patent and the results of the Experimental Report filed on 25 January 1989 clearly show that the problem underlying the patent in suit has been solved. Although, in the Appellant's view, the Experimental Report filed by Respondent OI on 20 January 1986 was open to criticism, the results reported therein, nevertheless, show an improvement in haze values for films prepared from the terpolymers obtained in accordance with the claimed process. The Appellant also maintained that only the properties of films obtained under the same conditions could be compared.

The Appellant acknowledged that the catalyst and polymerisation conditions used in the present process are disclosed in document (1). However, the last sentence of the third paragraph on page 9 would discourage the skilled person from using alpha-olefin having 3 or 4 carbon atoms and the teaching of this document, in particular comparative Example 6, would not induce the skilled person to move away from the monomers used in document (1) to the ones used in the present process. Furthermore, for the copolymers of document (1) to possess satisfactory properties they must have certain characteristics, identified as (i) to (iv). Thus according to this document not only is the choice of monomers important, but also the polymerisation conditions must be so selected that the product copolymers possess all four characteristics.

The Appellant also alleged that only certain parts of document (2') have been selected and then, with the

benefit of hindsight, combined with document (1). However the teaching of document (2') cannot be reconciled with the requirements of document (1) and, therefore, the teaching of these two documents cannot properly be combined.

- V. Respondent OI contended that the subjective problem as stated on page 2, lines 54 to 56 of the disputed patent has not been solved since, if the range of haze values for the copolymers disclosed in the Examples of document (1) are compared with the range of values for the terpolymers prepared in the Examples of the patent in suit, no improvement can be recognised and the mechanical properties of the terpolymers are considerably worse than those of the prior art copolymers. This Respondent also argued that, in view of the decision T 181/82, the results of Comparative Examples 3 and 4, those of the Experimental Report II filed by the Patentee on 25 January 1989 and those of his own Experimental Report filed on 20 January 1986 should be disregarded, since none of the prepared binary copolymers used for comparison could be considered to be known substances.

In this Respondent's opinion the objective problem underlying the disputed patent was to replace part of the expensive higher alpha-olefins in the copolymers disclosed in document (1) by the cheaper alpha-olefins having 3 and 4 carbon atoms. However, such terpolymers are known from document (2'), therefore the present process is an analogous process.

Examples 55 and 56, Table XIII and the disclosure in column 2, lines 28 to 34 and column 15, lines 42 to 45 of document (2') would direct the interest of the skilled person seeking to solve the objective problem underlying the disputed patent to this document. Since the statement

on page 9 of document (1) is only concerned with binary copolymers, it would not prevent him from combining the disclosure of these two documents.

- VI. Respondent OII agreed with Respondent OI's arguments and also contended that the improvement in haze values did not exist. The differences in haze values of the copolymers prepared by Respondent OI were within the experimental error of the method. Moreover, since the transparency of such copolymers is already very high, a slight increase in haze value would not make any noticeable difference to the transparency of the film.

Even if it is accepted that there is an improvement in haze value, this cannot be considered to be surprising since it is known from document (2') that terpolymers have good transparencies.

Characteristic (iv) of the copolymers of document (1) cannot be a distinguishing feature of the present terpolymers since these may also possess this characteristic.

- VII. The Appellant requested that the decision under appeal be set aside and the patent be maintained as granted. Alternatively, the Appellant requested that the patent be maintained on the basis of Claims 1 to 7 as granted and Claim 8 filed on 25 January 1989. As a further auxiliary request, the Appellant requested that the patent be maintained on the basis of Claims 1 to 7 as granted.

Both Respondents requested that the appeal be dismissed.

- VIII. At the conclusion of the oral proceedings, the decision was announced that the decision under appeal was set aside and that the patent was to be maintained as granted.

Reasons for the Decision

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

2. The patent in suit relates to a process for the preparation of ethylene copolymers by polymerising ethylene and alpha-olefins in the presence of a catalyst composed of a titanium catalyst component containing at least magnesium and titanium and an organoaluminium catalyst component in a hydrocarbon solvent at a temperature above the melting point of the copolymer formed to 240°C and a pressure of 196 kPa to 9807 kPa under such conditions that the said copolymer dissolves in the hydrocarbon solvent.
 - 2.1 Document (1), which may be considered to represent the closest prior art, discloses copolymers consisting essentially of ethylene and an alpha-olefin having 5 to 18 carbon atoms and possessing unique structural characteristics (cf. Claim 1). The copolymers may be prepared by polymerising ethylene and the alpha-olefin in the presence of the catalysts and under the polymerisation conditions used in the process of the disputed patent (cf. the paragraphs bridging pages 9 and 10 and pages 11 and 12).

Although the films prepared from these prior art copolymers were acceptable with respect to their impact and tear strength, it was considered that the transparency, as measured in terms of the haze values of the films, was not so satisfactory.

- 2.2 In the light of this prior art, the technical problem underlying the patent in suit may be seen in providing a process for the preparation of ethylene copolymers from which films may be manufactured having better haze values than those of films prepared from these prior art ethylene

copolymers. Although it is not necessary that the strength properties, such as the impact and tear strength, of the films be as high or higher than those of the prior art films, nevertheless, they must be above an acceptable minimum for the intended use of the films as packaging material.

According to the disputed patent this technical problem is essentially solved by random terpolymers consisting of more than 90 mole% to 99.5 mole% of ethylene, 0.2 to 9.8 mole% of an alpha-olefin having 3 or 4 carbon atoms and 0.2 to 9.8 mole% of an alpha-olefin having 5 to 18 carbon atoms obtained by polymerising the mixture of olefins using the polymerisation catalysts and polymerisation conditions disclosed in document (1).

- 2.3 In the Board's judgment, the results of Comparative Examples 3 and 4 in the published patent specification, which are confirmed by the results in the Experimental Report II submitted by the Appellant on 25 January 1989, render it plausible that this technical problem has been solved (about a 50% reduction in haze values).
- 2.4 The results of the experiments conducted by Respondent OI, which were filed on 20 January 1986, do not render this conclusion untenable, but would appear to support it. Although these comparative experiments are open to criticism on the grounds that different polymerisation conditions were used to prepare the compared binary and ternary copolymers and that the melt indices of the resulting copolymers are much lower than those of the polymers disclosed in both document (1) and the disputed patent, nevertheless the haze values of the ternary copolymers are better than those of the binary copolymers. Moreover, in the absence of any support for Respondent OII's allegation that these differences fall within the

experimental error of the method used to determine the haze values, the Board accepts that the ternary copolymers prepared in accordance with the claimed process have better haze values than those of the copolymers of document (1).

- 2.5 In the Board's view a true comparison cannot be made between the haze values disclosed in document (1) and those reported in the Examples of the disputed patent, since they were not obtained under identical conditions insofar as the films were of different thicknesses and the widths of the die slits of the machines used to form the films were not the same.
- 2.6 The Decision T 181/82 of 28 February 1984 (OJ, EPO, 1984, 401 to 414) sets out some of the prerequisites for comparative tests which are submitted as evidence to support the presence of an inventive step. One of the requirements laid down in this Decision is that only known compounds qualify for use in the comparison of compounds, including those compounds which are the inevitable result of the starting materials and the process applied thereto (cf. point 7). Applying this concept to the present case, it is found that not only the ethylene copolymers disclosed in the specific Examples of document (1) are considered to be known substances, but also those copolymers resulting from the copolymerisation of ethylene with the comonomers referred to in the second paragraph on page 9 of this document. Having regard to the fact that 1-hexene, 1-octene and 1-decene are preferred comonomers and 4-methyl-1-pentene is an especially preferred comonomer and that they are preferably copolymerised in an amount of 3 to 20% by weight of the copolymer, the copolymers of ethylene and the above-mentioned comonomers used in the Appellant's evidence (cf. comparative Examples 3 and 4 of the printed patent specification and the binary copolymers of

Experimental Report II) are considered to belong to the state of the art.

3. After examination of the cited prior art the Board has reached the conclusion that the subject-matter of the disputed patent is novel. Since novelty has been conceded and is no longer in dispute it is not necessary to consider this matter in detail.
4. It still remains to be examined whether the requirement of inventive step is met by the claimed subject-matter.
 - 4.1 As previously mentioned document (1) discloses copolymers of ethylene and alpha-olefins having 5 to 18 carbon atoms with unique structural characteristics. These copolymers, which are prepared by using the same catalysts and polymerisation conditions as the presently claimed process, provide films and sheets with improved properties compared with high pressure polyethylene and conventional ethylene copolymers.

According to this document, if an alpha-olefin having not more than 4 carbon atoms is selected as the comonomer, a copolymer having superior mechanical strength and/or transparency cannot be obtained (cf. lines 6 to 10 of the third paragraph on page 9). This statement would clearly discourage the skilled person seeking a solution to the technical problem underlying the disputed patent from using propene or 1-butene as comonomers.

The argument that this sentence should be construed as only applying to binary copolymers is not convincing. It is true that binary copolymers are in the foreground (cf. lines 1 and 5 from the bottom of page 11, page 12, lines 2, 6 and 30 and the majority of the Examples), nevertheless the teaching of the document is not res-

stricted to binary copolymers but extends to ternary and even quaternary copolymers (cf. Examples 5 and 6 and Claim 7). Therefore, the Board concludes that the skilled person reading this passage in the context of the whole document would be disinclined to contemplate using propene or 1-butene in any type of copolymer for the solution of the envisaged technical problem.

- 4.2 Document (2') discloses a process for the preparation of homogenous random partly crystalline copolymers of narrow molecular weight distribution having a homogeneity index of at least 75 by copolymerising ethylene and at least one alpha-olefin having 4 or more carbon atoms in an inert solvent therefor and for the copolymer to be prepared at a temperature of 40° to 100°C in the presence of a catalyst prepared by mixing an organoaluminium halide with a vanadium compound (cf. Claim 1). The homogeneity index as defined in this document is an empirical value of copolymer homogeneity that is calculated from the melting point of the copolymer and comonomer content using a derived equation (cf. column 6, lines 45 to 75).
- 4.3 According to this document the homogeneous copolymers exhibit a reduced haze level in extruded film, higher impact strength, reduced tendency towards delamination in extrude articles and better balance of physical properties in the machine and transverse direction of extrude film when compared with conventional heterogeneous copolymers (cf. column 2, lines 28 to 34). Table XIII of this document clearly illustrates the importance of homogeneity on the haze level, impact strength and balance of physical properties in the machine and transverse directions.
- 4.4 In order to obtain homogeneous copolymers with these advantageous properties this document places great importance on using vanadium containing catalysts having a

narrow range of compositions (cf. column 1, lines 50 to 54). Thus, the concentration of vanadium in the reaction zone is dependent upon the type of vanadium compound used and the aluminium to vanadium ratios vary depending on the type of vanadium compound and the number of carbon atoms in the alpha-olefin comonomer (cf. column 3, lines 24 to 44). Run Numbers 1 to 10, 30 to 32, 34, 36, 39 to 41, 44, 45, 48, 51 and 53, which describe the preparation of heterogenous copolymers having homogeneity indices of less than 75, clearly teach the necessity of adhering to these conditions with respect to catalyst composition and polymerisation conditions. There is no indication in this document that would suggest to the skilled person that homogeneous copolymers having the advantageous properties referred to in this document could be prepared by any other process than the one disclosed.

- 4.5 Run Numbers 55 and 56 (cf. Table VIII) describe the preparation of homogeneous terpolymers consisting of ethylene, 1-butene and 1-octene and ethylene, propylene and 1-octene respectively, which have compositions falling within the range of compositions referred to in the present Claim 1. These polymers are considered to be of practical interest because their physical properties are nearly equivalent to the correspondent octene copolymers and yet contain considerably less of the expensive octene comonomer (cf. column 15, lines 42 to 49).

Although the skilled person would immediately perceive the economic advantages provided by these terpolymers, he would, since it is central to the teachings of document (2'), conclude that homogeneous terpolymers having the favourable properties referred to in column 2, lines 28 to 34 could only be obtained by the process disclosed in this document.

In the Board's judgment, the skilled person seeking a solution to the technical problem underlying the patent in suit would not have combined the disclosures of documents (1) and (2') because of their inherent incompatibility. Thus, on the one hand, document (1) contains a warning against the use of propylene and 1-butene as comonomers and requires the presence of titanium-containing catalysts. Document (2'), on the other hand, permits the use of 1-butene as a comonomer and necessitates the presence of a vanadium-containing catalyst having narrow ranges of composition and polymerisation temperatures of between 40° to 100°C. Therefore, the combination of documents (1) and (2') could only have been made with a knowledge of the subject-matter of the disputed patent.

- 4.6 Therefore, in the Board's judgment, the cited prior art would not have suggested to the skilled person that the solution to the above-defined technical problem lay in random terpolymers having the compositions specified in the present Claim 1 prepared by polymerising the mixture of olefin in the presence of the polymerisation catalysts and under the reaction conditions disclosed in document (1).
5. In summary, the subject-matter of Claim 1 in accordance with the main request involves an inventive step. Dependent Claims 2 to 7, which relate to preferred embodiments of the process according to Claim 1, derive their patentability from this claim.
6. The novelty of the subject-matter of Claim 8 in accordance with the main request has not been contested by the Opponents (Respondents) during the opposition and subsequent appeal proceedings. It is true that two specific terpolymers (cf. Runs 55 and 56), from which melt-shaped articles are manufactured, have been disclosed in document (2'). But

these terpolymers were prepared using vanadium-containing catalysts whereas the presently claimed melt-shaped articles are made from terpolymers obtained by carrying out the polymerisation in the presence of titanium-containing catalysts. In the absence of any evidence to the contrary and having regard to the above-mentioned particularities of the process of document (2'), the Board accepts that the structures of terpolymers prepared by these different processes would not be identical.

In view of the unexpected decrease in haze level of the melt-shaped articles, the subject-matter of this claim also involves an inventive step.

7. In view of the above, the Appellant's auxiliary requests may be disregarded.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.
2. The European patent No. 0 010 428 is maintained as granted.

The Registrar:



M. Beer

The Chairman:



K. Jahn