DECISION of 11 June 2002

Case Number: T 0046/00 - 3.3.3
Application Number: 92900629.4
Publication Number: 0517868
IPC: C08F 2/06

Language of the proceedings: EN

Title of invention: Multi-stage process for producing polyethylene

Patentee: BOREALIS POLYMERS OY

Opponent: Basell Polyolefine GmbH
SOLVAY POLYOLEFINS EUROPE-BELGIUM (Société Anonyme)

Headword: -

Relevant legal provisions: EPC Art. 54, 56

Keyword: "Novelty - implicit disclosure (no)"
"Inventive step - obvious combination (no)"

Decisions cited: -

Catchword: -
Case Number: T 0046/00 - 3.3.3

**DECISION**

of the Technical Board of Appeal 3.3.3

of 11 June 2002

**Appellant:** Basell Polyolefine GmbH
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**Representative:**

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**Decision under appeal:** Decision of the Opposition Division of the European Patent Office posted 26 October 1999 rejecting the opposition filed against European patent No. 0 517 868 pursuant to Article 102(2) EPC.

**Composition of the Board:**

Chairman: R. J. Young
Members: P. Kitzmantel
U. J. Tronser
Summary of Facts and Submissions

I. Mention of the grant of European patent No. 0 517 868 in respect of European patent application No. 92 900 629.4 in the name of Borealis Polymers Oy, which had been filed on 20 December 1991 by Neste Oy as PCT/FI91/00406 (WO 92/12182), claiming a FI priority of 28 December 1990, was announced on 15 November 1995 on the basis of eight claims, Claim 1 reading as follows:

"1. Multi-stage process for producing polyethylene having a bimodal and/or broad molecular weight distribution in the presence of an ethylene polymerizing catalyst system in a multistep reaction sequence, in which the first reaction step is a liquid phase polymerization step and the second reaction step is formed by one or more gas-phase polymerization steps, characterized in that in the first reaction step ethylene is polymerized in a loop reactor (10) in an inert low-boiling hydrocarbon medium the residence time being at least 10 minutes, reaction mixture is discharged from the loop reactor (10) and at least the inert hydrocarbon mixture is removed from the reaction mixture and the polymer is transferred into one or more gas-phase reactors (30), where the polymerization is completed in the presence of ethylene and optionally hydrogen and a comonomer."

Claims 2 to 8 are dependent on Claim 1.

II. Notices of Opposition requesting revocation of the patent in its entirety on the grounds of lack of novelty and/or inventive step were filed by

HOECHST AG (later Hostalen PolyethylenGmbH; later
Elenac GmbH; now Basell Polyolefine GmbH) (Opponent I) on 8 August 1996, and

Solvay S.A., later Solvay Polyolefins Europe – Belgium (S.A.) (Opponent II) on 12 August 1996.

The following documents were inter alia cited during the opposition proceedings:

D1: US-A-4 368 291,

D5: JP-A-59/47209,

D6: English Translation of D5,

D7: Chandrasekhar et al., Indian Journal of Technology, vol. 26, pages 53 to 82 (1988), and


III. By its decision announced orally on 12 October 1999 and issued in writing on 26 October 1999, the Opposition Division rejected the oppositions.

It was held in that decision that the subject-matter of the patent in suit was novel over all citations and in particular over D6 because the disclosure of this document did not comprise the use of a loop reactor for the first stage slurry polymerisation, even less at a residence time of at least 10 minutes. Carrying out the slurry copolymerisation in a continuous manner, at an industrial scale and at a pressure of not more than 25 kg/cm² was in line with the only first stage process
exemplified in D6, i.e. with the polymerisation in a stirred vessel, and did not necessarily point towards the use of a loop reactor.

The claimed process which combined the loop and the gas phase reactor technologies was also inventive because it provided the possibility to produce a wide range of polyethylenes from LLDPE (linear low density polyethylene) to HDPE (high density polyethylene). This technical solution was not foreshadowed in any of the citations because neither had the skilled person an incentive to replace the stirred vessel of D6 by a loop reactor, nor to combine the loop reactor method with the gas phase process although both these methods had been known long before the present invention as was apparent from D7 and D8.

IV. On 30 December 1999 the Opponent I (Appellant) lodged an appeal against the decision of the Opposition Division and paid the appeal fee on the same day. The Statement of Grounds of Appeal was received on 25 February 2000.

V. The arguments presented by the Appellant in its written submissions (Statement of Grounds of Appeal and letter dated 11 April 2002) and during the oral proceedings held on 11 June 2002 may be summarized as follows:

(i) The subject-matter of Claim 1 of the patent in suit was anticipated by D6 which disclosed a process for the production of ethylene polymers comprising a slurry polymerisation in an inert hydrocarbon followed, after separation of the hydrocarbon, by a gas phase polymerisation. Since the term "slurry polymerisation"
encompassed only two alternatives, i.e. the CSTR (continuously stirred tank reactor) method and the loop reactor process, the latter being particularly appropriate for industrial scale production, and since Table 10 of D7 showed that the upper range of the pressure range of 5 to 50 kg/cm² disclosed in D6 was typical for loop reactors, it was evident that the loop reactor process was within the implicit disclosure of D6.

(ii) The information in Table 10 of D7 was relevant to the interpretation of the polymerisation reaction conditions in CSTRs and loop reactors because both technologies could be operated with the same diluents (light and heavy hydrocarbons) and with the same catalysts, including Ziegler-Natta catalysts.

(iii) Moreover, the decision under appeal was not correct in stating that D6 was restricted to the preparation of low to medium density copolymers because the products of the slurry polymerisation of Examples 1 to 3 of D6 exhibited densities of 0.946 to 0.948 and were, thus, typical HDPEs.

(iv) Nor was the argument of the decision under appeal convincing that D6 failed to disclose a residence time of the slurry polymerisation of at least 10 minutes because the preferred proportion of 40 to 60%, with respect to the final polymer, of the first stage polymer clearly showed that the duration of the slurry polymerisation was in excess of 10 minutes.
(v) As to the issue of inventive step the Appellant emphasized that the real challenge in the production of polyethylene copolymers were copolymers having a low and medium density, while HDPEs could easily be obtained by deleting comonomers. The provision of a process which permitted the preparation of HDPEs was not, thus, part of a real problem.

(vi) Furthermore, the slurry polymerisation of D6 allowed for a ratio hydrogen/ethylene of from 0.1 to 15 which showed that this document encompassed the preparation in the slurry polymerisation of a wide range of molecular weights (and thus melt flow rates (MFR)).

(vii) Therefore, the only problem to be solved by the alleged invention was the provision of an increased space time yield (enabled by better heat transfer and shorter residence times in the slurry reactor).

(viii) The solution of this problem was, however, known from D7 which taught that loop reactors provided a higher space time productivity than CSTRs.

(ix) But even if one were to accept that the problem underlying the alleged invention vis-à-vis D6 was the provision of an improved flexibility with regard to some parameters (inter alia molecular weight, MFR, density) of the polymerization products, the solution of such a problem was obvious over the newly cited document D14: J.N. Short "Low Pressure Ethylene
Polymerization Process" in "Transition Metal
Catalyzed Polymerizations - Alkenes and
Dienes", Roderic P. Quirc (Editor), MMI
Press Symposium Series, Volume 4, Part B,

which disclosed that loop reactors were suitable
for the production of a broad product spectrum.

(x) The latter conclusion was supported by the fact
that the ratio hydrogen/ethylene in the slurry
polymerisation stage exemplified in the patent
specification was from 155 to 250 only, while D6
disclosed a preferred range of from 300 to 10,000
showing thereby that D6 was able to provide an
even wider range of molecular weights (cf. Table
on page 4 of the Patentee's submission dated
24 April 1998).

(xi) The Appellant also drew attention to the fact that
the evidence for the achievement of a wide range
of molecular weights including very low values was
restricted to the use of supercritical propane as
diluent and was thus not valid for the whole scope
of granted Claim 1.

VI. The Respondent (Proprietor of the patent) presented its
counterstatements in a written submission dated 6 July
2000 and at the oral proceedings. They can be
summarized as follows:

(i) The subject-matter of Claim 1 was novel over D6
because, according to this document, the slurry
polymerisation was not carried out in a loop
reactor but in a CSTR system. Such a system was
suitable for use in industrial scale as was apparent from its world-wide exploitation. There was, thus, no reason to assume that D6 did not relate to the CSTR system which was even described in D14, the document newly cited by the Appellant.

(ii) Moreover, D6 stated that the pressure in the flush zone was from 2 to 15 kg/cm$^2$ and was about 1 to 10 kg/cm$^2$ lower than in the slurry polymerisation. Thus, the pressure at this stage must be in the range of 2 to 25 kg/cm$^2$ which was typical for CSTR.

(iii) Further evidence for the use in D6 of a CSTR system was the organoaluminum/titanium catalyst which could not be used in a loop system at such low pressures.

(iv) The subject-matter of the patent in suit was also inventive over D6 which was directed to specific titanium catalysts and did not provide a route to a system of great flexibility for a wide range of superior polymer products having a wide distribution in terms of density (including HDPE which was not within the disclosure of D6), MFR and molecular weight distribution.

(v) This conclusion was supported by a comparison of the worked Examples of the patent specification and of D6 which showed that the polymers of the slurry polymerisation of the patent covered a much wider range of densities and MFRs than those according to D6.
(vi) A combination of D6 with the disclosure of the loop reactor technology in D7, as suggested by the Appellant, was moreover not obvious because this technology was restricted to the use of a chromic acid/silica catalyst.

(vii) Furthermore, the use of a loop reactor was just one of many alternatives for carrying out the slurry polymerisation of D6. There were several other routes including the method according to D1. Furthermore, for the skilled person looking for an improvement of the process of D6 this document rather pointed towards the development of better catalysts, a tendency confirmed by D7 and D14.

(viii) Auxiliarily, the Respondent pointed to the fact that the use of supercritical propane was itself an inventive feature which offered a range of advantages, including very high MFRs of the slurry polymer.

VII. The Opponent II has not submitted any written comments during the appeal proceedings and, although duly summoned, did not attend the oral proceedings which were therefore carried out in its absence (Rule 71(2) EPC).

VIII. The Appellant requested that the decision under appeal be set aside and that the European patent No. 0 517 868 be revoked.

The Respondent requested that the appeal be dismissed (main request) and as an auxiliary request that the patent be maintained on the basis of Claims 1 to 7.
submitted at the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

2. Despite its late filing with the Statement of Grounds of Appeal only, document D14 is introduced into these proceedings because its submission has been occasioned by the emphasis in the decision under appeal that HDPE was within the density spectrum of the polymers to be produced.

3. Citations

3.1 Document D1

Claim 1 of this document relates to a low pressure ethylene polymerisation process using a mixed Ziegler catalyst comprising a first suspension (slurry) polymerisation stage in a C$_3$ to C$_6$ hydrocarbon, preferably in a stirred kettle (cf. Claim 6), followed by a gas-phase polymerisation stage.

3.2 Document D6

This document claims a method for manufacturing an ethylene/á-olefin copolymer in the presence of a catalyst comprising (A) a highly active titanium component which has a particle size diameter of 1 to 200 µm and a particle size distribution represented by a standard deviation ó of less than 2.1 and (B) an organic aluminum compound.
The polymerisation is carried out in a first slurry polymerisation zone in the presence of hydrogen to obtain a first-step ethylene/α-olefin copolymer which is isolated in powder form in an ensuing flush zone, the powder being then introduced into a gas phase polymerisation zone under conditions of forming a second-step ethylene/α-olefin copolymer having a molecular weight greater than that of the first step copolymer.

According to page 3, first paragraph it is possible by this method to produce on an industrial scale an ethylene copolymer having a wide molecular weight distribution and a moderate to low density (see also page 5, lines 4 to 8 and page 8, lines 19 to 25).

The slurry polymerisation is carried out in propane or butane at a temperature of from about 10 to 100°C, at a pressure of about 5 to 50 kg/cm and at a molar ratio hydrogen/ethylene of about 0.1 to 15. The intrinsic viscosities of the first stage copolymer may range from 0.1 to 3.0 l/g [in the Board's opinion "l/g" is a translation error and should in all probability read "dl/g"], its density may range from 0.910 to 0.950 and it may be obtained in an amount of 20 to 80 % by weight of the total end product (cf. page 20, last paragraph to page 22, two lines from bottom).

The second (gas-phase) polymerisation step is carried out in the essential absence of hydrogen to a copolymer having an intrinsic viscosity of about 1.3 to 15 [d]l/g and a density of 0.910 to 0.945 (paragraph bridging pages 24 and 25).

According to Example 1 the first step slurry
polymerisation is carried out in a 15 l polymerizer kettle equipped with an agitator (cf. page 26, lines 2 to 3 from bottom) and the same conditions were used in Examples 2 and 3. The exemplified densities are for the first step copolymer 0.948, 0.947 and 0.946 and for the final copolymer 0.939, 0.937 and 0.938 g/cm\textsuperscript{3} (cf. Table on page 32).

3.3 Document D7

Table 10 (page 67) of this review article summarises the main features of a number of commercial processes for the preparation of HDPE including two slurry processes using, respectively, a loop reactor and an autoclave. The loop process operates at 85 to 105°C, a pressure of 7 to 35 kg/cm\textsuperscript{2} and uses a chromic acid/silica catalyst, the autoclave process operates at 70 to 90°C, a pressure of 7 to 10 kg/cm\textsuperscript{2} and uses a high activity Ziegler-Natta catalyst.

Section 4.1 (pages 68 to 70) describes polyolefin polymerisations in a CSTR with highly active Ziegler-Natta catalysts in high ("heavy") and low ("light") boiling hydrocarbon diluents. It is mentioned on page 70 (left hand column, second paragraph) that LLDPE copolymers cannot be achieved in a heavy diluent process because of copolymer solubility. The ensuing paragraph of D7 relates to the vertical loop type reactor pioneered by Phillips Petroleum Company for the slurry polymerisation of polyethylene with a chromium trioxide catalyst on a silica-alumina support. At the end of this paragraph it is inter alia stated that "Phillips catalysts are not susceptible to molecular weight control by use of conventional chain transfer agents such as hydrogen".
The paragraph thereafter relates to loop reactor processes for the polymerisation of polypropylene including the use of a Ziegler catalyst. In this context it is stated in lines 16 to 20 of the right hand column: "The light diluent loop reactor process has all the advantages inherent to such processes, namely, high heat transfer coefficient, short residence time, low fouling characteristics and high space time yields ....".

On page 73 (last paragraph of right hand column) advantages and drawbacks of the stirred tank reactor are enumerated, among the latter being a decrease in heat transfer area, difficulties associated with mass transfer, heat removal and agitation with increasing vessel size as well as difficulty of maintaining narrow residence time distribution during continuous operation.

3.4 Document D14

Pages 653 to 655 relate to: "A. Slurry Polymerization In A Light Hydrocarbon – Phillips Loop Reactor Process". In the last paragraph on page 653 reference is made to the copolymerisation of ethylene in isopentane or isobutane at 85 to 110°C. On page 654, last paragraph it is set out that the Phillips loop reactor process uses supported chromium catalysts with extremely high activity.

In the last paragraph on page 654 (including lines 1 and 2 on page 655) several advantages of the Phillips loop reactor process are enumerated, among which: high reactor solids without high viscosities, excellent heat transfer capability and low fouling.
The second paragraph on page 655 again relates to the relatively short residence time of the Phillips process and goes on to state: "The same plant can produce a broad range of resins, from high melt index injection molding homopolymers of narrow molecular weight distribution to fractional melt index broad molecular weight distributions resin for extrusion or blow molding applications, and ranging from HDPE to LLDPE below 0.930 density".

On page 657, lines 2 to 5 reference is made some problems associated with the use of stirred reactors, among which limited heat transfer capability and fouling tendency.

Main request

4. Novelty

The claimed subject-matter is novel over D6 because the disclosure in this document of the slurry polymerisation does not unambiguously make available the use of a loop reactor.

This judgment arises from the following facts:

4.1 There is no explicit disclosure of a loop reactor in D6.

4.2 According to Examples 1 to 3 and according to Fig. 1 of D6 the slurry polymerisation was carried out in a "15 l polymerizer equipped with an agitator" (i.e. in a stirred vessel) (cf. page 26, lines 2 to 3 from bottom; page 30, lines 7 to 15; page 31, lines 1 to 5; page 33).
4.3 The arguments brought forward by the Appellant intended to prove that the operating conditions of the slurry polymerisation of D6 must be interpreted to require the use of the loop reactor technology are not convincing for the following reasons:

4.3.1 The fact that the experiments of D6 had been performed in a laboratory scale stirred vessel does not allow the conclusion that a change to industrial scale would necessitate the use of a loop reactor. Indeed, the slurry polymerisation of ethylene in a stirred vessel has been commercially exploited by the CSTR technology thus refuting the argument that the loop reactor technology was the only industrially used slurry polymerisation technique (cf. D7, page 68, left hand column: "4.1 Slurry phase processes" to right hand column, line 3 from bottom).

4.3.2 Nor is the Appellant's argument convincing that the disclosure of D6 comprised the use of a loop reactor because this would be the only explanation for the disclosure of pressure values in excess of 10 bar (i.e. about 5 to 50 kg/cm²: sentence bridging pages 20 and 21 of D6). Contrastingly, slurry polymerisations in a stirred kettle may be performed, according to D1 (column 3, lines 33 to 53) at pressures up to 30 bars. As to the upper limit of the pressure range of 50 kg/cm², there is no stringent connection of this value with the use of a loop reactor (according to Table 10 of D7 the upper pressure limit of the HDPE loop process is 35 kg/cm²). Rather this value can be explained by the drafter's ambition not to exclude possibly feasible borderline conditions.

4.3.3 Nor can anything be gained for the Appellant's case
from the use, according to D6, of propane or butane as diluents of the slurry polymerisation because document D1 shows that these compounds are used for the slurry polymerisation of ethylene in a stirred kettle (cf. Claim 1 and column 4, lines 4 to 6 of D1). Furthermore, propane is not contained in the list of hydrocarbon diluents for the loop reactor technology set out in Table II of D8. This evidence therefore shows that the use in D6 of propane or butane as slurry diluents is in agreement with the CSTR technology and does not lend itself to Appellant's speculation that D6 implied the loop reactor technology.

5. Closest prior art

There is agreement between the parties that D6 represents the closest prior art and the Board sees no reason to deviate from this opinion.

As set out in the preceding Section, the essential difference between the subject-matter of D6 and that of the opposed patent is that, according to the latter, the slurry polymerisation is performed in a loop reactor.

6. Problem and solution

6.1 The patent in suit states on page 3, lines 5 to 15:

"Accordingly it is an object of the invention to provide a process, which permits manufacturing polyethylene, which has bimodal and/or broad molecular weight distribution and has therefore excellent melt characteristics and extrusion or mould properties. This is because the use of a loop reactor compared to
conventional stirred reactors gives better heat transfer from the reaction mixture and therefore more even temperature profile and a higher slurry density and therefore better mixing. Further the use of loop reactor permits shorter residence times and as a consequence the catalyst has no time to inactivate considerably and the catalyst transferred to the gas-phase reactor is still very active. The use of a loop reactor also makes possible higher polymer melt flow index, because in a loop reactor higher hydrogen concentrations can be used and therefore it is possible to achieve a more narrow molecular weight distribution in the product of slurry phase. A short residence time in combination with the flexibility of a gas phase reactor permits to manufacture a product with lower density and more easier quality changes. Further the use of a loop reactor in combination with one or more gas-phase reactors allows the use of various catalysts and thereby flexibility in selection of catalysts."

[emphasis by the Board]

6.2 The following conclusions can be drawn from the worked Examples reported in the Table on page 6 of the patent specification:

6.2.1 Loop reactor stage

By variation of the reaction conditions (diluent propane or isobutane; reaction mixture in supercritical condition or not; catalyst feed; catalyst activity; temperature; pressure; ethylene concentration; hydrogen concentration; 1-butene concentration; residence time) it is possible to prepare ethylene homo- or copolymers having densities in the range of 929 to 978 kg/m³
(Examples 4 and 1, respectively), exhibiting a melt flow index MFR$_2$ in the range of 0.2 to 260 (Examples 8 and 2, respectively). A molecular weight distribution D(M$_w$/M$_n$) is only indicated for Examples 2 (D=3.5) and 4 (D=4.0).

This evidence thus shows that the loop reactor can be operated under very different conditions and may thus be used to prepare ethylene (co)polymers having low or high density, low or high molecular weight and a moderate molecular weight distribution.

In view of the multiplicity of differences in the reaction conditions between single examples, it is not possible to infer from this evidence any particular strategy, i.e. any information concerning the influence of a particular reaction parameter on the properties of the resulting first stage polymer.

6.2.2 Only the first stage (slurry) products of Examples 1, 4, 5 and 6 have been subjected to the second stage gas-phase polymerisation.

Again, these examples use different reaction conditions (production rate; temperature; ethylene concentration; hydrogen concentration; 1-butene concentration; 1-hexene concentration; residence time).

The resulting end products have densities in the range of 928 to 955 kg/m$^3$ (Examples 5 and 6, respectively), a melt flow MFR$_2$ of 0.7 or 0.8 (Examples 4 and 5, respectively (cf. submission of the then Applicant dated 23 November 1992); no values for Examples 1 and 6), and molecular weight distributions of 9 (Example 4), 12 (Example 1) and 23 (Example 6) (no
The only examples which thus allow a conclusion as to the influence of the gas-phase polymerisation on the product of the loop reactor polymerisation are Examples 4 and 5: according to Example 4 the density of 929 kg/m$^3$ of the loop reactor product was maintained after the gas-phase polymerisation, while the MFR$_2$ was lowered drastically from 150 to 0.7; according to Example 5 the density was lowered from 943 kg/m$^3$ to 928 kg/m$^3$ and the MFR$_2$ was lowered from 148 to 0.8. According to Example 4 the molecular weight distribution increased from 4 to 9.

Again, the simultaneous change of various reaction conditions makes it impossible to draw any conclusions as to the concrete influence of certain parameters on the results of the gas-phase polymerisation.

6.2.3 The conclusion which can be drawn from this experimental evidence with regard to the end-products of the claimed process is therefore that it allows the preparation of ethylene copolymers having bimodal/broad molecular weight distribution and low as well as high densities, which copolymers comprise portions of first stage slurry polymer of similar or higher density and much higher MFR (i.e. much lower molecular weight) than the end-product.

6.2.4 Contribution of the claimed subject-matter over the disclosure of D6

(i) From the aspects of the problem underlying the claimed subject-matter as set out in point 6.1 supra the afore-mentioned experimental evidence...
thus only demonstrates the provision of a bimodal and/or broad molecular weight distribution (with the consequential possibility to furnish polymers having excellent melt characteristics and extrusion and mould properties).

However, the solution of this aspect of the problem does not represent a contribution over D6 because this document already furnishes a method for producing ethylene copolymers having a wide molecular weight distribution including portions of high and low molecular weight (cf. point 3.2 supra). This follows from the range of the intrinsic viscosity of about 0.1 to 3.0 [d]l/g and equally from the range of the molar ratio hydrogen/ethylene of from about 0.1 to 15 (cf. page 21, last line to page 22, line 11 of D6) which shows that the molecular weight of the product of the slurry polymerisation of D6 may vary enormously, and may in fact be very low (cf. D7, page 63, left hand column, last sentence of penultimate paragraph: inverse relationship of molecular weight and square root of hydrogen partial pressure). Conversely, the gas-phase polymerisation of D6 may lead to polymers of much higher molecular weight which is reflected by the product's intrinsic viscosity of about 1.3 to 15 [d]l/g (cf. page 24, penultimate line to page 25, line 4).

This conclusion is not invalidated by the fact that the slurry products of the three examples of D6 do not essentially differ in their respective molecular weights but exhibit very
similar MFRs (and in turn molecular weights) because this simply results from their using the same molar ratio hydrogen/ethylene of 3.5 and the same monomer ratio 1-butene/ethylene of 0.03 (cf. page 26, line 4 from bottom to page 27, line 9; page 30, last paragraph; page 31, first paragraph; page 32, Table). These experimental data cannot therefore restrict the aforementioned generic disclosure which covers a broad range of possible molecular weights and molecular weight distributions, the latter property resulting from the molecular weight mixture caused by the two stages of the polymerisation: slurry and gas-phase.

Therefore the molecular weight distribution of the end-product does not qualify as a feature distinguishing the claimed subject-matter from the teaching of D6 and, consequently, this aspect cannot form part of the problem to be solved vis-à-vis this document.

(ii) As to the further asserted problem aspect of a more narrow molecular weight distribution of the product of the slurry stage, neither is there any evidence, nor is it credible, in the Board's judgment, that this objective is achieved vis-à-vis D6 because, as set out in the preceding paragraph, D6 may use much higher hydrogen/ethylene ratios and because the temperature of the slurry polymerisation of D6 may be up to 100°C (cf. page 20, lines 2 to 6 from bottom) while the upper temperature limit in the loop reactor of the patent in suit is 110°C (Claim 6).
It is, however, known from D7 (cf. page 64, left hand column, third paragraph) that "increased concentration of hydrogen (chain transfer agent) and increase in polymerization temperature leads to narrowing of MWD".

A narrower molecular weight distribution of the slurry stage product cannot thus be considered as an objectively existing aspect of the technical problem underlying the claimed invention vis-à-vis D6.

(iii) The same conclusion applies for the same reasons to the asserted problem aspect of providing a higher polymer melt flow index (i.e. a lower molecular weight) of the slurry stage product because the Appellant's contention of higher hydrogen concentrations in the loop reactor as compared with the CSTR is at variance with the disclosure of D6 which, as discussed above, allows for a molar ratio hydrogen/ethylene of up to 15 which is far in excess of the 1:1 molar ratio maximum according to Claim 3 of the patent in suit.

(iv) No other conclusion can be drawn with regard to the problem aspect concerning the asserted possibility of the claimed process to manufacture a final polymer with lower density than according to D6.

The density of a low pressure polyethylene copolymer is to a large extent dependent on the kind and amounts of the comonomers and in that respect the disclosure of the patent in suit is...
not distinguished from D6.

In fact, the Respondent did not elaborate in its submissions on this aspect but rather on the quite different allegation that, as opposed to D6, the patent in suit comprised the preparation of HDPE, a circumstance not as such emphasised in the specification but which can be inferred from the end-product densities of Examples 1 (density 948 kg/cm$^3$) and 6 (density 955 kg/cm$^3$).

However, the maximum density of the end-product according to D6 is 0.945 g/cm$^3$ (cf. page 25, lines 4 to 9) and is thus, above the value of 0.940 g/cm$^3$ (or 940 kg/m$^3$) which is normally considered in the art as lower density limit of HDPE. It is, moreover, apparent from said Examples 1 and 6 that their high densities correspond to very small comonomer contents: in both cases no comonomer was present in the slurry stage; in the gas-phase stage Example 1 added 0.8 mole% 1-butene, Example 6 added 0.9 mole% 1-hexene.

The adjustment of the density of the final polymer cannot thus be recognized as part of the problem underlying the patent in suit with respect to D6.

(v) The further asserted objective of a higher flexibility in the selection of catalysts has not been commented upon by the Appellant. While it is stated in the patent specification (page 3, lines 27 to 28) that "any polymerization catalyst suitable for the
production of ethylene polymers can be used", in Examples 1 to 8 only one catalyst was used. The constitution of this catalyst is not explicitly disclosed in the patent specification but only by reference to Example 1 of the PCT application FI90/00279 (page 5, lines 6 to 8). The actual scope of the flexibility of the catalyst selection cannot, thus, be judged objectively.

(vi) In contrast to the conclusions drawn in the preceding sub-points (i) to (v) it is perfectly credible, owing to the inherent advantages of the loop reactor technology, that the claimed polymerisation method including a loop reactor provides better heat transfer, higher slurry density, better mixing and shorter residence times than according to the stirred kettle method of D6 (cf. D7, page 70, right hand column, lines 16 to 20; D14, page 654, line 9 from bottom to page 655, second paragraph). This is particularly convincing because these advantages of the loop reactor correspond to respective deficiencies of the CSTR technique (D14, page 657, lines 2 to 5).

(vii) It is similarly credible in the Board's judgment, that, due to the shorter residence time in a loop reactor as compared with a CSTR, product quality changes may be performed more easily, i.e. more efficiently.

6.3 In the Board's judgment, in order to formulate the problem objectively underlying the present subject-matter vis-à-vis D6, the advantages actually achieved by the substitution of a loop reactor for the stirred
vessel used according to D6, i.e. better heat transfer, higher slurry density, better mixing, shorter residence times and more efficient quality changes (cf. sub-points 6.2 (vi) and (vii) supra) must be evaluated with respect to their effect on the entire polymerisation process. By that, one arrives at an objective problem formulation which is indeed the one chosen by the Appellant during the oral proceedings, namely the provision of a process for the polymerisation of ethylene (co)polymers having an improved space time yield, a definition which corresponds to the term "high space time reactor productivity" used in D7 (page 77, left hand column, last sentence).

7. Obviousness

7.1 This issue thus turns on the question whether the state of the art contains any information capable of inciting the skilled person to solve the afore-mentioned objective technical problem by substituting a loop reactor process for the stirred vessel process of the two stage polymerisation process of D6.

7.2 While both D7 and D14 contain statements as to a number of process advantages of the loop reactor over the CSTR system, particularly including the short residence time, the transfer of the loop reactor technology to the slurry polymerisation of D6, in order to solve the existing technical problem, is nevertheless non-obvious for the following reasons:

7.2.1 The polymerisation process of D6 requires a catalyst system which comprises (A) a highly active titanium component which has a particle size diameter of 1 to 200 µm and a particle size distribution represented by
a standard deviation \( \sigma \) of less than 2.1 and (B) an organic aluminum compound (cf. Claim 1).

The second paragraph on page 5 of D6 stresses the importance of the properties of the catalyst system in order to "... keep the gas phase polymerization operating smoothly and continuously for a long period of time ...".

According to D6 the consequence of catalyst particles which are smaller than the claimed diameter range may be: "agglutination of polymers" and "polymers are entrained by a waste gas system in the polymerizing bath"; the consequence of particles which are too big may be that the "flow state of the fluid bed" is "bad" and polymer may "lodge on walls ... making the uniform polymerization hardly possible". The same problems may result if the standard deviation \( \sigma \) of the particle size distribution is more than 2.1 (cf. page 11, last paragraph to page 12, second paragraph). The consequence of too big a \( \sigma \) value is, furthermore, illustrated by Comparative Example 1 according to which fine catalyst powder was entrained in the gas phase polymerizer leading to fouling and plugging of the gas phase reaction zone, the waste gas line and the cooler (cf. page 28, last line to page 30, line 6).

7.2.2 According to D7 the Phillips loop reactor process for the manufacture of polyethylene uses a catalyst which is prepared by impregnating a silica-alumina support with an aqueous solution of chromium trioxide. D7 is devoid of any disclosure suggesting a combination of this process with a gas-phase reactor polymerisation (cf. point 3.3 supra; D7: page 70, left hand column, lines 15 to 25).
7.2.3 It is not obvious in view of the existing technical problem to apply this teaching, which relates to the use of a catalyst system which is vastly different from the catalyst system of D6, to the slurry polymerisation of D6 because neither would the skilled person expect - in the absence of any information in that respect - that the Phillips catalyst would satisfy the requirements for a gas phase catalyst, nor could he surmise that the catalyst system of D6 would be applicable to the Phillips loop reactor system.

Rather the skilled person would expect that such a technology transfer would require a quite extensive research program which goes beyond ordinary routine adaptation of a few process parameters.

7.2.4 The above conclusion is supported by the information in D7, page 70, left hand column, lines 29 to 33 that Phillips catalysts were not susceptible to molecular weight control by use of conventional chain transfer agents such as hydrogen, because the use of hydrogen as chain transfer agent is essential for the slurry process of D6 (cf. page 22, lines 4 to 11; page 27, lines 4 to 9 (Example 1)).

7.2.5 A valid obviousness objection can also not be construed on the basis of the information in D7 directed to the use of conventional Ziegler catalysts for the loop reactor slurry polymerisation of propylene (cf. page 70, left hand column, line 34 to right hand column, line 32). While this passage inter alia states that "loop reactors constitute one of the most optimum designs of polymerization vessels for polyolefins" (right hand column, lines 25 to 27), it is conspicuous that there is no concrete suggestion therein of the
application of the loop reactor technology with Ziegler catalysts for the manufacture of polyethylene let alone ethylene copolymers. The relevance of the information in this passage to the applicability of the loop reactor technique in connection with Ziegler catalysts to the preparation of polypropylene only is underlined by the part of the sentence preceding the aforementioned quotation which qualifies the (Ziegler) catalysts as "high yield-high stereospecificity catalysts" (lines 20 to 23). It is self-evident that stereospecificity is not an issue with the polymerisation of ethylene.

7.2.6 A further warning against an extensive interpretation of this passage can be derived from the ensuing paragraph (page 70, right hand column, lines 28 to 32) where reference is made to a loop reactor process for the preparation of polypropylene with a "proprietary Solvay catalyst" followed by the information that this "process, however, requires catalyst deactivation and atactic recovery steps".

This shows the sensitivity of polymerisation systems, here the loop reactor system, to changes of the catalyst system. The extrapolation of a possible behaviour of such a catalyst system in a gas phase polymerisation stage to be carried out after the loop reactor stage is, of course, much more problematic.

7.2.7 Moreover, there is no information in D7 concerning the particle size distribution of the catalyst, a property whose importance is particularly emphasised in D6 with regard to the second stage gas-phase polymerisation which is not disclosed or suggested in D7.
7.2.8 In view of the afore-mentioned deliberations the skilled person was not in a position, on the basis of the information in D7, to expect that by a replacement of the stirred vessel by a loop reactor in the slurry polymerisation of D6 he would be able to successfully solve the existing technical problem.

7.2.9 No other conclusion is arrived at if D14 is considered instead of D7 because with regard to the operation of the loop reactor system this document also focuses on the Phillips system and does not add any more relevant information. In particular, D14 does not disclose the use of the loop reactor system for the preparation of ethylene copolymers with Ziegler catalysts. Furthermore, although referring to slurry (in light or heavy hydrocarbon), gas-phase and solution processes for the polymerisation of ethylene (cf. second paragraph of resumé on page 651) and discussing "unsolved problems" (pages 666 to 667), D14 does not envisage the combination of slurry and gas-phase polymerisation processes.

8. In the interests of completeness the Board observes that the Appellant's approach concerning the alleged obviousness of the replacement of the CSTR method of D6 by the loop reactor method of D7/D14 is based on the ex post facto assumption that, for the solution of the existing technical problem, a modification of the slurry polymerisation reactor was the only point of departure. The Appellant, however, failed to substantiate this allegation, which, in the Board's judgment, is anyway wrong because many ways may be conceived for the solution of the existing technical problem, e.g. modifications of the catalyst system and/or of the CSTR processing conditions including CSTR
cascades without a gas-phase reactor.

9. In summary, the grounds of opposition brought forward by the Appellant do not justify revocation of the patent as granted.

10. There is thus no need to consider the auxiliary request.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

E. Görgmaier R. Young