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D E C I S I O N
of 6 February 1997

Case Number: T 0954/93 - 3.3.3

Application Number: 83106661.8

Publication Number: 0100879

IPC: C08F 210/16

Language of the proceedings: EN

Title of invention:

Ethylene-hexene-1 copolymer and process to produce ethylene-monoolefin copolymers

Patentee:

PHILLIPS PETROLEUM COMPANY

Opponent:

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Union Carbide Chemicals and Plastics Company Inc.
SOLVAY & Cie, S.A., Bruxelles

Headword:

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Relevant legal provisions:

EPC Art. 54(2), 56

Keyword:

"Novelty (yes) - onus of proof not discharged" (Reasons 7.6)
"Inventive step (yes) - no recognition of technical problem in the prior art" (Reasons 8.3 to 8.5)
"High cost of experimentation to establish case irrelevant to burden of proof" (Reasons 7.6)
"Non appearance without notice of party reprehensible" (Reasons 2)

Decisions cited:

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Catchword:

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Case Number: T 0954/93 - 3.3.3

. D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 6 February 1997

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Decision under appeal: Interlocutory decision of the Opposition Division
of the European Patent Office posted 2 September
1993 concerning maintenance of European patent
No. 0 100 879 in amended form.

Composition of the Board:

Chairman: C. Gérardin
Members: R. Young
J. A. Stephens-Ofner

Summary of Facts and Submissions

- I. The mention of the grant of European patent No. 0 100 879, in respect of European patent application No. 83 106 661.8, filed on 7 July 1983 and claiming a US priority of 9 July 1982 (US 396 930) was announced on 20 May 1987 (Bulletin 87/21).
- II. Five Notices of Opposition were filed, respectively on 15 February 1988, by Norsolor (Opponent 01), on 12 February 1988, by Stamicarbon B. V. (Opponent 02), on 19 February 1988 by BASF (Opponent 03), also on 19 February 1988 by Union Carbide Corporation (Opponent 04), and, finally on 22 February 1988 by Solvay & Cie (Opponent 05). The grounds of Article 100(a) EPC were relied upon in each case, with Article 100(b) EPC additionally being relied upon in the case of Opponent 01. The oppositions were supported inter alia by the documents:
- D1: Finogienova et al., (considered in the form of its French translation): "Détermination de l'activité relative de l'éthylène et du hexène-1 dans la copolymérisation et répartition des chaînons dans les copolymères obtenus", Vysokomol. Soedin. Ser. B (1978), volume 20(6), pages 459 to 461;
- D5: US-A-4 011 382;
- D12: J. C. Randall, "Polymer Characterization by ESR and NMR", ACS Symposium Series 142, Edited by Woodward and Bovey, Am. Chem. Soc., Washington D.C., 1980, Chapter 6; and
- D13: US-A-3 887 494.

III. By a decision issued in writing on 2 September 1993 following oral proceedings held on 22 December 1992, the Opposition Division found that the patent in suit could be maintained in amended form, on the basis of a set of Claims 1 to 8 filed on 20 December 1991, independent Claims 1 and 3 of which read, respectively, as follows:

Claim 1:

"An ethylene-hexene-1 copolymer with a content of 1 - 6 mole % 1-hexene having a relative comonomer dispersity of 99% or higher, said copolymer having a maximum relative comonomer dispersity which is the sum of 100% plus the concentration of 1-hexene in mole percent."

Claim 3:

"A process to produce ethylene-monoolefin copolymers by contacting a gas mixture containing ethylene and another monoolefin with a silica/titania/chromium oxide catalyst, said silica and titania being coprecipitated, said catalyst having been activated by a method consisting of contacting the catalyst with free oxygen at a temperature in the range of from 177 to 1093°C, under gas phase polymerization conditions in a polymerization zone such as to produce an ethylene-monoolefin-copolymer, and recovering said ethylene-monoolefin-copolymer, characterized by one of the following four procedures:

- (a) contacting ethylene and 1-butene with said catalyst, wherein said 1-butene is used in a concentration in mole percent in the gas phase based on total olefins in the gas phase in the polymerization zone as 100 mole % which is defined by the following formula

$$CGB = \frac{CPB}{K4}$$

wherein CGB is the concentration of 1-butene in mole percent in the gas phase, CPB is the concentration of 1-butene in the copolymer and K4 is a factor in the range of 0.6 to 1.2,

- (b) contacting ethylene and 1-hexene with said catalyst, wherein said 1-hexene is used in a concentration in mole percent in the gas phase based on total olefins in the gas phase in the polymerization zone as 100 mole % which is defined by the following formula

$$CGH = \frac{CPH}{K6}$$

wherein CGH is the concentration of 1-hexene in mole percent in the gas phase, CPH is the concentration of 1-hexene in the copolymer and K6 is a factor in the range of 1.4 to 2.5,

- (c) contacting ethylene and 1-octene with said catalyst, wherein 1-octene is used in a concentration in mole percent in the gas phase based on total olefins in the gas phase in the polymerization zone as 100 mole % which is defined by the following formula

$$CGO = \frac{CPO}{K8}$$

wherein CGO is the concentration of 1-octene in mole percent in the gas phase, CPO is the concentration of 1-octene in the copolymer and K8 is a factor in the range of 4 to 7,

- (d) contacting ethylene and 4-methyl-1-pentene with said catalyst, wherein said 4-methyl-1-pentene is used in a concentration in mole percent in the gas phase based on total olefins in the gas phase in the polymerization zone as 100 mole % which is defined by the following formula

$$CGF = \frac{CPF}{K51}$$

wherein CGF is the concentration of 4-methyl-1-pentene in mole percent in the gas phase, CPF is the concentration of 4-methyl-1-pentene in the copolymer and K51 is a factor in the range of 1 to 1.4."

Claim 2 was directed to an elaboration of the product according to Claim 1, and Claims 4 to 8 were directed to further elaborations of the process according to Claim 3.

According to the decision, the claims were admissible in view of Articles 123(2) and (3) EPC and no objections under Articles 100(b) and (c) had been maintained.

As to novelty, none of the citations disclosed ethylene-hexene-1 copolymers having the properties claimed in Claims 1 and 2, nor a process for the production of ethylene-monoolefin copolymers of the features set out in Claim 3. In respect of citation D1, neither computer calculations nor calculations based on mere assumptions could be considered sufficiently substantiated to question novelty. Likewise, statements based on alleged inherent disclosure could not serve to deprive the claimed subject-matter of novelty.

As regards inventive step, none of the documents cited mentioned the problem solved by the patent in suit, namely the "super-random" distribution or comonomer dispersity. Consequently, even if the various disclosures were combinable with each other, for instance on the basis of belonging to the same narrow field of ethylene polymerisation, they could not serve to question the existence of an inventive step.

IV. Two Notices of Appeal were filed against the above decision filed, respectively on 4 November 1993 by Opponent 03 (Appellant 01), and on 5 November 1993 by Opponent 04 (Appellant 02), in each case together with payment of the prescribed fee.

The main arguments put forward by the Appellants in their Statements of Grounds of Appeal, filed on 27 December 1993 and 30 December 1993 respectively, may conveniently be divided into groups relating to the novelty (I) and inventive step (II) of the product claims (Claims 1 and 2) and to the novelty (III) and inventive step (IV) of the process claims (Claims 3 to 8). The arguments may be summarised as follows:

(I) Novelty of the product (Claims 1 and 2)

- (i) The catalyst used according to D1 corresponded to that used in Examples 3 and 4 of the patent in suit. Furthermore, the statement in D1 that "les hexène s'intègre dans le chaîne uniquement sous forme des chainous isolés" showed that the copolymer formed contained the hexene molecules in strictly isolated locations. This was an implicit disclosure, which was novelty destroying. The decision under appeal had been unjustified in dismissing this clear statement on the basis that it originated from "computer calculations", particularly since the same decision recognised novelty in respect of another parameter, which was itself based on a computer calculation. This violated the principle of neutrality enshrined in the EPC.
- (ii) The characterisation of the copolymers according to Claim 1 in terms of the relative monomer dispersity (RMD), which was an unusual parameter, gave rise to problems in any case, because the nuclear magnetic resonance (n.m.r.) analysis necessary for its determination involved a large number of variables and, even with a given spectrum, there were different ways of calculating the relevant parameter. Furthermore, n.m.r. methods were known to have a substantial error margin.

Consequently, the parameter RMD was unsuitable as a means of drawing a line between clearly not-novel ethylene-hexene-1 copolymers and those defined in Claim 1. In particular it would be difficult to demonstrate that a particular copolymer of the prior art fulfilled the requirements of Claim 1, since the relevant

parameter was not mentioned in prior art documents, and retrospective determinations of RMD values for the disclosed copolymers could be criticised on the basis that the skilled person would have carried out the determination of AMD differently.

It was not clear from the decision under appeal why novelty had been recognised on the basis of a parameter presenting problems of this kind.

- (iii) Document D12 had not been sufficiently considered. Although principally concerned with the n.m.r. investigation of polymers, its content went beyond this, on page 97, second paragraph, which stated that six ethylene/alpha olefin copolymers had been investigated, in which the concentration of alpha olefin was below 3%, which had as a consequence that "only isolated branches are produced". Thus D12 described a copolymer according to Claims 1 to 3.

- (iv) Finally, it was evident from the examples and comparative examples in the patent in suit that copolymerisation of ethylene with small quantities of hexene always led to products with a RMD of greater than or equal to 100%. Reference to Table IIA of the patent in suit had been made in this connection by the co-Opponent 04 (Appellant 02) during the opposition proceedings in their submission, filed on 19 February 1988. Furthermore, calculations showing this had been filed during the opposition proceedings by Opponent 03 (Appellant 01) in their submission, filed on 23 January 1991.

(II) Inventive step of the product (Claims 1 and 2)

Whilst it was not necessary to go into this in view of the lack of novelty argued above, it was not clear why the claimed copolymers, even if novel, involved an inventive step.

(III) Novelty of the process (Claims 3 to 8)

No specific arguments on this issue were presented in the Statements of Grounds of Appeal.

(IV) Inventive step of the process (Claims 3 to 8)

- (i) The acceptance, in the decision under appeal, of an inventive step in respect of the solution of the problem of high efficiency comonomer incorporation into the chain by the so-called "super-random" distribution of the monomers was not tenable, since this problem was only solved for ethylene/1-hexene copolymers, and therefore not justified, at least for the preparation of ethylene copolymers with comonomers other than 1-hexene.
- (ii) There was in any case no reduction in the amount of comonomer necessary for obtaining a polymer of a particular comonomer content.
- (iii) The K-values mentioned in Claim 3 were a reflection of the well known tendency of a comonomer, with decreasing vapour pressure, to become more soluble in the polymer already formed, and thus for a comonomer with higher molecular weight to be incorporated in the polymer to a greater extent.

(iv) Document D5 described a process for polymerisation of ethylene with alpha olefins in the gas phase, the only difference from the patent in suit being in the catalyst used. The catalyst used in the patent in suit was, however, described in D13, in which it was particularly recommended for the preparation of copolymers from ethylene and 1-olefins, amongst others 1-hexene. Since, furthermore, it was not disputed that D5 and D13 belonged to the same technical field, it was obvious for the skilled person to combine both citations and arrive at the claimed process.

V. The Respondent argued, in a submission filed on 17 January 1997, in essence as follows:

(I) Novelty of the product claims.

(i) The statements relied upon were not of a character to destroy the novelty of the claimed products, since they were based on a variety of assumptions, at least some of which were incorrect, or else were simply wrong. Clustering had always been a problem in this field. Consequently, there was no inherent disclosure of the claimed subject-matter.

(ii) The argument that n.m.r. measurements were difficult or inaccurate was incorrect and legally irrelevant. The patent in suit disclosed in detail how the comonomer dispersity was measured. There was thus no lack of clarity or the like.

(II) As regards inventive step in the product claims, no arguments had yet been adduced to question this. It was, however, quite surprising that ethylene-1-hexene copolymers of significant hexene content (1 to 6 mole %) existed which had extremely low clustering. A RMD of over 99% and even over 100% was totally unexpected, and indeed unheard of prior to the invention.

(III) Novelty of the process claims was uncontested.

(IV) Concerning the inventive step of the process claims, although it had been alleged that the claimed process would be obvious from a combination of D5 and D13, the use of the problem and solution approach led to the opposite result.

VI. Oral proceedings were held before the Board on 6 February 1997. The start of the proceedings was delayed by the non-appearance of Appellant 02 (originally Opponent 04), who, although having been duly summoned, had given no prior notice of an intention not to attend. Enquiries instigated by the Board were answered by a telephone call during the proceedings itself, in which the Chairman was informed that the party concerned had no further interest in the case. The proceedings were thus attended by Appellant 01 (originally Opponent 03), the Respondent (Patentee) and also a non-appealing party to the proceedings as of right (originally Opponent 05).

VII. The Appellants each requested that the decision under appeal be set aside and the patent be revoked in its entirety, as did the party to the proceedings as of right.

The Respondent requested that the appeals be dismissed.

Reasons for the Decision

1. The appeals are admissible.
2. *Procedural matters*

As stated in the Summary of Facts and Submissions, above, Appellant 02 failed to attend the hearing, contenting himself with informing the Board (at some late stage in the hearing) that he had lost interest in the proceedings, implying this to be a sufficient and adequate reason for failing to appear without due notice.

It is not uncommon for parties not to appear at hearings; frequently too, they lose interest for a variety of reasons. In such cases they routinely inform all concerned, in advance, that they will not be present. This is no more than the usual degree of courtesy owed to the other parties and the respect due to the Board as a Court of final appellate jurisdiction would seem to call for: a call clearly either unheeded or deliberately flouted by Appellant 02. The Board wishes to put on record its view that such conduct is reprehensible in the extreme and trusts that it will not be repeated.

3. *Amendments*

No objections were raised by the parties under Article 123(2) or (3) EPC to the text of the patent in suit on which the decision under appeal was based, and which underlies the present decision. The Board sees no objection either to this version.

4. *Sufficiency*

This ground of opposition was not specifically pursued in the written or oral proceedings. The Board sees no reason to question the sufficiency of the disclosure of the patent in suit.

A. The product (Claims 1 and 2).

5. *Clarity of Claim 1*

The copolymers according to Claim 1 of the patent in suit are characterised by a parameter, the "relative monomer dispersity", RMD, defined in the patent in suit as the ratio of the absolute comonomer dispersity (AMD) to the perfectly random comonomer dispersity or Bernouillian distribution (BMD).

The RMD is in effect a measure of the actual extent of clustering of comonomer units per average molecule of the polymer, compared with the level of clustering per average polymer chain calculated from statistical (Bernouillian) theory.

The objection that the parameter RMD is "unusual" is not in itself a ground of objection under the EPC. Nor is such an objection logically sustainable, in the Board's view, because the RMD is directly related to a fundamental physical characteristic of a copolymer, namely the distribution of comonomer units in the copolymer chain.

The objection that uncertainty would arise in the determination of whether, in practice, a particular polymer fulfilled or not the claimed requirements (section IV.(I)(ii), above), to the extent that it relates exclusively to polymers which may be prepared in the future, is purely a question of infringement,

with which the Board has no competence to deal. Insofar as it relates to the polymers of the prior art, it bears on the issue of novelty rather than clarity, and is consequently dealt with in under that section.

In neither case does any intrinsic lack of clarity arise. On the contrary, the parameter RMD defines the relevant measures of monomer distribution without any element of contradiction and in an understandable way.

Consequently, the subject-matter of Claim 1 is held to meet the requirements of Article 84 EPC in relation to clarity.

6. *The technical problem*

6.1 The patent in suit relates, in its product aspect, to the provision of an ethylene-1-hexene copolymer having from 1 to 6 mole % 1-hexene, and having inter alia a desirable balance of physical properties, especially an appropriately low density range. Such copolymers are known, for instance from D5, which is considered by the Board to represent the closest state of the art.

6.2 According to D5, there is provided a process for producing, at relatively high productivities, solid ethylene polymers having a density of <0.941 and a melt index of >0.0 to at least about 2.0, a relatively low content of n-hexane extractables and a low residual catalyst content, by copolymerising ethylene with sufficient quantities of C_3 to C_6 alpha-olefin monomer to provide the desired density in the polymer product. The process is carried out in a fluid bed at a temperature of about 30° to 105°C , under a pressure of less than 1 000 psi, by contacting the monomers with fluidized particles of an activated, supported

catalyst comprising, based on the total weight of the support and the catalyst, 0.05 to 3.0 wt % chromium, 1.5 to 9.0 wt% titanium, and >0.0 to 2.5 wt% of fluorine, the chromium and titanium being in the form of oxides after the activation (Claim 1; column 2, lines 56 to 66).

The copolymers contain a major mole percent (≥ 85 mol%) of ethylene and a minor mol% of one or more C_3 to C_6 alpha olefins, preferably propylene, butene-1, pentene-1 and hexene-1 (Claims 5 to 7 and column 3, lines 25 to 30).

The polymers have a density of about 0.900 to 0.925 for low density polymers and of about 0.926 to 0.940 for medium density polymers. The addition of progressively larger amounts of the comonomers to the polymers results in a progressive lowering, in approximately a linear fashion, of the density of the polymer. In order to achieve the desired density ranges, it is necessary to copolymerise enough of the $\geq C_3$ comonomers with ethylene to achieve a level of 1.0 to 15 mole % of the C_3 to C_6 comonomers in the copolymer. To achieve the same result, in terms of a given density, at a given melt index level, larger molar amounts of the comonomers would be needed in the order of $C_3 > C_4 > C_5 > C_6$. The amount of comonomer needed will depend on the particular comonomer(s) being employed and on the fluoride content of the catalyst (column 3, lines 30 to 50; column 6, line 56 to column 7, line 3).

There is furthermore provided a listing of the amounts, in mols of the various comonomers that must be copolymerised with ethylene in order to provide polymers having the desired density range at any given melt index. The listing also indicates the concentration, in mol%, of such comonomers which must be present in the gas stream of monomers which is fed to the reactor (column 7, lines 4 to 19).

Comonomer	mol % needed in copolymer	mol % needed in gas stream
propylene	3.0 to 15	6 to 30
butene-1	2.5 to 12	6 to 25
pentene-1	2.0 to 9.0	4 to 18
hexene-1	1.0 to 7.5	3 to 15

According to the examples, butene is copolymerised with ethylene using various Cr, Ti and F containing catalysts.

Thus it is clear from D5, that the density of the copolymer is directly related to the amount of comonomer incorporated in the ethylene polymer. In the case of hexene, however, the comonomer is considerably more expensive than ethylene.

6.3 Compared with this state of the art, the technical problem underlying the patent in suit may be seen as the search for a more cost effective way of achieving the advantages associated with low density in such a polymer, without sacrificing the desirable balance of relevant physical properties.

6.4 The solution proposed according to Claim 1 of the patent in suit is to provide an ethylene-1-hexene copolymer wherein the relationship of internal structure to the external physical properties has been shifted by restricting the extent of clustering of 1-hexene comonomer in the copolymer chain, resulting in an enhanced distribution of comonomer units along the copolymer chain, expressed in terms of a value of the RMD, of 99% or more.

At the levels claimed, it is possible to achieve a maximum of the desired chain interruptions caused by 1-hexene comonomer units, which are responsible for the desired density reduction, with a minimum consumption of the expensive 1-hexene comonomer.

6.5 It is evident, from the information given in the patent in suit itself, in particular in Table IIIA, that ethylene-1-hexene copolymers containing 1 to 6 mol% 1-hexene have been provided having a RMD within the claimed range, and indeed in excess of 100%, the latter values corresponding to a better distribution of comonomer units along the chain than the maximum predicted according to Bernouillian theory. Such polymers are termed "super-random" copolymers in the patent in suit. It can furthermore be seen that the copolymers have a useful range of melt index (Table IIIA, runs 1 and 2).

Consequently, it is credible that the technical problem has been effectively solved by the ethylene-1-hexene copolymers as claimed in the patent in suit.

7. *Novelty*

7.1 There is no reference in D5 to the RMD or to any comparable parameter of the polymers disclosed therein. Consequently, there is no explicit disclosure of the claimed polymers in D5.

7.1.1 As regards implicit disclosure, although there are general references in D5 to the production of ethylene-1-hexene copolymers, only the preparation of ethylene-1-butene copolymers is exemplified. Consequently, there is no disclosure of any ethylene-1-hexene copolymer in a form sufficiently characterised to permit, even in principle, the measurement of a value of RMD.

7.1.2 The argument, put forward by Appellant 01 at the oral proceedings, on the basis of the similarity of the various catalysts used according to Example II of the patent in suit to those disclosed in D5, is irrelevant in view of the absence of concrete disclosure of any particular ethylene-1-hexene copolymer in D5.

7.1.3 Hence, there is no implicit disclosure in D5 of the subject-matter of Claim 1 of the patent in suit.

7.2 According to D13, there is disclosed a method of preparing a silica-containing composition catalytically active for olefin polymerisation, comprising adding a titanium compound to a mineral acid; introducing an alkali metal silicate into the acid containing the titanium compound to form a hydrogel; aging and washing the latter to remove alkali metal; forming a xerogel from the hydrogel, and heating the xerogel also containing chromium to produce a catalytically active composition (Claim 1). The resulting catalyst is employed in any of the techniques which are employed when utilising catalysts

comprising chromium on a silica substrate (column 3, lines 58 to 63). In Example IV, the catalysts are used to copolymerise ethylene and butene-1, yielding a copolymer having a density of 0.927 and melt index of 5.4 (run III).

Whilst it is true that D13, which is referred to in the description of the patent in suit (page 2, last line), discloses cogel catalysts suitable for preparing the "super-random" ethylene-1-hexene copolymers according to Claim 1 of the patent in suit, there is only a general statement about the suitability of such catalysts to produce copolymers of ethylene with another 1-olefin, such as butene and hexene (column 2, lines 6 to 9), but no example of their use in a process in which ethylene-1-hexene copolymers are prepared, the only exemplification being of ethylene-butene-1, and this by a slurry process as opposed to the gas phase process provided by the patent in suit (Example IV).

Consequently, there is no disclosure of a ethylene-1-hexene polymer in a manner sufficiently characterised to yield, even in principle, a particular RMD value. The subject-matter of Claim 1 is thus novel in the light of the disclosure of D13.

7.3 According to D1, there is described the determination of the relative activities of ethylene and 1-hexene in copolymerisation, based on a kinetic method. According to this method, a differential equation derived from the copolymer composition is converted to a simple integral equation which is valid for high levels of monomer conversion during the reaction (page 1, third paragraph).

The ethylene and 1-hexene are copolymerised in solution in n-hexane at 140°C, using a chromic acid catalyst supported on a highly porous aluminosilicate, in a one litre stainless steel autoclave, without further addition of monomer. The monomer conversions are measured in terms of the lowering of their concentration during the copolymerisation, on samples extracted from the gas phase of the autoclave. The monomer content of the liquid phase is determined using phase equilibrium constants found in the literature for hydrocarbons at the temperatures of the system. The analyses were carried out by chromatography (page 2).

The values of the relative reactivities obtained could be used for evaluating the structure of the copolymer obtained, in terms of the distribution and alternation of the monomers in the copolymer chain, using methods of calculation according to which the structure of copolymers during synthesis depends principally on the product of the relative reactivities of the monomers participating. This indicates the formation of a statistical copolymer, in which a correct alternation of the units is impossible, and their position of incorporation in the chain has an uncertain character (paragraph bridging pages 3 and 4).

With an increase in the content of ethylene in the liquid phase, calculations show that the proportion of the sections containing the greatest number of ethylene units increases. The 1-hexene is integrated into the chain solely in the form of isolated units because the proportion of sections containing a 1-hexene unit varies within the limits of 0.999 - 0.995 for the different molar quantities of ethylene and 1-hexene (page 4).

The disclosure is not considered to be novelty destroying for the subject-matter of Claim 1, for the following reasons.

7.3.1 Firstly, there is no explicit reference to RMD. In this connection, the crucial reference to isolated units ("chaînes isolées") is unspecific as to what is meant by "units" ("chaînes"). If the reference is to the side chains, rather than the 1-hexene comonomers, then a "head-to-tail" incorporation of 1-hexene comonomer would result in side chains which were "isolated" compared with the situation of "head-to-head" incorporation, even if the comonomer molecules themselves were in a cluster. Furthermore, the "sections" are not further defined, in particular as to their relative length or the location of the 1-hexene units. Consequently, it is not clear what, if anything, the statement implies for the extent of isolation, in the sense of the RMD, of the comonomer units referred to.

Secondly, the statement is made in the context of the preceding sentence, which is evidently a description of what happens at a particularly advanced stage of the copolymerisation reaction. Thus, it is not even clear whether the statement itself relates to the resulting copolymers or simply to a particular stage of the copolymerisation reaction.

In view of these ambiguities, the Board finds that the statement is insufficiently precise to amount to an implicit disclosure of a particular RMD.

7.3.2 Quite apart from the above, the entire statement of results in D1 is derived from calculations based on certain parameters of the copolymerisation reaction, rather than any direct investigation of the polymer product itself. These calculations are in turn based on a

sequence of assumptions, in particular that the monomer activities in the samples extracted from the gas phase can be accurately related to the corresponding values in the liquid phase, and that the structure of the polymer formed can be predicted with the requisite degree of precision from these calculated values.

The validity of these assumptions has been challenged by the Respondent, in particular on the basis that the solubility of a participating monomer (of which activity is a function), although a factor for monomer distribution properties in the liquid phase, is not such a factor in the gas phase (submission filed on 17 January 1997, pages 2 and 3). No refutation of this argument or defence of the assumptions has, however, been forthcoming from the opposing parties.

In the Board's view, a calculation of the kind presented in D1 is no more reliable than the assumptions on which it is based. The assumptions in question have, however, in the light of the above, been convincingly shown to be, at the very least, unsafe. The conclusions drawn from such calculations disclosure are therefore not of a nature to reflect, in a reliable way, the nature of the reality behind the disclosure. Such matter cannot, in the Board's view, be taken into account for the purposes of Article 54(2) EPC.

Thus, the Board concurs with the finding of the decision under appeal in relation to the "calculations" in the disclosure of D1 (Section III., above).

- 7.3.3 In summary, D1 cannot be regarded as making, directly and unambiguously, a polymer of the RMD structure claimed in Claim 1 of the patent in suit available to the public in the sense of Article 54(2) EPC.
- 7.3.4 Consequently, the argument of implicit disclosure on the basis of a similarity between the catalyst referred to in D1 and those used according to the patent in suit (section IV.(I)(i), above) is irrelevant.
- 7.3.5 In other words, the subject-matter of Claim 1 is new in the light of D1.
- 7.4 According to D12 there is disclosed a method of characterising long chain branching in polyethylenes using a high field carbon-13 n.m.r. technique. In particular, according to page 97, paragraph 2, the C-13 n.m.r. spectra from a homologous series of six linear ethylene 1-olefin copolymers beginning with 1-propene and ending with 1-octene were investigated and reproduced in Figures 2 and 3. Also, the respective 1-olefin concentrations are less than 3%; thus, "only isolated branches are produced".
- 7.4.1 There is no explicit mention in D12 of RMD. The above statement concerning isolated branches, relied upon to demonstrate that an RMD in the claimed range was known from the prior art, was explained at the oral proceedings by the Respondent, who was accompanied by a co-worker of the author of D12 (also named as one of the inventors of the patent in suit), as having meant that the units were sufficiently isolated to enable a clear assignment of the peaks of interest in the spectra presented in D12. These did not particularly concern clustering, but rather the identification of branch lengths. It thus merely reflected a step in the

"learning process", which had not, at the date of writing of D12 (1979), advanced to the point of understanding the nature of comonomer cluster distribution.

- 7.4.2 Even if this explanation, which is convincing to the Board, had not been forthcoming, the statement referred to suffers from a number of the deficiencies already found in the similarly worded passage in D1 (cf. section 7.3 etc., above).
- 7.4.2.1 Firstly, it is a blanket statement made in respect of a number of copolymers and not directly in connection with a particular ethylene-1-hexene copolymer.
- 7.4.2.2 Secondly, even if it were interpreted as applying specifically to the ethylene-1-hexene copolymers, it is essentially qualitative in nature, in that it does not identify the extent to which the comonomer units are "isolated". It does not, therefore, amount to a disclosure of a particular RMD.
- 7.4.2.3 Finally, the statement does not arise from or reflect any objective investigation of the nature of the copolymer under consideration. On the contrary, it is in the nature of an assumption, since it is presented as the logical outcome of the low percentage of comonomer present. Consequently, and as in the case of D1, the statement cannot be regarded as reliable, or, therefore, as making a copolymer of the claimed structure directly and unambiguously available to the public, in the sense of Article 54(2) EPC (cf. Section 7.3.3, above).

7.4.3 The argument of Appellant 01 at the oral proceedings, that the statement was to be understood as a statement of fact corroborated by the n.m.r. spectrum shown in Figure 8 of D12, which showed no peaks indicative of comonomer clusters, is not convincing, because the statement on page 97 does not refer to Figure 8, but only to Figures 2 and 3.

7.4.4 There is in any case no disclosure of the preparation of the relevant copolymers in D12, let alone a concrete example which would have allowed the RMD to be concretely ascertained. This means that D12 cannot be held to disclose a ethylene-1-hexene copolymer of any particular RMD, let alone an RMD within the range claimed in Claim 1 of the patent in suit.

7.4.5 Consequently, the subject-matter of Claim 1 is novel in the light of the disclosure of D12.

7.5 The general argument of Appellant 01 at the oral proceedings, that clearly all ethylene-1-hexene copolymers of low 1-hexene content fulfilled the RMD requirement set out in Claim 1 of the patent in suit, was supported only by reference to the results shown by the copolymers according to Example II in Table IIA of the patent in suit (section IV.(I)(iv), above). These copolymers, although admittedly not made by a process as claimed in the patent in suit, have never been shown to belong to the state of the art in the sense of Article 54(2) EPC. It was, however, the onus of the Appellants to do this.

Consequently, the disclosure of Table IIA of the patent in suit does not support a finding of lack of novelty of the copolymers claimed in Claim 1 of the patent in suit.

7.6

To the extent that the argument goes beyond the basis referred to, and embraces all ethylene-1-hexene copolymers of low 1-hexene content, however prepared, it is no more than a speculative allegation devoid of any documentary or evidentiary support. In particular, there is a complete absence, in the submissions of the Appellants, of any attempt to establish, by comparative tests, the RMD values of ethylene-1-hexene polymers known to the state of the art.

Whilst the Board takes note of the argument of Appellant 01 during the oral hearing that comparative experiments would be prohibitively expensive for a party not already operating a gas phase copolymerisation process of the kind required, this consideration cannot, in itself, reverse the onus of proof, putting the Patentee in the position of having to demonstrate that particular levels of clustering were encountered in this or that known polymer; although it goes without saying, that an Applicant or Patentee is under the greatest obligation of good faith in presenting the nature of his invention to the public.

In this connection, it is clear from the general submissions of the Respondent, which have not been directly contradicted, that clustering had always been a problem in the art, and that experience had shown that the particular levels of RMD claimed were never attainable in a conventional liquid phase process such as mentioned in D1 (submissions of the Respondent filed on 17 January 1997, page 4, in relation to D12; and on 23 July 1990, passage bridging pages 2 and 3).

It was, however, for the Appellants to demonstrate the opposite. This they have failed to do.

7.7 Consequently, the subject-matter of Claim 1 novel. The subject-matter of dependent Claim 2 is, by the same token, also novel.

8. *Inventive step*

Whilst the closest state of the art, D5, is concerned inter alia with an efficient incorporation of comonomer into ethylene polymers, this is merely presented as an aspect of the efficiency of the process, bearing in mind that 1-hexene comonomer is less reactive than ethylene (section 6.2, above). In particular, there is no suggestion of obtaining advantages associated with low density whilst using a smaller amount of comonomer relative to ethylene, let alone of controlling the distribution of the comonomer to achieve this purpose. Thus, there is no hint of a recognition of the technical problem underlying the patent in suit in the disclosure of D5, let alone of the solution proposed according to Claim 1.

8.2 The same considerations apply to D13, which is primarily concerned with the operability of the catalyst and does not mention the efficiency of incorporation of the comonomer.

8.3 The disclosure of D1 has been found, on its true interpretation, not to disclose any particular comonomer distribution (section 7.3, above). Even if, however, the references to isolated units were taken at face value, the disclosure of D1 is by its nature exclusively a structural investigation. There is nothing to relate the structures reported to any particular physical property such as density, let alone any advantages which might be associated therewith.

Consequently, there is equally no recognition in D1 of the technical problem. There is, therefore, no logically perceptible route to the proposed solution.

8.4 The same remarks apply with equal force, and for analogous reasons, to the disclosure of D12.

8.5 Consequently, neither the technical problem nor its solution is obvious from any of the documents cited. In other words, the subject-matter of Claim 1 involves an inventive step. The subject-matter of Claim 2 by the same token also involves an inventive step.

B. The process (Claims 3 to 8).

9. *The technical problem*

9.1 The patent in suit relates, in its process aspect, to a method of producing ethylene-monoolefin copolymers by contacting a gas mixture containing ethylene and another monoolefin with a silica/titania/chromium oxide catalyst, the catalyst having been activated by contacting the catalyst with free oxygen at a temperature in the range from 177° to 1093°C, under gas phase polymerisation conditions in a polymerisation zone to produce the ethylene-monoolefin copolymer with an efficient incorporation of monoolefin comonomer. Such a process is, however, known from D5, which is considered to represent the closest state of the art (cf. Section 6.2, above).

9.2 According to D5, the efficiency of comonomer incorporation is determined by the choice of comonomer, in the order $C_3 > C_4 > C_5 > C_6$, and the fluorine content of the catalyst (column 3, lines 43 to 50; column 6, lines 60 to 63). Furthermore, the listing of the amount, in mols, of the various comonomers that must be present in the gas stream to

provide polymers having a particular content of comonomer shows that 6 to 25 mol% of 1-butene is required in the gas stream to provide 2.5 to 12 mol% of the same comonomer in the copolymer. This corresponds to a minimum ratio of comonomer content of gas phase/comonomer content in the resulting copolymer = $6/2.5$ or 2.4. The ratios for the incorporation of 1-hexene are higher than this.

- 9.3 Compared with this state of the art, the technical problem may be seen as the search for a process capable of providing a further improved efficiency of incorporation of monoolefin comonomer.
- 9.4 The solution proposed according to Claim 3 of the patent in suit is to modify the process of D5 by (i) replacing the catalyst by one in which fluorine may be absent and the silica and titania have been cogelled, and (ii) controlling the concentration of the comonomer in the gas phase relative to its concentration in the resulting comonomer according to a relationship including a constant K which is defined for each of the comonomers 1-butene, 1-hexene, 1-octene and 1-methylpentene in Claim 3.
- 9.5 It can be seen from the from the values of the constants K recited in Claim 3 of the patent in suit that the efficiency with which the respective comonomer is incorporated in the copolymer corresponds to a molar concentration of comonomer in the gas phase equal to, or less than that in the copolymer, except in the case of the comonomer 1-butene, in which the ratio can be as high as $1/0.6 = 1.66$. These values are corroborated by the results of the examples in the patent in suit, in particular in Example I, runs 29 and 30 as shown in Table IIB in relation to a 1-butene comonomer; Example II, runs 1 and 2 as shown in Table IIIA in relation to a 1-hexene comonomer; and in

Example IV as shown in Table IV in relation to the remaining comonomers. The value of 1.66 referred to above, which corresponds to the least favourable incorporation rate efficiency of any of the comonomers defined in Claim 1, is nevertheless considerably higher than the rate of incorporation for the same comonomer listed in D5.

9.6 Consequently, the Board finds it credible that the problem is effectively solved by the claimed measures.

10. *Novelty*

The novelty of the process according to Claim 3 was not challenged. Consequently, the subject-matter of Claim 3 is held to be novel. Hence, the subject-matter of the dependent Claims 4 to 8 is also novel.

11. *Inventive step*

11.1 There is no hint to the solution of the stated problem in the disclosure of D5.

11.2 The argument that the constants of incorporation of the various comonomers defined according to Claim 3 of the patent in suit are simply the inevitable result of operating the gas phase process with the respective comonomer, is not supported by the disclosure of D5 itself, which teaches the necessity of establishing a considerable excess of the respective comonomer in the gas phase over that which is obtained in the resulting copolymer. Seen against this background, the possibility, given by the process according to the patent in suit, of having a higher molar concentration of comonomer incorporated in the copolymer than is present in the surrounding gas phase, is indeed a surprising result.

- 11.3 The argument that there was no absolute reduction in the amount of comonomer to be supplied for obtaining a polymer of a particular comonomer content is beside the point, since the technical problem relates to the efficiency of comonomer incorporation and not the absolute amount consumed. The increase in such efficiency, however, opens the possibility of reducing the relative content of comonomer in the gas phase, which brings further advantages in terms of less demanding process operating parameters (patent in suit, page 5, lines 13 to 15).
- 11.4 The further argument, that the K-values are simply a reflection of the known tendency for a comonomer of higher molecular weight to be incorporated to a greater extent is not convincing, since the efficiency of comonomer incorporation implied by the K-values represents a further increase over what was known from the closest prior art (sections 9.3 to 9.6, above).
- 11.5 Even though the disclosure of D13 briefly mentions the production of ethylene-1-hexene copolymers, it is not primarily concerned with efficiency of comonomer incorporation in a gas phase process, but rather with catalyst operability in a slurry process. Consequently, even if the skilled person had considered applying the catalyst according to D13 in a gas phase process such as that disclosed in D5, he would not have done so in any expectation of solving the stated problem. On the contrary, the teaching in D5 that the efficiency of comonomer incorporation depends directly on the fluorine content of the catalyst (column 6, lines 60 to 63) would discourage him from applying a catalyst of the kind disclosed in D13, which does not contain fluorine.
- 11.6 The remaining documents are more remote.

11.7 Consequently, the subject-matter of Claim 3 does not arise in an obvious way from the prior art. In other words, it involves an inventive step. The subject-matter of dependent Claims 4 to 8 by the same token also involves an inventive step.

Order

For these reasons it is decided that:

The appeals are dismissed.

The Registrar:


E. Gorgmaier

The Chairman:


C. Gérardin

