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DECISION
of 21 June 2001

Case Number: T 0314/99 - 3.3.3

Application Number: 84303805.0

Publication Number: 0129368

IPC: C08F 10/00

Language of the proceedings: EN

Title of invention:
Process and catalyst for polyolefin density and molecular weight control

Patentee:
ExxonMobil Research and Engineering Company

Opponent:
Targor GmbH (Former Opponent)
Fina Research S.A.

Headword:
-

Relevant legal provisions:
EPC Art. 54(2), 56, 82, 114(2), 134(7)

Keyword:
"Authorisation of legal practitioner (yes)"
"Late submitted documents - not admitted"
"Public availability of prior art ("Diplomarbeit") - no, inadequate evidence"
"Unity of invention (no) - no issue in opposition"
"Inventive step (no)- part-invention obvious"

Decisions cited:
G 0001/91, T 0184/82, T 0013/84, T 1002/92

Catchword:

Where conceptual non-unity arises between different embodiments covered by a claim, this may necessitate the formulation of corresponding partial problems, the respective solutions of which must be assessed separately for inventive step. (Reasons 9).

Europäisches Patentamt
Beschwerdekammern

European Patent Office
Boards of Appeal

Office européen des brevets
Chambres de recours

Case Number: T 0314/99 - 3.3.3

DECISION
of the Technical Board of Appeal 3.3.3
of 21 June 2001

Appellant: ExxonMobil Research and Engineering Company
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Decision under appeal: **Interlocutory decision of the Opposition Division of the European Patent Office posted 13 July 1999 concerning maintenance of European patent No. 0 129 368 in amended form.**

Composition of the Board:

Chairman: R. Young
Members: P. Kitzmantel
J. C. M. De Preter
C. G. Idez

C. Rennie-Smith

Summary of Facts and Submissions

I. Mention of the grant of European patent No. 0 129 368 in respect of European patent application No. 84 303 805.0 in the name of Exxon Research and Engineering Company (now "ExxonMobil Research and Engineering Company"), which had been filed on 5 June 1984 claiming a US priority of 6 June 1983, was announced on 26 July 1989 on the basis of 5 claims, of which independent Claims 1 and 4 read as follows:

"1. A catalyst suitable for the polymerisation of an olefin comprising (a) the carbene represented by the formula

$Cp_2Zr{=}CH_2P(C_6H_5)_2CH_3$

and its derivative of the formula

$Cp_2ZrCH_2CH(CH_3)CH_2$
.))))))))))-

and the carbene represented by the formula

$Cp_2Ti{=}CH_2.Al(CH_3)_2Cl$

and the derivatives of this carbene

$Cp_2Ti{=}CH_2.Al(CH_3)_3$, $(Cp_2TiCH_2)_2$,
$Cp_2TiCH_2CH(CH_3)CH_2$, $Cp_2Ti{=}CH_2.AlR'''_2Cl$,
.))))))))))-

wherein Cp is a cyclopentadienyl or substituted cyclopentadienyl radical, and R"' is an alkyl, aryl or alkylaryl radical having 1-18 carbon atoms; or a compound of the formula

$(C_5R'_m)_pR''_s(C_5R'_m)MeQ_{3-p}$ or $R''_s(C_5R'_m)_2MeQ'$,

wherein Me is a Group 4b, 5b or 6b metal, $(C_5R'_m)$ is cyclopentadienyl or substituted cyclopentadienyl, each R', which can be of the same or different, is hydrogen, an alkyl, alkenyl, aryl, alkylaryl or arylakyl radical having from 1 to 20 carbon atoms or two R' substituents together form a fused C_4-C_6 ring, R" is a C_1-C_4 alkylene radical, a dialkyl germanium or silicone or an alkyl phosphine or amine radical bridging two $(C_5R'_m)$ rings, each Q which can be the same or different, is aryl, alkyl, alkenyl, alkylaryl or arylakyl radical having from 1 to 20 carbon atoms or halogen, Q' is an alkylidene radical having from 1 to 20 carbon atoms, s is 0 or 1, p is 0, 1 or 2; provided that s is 0 when p is 0; m is 4 when s is 1; m is 5 when s is 0; and that at least one R' is a hydrocarbyl radical when s=0 and Q is an alkyl radical or halogen and (b) an alumoxane."

"4. A process for polymerising one or more olefins which comprises conducting the polymerisation in the presence of a catalyst system as claimed in any one of claims 1 to 3."

Claims 2 and 3 were dependent on Claim 1, and Claim 5 was dependent on Claim 4.

II. Notices of Opposition requesting revocation of the patent in its entirety on the grounds of Articles 100(a) and (b) EPC were filed by

Hoechst AG (later Targor GmbH) (Opponent I) on 21 April 1990, and by Fina Research S.A. (Opponent II) on 25 April 1990.

The oppositions were supported *inter alia* by the documents

D1: EP-A-0 035 242,

D2: DE-A-3 127 133,

D3: H. Höcker and K. Saeki, Die Makromolekulare Chemie, 148, 107 to 118 (1971), and

B2: T.J. Katz and N. Acton, Tetrahedron Letters No. 28, 1970, Pergamon Press, pages 2497 to 2499.

During the first oral proceedings before the Opposition Division held on 9 June 1992 novelty objections were raised for the first time on the basis of the newly cited documents

D4: J.Herwig and W.Kaminsky, Polymer Bulletin 9, 464 to 469 (1983) (referred to in CA 98, No. 20, 161589u, 16 May 1983), and

D5: Jens Herwig, Dissertation, Hamburg 1979.

In view of their apparent relevance these documents were admitted into the opposition proceedings.

III. In a written submission under Article 115 EPC dated 14 March 1995 Spherilene S.r.l. presented arguments as to novelty and inventive step of the claimed subject-matter relying *inter alia* on documents

D6: K.Külper, Diplomarbeit, Hamburg, 1981, D7: W.Marconi et al., La Chimica, vol. 44 (1962), 235 to 240 (with English translation), and

D8: EP-A-0 069 951 (claims the priority of D2).

IV. In its interlocutory decision announced at the second oral proceedings held on 15 April 1999 and issued in writing on 13 July 1999 the Opposition Division maintained the patent on the basis of the following set of three claims of the then third auxiliary request (Annex V of the said decision):

"1. A catalyst suitable for the polymerisation of an olefin comprising (a) a compound of the formula

$$(C_5R'_m)_pR''_s(C_5R'_m)MeQ_{3-p} \text{ or } R''_s(C_5R'_m)_2MeQ',$$

wherein Me is zirconium, $(C_5R'_m)$ is cyclopentadienyl or substituted cyclopentadienyl, each R' which can be the same or different, is hydrogen, an alkyl, alkenyl, aryl, alkylaryl or arylalkyl radical having from 1 to 20 carbon atoms or two R' substituents together form a fused C_4-C_6 ring, R" is a C_1-C_4 alkylene radical or a dialkyl silicon radical bridging two $(C_5R'_m)$ rings, each Q which can be the same or different, is aryl, alkyl, alkenyl, alkylaryl or arylakyl radical having from 1 to 20 carbon atoms or halogen, Q' is an alkylidene radical having from 1 to 20 carbon atoms, s is 0 or 1, p is 1; m is 4 when s is 1; and m is 5 and $(C_5R'_m)$ is mono-alkyl, di-alkyl, tri-alkyl, or tetra-alkyl substitued cyclopentadienyl when s is 0; and (b) an alumoxane.

2. A process for polymerising one or more olefins which comprises conducting the polymerisation in the

presence of a catalyst system as claimed in claim 1.

3. A process according to claim 3 wherein the olefin is ethylene or an á-olefin having from 3 to 8 carbon atoms per molecule."

V. The independent claims of the other requests referred to in the decision of the Opposition Division differ essentially from Claim 1 of the above-quoted third auxiliary request in the following respects:

(i) None of these claims comprises the feature "and m is 5 and ($C_5R'_m$) is mono-alkyl, di-alkyl, tri-alkyl, or tetra-alkyl substituted cyclopentadienyl when s is 0".

(ii) Claim 1 of the main request (Annex II of the decision) also differs from Claim 1 of the third auxiliary request in that:

- Me in the formula of compound (a) is a Group 4b metal,
- R" may additionally be a dialkyl germanium or an alkyl phosphine or amine radical bridging two ($C_5R'_m$) rings,
- p may also be 0,
- s is 0 when p is 0,
- m is 5 when s is 0,
- compounds wherein Me is titanium or all of the cyclopentadienyl radicals are unsubstituted are

excluded.

(iii) Claim 1 of the first auxiliary request (Annex III of the decision) differs further from Claim 1 of the third auxiliary request in that:

- compounds wherein all of the cyclopentadienyl radicals are unsubstituted are excluded (cf. text of the claim at point VIII below).

(iv) Claims 1 of the second and fourth auxiliary requests (Annexes IV and VI of the decision) furthermore differ from Claim 1 of the third auxiliary request in that:

- s may only be 1 (i.e. only bridged compounds are covered).

(v) Claim 3 of the second auxiliary request (Annex IV of the decision) differs from Claim 1 of the third auxiliary request in that

- it relates to a process for copolymerizing ethylene with an á-olefin having from 3 to 8 carbon atoms per molecule in the presence of a catalyst system whose characterisation differs from that of Claim 1 of the third auxiliary request in that (apart from the difference in point (i) supra)

- compounds wherein all of the cyclopentadienyl radicals are unsubstituted are excluded.

VI. The decision under appeal held *inter alia*

(i) that document D6, the Diplomarbeit of Klaus Külper, should be admitted into the proceedings, although it was submitted after the opposition period by a third party under Article 115 EPC;

(ii) that this document formed state of the art because, in the light of the testimony given by Mrs Oetken, the Head Librarian of the Hamburg University Chemistry Department Library since 1967, in the case Exxon vs. Mobil before the United States District Court for the Southern District of Texas, Houston Division (hereinafter "the US proceedings"), it was registered in the "logbook" (cf. point X (iii-1) below) of the Chemistry Department Library not later than January 1982 and, from that date on, it could be inspected on request by any member of the public;

(iii) that D6, which disclosed a pentamethyl substituted zirconocene/aluminoxane catalyst system for the polymerisation of ethylene, anticipated the subject-matter of the main and first auxiliary requests;

(iv) that, because of D6's further reference to the use of unsubstituted zirconocenes for the copolymerisation of ethylene with hexene-1, it also rendered obvious the process of Claim 3 of the second auxiliary request;

(v) but that the subject-matter of the third auxiliary request was novel and inventive, because the available state of the art did not disclose zirconocene/aluminoxane catalyst systems having bridged cyclopentadienyl radicals, nor did it suggest the improved catalytic activity of unbridged catalyst systems, whose cyclopentadienyl radicals were mono-, di-,tri-, or tetra-alkyl substituted.

VII. Notices of appeal against that decision were filed by

- the Patentee on 21 September 1999,
- Opponent I on 3 September 1999, and
- Opponent II on 23 July 1999.

The appeal fee of the Patentee and of Opponent I were paid together with the respective Notices of Appeal, and that of Opponent II was paid on 6 August 1999.

The respective Statements of Grounds of Appeal were filed with submissions dated 17 November 1999 (Patentee), 22 November 1999 (Opponent I) and 22 November 1999 (Opponent II).

By a letter dated 15 November 2000, the Patentee requested that the appeal proceedings be expedited, since the patent in suit was the subject of infringement proceedings before an English court, the latter proceedings having been stayed pending determination of the present appeal, and the English judge having required that such a request be made.

Summons to attend oral proceedings were dispatched by the Board on 28 December 2000.

Notification of the withdrawal of the opposition of Opponent I was received on 18 June 2001. Opponent I thus ceased as of that date to be a party to the substantive issues in the appeal proceedings.

Notification of the withdrawal of the appeal of Opponent II was received on 20 June 2001. Opponent II thus became as of that date the Respondent to the Patentee's appeal.

VIII. At the oral proceedings held on 21 June 2001 the Patentee submitted as its sole request a corrected version of Claims 1 to 3 of the former first auxiliary request (Annex III of the decision under appeal), the only correction being the change in Claim 3 of the erroneous reference "according to Claim 3" to "according to Claim 2".

Claim 1 of this request reads:

"1. A catalyst suitable for the polymerisation of an olefin comprising (a) a compound of the formula

$$(C_5R'_m)_pR''_s(C_5R'_m)MeQ_{3-p} \text{ or } R''_s(C_5R'_m)_2MeQ',$$

wherein Me is zirconium, $(C_5R'_m)$ is cyclopentadienyl or substituted cyclopentadienyl, each R' which can be the same or different, is hydrogen, an alkyl, alkenyl, aryl, alkylaryl or arylalkyl radical having from 1 to 20 carbon atoms or two R' substituents together form a fused C_4-C_6 ring, R" is a C_1-C_4 alkylene radical or a dialkyl silicon radical bridging two $(C_5R'_m)$ rings, each Q which can be the same or different, is aryl, alkyl, alkenyl, alkylaryl or arylakyl radical having from 1 to 20 carbon atoms or halogen, Q' is an alkylidene radical having from 1 to 20 carbon atoms, s is 0 or 1, p is 1; m is 4 when s is 1 and m is 5 when s is 0, excluding compounds wherein all of the cyclopentadienyl radicals are unsubstituted; and (b) an alumoxane."

IX. The arguments presented by the Patentee in its written and oral submissions, insofar as they are still relevant after the withdrawal by Opponent I of its opposition and by Opponent II of its appeal, may be summarized as follows:

(i) All amendments to the claims are supported by the original disclosure;

(ii) The opposition ground of Article 100(b) EPC should not be admitted;

(iii) The newly cited documents (submitted with the respective Statements of Grounds of Appeal of Opponent II (D9 to D11) and Opponent I (D12); cf. point X (iv) below), namely:

D9: US-A-2 924 593,

D10: Makromol.Chem. Rapid Comm. 4, 417-21, 1983,

D11: Proc. IUPAC Macromol. Symp. 28th, 247, and

D12: "Advances in Polyolefins", ed. R.B.Seymour and T. Chang, New York 1987, pages 361-371

should not be admitted, because

- no justification had been given for their late submission,

- these documents were already known to the Opponents from the US proceedings by the time of the first instance proceedings,

- D9 was not relevant, in that it was published in 1960 and was, thus, part of a research line which had led to the development of conventional Ziegler-Natta type catalysts,

- it had not been established that D10 and D11 were published before the priority date of the patent in suit, and

- D12 was published after the priority date of the patent in suit.

(iv) In order to decide on the public availability of Külper's Diplomarbeit D6 the following evidence was *inter alia* to be considered:

"Diplomprüfungsordnung":
Diplomprüfungsordnung für Studierende der Chemie of 28 May 1969 according to "Amtlicher Anzeiger, Teil II des Hamburgischen Gesetz- und Verordnungsblattes", Nr. 123, pages 805 to 808, 1 July 1969) filed as Exhibit C with Patentee's submission dated 25 August 1995,

"German Copyright Act":
Excerpt of English translation entitled "Act Dealing with Copyright and Related Rights (Copyright Act)" filed as Exhibit P-2 of the Patentee's submission dated 17 November 1999,

"Declaration Oetken 1":
Declaration of Gerda Oetken, Dipl.-Bibl., Head Librarian of the Library of the Department of Chemistry of the University of Hamburg dated 17 October 1994, filed by Spherilene with its

Article 115 EPC intervention dated 14 March 1995,

"Declaration Oetken 2":
Declaration of the said Gerda Oetken dated 28 June 1995, filed with Patentee's submission dated 17 January 1996,

"Declaration Kaminsky 1":
Declaration of Prof. W. Kaminsky, Institute for Technical and Macromolecular Chemistry of the University Hamburg, dated 31 October 1996, filed with Opponent I's submission dated 13 November 1996,

"Declaration Kaminsky 2":
Declaration by Prof. W. Kaminsky dated 16 June 1998 in the US proceedings (Annex V of Patentee's submission dated 15 March 1999),

"Transcript Oetken":
Excerpt from the transcript of the examination of Mrs Oetken on 5 August 1998 in the US proceedings (Annex L of Mr Pietzcker's observations dated 14 December 1998 attached to the Patentee's submission dated 15 December 1998),

"Transcript Kaminsky 1":
Excerpt from the transcript of the deposition of Prof. W. Kaminsky on 20 October 1997 in the US proceedings (Annex K of Mr Pietzcker's observations dated 14 December 1998 attached to the Patentee's submission dated 15 December 1998),

"Transcript Kaminsky 2":
Excerpt from the transcript (pages 1 to 4, 69 to

76) of the examination of Prof. W. Kaminsky on 31 July 1998 in the US proceedings (Annex U of Patentee's submission dated 15 March 1999),

"Transcript Kaminsky 2*":
Excerpt of the transcript (pages 8, 53, 54, 55, 71, 72) of the said examination of Prof. W. Kaminsky on 31 July 1998 in the US proceedings (filed with Opponent I's submission dated 15 January 1999),

"US court opinion":
"Corrected Memorandum Opinion" of the Court in the US proceedings dated 10 September 1998 (Annex M of Mr Pietzcker's observations dated 14 December 1998 attached to the Patentee's submission dated 15 December 1998), and

"Decision Bundespatentgericht":
Decision of the German Bundespatentgericht of 6 December 1983 12W (patent) 19/83, referred to in "Mitteilungen der deutschen Patentanwälte, 75. Jg./1984, pages 148 to 149" (Annex O of Mr Pietzcker's observations dated 14 December 1998 attached to the Patentee's submission dated 15 December 1998).

(v) Taking the afore-mentioned evidence into account, Külper's Diplomarbeit D6 could not, in the Patentee's opinion, be considered as an applicable prior art document for the following reasons:

(v-1) A Diplomarbeit is protected by the German Copyright Act, according to which its publication is subject to authorization by the author, which in the present case was not given;

(v-2) The only copy of D6, which was in the possession of the Hamburg University Chemistry Department Library, was locked away in a cupboard in a section of the library (hereinafter called "archive") which is not a part of the public service area; access to documents in that cupboard was only possible on specific request to the Head Librarian, Mrs Oetken;

(v-3) On a fair assessment of probabilities, conclusive evidence was lacking that the public had been aware of D6 before the relevant priority date, since neither

(v-3.1) the date of D6's entry into the library's "logbook" (cf. point X (iii-1) below), nor

(v-3.2) the date of publication of the "Jahresbericht" (cf. point X (iii-1) below), wherein D6 was referred to with its full title, could be established with any certainty;

(v-4) The testimony of Mrs Oetken, as contained in the "Transcript Oetken", namely that she would have handed out D6 to anyone asking to see it, was unreliable,

(v-4.1) because it related to a situation 16 years previously and because the vagueness of Mrs Oetken's recollection of the facts surrounding D6 was conspicuous from the two different dates (8 September 1981, and 8 October 1981) she indicated, respectively, in "Declaration Oetken 1" and Declaration Oetken 2", and (v-4.2) because Mrs Oetken was not a neutral witness, since she had a professional relationship with Prof. Kaminsky, who himself had a personal interest in the outcome of the case, as reflected by his status as inventor of D8, a document of Hoechst AG (later Targor GmbH), which was Opponent I in the present case.

(v-5) Even if Mrs Oetken's willingness to hand out D6 was accepted, this did not make D6 prior art in the sense of Article 54(2) EPC,

(v-5.1) because, unless D6 was actually provided for inspection, such circumstances (the locked cupboard, supplying only on request) did not fulfil the requirement of <u>making</u> available and

(v-5.2) because such willingness to disclose involved an unlawful or even criminal behaviour which could not be treated as a lawful disclosure; in this respect the Patentee requested that the following questions be referred to the Enlarged Board of Appeal:

"1. If public access to a document can only result from criminal acts, and if there is no specific evidence in the case that any criminal act was in fact commited, can one make a finding of public access on the basis that a single person (here: the keeper of the document) years after the relevant time period states that she might have been prepared to commit the criminal act?

2. If the keeper of an unpublished document does not have the right to communicate its content to a third party without consent of another (e.g. author or

copyright owner), is the document nevertheless made available to the public if the keeper is willing to communicate its content at the request of a third party without that consent?

3. If the answer to question 2 is "yes", when is the document made available to the public:
a) on receipt by the keeper?
b) at such time, after receipt, when the keeper becomes willing to communicate the content of the document in his or her keeping?
c) only when the contents of the document are communicated to a third party?"

(vi) The closest prior art was, in the Patentee's view, represented by documents D1, D2 or D8. Vis-à-vis this state of the art the subject-matter of Claim 1 of the only request (cf. point VIII supra) was non-obvious,

(vi-1) because there was no relevant prior art relating to those zirconocene catalyst compositions comprising bridged ($s = 1$) cyclopentadienyl radicals,

(vi-2) because the available prior art did not suggest that unbridged ($s = 0$) zirconocene catalyst compositions comprising two cyclopentadienyl radicals, which had at least one and up to four substituents R', would provide enhanced catalyst activities leading to polyolefins of higher molecular weight at conventional polymerisation temperatures, and

(vi-3) because there was also no hint in the prior art that such unbridged zirconocene catalyst compositions, whose cyclopentadienyl radicals had five substituents, could be used together with differently substituted zirconocenes for the preparation of reactor blends of copolymers of ethylene with á-olefins, because in this event their low comonomer insertion capabilities were useful for tailoring the density of the copolymer blends.

(vii) In the Patentee's opinion D5 was not an appropriate starting point for the assessment of inventive step, as it was concerned with the influence of a methyl substitution of the cyclopentadienyl radicals of titanocene catalysts on the stereoregulation of the propene insertion.

X. The arguments presented by the Opponents in their written submissions, as far as they are still relevant after the withdrawal by Opponent I of its opposition and by Opponent II of its appeal, may be summarized as follows:

(i) Claim 1 contravened Article 123(2) EPC, because it related to a selected sub-group of zirconium compounds that was not originally disclosed; moreover, there was no basis in the original application for the amendment of the term "dialkyl germanium or silicon**e**" [emphasis by the Board] into "dialkyl germanium or silicon".

(ii) The patent in suit lacked a disclosure which was enabling within the whole claimed scope.

(iii) D6 represented applicable prior art,

(iii-1) because the public was informed of its existence before the priority date of the patent in suit by the following two documents:

"logbook":

colour copy of the notebook ("Eingangsbuch") of Diplomarbeiten 1 to 1229 of the Chemistry Department Library of the University of Hamburg (Annex N of Mr Pietzcker's observations dated 14 December 1998 attached to the Patentee's submission dated 15 December 1998), and

"Jahresbericht":

annual report 1981 of the Department of Applied Chemistry in the Institute for Inorganic and Applied Chemistry of the University of Hamburg (filed with Opponent I's submission dated 15 January 1999),

(iii-2) because it was established by the testimony of both Mrs Oetken and Prof. Kaminsky that D6, although having been stored in the archive in a locked cupboard, had been available upon request to any member of the public,

(iii-3) because the public dissemination of D6 was not prohibited by the German Copyright Act, which did not cover Diplomarbeiten,

(iii-4) because, even if D6 was covered by the German Copyright Act, Mrs Oetken's willingness to hand out D6 to anyone asking for it could not be considered as a contravention of that Act, since by allowing his Diplomarbeit to be subject to the routine procedures of the Chemistry Department Library Mr Külper had tacitly consented to the said conduct of Mrs Oetken, and finally,

(iii-5) because, even on the assumption that Mrs Oetken's attitude was in contravention of the German Copyright Act, this had no impact on the fact that by her "willingness" D6 had indeed been made available.

(iv) For the assessment of inventive step the newly cited documents D9, D10, D11 and D12 should be taken into account (cf. point IX (iii) supra).

(v) The closest prior art was represented by D5 and/or D9, which both disclosed metallocene catalyst compositions comprising substituted cyclopentadienyl radicals.

(vi) The claimed subject-matter was obvious over D5, because the skilled person looking for metallocene catalysts having enhanced activity would have been prompted by the information in D8, according to which zirconocenes were more active than titanocenes, to replace titanium by zirconium in the otherwise identical catalyst compositions of D5.

(vii) Similarly, it did not involve an inventive step to use alumoxanes as cocatalysts in the zirconocene catalyst compositions according to D9, because it was known from several documents, including D4, D5, D6 and D8, that alumoxanes were better cocatalysts for metallocenes.

(viii) But even when starting from the zirconocene catalyst compositions according to D1, D2 or D8, which comprised unsubstituted cyclopentadienyl radicals, an inventive step could not be recognized, because the data reported in the Experimental Reports of Opponent I filed with its submission dated 20 April 1990 and with the Statement of Grounds of Appeal showed that the use of bridged or substituted cyclopentadienyl radicals did not lead to enhanced polymerisation activities and/or enhanced molecular weights of the thus prepared polyolefins.

(ix) Against this background the claimed catalyst system was obvious, because rather than establishing a prejudice against the use of metallocenes with alkyl substituted cyclopentadienyl radicals the skilled person looking for <u>further</u> similar catalyst systems would be prompted by the other citations on file (D3 to D7) to use such substituted zirconocenes.

XI. The Patentee requested that the decision under appeal be set aside and that the patent be maintained on the basis of Claims 1 to 3 as submitted during the oral proceedings, and further that documents D9 to D12 be disregarded.

Opponent II made no request.

Reasons for the Decision

1. The appeal of the Patentee is admissible.

In view of the withdrawal of Opponent II's appeal, there is no need to decide on its admissibility as originally requested by the Patentee.

2. *Representation by a legal practitioner (Article 134(7) and Rule 101 EPC)*

The Patentee was represented by the firm Uexküll & Stolberg, and particularly by its partner Mr Franck acting under General Authorisation Nr. 23259, which includes the right to grant sub-authorisations (cf. submission dated 18 June 1991 and Register of General Authorisations of the EPO).

In its submission dated 21 June 2001 Uexküll & Stolberg informed the EPO that the Patentee appointed the legal practitioner (Rechtsanwalt) Dr. Rolf Pietzcker as an additional representative for the opposition and appeal proceedings.

Upon the Board's observation at the oral proceedings that this statement was neither a proper authorization by the Patentee, because it was not signed by the Patentee company, nor was it by its very wording a proper sub-authorization by the Patentee's main representative Uexküll & Stolberg, Mr Franck requested that the said submission be considered as a sub-authorisation, which was accepted by the Board.

The Board is accordingly satisfied that the authorisation required for legal practitioners as set out in Article 2 of the "Decision of the President of the European Patent Office dated 19 July 1991 on the filing of authorisations" (cf. OJ EPO 1991, 489) is fulfilled.

3. *Article 123(2) and (3) EPC*

In the Board's judgment the multiple restrictions in Claim 1 (selection of zirconium only, restrictions of the meanings of R", p and proviso) as well as the proviso "excluding compounds wherein all of the cyclopentadienyl radicals are unsubstituted" are sufficiently supported by the general description of the original application and by the compounds which are specifically exemplified therein (cf. page 7, lines 1 to 11; page 8, lines 1 to 20; Examples).

The same applies to the amendment in the definition of the radical R" of the term "silicone", which designates compounds comprising -O-Si-O- bonds, to "silicon", which designates the chemical element Si. The latter amendment amounts to the correction of an error, which is apparent from the placing of this term next to the name of the chemical element "germanium" and the exemplified compounds <u>dimethylsilyl</u>dicyclopentadienyl zirconium dimethyl/chloride (original application, page 8, lines 15 to 17; Example 11).

By these restrictions the scope of Claim 1 is also narrower than that of its granted version.

The claims of the sole request thus meet the requirements of Article 123(2) and (3) EPC.

4. *Late filed documents (Article 114(2) EPC)*

In their Statements of Grounds of Appeal the Opponents referred for the first time to documents D9, D10, D11 and D12 (cf. points IX (iii) and X (iv) supra).

Neither of the Opponents gave any reasons for the late submission of these documents.

4.1 D9 opens a completely new line of argument insofar as it relates to a catalyst composition comprising zirconocenes having methyl-substituted cyclopentadienyl radicals for the polymerisation of ethylene, which differs from the compositions according to the patent in suit by the use of a different cocatalyst. D9 is devoid of any information concerning the impact of the methyl substitution on the catalyst's activity and/or molecular weight.

It is apparent to skilled persons that, in the years following the publication of D9 (publication date: 9 February 1960), scientific research in the field of low pressure polyolefins led to the development of Ziegler-Natta catalyst systems, comprising e.g. trialkyl aluminium and transition metal compounds, including titanium and zirconium compounds having organic ligands. From the publication history chart, based on a computer search in Chemical Abstracts, which was filed by the Patentee as Attachment II with its submission dated 15 May 1992, it is evident that the use of metallocenes having substituted ligands (e.g. cyclopentadienyl) was not an issue before the priority date (6 June 1983) of the patent in suit. Put in this historical context, the isolated publication in the year 1960 of a catalyst system comprising a zirconocene with methyl substituted cyclopentadienyl ligands cannot be regarded as a disclosure which the skilled person would *prima facie* consider to be of any merit for the further development of olefin polymerisation catalysts at the priority date of the patent in suit.

Accordingly, and in application of the principles set out in T 1002/92 (OJ EPO 1995, 605, particularly Reasons 3.4) the Board decided not to admit D9 for consideration in this appeal (Article 114(2) EPC).

In arriving at this decision the Board has considered that D9 was probably not new to the Patentee, because according to Opponent II's Statement of Grounds of Appeal (pages 5 and 6, point 3.2.3) its Australian counterpart (AU-A-220 436) was "one of the key documents in that litigation in which the proprietor is the plaintiff and Mobil is the defendant". This, however, neither changes the facts which led to the decision not to admit D9, nor does it discharge the Opponents from their duty to submit their case, including new documents, at the earliest possible date. It is noted in this respect that apparently not only the Patentee but also the Opponents kept abreast of the proceedings in the afore-mentioned US-case Exxon vs. Mobil and that they should therefore have been aware of the significance of D9 during the first instance proceedings (cf. points IX (iii) supra).

4.2 Document D10 bears a date of receipt ("Eingangsdatum") of 21 February 1983 and refers to a presentation at the "Hamburger Makromolekulares Symposium vom 4.-6. Oktober 1982". However, D10 does not carry a publication date and, according to the statement of Opponent II at the oral proceedings before the Board, it could not be established that D10 was published before 6 June 1983, the priority date of the patent in suit.

In this event, and because it cannot be taken for granted that the relevant content of D10 had actually been presented at the "Hamburger Makromolekulares Symposium" (the format of D10 is certainly not that of an oral presentation), this document is not considered as prior art according to Article 54(2) EPC. It is not, therefore, admitted for consideration in this appeal (Article 114(2) EPC).

4.3 Document D11 does not carry a printed date. Certain handwritten information thereon suggests 1982 as the year of publication, but another handwritten statement thereon reads "10-08-93" and yet a third handwritten statement, albeit struck out, reads "11-4-83".

It is, thus, unclear when D11 was actually published and, accordingly, it is not admitted for consideration in this appeal (Article 114(2) EPC).

4.4 Document D12 was published in 1987 and refers to "Proceedings of the ACS International Symposium of recent Advances in Polyolefins, held September 8-13, 1985, in Chicago, Illinois".

Since it was thus not published before the present priority date it is not admitted for consideration in this appeal (Article 114(2) EPC).

5. *Public availability of document D6 (Article 54(2) EPC)*

5.1 This document is the diploma thesis (hereinafter "Diplomarbeit") of Klaus Külper which he carried out from October 1980 to September 1981 at the Institute of Inorganic and Applied Chemistry of the University of Hamburg under the supervision of Prof. Dr. W. Kaminsky.

It is agreed by the parties that D6 was never supplied to any member of the public before 6 June 1983.

5.2 According to § 19 of the "Diplomprüfungsordnung" Diplomarbeiten have to be submitted in two copies to the examining authority ("Prüfungsausschuß") and pursuant to § 20 of this regulation they are to be assessed ("beurteilt") by the supervising professor and a second assessor.

5.4 It is undisputed by the parties that one copy of the Diplomarbeit was then sent to the Chemistry Department Library. On the basis of the available evidence, however, the exact dates of this delivery cannot be established:

5.4.1 According to the "Declaration Kaminsky 1" this happened a few days after he had sent it to the examining authority ("Prüfungsamt") on 24 September 1981.

5.4.2 According to the "Declaration Oetken 1" D6 was sent to the Chemistry Department Library of the University of Hamburg by the "Prüfungsamt" on 8 September 1981 and was generally available [in that library] from this point in time for interested readers.

5.4.3 According to the "Declaration Oetken 2" D6 was generally available in the Chemistry Department Library of the University of Hamburg from 8 October 1981 onwards.

5.4.4 Thus there are several different dates on which the copy of D6 is alleged to have been dispatched to and to have arrived in the Chemistry Department Library of the University of Hamburg, two of them in separate written declarations by the same person (Mrs Oetken).

Since it has not been disputed by the Patentee that D6 did in fact arrive in the archive of the Chemistry Department Library of the University of Hamburg before the relevant priority date, and in view of the fact that all the various dates in the corresponding declarations are before the end of 1981, the Board is prepared to take the view, on the balance of probabilities, that D6 arrived in the archive before the relevant priority date (6 June 1983) and indeed some time towards the end of the year 1981.

5.5 However, in the Board's judgment D6 did not by its mere arrival in the archive become publicly available, since that did not mean it was as of that point in time catalogued or otherwise prepared for the public to acquire knowledge of it, and because without such means of information the public would remain unaware of its existence.

This conclusion is in accordance with the "Decision Bundespatentgericht" (cf. point IX (iv) supra), which concerned the arrival of a dissertation in the archive of the Library of the Massachusetts Institute of Technology (MIT), in which it was found that this dissertation had not been made available to the public, even though it carried a date stamp showing its date of arrival in the archive, because it could not be assumed that it had been entered into the library catalogue within the period of time ending with the relevant priority date in that case.

5.6 In summary, the possibility that the public could acquire knowledge or awareness of the existence of D6 is a precondition of its public availability before the priority date of the patent in suit.

Two means existed, in the Opponents' view, for the public to acquire such knowledge or awareness, namely the "logbook" (point X (iii-1) supra) and the "Jahresbericht" (yearbook 1981, ibid).

5.6.1 The "logbook"

(i) As set out in the decision under appeal, this was a handwritten note book having the title "Diplomarbeiten", in which the diploma degree papers received in the archive were entered by the librarians. Under a serial number for each Diplomarbeit, the name of the graduate, the title of the work and the year in which it was submitted were entered into the "logbook". D6 was registered under the No. 877. Beside and below the original entry two notes made in a different handwriting from the original one indicate 8 October 1981 as the date of arrival of D6. On the page containing the serial Nos. 873 to 881 a date (28.1.1982) is noted on the top of the page (next to the serial No. 873). Five pages later, at the bottom of the page containing the entries Nos. 913 to 919, there is a note saying "1981 gez. bis 919" ("gez." being interpreted by the parties to mean "counted" (=<u>gez</u>ählt)).

(ii) These facts have to be seen, according to the decision under appeal, in the light of the evidence of Mrs Oetken, the Head Librarian of the Chemistry Department Library since 1967, ("Transcript Oetken"). According to her evidence, it usually took about three to four weeks from the time that a Diplomarbeit was submitted to the examining authority until it was actually received in the library ("Transcript Oetken", pages 304 and 321). The diploma degree papers were not always immediately registered after their arrival, but only some time later. At the end of 1981, the assistant librarian had a pile of diploma degree papers on his desk ("Transcript Oetken", page 305). After having been urged several times by Mrs Oetken, the assistant recorded these papers in the "logbook" at the end of January 1982 ("Transcript Oetken", page 305). The entry No. 877 was made at or near the date indicated at the top of the page, 28.1.1982 ("Transcript Oetken", page 303). In order to compile statistics for the year 1981, all the entries of the papers received in 1981 were counted by the assistant librarian and recounted by Mrs Oetken ("Transcript Oetken", page 306).

(iii) Whilst this was sufficient to convince the Opposition Division that entry No. 877 was made not later than January 1982 (Reasons for the Decision, page 5) the Board cannot share this view for the following reasons:

(iv) It is necessary to consider the fundamental nature of the evidence presented:

(iv-1) The "logbook" itself is not an official publication of the library. There has been no testimony to the effect that the "logbook" itself was laid out for inspection on the shelves of the publicly accessible part of the library. On the contrary, it is evident from the testimony of Mrs Oetken that it was located in the archive where an interested person would have to ask for it. Leaving aside the question of whether there was any barrier here to public accessibility (since this has not been alleged in relation to the "logbook" itself), it is clear that the "logbook" was an essentially internal document of the library staff. Its purpose was purely to maintain an inventory of the diploma degree papers. It was thus neither intended for nor, in its original form, capable of functioning as a device for establishing publication dates. Yet this is precisely the function which has been imposed on it by the Opponents' submissions. The "logbook" acquires its new and different character solely by virtue of the annotations referred to, since it has been alleged that these establish the date on which the relevant Diplomarbeit was entered in the "logbook" and therefore the date on which the public could gain knowledge of its existence.

(iv-2) Closer examination of the annotations, however, reveals the following:

(iv-2.1) According to Mrs Oetken, she counted the 1981 entries and the note "28.1.82" was written by the library assistant, as she would know from the handwriting, at the beginning of 1982 ("Transcript Oetken", page 331, lines 2 to 13). It was the appearance, after entry No. 919, of the note in different handwriting "1981 gez. bis 919" which clearly weighed with the Opposition Division in reaching its conclusion that entry No. 877 was made not later than January 1982. The handwriting of this latter note, however, as well as the visual appearance of the stroke of the pen of all the subsequent "year" notes is the same (but for the year "1983" on the top of the page starting with entry No. 995, but including the notes "1983" on top of the pages of the entry Nos. 972 to 994). It cannot, therefore, be concluded with certainty that the relevant entries had actually been made before

6 June 1983 and it cannot be ruled out that they had been added at a later time, when for one reason or another the time frame became relevant.

(iv-2.2) Furthermore, none of the entries on the aforementioned pages, except for the first one (No. 873) indicates a date of entry, nor do these entries, especially the D6 entry No. 877, comprise ditto marks, which could confirm that the entry date of 28 January 1982 for No. 873, was intended to apply also to the subsequent entries (as is the case e.g. for the entries 860 to 872).

(iv-2.3) For determining the entry date one can also not rely with certainty on the year in which a Diplomarbeit was finished, which is regularly indicated together with the designation of the subject-matter concerned, because a chronological order is not strictly applied: e.g. the entries Nos. 898, 914 and 919, which relate, respectively, to Diplomarbeiten from 1980, 1978 and 1978, are to be found among the 1981 Diplomarbeiten and similarly the entries Nos. 927 and 930, which both relate to Diplomarbeiten from 1981 are to be found among the 1982 Diplomarbeiten.

(iv-2.4) The notes "1981 gez. bis 919" and "28.1.1982" cannot, therefore, establish that the entries up to 919 and including No. 877 had been made at the beginning of the year 1982 or even before 6 June 1983.

(iv-2.5) Furthermore, the handwriting and the visual appearance of the stroke of the pen do not essentially change from entry No. 873 to entry No. 990, the latter entry concerning a Diplomarbeit from 1983, whose entry into the "logbook" cannot have been made before sometime during the year 1983 at the earliest. This also casts doubt on the reliability of Mrs Oetken's recollection of the time of the entry of D6 into the "logbook".

(iv-2.6) This doubt is reinforced by Mrs Oetken's admission that the additional date information added to entry No. 877 in connection with a further, differently handwritten entry No. 877a, had been made later from her personal memory ("Transcript Oetken", page 320, lines 13 to 16).

(iv-2.7) Moreover, as was set out on page 23, second paragraph of the "US court opinion", the "logbook" contains further date entries for the Nos. 876, 979 892 and 895, which apparently had been added at the same time and by the same person as those of entry No. 877a.

(iv-3) Thus, it is apparent, firstly, that the annotations, far from being contemporary with the entries themselves, are later additions and secondly, far from being systematically made throughout the "logbook", they are idiosyncratically concentrated around the entry of interest in the present case, namely entry No. 877, corresponding to D6. Furthermore, their content corresponds to the substance of one of the declarations of Mrs Oetken herself ("Declaration Oetken 2").

Since, furthermore, the annotations were all evidently made either by Mrs Oetken herself or under her direct supervision, the evidence of the "logbook", insofar as it relates to a date on which a particular entry was actually made, must be regarded as *ex post facto* re-construction originating from the witness herself.

The Board, which has only been presented with extracts and "snippets" from the proceedings before the US court, has not had the opportunity to form the kind of direct impression of this evidence which led that court, in its "US court opinion" to the conclusion,

"In this Court's view these dates appear to have been added at the same time and by the same person that added the date October 8, 1981, for the Külper Diplomarbeit.

It is apparent that these diplomarbeiten are also significant in the polymer science area and that the dates were added for improper motives, perhaps to support a basis for scientific or legal arguments such as those made in this case."

Nevertheless, it is clear that such evidence cannot be regarded, in an objective sense, as having a probative value greater than that of the corresponding declarations themselves.

The latter have, however, been vigorously contested by the Patentee, the sole remaining Appellant in the present proceedings, which urged the objective unsuitability of the documents presented as a means of discharging the Opponent's burden of proof, especially in view of the gulf of time separating the events testified to from the declarations themselves, which were in any case mutually inconsistent, and the lack of independence of Mrs Oetken as a witness (sections IX v-4.1 and 4.2 supra).

In the absence of further submissions or evidence to refute these arguments, the Board can come to no other conclusion than that the evidence of the annotated "logbook" is unsafe. In particular, it cannot be regarded as having sufficient probative value to discharge the Opponents' onus of proof to establish the date of availability to the public of D6 through its entry in the "logbook" with the serial No. 877.

5.6.2 "Jahresbericht"

(i) It was contended by the Opponents that the "Jahresbericht 1981", which contained a reference to D6, was made available to the public before 6 June 1983 by its display on a shelf of the library and/or by its distribution to guests, etc. of Prof. Kaminsky's institute. The copy of the "Jahresbericht" which was filed by Opponent I does not indicate a date of publication; the poorly legible stamp it carries on page 1 comprises the word "chemistry department" ("Fachbereich Chemie").

(ii) The Opponents relied on the following evidence:

(ii-1) According to the "Transcripts Kaminsky 2 and 2*" (page 53, line 10 to page 54, line 1) the preparation of annual reports of the Chemistry Department of the University of Hamburg started in 1978 and, according to paragraph 32 of the "Declaration Kaminsky 2" it was regular practice to provide guests, research sponsors and guest lecturers with copies of these annual reports.

(ii-2) While Prof. Kaminsky stated in paragraph 30 of the afore-mentioned declaration that the "Jahresbericht 1981" was prepared by himself, he testified before the US court that it was indeed Prof. Zachmann, the director of the institute [Chemistry Department], who was responsible for its preparation and that he contributed only "the part about the metallocene work and about my research group" (page 54, line 16 to page 55, line 4 and page 71, line 24 to page 72, line 2 of the said transcripts).

(ii-3) On page 72, lines 3 to 24 of said transcripts Prof. Kaminsky stated that, in his memory, the "Jahresbericht 1981" was published in March 1982, because a time span of three months from the end of the reporting year was regular practice ("Because we have always three months back the publication of such a report in that time").

(ii-4) On page 73, lines 1 to 13 of said transcripts Prof. Kaminsky admitted, however, that the Jahresbericht 1996 had not yet been made available "a year and a half later" [i.e. at the time of the US court deposition on 31 July 1998] and explained that this was an exceptional situation due to the death of Prof. Zachmann.

(ii-5) Mrs Oetken stated that from "around 1980 or a little bit earlier" the year-end reports prepared by Prof. Kaminsky and sent to her by him were then put on a special shelf in the reading room of the Chemistry Library where such reports were kept. However, she could not remember putting the "Jahresbericht 1981" on that shelf ("Transcript Oetken", page 310, line 13 to page 311, line 25).

(iii) For the following reasons the above evidence is not able to support the Opponents' contention that the public could acquire knowledge of the existence of D6 by the "Jahresbericht" before the relevant date (6 June 1983):

(iii-1) Both Mrs Oetken and Prof. Kaminsky rely in their statements on the regular practice of the Department. However, such practice only began in 1978, and could not be confirmed to exist in the years 1995 and 1996. It cannot, therefore, be considered as solidly established, or, thus, as being a reliable basis for interpolation of facts, for which no specific confirmation exists.

(iii-2) In this connection, no such specific confirmation exists for the dates on which the "Jahresbericht" was completed, printed and put on the special library shelf. Prof. Kaminsky and Mrs Oetken, the only witnesses produced by the Opponents, rely on little more than speculation: Prof. Kaminsky only refers to the alleged regular practice and Mrs Oetken even admitted that she had no concrete recollection of putting the "Jahresbericht" on the shelf (cf. sub-points ii-3 and ii-5 supra).

(iv) In the Board's judgment, therefore, it has not been established, on the balance of probabilities, that the "Jahresbericht" in question was made available to the public before 6 June 1983.

5.7 Since, thus, the only means referred to by the Opponents to underpin their contention of the possibility of the public being aware of the existence of D6, namely the "logbook" and the

"Jahresbericht", are, on the balance of probabilities, not convincing, and since such a possibility is a precondition for the public availability of D6, it cannot be decided otherwise than that this document is not part of the state of the art as defined in Article 54(2) EPC.

The Board notes, with regard to the completeness of the Opponents' evidence concerning the facts surrounding both the entry of D6 into the "logbook" and the publication of the "Jahresbericht", that in both cases they failed to corroborate their various contentions by the production of witnesses who inspected the "logbook" (e.g. students in preparation of their theses or Mr Külper, the author of D6 himself) and/or who read or received a copy of the "Jahresbericht" (guests, sponsors, lecturers), although one would expect that such witnesses would exist and could be traced.

5.8 In view of this conclusion there is no need to deal with the Opponents' contentions concerning the possibility of the public obtaining access to the copy of D6, which was locked up in the cupboard, and/or the lawfulness of Mrs Oetken's alleged willingness to hand out this copy to anyone who asked for it. Consequently, there is also no need to consider further the Patentee's requests for questions to be referred to the Enlarged Board of Appeal.

6. *Citations*

6.1 Document D1

Claim 1 of this document relates to a process for the preparation of homo- or copolymers of ethylene and propylene in the presence of a Ziegler catalyst system, which is free of halogen and is constituted by a cyclopentadienyl compound, which comprises a transition metal, preferably zirconium, and an alumoxane. According to Claim 2 bis(cyclopentadienyl) zirconium methyl may be used.

It is stated in D1 that the use of alumoxane provides better productivities (expressed by weight ratio of yield to amount of transition metal) than the known use of aluminum trialkyl/water (page 2, first paragraph; page 4, last paragraph; page 9, Example 2).

Furthermore, D1 discloses that polymers with higher molecular weights may be obtained at relatively low reaction temperatures than at higher temperatures (paragraph bridging pages 5 and 6; page 13, Table II).

6.2 Document D2

Claim 1 of this document relates to a process for the preparation of polyolefins in the presence of a catalyst system comprising (a) a compound of the formula (cyclopentadienyl)$_2$MeRHal, wherein R is cyclopentadienyl, a C_1-C_6 alkyl radical or halogen, Me is titanium or zirconium and Hal is halogen, and (b) certain alumoxanes.

Examples 1 and 2 (manuscript page 7) illustrate the temperature dependence of the polymerisation of ethylene in the presence of (cyclopentadienyl)$_2$ZrCl$_2$

and methylalumoxane: the viscosity average molecular weight of the resulting polyethylene is 91 000 at 90°C (Example 1), but 1 000 000 at 20°C (Example 2).

6.3 Document D3

This paper relates *inter alia* to the polymerisation of ethylene in the presence of a catalyst system comprising (cyclopentadienyl)$_2$TiCl$_2$ and $(C_2H_5)_2AlCl$ and studies the effects of methyl and ethyl substitution of the cyclopentadienyl radicals with respect to aging and molecular weight distribution (cf. page 107, summary).

Figure 6 on page 111 compares the molecular weight distributions obtained in the presence of a titanocene whose two cyclopentadienyl radicals are unsubstituted (Figure 6a) and of a titanocene whose two cyclopentadienyl radicals are each mono-methyl substituted (Figure 6b). These graphs are commented on at page 112, lines 3 to 5: "In Fig. 6b, however, the low molecular weight oligomers are clearly more emphasized than in Fig. 6a as is to be expected for the lower initiation rate."

6.4 Document D4

This paper relates to halogen-free Ziegler catalysts comprising bis(cyclopentadienyl) titanium dimethyl and methylalumoxane (cf. Summary).

On page 468 (penultimate paragraph, first sentence) D4 sets out: "When the Cp-ligand of the $Cp_2Ti(CH_3)_2$ is exchanged for the larger CH_3-Cp-ligand, activity decreases, retaining the same tacticity of the produced polypropylene."

6.5 Document D5

Section 5.5 of this dissertation is entitled "Variation of the steric environment of Ti by using $(CpCH_3)_2Ti(CH_3)_2$ instead of $Cp_2Ti(CH_3)_2$"[translation from German].

This variation of the cyclopentadienyl ligand of the titanocene/methylalumoxane catalyst system was carried out in order to verify the assumption that the methyl substitution would lead to a higher stereoregularity of the propylene insertion (page 64, lines 6 to 12 from bottom).

However, while this expected effect was not observed, it was found that the methyl substituted catalyst was less active than the unsubstituted one by a factor of 5 to 10 in the case of the polymerisation of ethylene, by four powers of 10 in the case of the polymerisation of propylene, and considerably hindered the propene insertion (page 68, lines 1 to 10).

6.6 Document D7 (translation from Italian)

This paper is entitled "Polymerisation of Mono- Di-Olefins Catalysed by Titanium-Indenyl Derivatives Together with Metal-Alkyls."

According to the first full paragraph below the formula on page 1, aluminium trialkyls may be used as metal-alkyls.

The first six lines of the "Conclusions" on page 9 read: "The bisindenyl titanium halides together with appropriate organometallic derivatives constitute a class of soluble catalysts suitable for the polymerisation of ethylene and conjugated diolefins.

For the first of these monomers, the catalytic activity is slightly less than shown using analogous cyclopentadienyl derivatives of titanium and is not sensitively influenced by the nature of the halogen bonded to the titanium."

6.7 Document D8

This document claims the priority of D2, but contains *inter alia* the following additional information [translation from German]:

"It is also surprising that when using bis(cyclopentadienyl) zirconium halogenide ... a significantly better activity was found than when using the analogous titanium compound. At the polymerisation temperature of 70°C, which is important in practice, at which bis(cyclopentadienyl) titanium compounds already decompose and consequently cannot be used, activities are achieved according to the inventive method using zirconium compounds, which are higher by a factor of 10 and more than those which may be obtained with the corresponding titanium compounds as catalyst component at a temperature of 20°C. But also at the same or a comparably low polymerisation temperature the activity of the zirconium catalysts of the invention is higher than that of catalysts, which comprise the corresponding titanium compound as heavy metal component." (page 4, line 31 to page 5, line 12).

"For the high activity, which is achieved with the method according to the invention, is important, apart from the choice of the heavy metal component, also the use of certain alumoxanes as cocatalyst." (page 5, lines 21 to 24)

6.8 Document B2

This document discloses on page 2498, third paragraph that methylene(biscyclopentadienyl)titanium dichloride can replace biscyclopentadienyl titanium dichloride as component of a Ziegler catalyst together with diethylaluminum chloride in the polymerisation of ethylene.

7. *Novelty*

The novelty of the subject-matter of Claim 1 was only contested with respect to document D6, which has been held by the Board not to belong to the state of the art according to Article 54(2) EPC (point 5.7 supra).

None of the remaining documents in the proceedings discloses all features of present Claim 1 and the Board is, therefore, satisfied that its subject-matter is novel.

8. *Closest state of the art*

In the Board's judgement the closest prior art is represented by the catalyst systems which are disclosed in D1, D2 and D8, because these systems differ from those according to present Claim 1 only by the missing substitution of the cyclopentadienyl radicals of the zirconocene component and because D1 was chosen as starting point of the alleged invention by the Patentee itself (cf. patent in suit: page 2, lines 26 to 52, Examples 1 and 9; original application: page 2, line 12 to page 3, line 18; Examples 1 and 10).

The Opponents' opinion that the titanocene/alumoxane catalyst systems according to D5, which comprise methyl substituted cyclopentadienyl radicals, would be a more appropriate starting point for the assessment of inventive step of the claimed subject-matter is, therefore, at variance with the facts. Rather, the skilled person looking for effective catalyst systems for the polymerisation of olefins would be dissuaded from using titanocenes having methyl substituted cyclopentadienyl radicals because these can neither provide the expected stereoregulation of the propene insertion nor are they as active as the unsubstituted metallocene catalyst systems (cf. point 6.5 supra).

9. *Problem and solution*

9.1 According to the patent in suit (page 2, lines 49 to 52; page 3, lines 13 to 18 of the original application) the alleged invention is concerned with the provision of homogeneous catalysts which can be usefully employed to produce high molecular weight polymer products at conventional polymerisation temperatures and which are able to control molecular weight and density of the polymer product without resorting to temperature control or hydrogen.

9.2 According to Claim 1 this problem is to be solved by certain cyclopentadienyl radical comprising zirconocene/alumoxane catalyst systems, whose two cyclopentadienyl substituents are either bridged (s = 1) or are unbridged (s = 0) but substituted in a certain way, e.g. with alkyl groups.

9.3 The evidence produced by the Patentee demonstrates that not all aspects of the problem as set out in point 9.1 supra are equally solved for all embodiments of a catalyst system as covered by present Claim 1.

Whereas on the one hand embodiments relating to catalyst systems the cyclopentadienyl radicals of whose zirconocene component carry one to four substituents, when used in the homopolymerisation of ethylene, provide enhanced activities and enhanced molecular weights, on the other hand embodiments wherein the cyclopentadienyl radicals carry five substituents, when used in the copolymerisation of ethylene with other á-olefins, provide low activities and low molecular weights as well as low comonomer insertion. Furthermore, embodiments wherein the cyclopentadienyl radicals are bridged, when used in the copolymerisation of ethylene with other á-olefins, provide high comonomer insertion levels.

This is demonstrated as follows:

9.3.1 Tables I and II of the patent specification show that zirconocenes whose unbridged cyclopentadienyl radicals are monomethyl, monoethyl or mono-â-phenylpropyl substituted (Examples 2, 3, 4, 7, 8) provide enhanced molecular weights (Mw and Mn) as well as enhanced activities, as compared to catalyst systems comprising zirconocenes without any substitution of the cyclopentadienyl radicals (Examples 1 and 6).

The same effects are demonstrated in Table B of Attachment I of Patentee's submission dated 15 May 1992 (first filed during the examination stage with the submission dated 18 April 1986 as part of the Declaration of H.C. Welborn, re-submitted as Annex E2 with Patentee's submission dated 13 June 2000).

9.3.2 Examples 5 and 6 of the patent specification show that these effects are not obtained when zirconocenes having pentamethyl substituted cyclopentadienyl radicals are used.

Moreover, as illustrated by a comparison of Examples 9 and 11 of the patent specification, the use of the latter zirconocenes in the copolymerisation of ethylene with propene leads to very low propene insertion (Example 11: 3.6 mole% propene) as compared with the use of zirconocenes without substitution of the cyclopentadienyl radicals (Example 9: 31 mole% propene).

Similarly Tables C and D of Attachment I (cf. point 9.3.1 supra) show that, when used in the copolymerisation of ethylene with propene or 1-butene, zirconocenes having two pentamethyl substituted cyclopentadienyl radicals, provide low activities, low molecular weights and low comonomer insertion levels (as demonstrated by high R-values).

9.3.3 Table E of said Attachment I comprises results of the copolymerisation of ethylene and 1-butene for two catalysts having dimethylsilyl bridged cyclopentadienyl or tetrahydrocyclopentadienyl radicals. In both cases enhanced polymerisation activities and reduced R-values are reported (demonstrating a high comonomer insertion level) as compared with a catalyst system comprising a zirconocene having unsubstituted cyclopentadienyl radicals.

9.3.4 It is, thus, apparent that the three different embodiments covered by Claim 1, namely

- catalyst systems the zirconocene component of which comprises one- to tetrasubstituted cyclopentadienyl radicals (= "embodiment 1"),

- catalyst systems the zirconocene component of which comprises pentasubstituted cyclopentadienyl radicals ("embodiment 2"), and

- catalyst systems the zirconocene component of which comprises bridged cyclopentadienyl radicals ("embodiment 3")

do not belong to the same "single general inventive concept" (Article 82 EPC).

However, lack of unity is not an issue in opposition (or opposition appeal) proceedings (G 1/91, OJ EPO 1992, 253).

9.3.5 It is, however, a consequence of this conceptual lack of unity that different aspects of the problem set out in point 9.1 supra apply to said "embodiments 1, 2 and 3".

(i) "Embodiment 1" is, thus, concerned with the provision of homogeneous catalysts which can be usefully employed to produce high molecular weight homopolymer products at conventional polymerisation temperatures;

(ii) "Embodiment 2" is concerned with the provision of homogeneous catalysts which can be usefully employed in the copolymerisation of ethylene with other á-olefins and which are able to control molecular weight and density of the polymer product by low comonomer insertion levels, and

(iii) "Embodiment 3" is concerned with the provision of homogeneous catalysts which can be usefully employed in the copolymerisation of ethylene with other á-olefins and which are able to control molecular weight and density of the polymer product by high comonomer insertion levels.

9.3.6 It can be concluded from the experimental results referred to, respectively, in points 9.3.1 to 9.3.3 supra that the afore-mentioned partial problems (i), (ii) and (iii) are effectively solved, respectively, by "embodiments 1, 2 and 3".

The counter-experiments provided by Opponent I ("Versuchsbericht" attached to Opponent I's Statement of Grounds of Appeal), which are intended to deny the effective solution of partial problem (iii) by "embodiment 3", are not sufficient, in the Board's judgement, to question the Patentee's experimental results because, by selecting different reaction conditions and catalyst compositions (particularly

Al/Zr ratio), Opponent I failed to discharge its burden of proof. No valid conclusion may, therefore, be drawn from the different results reported for the zirconocene catalyst $Me_2Si(Ind)_2ZrCl_2$ in the opponent's "Versuchsbericht" and in Table E of the afore-mentioned Attachment I.

Concerning those counter-experiments which the Opponent filed with its submission dated 20 April 1990, these relate solely to the use of zirconocenes having bridged cyclopentadienyl radicals for the homopolymerisation of ethylene and do not allow any conclusion with respect to the efficiency of these catalysts for the copolymerisation of ethylene with á-olefins. They are, thus, irrelevant with regard to the afore-mentioned partial problem (iii).

With regard to those counter-experiments the Board notes that these have been presented by Opponent I with the double purpose of attacking the compliance of the claimed subject-matter with the requirements of Article 56 EPC and with Article 83 EPC (opposition ground Article 100(b) EPC). The latter objection is, however, without substance, since those counter-experiments do not challenge the enabling character of the patent specification, but only the degree of improvement which may be achieved (activity, molecular weight). Any possible objection under Article 100(b) EPC must, therefore, be rejected as unsubstantiated. In view of the outcome of this appeal this issue is, however, of no relevance.

9.3.7 In the Board's judgement it clearly results from the statements in D3, D4, D5 and D7 (cf. points 6.3, 6.4, 6.5 and 6.6 supra), which point to the reduced activities and/or molecular weights obtained by the use of metallocene catalysts having methyl or indenyl substituted cyclopentadienyl radicals, that the present solution ("embodiment 1") of the afore-mentioned partial problem (i) is non-obvious.

This conclusion is not invalidated by the fact that all these documents make use of titanocenes, not of zirconocenes, because Ti and Zr belong to the same group IVb of the Periodic Table and thus have homologous properties, any differences resulting mainly from the larger diameter of Zr. A person skilled in the art would, therefore, expect that the substitution of ligands of the metal atom, cyclopentadienyl radicals inclusive, would in principle have the same consequences for titanocenes and zirconocenes.

9.3.8 The same conclusion of non-obviousness applies to the solution of the afore-mentioned partial problem (iii) by "embodiment 3", because there is no prior art which would suggest that catalyst systems comprising an alumoxane component and a zirconocene component, which comprises bridged cyclopentadienyl radicals, would be able to provide ethylene copolymers having increased comonomer levels (cf. point 9.3.3 supra).

The methylene bridged titanocene Ziegler catalyst systems used according to B2 for the homopolymerisation of ethylene (cf. point 6.8 supra) differ from the catalyst systems according to "embodiment 3" by the use of a different transition metal (Ti in lieu of Zr) and of a different cocatalyst (diethylaluminium chloride in lieu of alumoxane) and are not disclosed to be useful in the copolymerisation of ethylene with á-olefins. Document B2 cannot thus provide the skilled person with any information with respect to the claimed solution of the afore-mentioned partial problem (iii).

9.3.9 By contrast, the solution of the afore-mentioned partial problem (ii) by "embodiment 2" does not, in the Board's judgement, involve an inventive step because on the one hand the skilled person was aware from documents D3, D4, D5 and D7 that the substitution of the cyclopentadienyl radicals of metallocenes with alkyl (e.g. methyl) groups was bound to reduce the catalyst's activity and its ability to promote the preparation of high molecular weights (cf. point 9.3.7 supra); and on the other hand the skilled person was aware from D5 (particularly page 68, lines 6 to 10) that the mono-methyl substitution of the cyclopentadienyl radicals of the compound $(cyclopentadienyl)_2 Ti(CH_3)_2$ hinders, in the course of its homopolymerisation, the insertion of propene.

Concerning the relevance of the transition metal used, Ti or Zr, the afore-mentioned considerations apply (cf. point 9.3.7 supra).

The skilled person would thus expect that the same substitution-related effects would, in the copolymerisation of ethylene and propene, lead to reduced levels of propene insertion, as is indeed the case for the catalyst component whose cyclopentadienyl radicals are pentamethyl substituted (cf. point 9.3.2 supra).

The subject-matter of "embodiment 2" does not, therefore, involve an inventive step, because this solution of the afore-mentioned partial problem (ii) is obvious over the cited prior art (D3, D4, D5 and D7).

This conclusion is not invalidated by the non-applicability of the analogous reasoning (i.e. expectation of inferior performance) to the one- to tetra-methyl substituted zirconocene species, which, in clear contrast, are more active and provide higher molecular weights than its unsubstituted counterparts (cf. point 9.3.7 supra), because this surprising performance does not belong to the state of the art. The argument that the skilled person would expect from the penta-methyl substituted species the same performance as that of the one- to tetra-methyl substituted species, with the consequence that the non-fulfilment of this expectation had to be considered as evidence for the non-obviousness of this "embodiment 2", must, therefore, fail, because it would rely on an *ex post facto* analysis.

The Patentee's alternative argument, namely that the non-obviousness of this "embodiment 2" could be justified by the possible use of such penta-substituted zirconocenes in combination with other zirconocenes for the preparation of so-called in-situ polymerised "reactor blends" of copolymers having quite different comonomer insertion levels must be disregarded, because this problem cannot be deduced from the application as a whole in the form originally filed, even when considered in the light of the available prior art. Therefore, the technical advantage allegedly afforded by the possibility to prepare "reactor blends" cannot be taken into account for the assessment of inventive step of "embodiment 2" (cf. T 184/82, OJ EPO 1984, 261; T 13/84, OJ EPO 1986, 253).

9.3.10 With respect to the requirements of Article 56 EPC, the inventiveness of the subject-matter of a claim must be denied as a whole in the event that only one of its embodiments is obvious. Since, in the present case, "embodiment 2" has been found to be obvious, it follows that Claim 1 does not meet the requirements of that Article.

9.3.11 Since the further Claims 2 and 3 belong to the same set as Claim 1, they must share the latter's fate.

9.3.12 The Patentee's sole request must, therefore, be denied altogether.

10. The Patentee's appeal must thus be rejected, with the consequence that the patent will be maintained in the amended version which was accepted by the appealed interlocutory decision of the Opposition Division.

Before that decision is executed, the Patentee may wish to request that the dependancy of Claim 3 be corrected under Rule 89 EPC (from "according to claim 3" to "according to claim 2"). In this respect the requirements of Rule 58 EPC are to be observed.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

E. Görgmaier R. J. Young