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Aktenzeichen / Case Number / N<sup>o</sup> du recours : T 547/89 - 3.3.3

Anmeldenummer / Filing No / N<sup>o</sup> de la demande : 79 100 975.6

Veröffentlichungs-Nr. / Publication No / N<sup>o</sup> de la publication : 0 004 651

Bezeichnung der Erfindung: The use of ethylene hydrocarbon copolymers for  
Title of invention: molding purposes  
Titre de l'invention :

Klassifikation / Classification / Classement : C08L 23/08

### ENTSCHEIDUNG / DECISION

vom / of / du 21 August 1990

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /  
Titulaire du brevet :

Union Carbide Corporation

Einsprechender / Opponent / Opposant :

01) BASF Aktiengesellschaft  
02) BP Chemicals Limited

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé : "Inventive step (confirmed)"

Leitsatz / Headnote / Sommaire

Europäisches  
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Beschwerdekammern

European Patent  
Office

Boards of Appeal

Office européen  
des brevets

Chambres de recours



Case Number : T 547/89 - 3.3.3

**DECISION**  
of the Technical Board of Appeal 3.3.3  
of 21 August 1990

Appellant :  
(Opponent 01)

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Decision under appeal :

Interlocutory decision of the Opposition Division of  
the European Patent Office dated 21 June 1989  
concerning maintenance of European Patent  
No. 0 004 651 in amended form.

Composition of the Board :

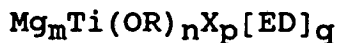
Chairman : F. Antony  
Members : C. Gérardin  
J. Stephens-Ofner

## Summary of Facts and Submissions

- I. The mention of the grant of the patent No. 4 651 in respect of European patent application No. 79 100 975.6 filed on 30 March 1979 and claiming two priorities of earlier applications in the United States of America, was published on 27 March 1985 on the basis of three claims.

Claim 1 read as follows:

"Use of low density ethylene hydrocarbon copolymer containing  $\geq 90$  mol percent of ethylene and  $\leq 10$  mol percent of at least one  $C_3$  to  $C_8$   $\alpha$ -olefin and having a density of 0.91 to 0.94 g/cm<sup>3</sup>, a molecular weight distribution  $M_w/M_n$  in the range of from 2.7 to 4.1, a total unsaturation group content of from 0.1 to 0.3 C=C/1000 carbon atoms, and a secant modulus of  $\geq 2100$  to  $\leq 5600$  bar ( $\geq 30,000$  to  $\leq 80,000$  psi), and obtainable by copolymerizing ethylene with one or more  $C_3$  to  $C_8$   $\alpha$ -olefin monomers at a temperature of from about 30° to 115°C in a gas phase containing from 0 to 2.0 moles of hydrogen per mol of ethylene in the presence of particles of an activated precursor composition wherein said precursor composition has the formula



wherein

m is  $\geq 1.5$  to  $\leq 56$

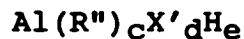
n is 0 or 1

p is  $\geq 6$  to  $\leq 116$

q is  $\geq 2$  to  $\leq 85$

R is a C<sub>1</sub> to C<sub>14</sub> aliphatic or aromatic hydrocarbon radical, or COR' wherein R' is a C<sub>1</sub> to C<sub>14</sub> aliphatic or aromatic hydrocarbon radical,  
 X is selected from the group consisting of Cl, Br, I or mixtures thereof;

ED is a liquid organic electron donor compound in which said precursor composition and the Ti and Mg components thereof are soluble and which is selected from the group consisting of alkyl esters of aliphatic and aromatic carboxylic acid, aliphatic ethers, cyclic ethers and aliphatic ketones,  
 said precursor composition having an average particle size of 10 to 100 μm, a bulk density of 0.29 to 0.53 g/cm<sup>3</sup>, and being mechanically mixed or blended with at least one solid inert carrier material in a ratio of 0.033 to 1 part of precursor composition per part by weight carrier material, and being activated with an activator compound having the formula



wherein X' is Cl or OR''', R'' and R''' are the same or different and are C<sub>1</sub> to C<sub>14</sub> saturated hydrocarbon radicals,  
 d is 0 to 1.5,  
 e is 0 or 1, and

$$c+d+e=3$$

which polymerization is conducted continuously in a gas phase fluid bed reaction zone under a pressure of <69 bar (<1000 psi) and said precursor composition is partially activated with >0 to <10 mols of activator compound per mol of titanium compound in said precursor composition in a hydrocarbon slurry prior to feeding of the

precursor composition to said reaction zone, dried to obtain a free-flowing solid particulate material, and then being completely activated in said reaction zone with  $\geq 10$  up to a total of  $\leq 400$  mols of activator compound per mol of titanium compound in said precursor composition, said complete activation being effected by spraying additional activator compound into the reactor in the form of a solution in a hydrocarbon containing 2 to 30 weight percent of the activator compound, or absorbed on a carrier material containing 10 to 50 weight percent of the activator compound;

for preparing articles by injection molding, roto molding and blow molding."

- II. On 16 November 1985 Opponent 1 (the Appellant) filed a notice of opposition to the granted patent and requested revocation thereof on the ground of lack of inventive step.

Opponent 2 lodged an opposition on 23 December 1985 against the grant of the patent and requested revocation for non-compliance with the requirements of Article 100a) and b) EPC.

These various objections, which were emphasised and elaborated in later submissions, were based mainly on the following documents:

- (7) FR-A-2 347 388
- (11) US-A-4 076 698.

- III. By a decision dated 21 June 1989, the Opposition Division held that there were no valid grounds of opposition to the maintenance of the patent in amended form on the basis of a

new set of two claims submitted during oral proceedings held on 9 December 1988, the amendments to Claim 1 consisting in the replacement of the expression "at least one C<sub>3</sub> to C<sub>8</sub>  $\alpha$ -olefin" by "propylene and/or butene-1" and in the incorporation of a melt index value of 7 to 100 g/10 min in the definition of the copolymer.

The Opposition Division held that the disclosure of the patent in suit was sufficient and that the claimed subject-matter was novel. As to inventive step, it stated that no hint was given in document (11), regarded as the closest state of the art, that copolymers of ethylene with propylene and/or butene-1 would be suitable for preparing articles by injection moulding or roto moulding having high stress crack resistance, improved low temperature properties and good stiffness. Likewise, document (7) taught that the higher  $\alpha$ -olefins, i.e. those having more than four carbon atoms, had a beneficial effect on stress crack resistance, which would not lead to the copolymers used in the patent in suit.

IV. The Appellant thereafter filed a notice of appeal on 17 August 1989 and paid the prescribed fee at the same time. The arguments presented in the Statement of Grounds of Appeal filed on 24 October 1989 and during oral proceedings held on 21 August 1990 can be summarised as follows:

Document (11) explicitly taught that copolymers of ethylene with propylene and butene-1 showed good stress crack resistance and impact strength, although high crystallinity could be obtained only with higher  $\alpha$ -olefins. This clearly suggested the suitability of copolymers of ethylene with propylene or butene-1 for the moulding applications claimed by the Respondent (Patentee).

In addition, the Appellant relied on some of the late filed documents submitted in the course of the opposition procedure, as well as on several additional documents filed together with the Statement of Grounds of Appeal and lastly on a new document (US-A-3 645 992), introduced as late as the oral proceedings.

- V. In the counterstatement of appeal and during oral proceedings the Respondent put forward essentially the following arguments:

Document (11) did not teach the use of copolymers of ethylene with propylene and/or butene-1 for preparing moulded articles having the desired mechanical properties, but rather that these two comonomers would not be very effective for this purpose. From the experimental data, one could only conclude that propylene and butene-1, unlike the higher  $\alpha$ -olefin comonomers, did not substantially improve the mechanical properties of ethylene polymers. Having regard to document (7), this reference merely confirmed the teaching of document (11) in that it specified that higher  $\alpha$ -olefins should be employed if copolymers with a high stress crack resistance were to be produced.

- 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.

#### Reasons for the Decision

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

2. The Board has considered the documents cited after expiration of the nine-month opposition period in order to determine their relevance, namely their evidential weight compared with that of the documents filed in time, and has found that none of them was relevant in the above sense. It has, therefore, decided to disregard them pursuant to Article 114(2) EPC.
  
3. As was stated in point III above, the wording of Claim 1 differs from the use claim as granted by a more specific definition of the  $\alpha$ -olefin comonomer and by the incorporation of a melt index value of 7 to 100 g/10 min, thus is more limited (Article 123(3) EPC). The choice of propylene and/or butene-1 as comonomer(s) is disclosed on page 3, lines 25/26 of the description of the patent specification, corresponding to page 4, lines 6 to 10 of the application as originally filed. The melt index range, which results from the combination of the lower limit of the preferred melt index range with the upper limit of the basic melt index range, is supported by the disclosure on page 4, lines 6/7, respectively page 7, lines 7 to 9.

As far as Claim 2 is concerned, the only difference concerns the melt index range which now corresponds to the preferred melt index range mentioned above and is thus admissible for the same reasons.

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

4. The patent in suit concerns the use of ethylene hydrocarbon copolymers for moulding purposes. A similar subject-matter is disclosed in document (11), which the Board, like the Opposition Division, regards as the closest state of the art. This document describes copolymers of ethylene containing 3 to 7% by weight of at least one normal

aliphatic  $\alpha$ -olefinic hydrocarbon with 5 to 10 carbon atoms and having, among other properties, a melt index within the range of 0.3 to 20 and a density of 0.93 to 0.94 (Claim 1). These copolymers, which are prepared by using conventional catalyst systems based on titanium tetrachloride (Examples 13 to 20), can be injection moulded into various articles showing a desirable combination of mechanical properties, especially good impact toughness and stress crack resistance (column 2, lines 42 to 47). However, the low temperature properties of these articles cannot be regarded as satisfactory.

In the light of this shortcoming, the technical problem underlying the patent in suit can be regarded as providing an ethylene copolymer which, when used in injection moulding operations, confers improved low temperature properties upon the moulded articles, without impairing their impact toughness and stress crack resistance.

This problem is solved according to Claim 1 of the patent in suit by using a copolymer of ethylene with minor amounts of propylene and/or butene-1, having a melt index of 7 to 100 g/10 min, a molecular weight distribution Mw/Mn in the range of 2.7 to 4.1, a total unsaturation group content of from 0.1 to 3 double bonds per 1000 carbon atoms, and a secant modulus of from 2100 to 5600 bar, and which has been obtained by means of a specific catalyst system activated in two steps.

In view of the properties mentioned in the examples of the patent in suit, especially in Tables II, IV and VIII, in which the beneficial effects of the above combination of compositional and operative features on impact strength at -40 and -51°C are illustrated, the Board is satisfied that the above-defined technical problem has been effectively

solved. These advantages have not been disputed by the Appellant.

5. After examination of the cited documents, the Board has reached the conclusion that the solution as claimed by the Appellant is not disclosed in any of them and that the subject-matter of the patent in suit is, therefore, novel. Since the issue of novelty has not been in dispute at the appeal stage, it is not necessary to consider this matter in detail.
6. It still remains to be examined whether the claimed subject-matter involves an inventive step with regard to the cited documents.
  - 6.1 The fact that the description of document (11) gives a somewhat broader definition of suitable comonomers by suggesting a range of from 4 to 18 carbon atoms, and making a specific reference to butene-1 (column 3, lines 15 to 37, in particular line 24), and even indicates some mechanical properties of copolymers of ethylene with propylene and butene-1 (Tables II, V and VI), raises the question whether, as the Appellant argued, the literal teaching of this document implicitly extends to these two comonomers and thereby points to the solution claimed by the Respondent.
    - 6.1.1 The passage referred to by the Appellant (column 3, lines 19 to 25) indicates that the  $\alpha$ -olefins which contain more than 4 carbon atoms, preferably from 5 to 18 carbon atoms, per molecule, are the most outstanding in producing resins with high impact strength and excellent stress crack resistance, while at the same time having relatively high crystallinity. It is further specified that, whilst propylene is not effective, butene-1 shows only marginal effectiveness in this respect; on the other hand,  $\alpha$ -olefins

with more than 18 carbon atoms are not only of low commercial interest because of their cost, but produce branches which are sufficiently long to crystallise per se and behave in a manner similar to the polymer backbone, which has an overall detrimental effect on the mechanical properties of the copolymer. A graph plotting these properties against the number of carbon atoms in the  $\alpha$ -olefin comonomer goes thus through a maximum.

From the foregoing, in the Board's view, there can be no doubt that a good balance of mechanical properties can only be obtained with copolymers of ethylene and  $\alpha$ -olefins having 5 to 18 carbon atoms.

6.1.2 This conclusion is confirmed by other passages of the description of this document as well as by experimental results reported therein.

It is first recalled (column 4, lines 17 to 34) that certain film and substrate coating applications for ethylene polymers require low liquid and vapour transmission rates, high stiffness and adequate impact toughness, for which neither linear polyethylenes, nor conventional branched polyethylenes qualify. By contrast, ethylene copolymers containing a small amount of  $\alpha$ -olefin with more than 5 carbon atoms as comonomer exhibit high crystallinity and impact toughness. It is explicitly specified that the effect resulting from the use of butene-1 as comonomer would be marginal.

Similarly, the data in Table V, wherein the stress crack resistance of various ethylene copolymers is rated, show that only copolymers of ethylene with  $\alpha$ -olefins having at least 5 carbon atoms have sufficient stress crack resistance to qualify for those wire coating applications in which both stress crack resistance and high

crystallinity are required (column 9, lines 16 to 33). The number of hours to failure at 100°C (stress crack resistance test) provides evidence that ethylene-butene-1 copolymer (106 hours) has a behaviour much closer to linear polyethylenes (8 to 104 hours) than to ethylene-pentene-1 copolymer (640 hours) or ethylene-decene-1 copolymer (at least 860 hours).

Likewise, the data given in Table VI demonstrate that only the ethylene- $\alpha$ -olefin copolymers produced with  $\alpha$ -olefins containing 5 or more carbon atoms have the stress crack resistance and stiffness required for moulding applications like injection moulding, blow moulding and vacuum forming (column 10, line 25 to column 11, line 4). As above, it is underlined that shorter and longer branched copolymers do not have sufficient stress crack resistance. The figures of Table VI for the failures of the unnotched samples in air at 50 and 100°C clearly show that ethylene-butene-1 copolymers, although admittedly better than the various linear polyethylenes, under the stated test conditions are less satisfactory than ethylene copolymers with decene-1, tetradecene-1 and octadecene-1.

6.1.3 From the foregoing, in the Board's view, there can be no doubt that the teaching of document (11) points towards the use of copolymers of ethylene and  $\alpha$ -olefins having 5 to 18 carbon atoms.

6.2 A similar conclusion can be drawn from document (7), which is concerned first of all with a process for the production of ethylene copolymers (Claim 1). These copolymers show specific properties which make them particularly suitable for injection moulding, blow moulding, extrusion moulding and similar applications (page 9, lines 31 to 38). Although propylene and butene-1 are quoted as suitable comonomers from a merely preparative point of view, higher  $\alpha$ -olefins

are particularly preferred when environmental stress crack resistance and low temperature impact strength are to be improved (page 5, lines 28 to 35). In the Board's view, this teaching points unambiguously in a direction opposite to the solution claimed by the Respondent.

6.3 Quite apart from the compositional aspect of the copolymer, it is also necessary to consider the operative features of its process of preparation, since Claim 1 of the patent in suit is drafted as an use claim of a product characterised by its process of preparation. In this respect, without even considering possible differences in the process features between the patent in suit and the prior art illustrated by documents (7) and (11), the Board notes that a critical feature of the claimed subject-matter is the method of activation of the catalyst. As mentioned in the patent specification (page 5, line 55 to page 6, line 7), in order to be suitable for the preparation of the copolymers presently used, the catalyst must be activated in two steps; more specifically, the precursor composition is first partially activated outside the polymerisation reactor in a hydrocarbon solvent slurry, then, after removal of this solvent by drying, fed to the polymerisation reactor where activation is completed by treatment with additional activator compound. This essential feature is not suggested by either of the above prior art documents.

6.4 The combination of compositional and operative features according to Claim 1 of the patent in suit confers further unexpected advantages to ethylene copolymers when used in injection moulding, roto moulding and blow moulding operations. In addition to the superior stress crack resistance and improved low temperature properties aimed at according to the definition of the problem underlying the patent in suit, the copolymers of ethylene and propylene

and/or butene-1 have high surface gloss as well as low warpage and are further essentially free from dull swirls (patent specification, page 2, lines 38 to 40). These properties are particularly advantageous in the case of the articles envisaged in the patent in suit, namely lids, closures, food containers, bottles, dishpans and toys (page 2, lines 5 to 8). This has not been disputed by the Appellant.

- 6.5 In conclusion, for all the above reasons, the combination of features according to Claim 1 cannot be regarded as obvious with respect to the prior art cited and involves an inventive step.
7. Claim 1 being allowable, the same applies to dependent Claim 2, which is directed to the use of a preferred ethylene copolymer and whose patentability is supported by that of the main claim.

#### Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

E. Görgmaier

F. Antony