BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS | BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE | CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

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File Number: T 248/90 - 3.3.3

Application No.: 84 103 441.6

Publication No.: 0 120 503

Title of invention: Preparation of low density, low modulus ethylene copolymers in a fluidized bed

Classification: C08F 210/16

D E C I S I O N
of 4 April 1991

Proprietor of the patent: UNION CARBIDE CORPORATION

Opponent: BASF Aktiengesellschaft, Ludwigshafen

Headword:

EPC Article 56

Keyword: "Inventive step (confirmed)"

Headnote

Case Number : T 248/90 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 4 April 1991

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Decision under appeal :
Decision of Opposition Division of the European Patent Office dated 31 January 1990 rejecting the opposition filed against European patent No. 0 120 503 pursuant to Article 102(2) EPC.

Composition of the Board :

Chairman : S. Schoedel
Members : C. Gérardin
R. Schulte

Summary of Facts and Submissions

I. The mention of the grant of the patent No. 120 503 in respect of European patent application No. 84 103 441.6 filed on 28 March 1984 and claiming the priorities of 29 March 1983 and 13 March 1984 from two earlier applications in the United States, was published on 1 April 1987 on the basis of thirteen claims.

Claim 1 read as follows:

"A continuous process for producing ethylene copolymers having a density of less than 0.91 g/cm^3 and a 1% secant modulus of less than 140,000 kPa in a fluidized bed without particle agglomeration, said copolymers containing no more than 94 mol percent of polymerized ethylene and at least 6 mol percent of polymerized alpha olefin containing from 3 to 8 carbon atoms, which comprises continuously contacting, in a fluidized bed reaction zone, at a temperature of from 10°C up to 80°C and a pressure no greater than 7000 kPa, a gaseous mixture containing (a) ethylene and at least one higher alpha olefin containing from 3 to 8 carbon atoms, in a molar ratio of such higher alpha olefin to ethylene of from 0.35:1 to 8.0:1, and (b) at least 25 mol percent of at least one diluent gas, with particles of a catalyst system comprising a precursor composition having the formula

$$Mg_mTi(OR)_nX_p[ED]_q$$

wherein

R is an aliphatic or aromatic hydrocarbon radical containing from 1 to 14 carbon atoms, or COR' wherein R'

is an aliphatic or aromatic hydrocarbon radical containing from 1 to 14 carbon atoms,

X is selected from the group consisting of Cl, Br, I, and mixtures thereof,

ED is an organic electron donor compound selected from the group consisting of alkyl esters of aliphatic and aromatic acids, aliphatic ethers, cyclic ethers and aliphatic ketones,

m is 0.5 to 56,
n is 0, 1 or 2,
p is 2 to 116, and
q is 2 to 85,

said precursor composition being diluted with an inert carrier material and completely activated with an organoaluminum compound having the formula

$$Al(R")_dX'_eH_f$$

wherein

X' is Cl or OR"',
R" and R"' are saturated hydrocarbon radicals containing from 1 to 14 carbon atoms,
e is 0 to 1.5,
f is 0 or 1, and
d+e+f=3,

said activator compound being employed in an amount such as to provide a total aluminium:titanium molar ratio in said reaction zone from 10:1 to 400:1."

Claims 2 to 13 were dependent claims directed to preferred embodiments of the main claim.

II. On 22 December 1987 the Opponent filed a notice of opposition against the grant of the patent and requested revocation thereof for non-compliance with the requirements of Articles 52 to 57 EPC. It was first objected in general terms that the subject-matter of the patent in suit was not novel. Regarding the issue of inventive step it was then argued on the basis of the teaching of EP-A3-4 651 (document (1)) and EP-A3-4 646 (document (2)), wherein the main features of the claimed process were disclosed, that low temperatures were commonly used for the production of low density polymers.

These objections were emphasised and elaborated in a later submission, wherein it was referred to following additional documents

(3) GB-A-2 066 274
(4) DE-A-2 942 367
(5) Polyolefins - Structure and Properties by Herman V. Boenig, Elsevier Publishing Company, Amsterdam 1966, pages 118 and 119,

from which it appeared that the value of the 1% secant modulus specified in Claim 1 was in fact common and the use of large amounts of hydrogen as diluent gas well known in the art.

III. By decision of 31 January 1990 the Opposition Division rejected the opposition. It was first stated in that decision that the statement, according to which in its general aspect the subject-matter of the patent in suit was not novel, did not constitute an objection of lack of novelty. In this respect, it was specified that, although document (2) mentioned the use of up to 2 mols of hydrogen

per mol of ethylene, this feature was only optional; moreover, this teaching was directed to the preparation of ethylene homopolymers having a density outside the range envisaged by the Patentee. Further, in view of the upper limit of 0.91 g/cm^3 for that parameter in Claim 1 of the patent in suit, only documents (3) and (4) were to be considered for the issue of inventive step; in both documents, however, the use of hydrogen was only optional, which did not suggest the criticality of large amounts thereof in order to prevent formation of polymer agglomerates and sustain continuous polymerisation.

IV. The Appellant (Opponent) thereafter filed a notice of appeal on 28 March 1990 and paid the prescribed fee at the same time. The arguments presented in the Statement of Grounds of Appeal filed on 31 May 1990 and during oral proceedings held on 4 April 1991 concentrated first on the scope of document (1). The restrictive interpretation made by the Opposition Division of that citation could not be accepted, since a density of 0.9 g/cm^3 was actually mentioned for ethylene copolymers in Claim 3, which made that teaching relevant for the solution of the technical problem underlying the patent in suit; furthermore, amounts of hydrogen corresponding to 26.95, 27.1 and 28.6 mol % per mol of monomer were reported in Examples 2, 12 and 13. Consequently, the claimed subject-matter was a mere combination of that feature with the teaching of document (4). Nor could the range of values of the 1% secant modulus specified in Claim 1 of the patent in suit be regarded as inventive, since it was known from document (5) to control the stiffness of ethylene copolymers by density.

V. The arguments presented by the Respondent (Patentee) in the counterstatement of appeal filed on 17 December 1990 and during oral proceedings can be summarised as follows.

Firstly, it was noted that novelty was no longer under attack. Secondly, regarding the question of inventive step, the reference to Examples 2, 12 and 13 in document (1) was inappropriate, since, on the one hand, the amount of comonomer used therein was too low to give rise to copolymers having a density lower than 0.91 g/cm^3, and, on the other hand, an increase of the concentration of comonomer in order to meet that requirement would have resulted in polymer agglomeration. Further, the figure of 0.90 g/cm^3 was rather inconsistent with the general description of that citation, although it may have been disclosed as such in the original US application. As to the inert gas referred to in documents (3) and (4), it was only present during the pre-treatment of the catalyst with olefin; this could not be equated with the use of inert gas during polymerisation.

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

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 patent in suit concerns a continuous process for the preparation of low density, low modulus ethylene copolymers in a fluidised bed. A similar subject-matter is disclosed in document (4), which the Board, like the Opposition Division, regards as the closest state of the art. That citation describes a process for the preparation of copolymers of ethylene and 8 to 60 mol percent of butene-1 having a density of 0.85 to 0.91 g/cm^3; the reaction is carried out in vapour phase at a temperature

of from 20 to 110°C in the presence of a catalyst comprising a solid substance, an organoaluminium compound as well as, optionally, an organocarboxylic acid ester, the solid substance containing a magnesium-containing inorganic solid carrier and a titanium compound and/or vanadium compound (Claims 1 and 11 in combination with page 11, paragraph 5). In practice, hydrogen is fed to the polymerisation reactor together with the two monomers in such proportions that the gaseous mixture contains 65 to 75 mol percent of ethylene, 15 to 25 mol percent of butene-1 and 10 mol percent of hydrogen (Claim 12 and Examples 1 to 4). This combination of features, whereby a high productivity is achieved while simultaneously the difficulties caused by polymer deposition and agglomeration within the reactor are overcome to a large extent, makes it possible to prepare ethylene copolymers having a desirable combination of properties, such as transparency, outer appearance, luster, flexibility and rubber-like elasticity (page 8, paragraph 3 to page 9, paragraph 2). Although the description underlines that the polymerisation reaction proceeds extremely smoothly (page 8, paragraph 3, last sentence) and the working examples even refer to a continuous process, in practice that reaction is stopped after ten hours to allow separation of the resulting polymer by means of a dry cyclone.

In the light of this prior art teaching the technical problem underlying the patent in suit can thus be seen in modifying the above process in order to allow uninterrupted polymerisation for weeks at a time, without impairing the above-mentioned properties of the resulting copolymers.

According to the patent in suit, this problem is solved by using a gaseous mixture containing at least 25 mol percent

of at least one diluent gas and by carrying out the polymerisation reaction in a fluidised bed.

In view of the experimental results in the patent specification, especially the data in Tables 1 to 5, which show the criticality of the combination of features and parameters as specified in Claim 1, the Board is satisfied that the above-defined technical problem has been effectively solved. These results have not been disputed by the Appellant.

3. 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 Appellant has not raised the issue of novelty at the appeal stage and even explicitly acknowledged that the requirement of novelty was met, it is not necessary to consider this matter in detail.

4. It still remains to be examined whether the claimed subject-matter involves an inventive step with regard to the cited documents.

4.1 The optional features disclosed in document (4) cannot lead to the solution according to the patent in suit. One of these alternative embodiments comprises contacting the foregoing catalyst system with an α-olefin before using it in the vapour phase polymerisation reaction, whereby the polymerisation activity can be improved to a large extent and the operation can be performed more stably (page 13, paragraph 1). Additionally, hydrogen or another inert gas, such as nitrogen, argon or helium, may be present during that pre-treatment (page 13, paragraph 3, last sentence). However, there is no mention of a suitable amount of hydrogen or inert gas, nor any indication about possible

advantages resulting from that specific process feature. It follows that, even if the skilled man decided to operate along that alternative, hydrogen would be partly incorporated into the catalyst and partly combined with the monomer feed, which cannot suggest the process feature defined under (b) of Claim 1 of the patent in suit.

The second feature to be considered is the polymerisation process itself. As noted above in point 2, the examples in document (4) show that the polymerisation reaction is interrupted after ten hours. Whereas the Appellant put forward that this is a mere checking operation in order to make sure that the polymer does not adhere to the inner wall, stirrer and polymer withdrawing pipe, and that this interruption does not affect the continuity of the process, the Respondent pointed out that it was in fact necessary to separate the polymer already formed, whether there was adhesion or not, and that this was actually the purpose of the dry cyclone (page 14, Example 1, paragraph 2). In the Board's view, this difference of interpretation by the parties is a minor point with regard to the fact that the polymer is said to be not only low in density, but highly sticky (page 8, paragraph 3). For the skilled man, this would not be an incentive to operate under the same conditions in a fluidised bed.

For these reasons, none of the alternatives envisaged in document (4) would suggest the solution claimed in the patent in suit.

4.2 Similar considerations apply to the teaching of document (3), which is concerned with a very similar process for the polymerisation in vapour phase of ethylene and 4 to 250 mol percent of an α-olefin giving rise to copolymers of density of 0.85 to 0.91 g/cm^3 (Claim 1). Like above, the pre-treatment of the catalyst with

ethylene and/or an α-olefin can be performed in the presence of hydrogen or another inert gas, such as nitrogen, argon or helium (page 4, lines 15 to 22 and 58 to 62). Additionally, hydrogen may be present in the vapour phase in concentration up to 5 mol percent, this limit being critical for the general properties of the polymers (page 4, lines 105 to 110). This embodiment is illustrated in Example 2, according to which the gases fed to the autoclave comprise 75 mol percent of ethylene, 23 mol percent of butene-1 and 2 mol percent of hydrogen. Further, as in the case of document (4), the polymerisation reaction is interrupted after ten hours (see Examples 1 to 6) and emphasis is put on the stickiness of the polymer formed as well as on the low polymer adhesion to the reactor and agglomeration of polymer particles (page 2, lines 74 to 81).

Thus, although this teaching is less restrictive than that of document (4) in that it is not limited to butene-1 as comonomer, but extends to the copolymerisation of any α-olefin with ethylene, the limit of 5 mol percent of hydrogen in the vapour phase not to be exceeded does not suggest the large amounts of inert gas according to the claimed solution. Likewise, the stickiness of the particles speaks against the preparation of such polymers in a fluidised bed.

It follows that document (3) cannot contribute to the solution of the above-defined technical problem.

4.3 During oral proceedings the Appellant pointed out for the first time that document (1) was not limited, as generally admitted hitherto, to ethylene copolymers having a density

within the range of 0.91 to 0.94 g/cm^3 as specified in Claim 1 and the description, but according to independent Claim 3 encompassed polymers having a density as low as 0.90 g/cm^3. This made that teaching relevant for the solution of the above-defined problem.

This document, whose Applicant is the Respondent, describes the preparation of copolymers of ethylene and 1 to 10 mol percent of an α-olefin having 3 to 8 carbon atoms in a fluid bed process, wherein the catalyst system and operative features (page 6, paragraph 2 to page 21, paragraph 2) correspond to a large extent to the definitions given in the patent specification. The data in the Table on page 16 show that the requirements regarding the comonomer: ethylene molar ratio in the gas stream and in the polymerised units as specified in the patent in suit are met as well, at least in some cases. Further, it is mentioned that up to 2 moles of hydrogen per mole of monomer in the gas stream may be used and that any gas inert to the catalyst and the reactants may be present as well (page 19, lines 24 to 31). According to Claim 1, the density of the resulting polymers is within the range of 0.91 to 0.94 g/cm^3; according to independent Claim 3, that parameter can vary from 0.90 to 0.94. This discrepancy is attributed by the Respondent to the fact that the whole set of claims, in particular Claims 1 and 3, corresponds to the original version in the US application, wherein two different ranges of density were defined independently. This explanation as well as the value of 0.90 being plausible, it will be assumed hereinafter that ethylene copolymers having a density as low as 0.90 g/cm^3 are envisaged in document (1).

The experimental data in Tables A and B, pages 33 and 34, do not show any relation in the sense suggested by the Appellant between the amount of hydrogen in the monomer

gas stream and the density of the copolymer. Firstly, there is no single run in which a polymer having a density lower than 0.91 g/cm^3 is produced which could be regarded as a reference. Secondly, in contradistinction to the Appellant's contention, one observes in Runs 2, 12 and 13 in Example 1 that the highest hydrogen:ethylene ratios correspond to the highest densities; this finding cannot be an incentive to operate with large amounts of hydrogen in order to produce low density polymers. If, on the other hand, one follows the Appellant's alternative argument, according to which density is determined primarily by the comonomer:ethylene ratio, then no conclusion can be drawn about the amount of hydrogen from Tables A and B, since the reaction conditions in the various runs differ by more than one parameter, so that no useful comparison can be made. This means that in any case, i.e. even if one adopts for document (1) the interpretation the most favourable for the Appellant, no useful conclusion can be drawn for the solution of the above defined technical problem.

4.4 As far as document (2) is concerned, it cannot suggest the claimed subject-matter, as noted by the Opposition Division, since it is directed to the preparation of ethylene homopolymers having a density of 0.958 to 0.972 g/cm^3.

4.5 It remains to consider the 1% secant modulus parameter which together with density characterises the ethylene copolymers produced by the claimed process.

During oral proceedings the Appellant argued on the basis of document (5), especially the Table and the diagram of Figure 15 on page 119, that modulus was closely related to density, i.e. it decreased with the latter; consequently, the value of 140 000 kPa for the 1% secant modulus was regarded as normal for polymers having a density as low as

0.91 g/cm^3 or less. In the Board's view, however, the correlation between modulus and density on Figure 15 is not very accurate in the case of low densities, since the five copolymers having densities between about 0.918 and 0.932 g/cm^3 have a comparable modulus. Further, the Appellant, who as Opponent has the onus of proof, has not provided any evidence that copolymers having the required density, such as those described in document (4), would also meet the requirement regarding the 1% secant modulus. Nor could the Respondent provide additional information in this respect.

This has led the Board to not rely on that parameter for the definition of the technical problem underlying the patent in suit and to adopt a less ambitious line based on resulting ethylene copolymers having properties more or less equivalent to those disclosed in document (4), i.e. the line of an alternative process. It follows that any improvement regarding the stiffness of these polymers would be an additional technical effect in favour of an inventive step. Irrespective of such an effect, however, the correspondence between the polymerisation temperature and the secant modulus as illustrated in Figure 1 of the patent specification cannot be inferred from the prior art; in particular, the definition of an operable region, within which the secant modulus may be determined for each temperature, must be regarded as a surprising result contributing to the inventiveness of the claimed process.

4.6 In conclusion, for these various reasons, the subject-matter of Claim 1 involves an inventive step.

5. Claim 1 being allowable, the same applies to dependent Claims 2 to 13, which are directed to preferred embodiments of the subject-matter of Claim 1 and whose inventiveness 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 S. Schödel