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 434/90 - 3.3.3

Application No.: 80 900 786.7

Publication No.: 0 027 474

Title of invention: Process for producing polyolefin

Classification: C08F 10/00

D E C I S I O N
of 2 February 1993

Applicant: Mitsui Petrochemical Industries, Ltd.

Opponent: Hoechst Aktiengesellschaft, Frankfurt

Headword:

EPC Article 56

Keyword: "Inventive step - denied"

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number : T 434/90 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 2 February 1993

Appellant : (Proprietor of the patent)
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Decision under appeal :
Decision of the Opposition Division of the European Patent Office dated 22 March 1990 revoking European patent No. 0 027 474 pursuant to Article 102(1) EPC.

Composition of the Board :

Chairman : F. Antony
Members : C. Gérardin
F. Benussi

Summary of Facts and Submissions

I. The mention of the grant of the patent No. 0 027 474 in respect of European patent application No. 80 900 786.7 filed on 23 April 1980 and claiming the priority of 23 April 1979 from an earlier application in Japan, was published on 12 August 1987 on the basis of 15 claims, Claim 1 reading as follows:

"A process for preparing a polyolefin having predetermined properties by polymerizing at least one olefin in the presence of a Ziegler catalyst and hydrogen in an inert aliphatic hydrocarbon solvent in a gas/liquid phase reactor having a continuous gas phase space above the liquid phase which is characterized by:

a) introducing said catalyst, olefin and hydrogen into the reactor;

b) analysing olefin and hydrogen concentrations in the gas phase within the reactor by gas chromatography;

c) generating input signals representative of said detected concentrations;

d) comparing said input signals with preset values corresponding to said predetermined properties;

e) generating control signals as a function of deviations between said input signals and said preset values; and

f) controlling the feed rates of said hydrogen and said olefin responsive to said control signals."

Claims 6 and 11 are formally independent process claims, which essentially recite all the process features of Claim 1 and wherein additionally the control of the process is achieved by means of a computer. The other Claims 2 to 5, 7 to 10 and 12 to 15 are dependent claims directed to preferred embodiments of the process according to Claims 1, 6 and 11, respectively.

II. On 10 May 1988 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 a later submission, was based essentially on the following documents:

(1) US-A-3 691 142,
(3) US-A-4 003 712, and
(6) Journal of Chromatographic Science, Volume 7, 1969, pages 321 and 322.

III. By a decision issued on 22 March 1990 the Opposition Division revoked the patent on the ground that the subject-matter of the three independent process claims did not involve an inventive step. More specifically, it was stated in this decision that the control of the properties of polyolefins prepared in a fluidised bed reactor on the basis of an analysis of the monomer and hydrogen concentrations in the gas phase was known from document (3); the use of gas chromatography to carry out gas analysis being described in document (6), the combination of these two teachings could not be regarded as inventive.

IV. The Appellant (Patentee) thereafter filed a Notice of Appeal against this decision on 22 May 1990 and paid the prescribed fee at the same time. In the Statement of Grounds of Appeal filed on 11 July 1990 and in a subsequent statement as well as during oral proceedings requested by both parties and held on 2 February 1993 the Appellant argued that in document (3) the control of the gas composition was self-evident, since there the polymerisation was performed in the gas phase. Whereas however the reaction conditions in the gas phase were well

known in the art, there were unknown factors which caused a disturbance in the gas/liquid phase reaction; in particular, there was no direct relationship between the concentrations of the monomers in the liquid phase and in the gas phase once the polymerisation reaction had started. Moreover, the process disclosed in document (3) aimed at the preparation of polyolefins having low catalyst residues, which was different from the control of the properties of these polymers. The teaching of document (6) was not combinable therewith, since there was no hint that gas chromatography would be suitable to analyse the gas composition in a gas/liquid phase polymerisation process.

V. In its written and oral submissions the Respondent (Opponent) maintained that a correlation between the gas phase and the liquid phase compositions could well be inferred from document (3). Further, the use of gas chromatography to analyse the gas phase could not be regarded as an inventive feature, since document (6) described such a method to analyse a gas mixture containing olefins. Besides, document (11) = Journal of American Chemical Society, 83, 1961, 2654 to 2658, cited for the first time in the Counterstatement of Appeal, not only showed that in a gas/liquid polymerisation process the concentration of monomers in the liquid phase and, thereby, the properties of the polymers could be controlled via the partial pressure of these monomers in the gas phase, but even specified that gas chromatography was suitable to analyse the gas composition. Document (1) provided evidence that in a continuous gas/liquid phase polymerisation process a control of the reaction was possible by means of the gas phase pressure.

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

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 admissible.

2. The arguments presented by the Appellant during oral proceedings to demonstrate that document (3) could not, either in isolation, or in combination with document (6), lead to the process as defined in any of Claims 1, 6 or 11, brought to light that this citation is not, in the Board's view, an optimal starting point for the definition of the technical problem underlying the patent in suit. As explained by the Appellant to the Board's satisfaction, in the fluid bed reactor system described therein the perturbations caused by the percolation of gas through the bed affect the catalyst particles as well, which results in a non-negligible retention of catalyst by the polymer product. The major concern of the process of document (3) is thus to limit the amount of the catalyst residues in the polymer.

Consequently, it seems more consistent and systematic to start from a document dealing like the patent in suit with a gas/liquid phase polymerisation process, such as document (1), and to follow the line of argumentation adopted by the Respondent, which also took into consideration the teaching of the late-filed document (11). For this reason, the Board has decided to admit the latter citation into the procedure (Article 114(1) EPC).

3. The patent in suit concerns a process for producing polyolefin in a gas/liquid phase reactor. Such a process

is described in document (1), which the Board for the above-mentioned reasons regards as the closest state of the art. More specifically, this citation discloses an automatically controlled continuous process for manufacturing ethylene copolymers, wherein ethylene and at least one other α-olefin are copolymerised in the presence of a catalyst in a hydrocarbon solvent; this reaction is carried out in a gas/liquid phase reactor with a control system for various process parameters, whereby reactor fouling can be avoided (column 1, lines 5 to 61; column 3, lines 15 to 26). The catalyst is a conventional coordination catalyst system, in particular a system comprising vanadium tris(acetylacetonate) and diisobutyl aluminium monochloride, the ratio vanadium to aluminium being generally within the range 1:2 to 1:20 (column 3, lines 43 to 66). The most important parameter to be controlled is the partial pressure of ethylene, since, on the one hand, the latter is known to change much faster than the partial pressure of the other monomer(s), and, on the other hand, an unbalanced ethylene: comonomer(s) ratio in the liquid phase causes the formation of ethylene rich insoluble copolymers which can foul the reactor (column 1, lines 15 to 37; column 2, lines 32 to 37). The partial pressure of ethylene in the gas phase can be determined from one single pressure signal corresponding to the pressure of gas phase in the reactor, which is the total of the partial pressures of the various monomers (column 4, lines 26 to 31). In practice, the control is achieved by a dual system which comprises a primary control system that maintains the reactor conditions within a predetermined routine operating range, and a secondary control system that controls the amount of ethylene being fed to the reactor (column 1, line 65 to column 2, line 24; Example). Although this method insures an homogeneous composition of the polyolefins, whereby the formation of insoluble products can be avoided, it suffers

from two major shortcomings. The first is that specific product parameters, like melt index and density, cannot be controlled; the second is that the control system is rather complicated in that it operates in two steps.

In the light of these shortcomings the technical problem underlying the patent in suit can thus be seen to be in the definition of an improved gas/liquid phase polymerisation process, wherein melt index and density of the polyolefins can be constantly adjusted according to predetermined values in an effective manner.

According to each of the three independent process claims this problem is to be solved by (i) feeding the reactor with a stream of olefin(s) and hydrogen (cf. feature a) in the claims), (ii) analysing the concentration thereof in the gas phase within the reactor by gas chromatography (cf. features b) and c) in the claims), (iii) comparing these figures with preset values corresponding to predetermined properties (cf. features d) and e) in the claims), and (iv) adjusting the feed rates of olefin(s) and hydrogen according to these deviations (cf. feature f) in the claims).

In view of the experimental results in the patent in suit, in particular of Figures 2 and 6 which show that polyolefins having uniform melt index and uniform density can be produced quite steadily by the claimed processes, the Board is satisfied that the above combination of features (i) to (iv) provides an effective solution to the above-defined technical problem.

4. After examination of the documents relied upon by the Respondent, the Board has come to the conclusion that this technical teaching is not disclosed in any of them and that the subject-matter of the patent in suit as defined

in any of Claims 1, 6 and 11 is, therefore, novel. Since the issue of novelty has not been disputed, it is not necessary to consider this matter in detail.

5. It still remains to be decided whether that subject-matter involves an inventive step having regard to the teaching of the documents relied upon by the Respondent.

5.1 Since the main controversial issue during oral proceedings concerned the interaction between gas phase and liquid phase in a gas/liquid phase polymerisation process, i.e. the question whether at the priority date of the patent in suit it was known in such a process to control the properties of a polyolefin via the results of gas phase analysis, it seems appropriate as a preliminary remark to observe that document (1) provides a positive answer.

As stated above, one of the advantages of the process described therein is the fact that the pressure of the gas phase can be used to adjust the amount of the main monomer in the reactor to avoid reactor fouling, which is nothing else than the control of the composition of the copolymer. This citation teaches thus that a specific property of a polymer can be controlled by analysis of the gas phase in the course of a gas/liquid phase polymerisation reaction, thus in a dynamic state. It follows that the general concept on which the patent in suit is based was already known well before the priority date of the patent in suit.

This conclusion makes superfluous further theoretical considerations about the validity of Henry's Law in the present case, in particular about the differences between the steady state, as it exists after introduction of the monomer(s) and the catalyst system into the reactor, but before the beginning of the polymerisation reaction, and

the dynamic state during this reaction, wherein the monomer(s) is(are) continuously transferred from the gas phase to the slurry phase. As argued by the Respondent, in view of the teaching of document (1), this difference is artificial in the present case, for in a continuous process the various parameters of the process are kept as constant as possible, which involves, first, that the amount of monomer(s) polymerised in the liquid phase is compensated by an equal amount of monomer(s) transferred from the gas phase, and, secondly, that the composition in the gas phase is constantly adjusted by a corresponding feed of gas monomer(s); this means that the dynamic state should be properly regarded as an equilibrium state.

It follows that the argument of a prejudice based on the thermodynamic principles of Henry's Law alleged by the Appellant cannot be accepted, and that, consequently, documents (3) and (11) will be interpreted in the light of the above conclusion.

5.2 These two documents provide evidence that the specific features (i) to (iv) referred to in the penultimate paragraph of point 3 above are in fact well known in the art.

5.2.1 This applies in the first place to gas chromatography (feature (ii)), which is used in document (11) to analyse the gas phase in a gas/liquid phase polymerisation process.

This citation can be regarded as a study of the relative reactivity of ethylene and propylene in the presence of various catalyst systems; from the embodiments illustrated in the experimental section on page 2654 it appears that this study was carried out in the framework of batch processes. However, as correctly pointed out by the

Respondent, the critical teaching of document (11) is the correlation which exists between the partial pressure of the monomers in the gas phase and their concentration in the liquid phase (page 2655, Figure 2 and column 2, paragraph 2, lines 1 to 14) and, thereby, the correlation between the monomer composition in the gas phase determined by gas chromatography and the polymer composition (page 2656, Table I), which in turn is responsible for the specific properties of that polymer. As further argued by the Respondent, the skilled person would not restrict this correlation between the monomer composition in the gas phase and the properties of the resulting polymer to batch processes, but would self-evidently regard it as valid in continuous processes as well.

There was thus a strong incentive for the skilled person, faced with the above-defined problem, to use gas chromatography as a method of analysis of the gas phase and, thereby, arrive at feature (ii) without inventive contribution.

5.2.2 This applies as well to the use of hydrogen (feature (i)), the control of the operating conditions (feature (iii)) and the subsequent adjustment of the gas streams (feature (iv)), which are all mentioned in document (3).

In the process described therein, at least one olefin, in particular ethylene, is polymerised in the presence of hydrogen and a silyl chromate/ethoxydiethylaluminium catalyst system in a fluid bed reactor; the gas phase above the bed is subjected to a continuous control of its composition by means of a gas analyser, whereby the process can be operated on a continuous basis according to predetermined conditions and polyolefins of predetermined

properties can be prepared (Claim 1 in conjunction with column 2, lines 28 to 31 and 60 to 63; column 3, lines 21 to 39; column 5, lines 19 to 31 and Example 1). In particular, it has been found that the melt index of polyolefins increases with the hydrogen concentration in the gas stream, thus together with the ratio hydrogen: monomer(s) in the gas phase (column 3, lines 16 to 20; column 6, lines 11 to 24; column 10, lines 23/24; Example 16, Table IV) and, further, that the use of propylene as comonomer together with ethylene lowers the density of the polyolefin (Tables I and II, Examples 2 to 8).

Although these features have been disclosed in the framework of a gas phase polymerisation process, in view of the above-noted correlation, they would be considered by the skilled person for the solution of the technical problem underlying the patent in suit. The teaching of document (3) provides thus an incentive for the skilled person to operate along the same lines, i.e. to use hydrogen to increase the melt index and a comonomer to lower the density (feature (i)), to subject the gas phase to a continuous control by means of a gas analyser (feature (iii)) and to adjust the composition of the gas by signals sent from the gas analyser (feature (iv)).

5.2.3 The combination of features in the three independent process claims, i.e. the combination of feature (ii), on the one hand, with features (i), (iii) and (iv), on the other hand, does not provide other advantages than those directly derivable from the teaching of documents (3) and (11). In the absence of such a surprising effect, the technical problem underlying the patent in suit cannot be defined in more ambitious terms, so that the solution as defined in these three claims must be regarded as obvious.

5.3 For this reason the subject-matter of the three independent process Claims 1, 6 and 11 does not involve an inventive step.

6. In the absence of a separate request directed to the specific features mentioned in the dependent Claims 2 to 5, 7 to 10 and 12 to 15, the latter must fall with the respective independent claims, since a request can only be considered as a whole. Besides, no argument in favour of the inventiveness of any of these features has been provided by the Appellant.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar: **The Chairman:**

E. Görgmaier F. Antony