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**D E C I S I O N**  
**of 6 February 2001**

**Case Number:** T 0457/98 - 3.3.3

**Application Number:** 89909697.8

**Publication Number:** 0490888

**IPC:** C08F 10/00

**Language of the proceedings:** EN

**Title of invention:**

Catalyst and process for the polymerization of olefins

**Patentee:**

FINA RESEARCH S.A.

**Opponents:**

Exxon Chemical Patents Inc.  
TARGOR GmbH

**Headword:**

-

**Relevant legal provisions:**

EPC Art. 54, 83

**Keyword:**

"Disclosure - sufficiency (yes) - no undue burden"  
"Novelty (no) - prior specific disclosure"

**Decisions cited:**

G 0002/88

**Catchword:**

-



Case Number: T 0457/98 - 3.3.3

**D E C I S I O N**  
**of the Technical Board of Appeal 3.3.3**  
**of 6 February 2001**

**Appellant:** FINA RESEARCH S.A.  
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**Representative:**

**Respondent:** Exxon Chemical Patents Inc.  
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(Opponent 2) TARGOR GmbH  
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**Representative:** Lederer, Franz, Dr.  
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**Decision under appeal:** Decision of the Opposition Division of the  
European Patent Office posted 3 March 1998  
revoking European patent No. 0 490 888 pursuant  
to Article 102(1) EPC.

**Composition of the Board:**

**Chairman:** B. ter Laan  
**Members:** C. Idez  
V. Di Cerbo

## Summary of Facts and Submissions

- I. Mention of the grant of European patent No. 0 490 888 in respect of European patent application No. 89 909 697.8, filed on 8 September 1989 as international patent application No. PCT/BE89/00043, published under No. WO 91/03500, was published on 18 May 1994 (Bulletin 94/20) on the basis of seventeen claims, Claim 1 reading:

"Use of an alumoxane solution containing at least 20 wt % of alumoxane as cocatalyst for the polymerization or copolymerization of olefins with a homogeneous metallocene-type catalyst of general formula  $(Cp)_mR_nMQ_k$  wherein each Cp is the same or different and is a cyclopentadienyl or a cyclopentadienyl substituted by one or more hydrocarbyl radical such as alkyl, alkenyl, aryl, arylalkyl or alkylaryl radical having from 1 to 20 carbon atoms, R is a structural bridge between two Cp rings, M is a transition metal selected from groups 4 or 5, each Q is the same or different and is a hydride or a hydrocarbyl group having from 1 to 20 carbon atoms or an alkenyl group having from 2 to 20 carbon atoms or a halogen,  $m=1-3$ ,  $n=0$  or 1, and k is such that the sum of  $m+k$  is equal to the oxidation state of M."

Claims 2 to 4 referred to preferred embodiments of the use according to Claim 1.

Claim 5 read:

"Homogeneous catalyst system for olefin polymerization or copolymerization, said system comprising (i) a metallocene-type catalyst of general formula  $(Cp)_mR_nMQ_k$  wherein each Cp is the same or different and is a

cyclopentadienyl or a cyclopentadienyl substituted by one or more hydrocarbyl radical such as alkyl, alkenyl, aryl, arylalkyl or alkylaryl radical having from 1 to 20 carbon atoms, R is a structural bridge between two Cp rings, M is a transition metal selected from groups 4 or 5, each Q is the same or different and is a hydride or a hydrocarbyl group having from 1 to 20 carbon atoms or an alkenyl group having from 2 to 20 carbon atoms or a halogen,  $m=1-3$ ,  $n=0$  or 1, and k is such that the sum of  $m+k$  is equal to the oxidation state of M, and (ii) an alumoxane cocatalyst, said alumoxane being provided under the form of a solution of at least 20 wt%."

Claims 6 to 8 were directed to preferred embodiments of the catalyst system according to Claim 5.

Claim 9 read:

"Process for producing a homogeneous catalyst system for olefin polymerization and copolymerization, said system comprising (i) a metallocene-type catalyst of general formula  $(Cp)_mR_nMQ_k$  wherein each Cp is the same or different and is a cyclopentadienyl or a cyclopentadienyl substituted by one or more hydrocarbyl radical such as alkyl, alkenyl, aryl, arylalkyl or alkylaryl radical having from 1 to 20 carbon atoms, R is a structural bridge between two Cp rings, M is a transition metal selected from groups 4 or 5, each Q is the same or different and is a hydride or a hydrocarbyl group having from 1 to 20 carbon atoms or an alkenyl group having from 2 to 20 carbon atoms or a halogen,  $m=1-3$ ,  $n=0$  or 1, and k is such that the sum of  $m+k$  is equal to the oxidation state of M, and (ii) an alumoxane cocatalyst, characterized in that said alumoxane is provided under the form of a solution of

at least 20 wt%."

Claim 10 to 12 referred to preferred embodiments of the process according to Claim 9.

Claim 13 read:

"Process for the polymerization or copolymerization of olefins in the presence of a catalyst system comprising (i) a metallocene-type catalyst of general formula  $(Cp)_mR_nMQ_k$  wherein each Cp is the same or different and is a cyclopentadienyl or a cyclopentadienyl substituted by one or more hydrocarbyl radical such as alkyl, alkenyl, aryl, arylalkyl or alkylaryl radical having from 1 to 20 carbon atoms, R is a structural bridge between two Cp rings, M is a transition metal selected from groups 4 or 5, each Q is the same or different and is a hydride or a hydrocarbyl group having from 1 to 20 carbon atoms or an alkenyl group having from 2 to 20 carbon atoms or a halogen,  $m=1-3$ ,  $n=0$  or 1, and k is such that the sum of  $m+k$  is equal to the oxidation state of M, and (ii) an alumoxane cocatalyst, characterized in that said alumoxane is provided under the form of a solution of at least 20 wt%."

Claims 14 to 17 were directed to preferred embodiments of the process according to Claim 13.

- II. On 10 February 1995 and 14 February 1995 two Notices of Opposition against the granted patent were filed, in which the revocation of the patent in its entirety was requested on the grounds set out in Articles 100(a) EPC (both Opponents) and 100(b) EPC (Opponent II).

The oppositions were, *inter alia*, supported by the following document:

D1 EP-A-0 302 244

III. By a decision taken on 4 February 1998 and issued in writing on 3 March 1998 the Opposition Division revoked the patent. That decision was based on the set of claims as filed; Claim 13 was however amended by the Appellant (see the letters dated 9 January 1997 and 21 January 1998), the amendment being the insertion of the word "homogeneous" in the first sentence, between "...in the presence of a" and "catalyst system comprising...".

The Opposition Division held that the requirements of Article 83 were complied with, but that the claimed subject-matter lacked novelty (Article 54 EPC).

- (a) In view of the worked examples of the patent in suit and since the skilled person would, in the word "catalyst", read only those compounds that actually functioned as such, the requirements of Article 83 EPC were fulfilled.
- (b) Regarding novelty, in the examples of D1 all the features of the independent claims were present except for the concentration of the aluminoxane solution, which was less than 20 wt.%. However, in the description of D1 the use of aluminoxane solutions having a concentration of 30% or even of saturated solutions was described. D1 described a two-step process for the preparation of the catalyst, but the wording of the present claims did not exclude such a two-step process, so that the claimed subject-matter was not novel.

IV. On 6 May 1998 the Appellant (Proprietor) lodged an

appeal against the above decision and paid the prescribed fee simultaneously. With the Statement of Grounds of the Appeal, which was filed on 2 July 1998, the Appellant stated that the claimed subject-matter was novel.

At the oral proceedings held on 6 February 2001, only the Appellant was present. During the discussion he replaced the claims on file with two sets of 13 and 9 claims, respectively, as the main and an auxiliary request. Claim 1 of the main request read:

"Homogeneous catalyst system for olefin polymerization or copolymerization, said system consisting of (i) a metallocene-type catalyst of general formula  $(Cp)_mR_nMQ_k$  wherein each Cp is the same or different and is a cyclopentadienyl or a cyclopentadienyl substituted by one or more hydrocarbyl radical such as alkyl, alkenyl, aryl, arylalkyl or alkylaryl radical having from 1 to 20 carbon atoms, R is a structural bridge between two Cp rings, M is a transition metal selected from groups 4 or 5, each Q is the same or different and is a hydride or a hydrocarbyl group having from 1 to 20 carbon atoms or an alkenyl group having from 2 to 20 carbon atoms or a halogen,  $m=1-3$ ,  $n=0$  or 1, and k is such that the sum of  $m+k$  is equal to the oxidation state of M, and (ii) an alumoxane cocatalyst, said alumoxane being provided under the form of a solution of at least 20 wt% in benzene, toluene or xylene."

Claims 2 to 4 were directed to preferred embodiments of the catalyst system according to Claim 1.

Claim 5 read:

"Process for producing a homogeneous catalyst system

for olefin polymerization and copolymerization, said system consisting of (i) a metallocene-type catalyst of general formula  $(Cp)_mR_nMQ_k$  wherein each Cp is the same or different and is a cyclopentadienyl or a cyclopentadienyl substituted by one or more hydrocarbyl radical such as alkyl, alkenyl, aryl, arylalkyl or alkylaryl radical having from 1 to 20 carbon atoms, R is a structural bridge between two Cp rings, M is a transition metal selected from groups 4 or 5, each Q is the same or different and is a hydride or a hydrocarbyl group having from 1 to 20 carbon atoms or an alkenyl group having from 2 to 20 carbon atoms or a halogen,  $m=1-3$ ,  $n=0$  or 1, and k is such that the sum of  $m+k$  is equal to the oxidation state of M, and (ii) an alumoxane cocatalyst, characterized in that said alumoxane is provided under the form of a solution of at least 20 wt% in benzene, toluene or xylene."

Claims 6 to 8 referred to preferred embodiments of the process according to Claim 5.

Claim 9 read:

"Process for the polymerization or copolymerization of olefins in the presence of a homogeneous catalyst system consisting of (i) a metallocene-type catalyst of general formula  $(Cp)_mR_nMQ_k$  wherein each Cp is the same or different and is a cyclopentadienyl or a cyclopentadienyl substituted by one or more hydrocarbyl radical such as alkyl, alkenyl, aryl, arylalkyl or alkylaryl radical having from 1 to 20 carbon atoms, R is a structural bridge between two Cp rings, M is a transition metal selected from groups 4 or 5, each Q is the same or different and is a hydride or a hydrocarbyl group having from 1 to 20 carbon atoms or an alkenyl group having from 2 to 20 carbon atoms or a halogen,



m=1-3, n=0 or 1, and k is such that the sum of m+k is equal to the oxidation state of M, and (ii) an alumoxane cocatalyst, characterized in that said alumoxane is provided under the form of a solution of at least 20 wt% in benzene, toluene or xylene."

Claims 10 to 13 were directed to preferred embodiments of the process according to Claim 9.

The claims of the auxiliary request, with appropriate renumbering, correspond to Claims 5 to 13 of the main request, so that the two independent Claims 1 and 5 of the auxiliary request refer to a process for producing a homogeneous catalyst system as defined in Claim 5 of the main request, and a process for the polymerization or copolymerization of olefins in the presence of a homogeneous catalyst system as defined in Claim 9 of the main request.

The arguments of the Appellant, given in writing and during the oral proceedings, can be summarized as follows:

D1 disclosed a two-step process for the (co)polymerisation of olefins, i.e. a preactivation and the polymerisation reaction, the first step being carried out with an aluminoxane solution the concentration of which could range from 1% to saturation and was preferred to have a concentration of 5 to 30 wt%, the second step requiring an aluminoxane activator the concentration of which was not disclosed. In all the examples the aluminoxane was introduced in two steps, in a solution having a concentration of between 5 and 10 wt%. The solution used in the second step was not necessarily the same as that used in the

first step, so that there was no disclosure that all the aluminoxane was provided under the highly concentrated form required by the patent in suit. Any dilution by reactor contents should not be taken into account in view of the restriction that the solution must be in benzene, toluene or xylene. Therefore, the claimed catalyst system differed from the one described in D1 in the lower amount of solvent. Also, D1 was not limited to homogeneous catalysts.

V. Respondent 1 did not reply to the Statement of Grounds of Appeal, whereas Respondent 2 argued in writing along the following lines:

- (a) Regarding Article 83 EPC, the definition of the metallocene catalyst included many compounds that did not have any catalytic activity and hence could not possibly serve as catalysts. It was not feasible for the skilled person to try out all the possibilities encompassed by the patent in suit and see if they possessed any catalytic activity.
- (b) As regards novelty, D1, Example 1, disclosed an aluminoxane solution as a cocatalyst for the polymerisation of an olefin. That solution could, according to the description, be saturated or have a preferred concentration of 5 to 30 wt%. Since it was customary practice at the EPO to combine the disclosure of an example with a specifically mentioned limit of a range, D1 disclosed all the features of the claimed subject-matter. Furthermore, the patent in suit was not restricted to a one-step process.

VI. The Appellant requested that the decision under appeal

be set aside and the patent be maintained on the basis of the main request or, alternatively, on the basis of the auxiliary request, both filed during oral proceedings.

Respondent 2 requested that the appeal be dismissed.

## Reasons for the Decision

I. The appeal is admissible.

### *Main request*

### *The wording of the claims*

2. Claims 6 to 19 as originally filed form the basis of the present main request, the claims being appropriately renumbered.

2.1 Claims 1, 5 and 9 differ from Claim 6, 10 and 14 as originally filed, respectively, in that

(a) the catalyst system now **consists of** a metallocene and an aluminoxane solution and

(b) the aluminoxane solution is now defined as being in benzene, toluene or xylene.

Claim 9 additionally differs from original Claim 14 in that the catalyst system is restricted to being homogeneous.

2.1.1 The meaning of the word "comprising" is generally

interpreted as encompassing all the specifically mentioned features as well optional, additional, unspecified ones, whereas the term "consisting of" only includes those features as specified in the claim. Therefore, "comprising" includes the term "consisting of", so that the amendment from the former into the latter term does not extend beyond the content of the application as originally filed.

2.1.2 The solvent of the aluminoxane solution is indicated on page 3, line 4, where benzene, toluene and xylene are mentioned as suitable organic solvents for the aluminium trialkyl from which the aluminoxane is prepared. In all examples toluene is used as the solvent for the aluminoxane.

2.1.3 The introduction of the word "homogeneous" in Claim 9 is in conformity with the wording of the other independent claims as well as the original description, page 1, second and third paragraph.

2.2 For the above-indicated reasons, the claims of the main request do not contravene the requirements of Article 123(2) EPC.

3. Since, as can be seen from point 2.1 above, the amendments all amount to restrictions, the scope of protection is not extended, so that Article 123(3) EPC is also complied with.

4. The amendments to the claims do not introduce any unclarities. Accordingly, the requirements of Article 84 EPC are complied with.

*Sufficiency of disclosure*

5. Catalyst systems based on metallocene compounds had been known for quite some time at the priority date of the patent in suit. Therefore, at that date, the skilled person would have had some knowledge about those compounds and their catalytic properties.

In the patent specification reference is made to a number of patent applications using metallocenes as catalyst components (page 3, lines 17, 23 and 29; page 3, line 29). Also, the metallocenes suitable for the present polymerisation are described more specifically on page 4, lines 6 to 21, and in the examples. Hence there can be no doubt that the skilled person would not need to try out all the possibilities encompassed by the patent in suit and see if they possessed any catalytic activity, in order to prepare the products and carry out the processes defined in the claims of the patent in suit.

Therefore, the requirements of Article 83 are fulfilled.

### *Novelty*

6. The only document cited against novelty was D1.
  - 6.1 D1 describes a process for the preparation of a 1-olefin polymer by polymerisation of a 1-olefin having the formula  $R-CH=CH_2$ , wherein R is an alkyl group with 1 to 28 C-atoms, at a pressure of from 0.5 to 60 bar, in solution, in suspension or in the gas phase, in the presence of a catalyst which consists of a metallocene as a transition metal compound and

an aluminoxane as an activator, characterised in that the polymerisation is carried out in the presence of a catalyst the transition metal component of which, before polymerisation, is preactivated with an aluminoxane of a specific linear type and/or one of a specific cyclic type, at a temperature of -78 to 100°C, during a period of 5 minutes to 60 hours, and in which the activator is also an aluminoxane of one of the specified formulae (Claim 1).

The preactivation is carried out in solution, preferably such that the metallocene is solved in a solution of the aluminoxane in an inert hydrocarbon, in particular toluene (page 6, lines 38 to 41). The concentration of the aluminoxane is from 1 wt% to saturation, preferably from 5 to 30 wt% (page 6, lines 42 and 43). The second component of the catalyst, the activator, is an aluminoxane of one of the formulae specified for the preactivation. Preferably, the same aluminoxane is used as in the preactivation step (page 6, lines 55 to 57). Although that passage does not refer to the concentration of the aluminoxane in the second step, in all the examples the same solution is used in both the preactivation step as well as for the activator.

In particular, in Example 1, 9 mg (0.02 mmol) bisindenyl dimethylsilyl zirconium dichloride is solved in a 25 cm<sup>3</sup> solution of methyl aluminoxane (20 mmol Al) in toluene and preactivated by having it stand for 15 minutes. This orange-red solution is then added to the reactor in which liquid propylene and 50 cm<sup>3</sup> of a methyl aluminoxane solution (40 mmol Al) already are present and have been stirred for 15 minutes at 30°C before the metallocene containing

catalyst component is added. The other examples describe basically the same process. In the comparative examples several other embodiments are compared to the worked examples.

- 6.2 Claim 1 of the main request refers to a homogeneous catalyst system, said system consisting of a metallocene component and an aluminoxane component, the latter being provided in a concentration of at least 20 wt% in benzene, toluene or xylene.

Claim 5 of the main request refers to a process for producing a homogeneous catalyst system, said system consisting of a metallocene component and an aluminoxane component, the latter being provided in a concentration of at least 20 wt% in benzene, toluene or xylene.

- 6.2.1 By amending the term "comprising" into "consisting of" and adding the requirement that the aluminoxane solution should be in benzene, toluene or xylene, according to the Appellant, the claimed subject-matter was redefined in the sense that now all the aluminoxane should be added at once in the highly concentrated form. Any aluminoxane present in the reactor would not have that high concentration, so that the activator in D1 would not fall within the claimed scope. The resulting product would differ from known ones in that less solvent would be present.

- 6.2.2 The Board cannot follow that argument. The present wording of Claim 1 is a product-by-process claim, the only process feature being that the aluminoxane should be provided in the form of a solution of at

least 20 wt% in benzene, toluene or xylene. That process feature, embodied in the verb "provided", does not say anything about the manner of adding the aluminoxane solution to the polymerization system. There is no requirement that it should be added all at once or that it should be added to the reactor separately or that it should be added to the metallocene component before the combined components are added to the reactor, nor is there any restriction regarding the form in which the metallocene should be added to the polymerization system e.g. as a solid or, like the aluminoxane, in the form of a solution. In particular, Claim 1 does not require to solve all of the metallocene component in all of the aluminoxane solution before adding the combined components to the reactor, as is done in all the examples.

The reference to the solvent does not indicate anything relevant in that respect. It merely limits the possible solvents to be used for the aluminoxane solution. However, it does not exclude that that solution is added to the reactor partly or in whole before being brought in contact with the metallocene component, which itself may be solid or in solution.

The term "... said system consisting of ..." does not change the above interpretation, since it can only refer to the product features of the catalyst system. "Consisting of" merely indicates that no other catalyst component may be present in the system so described; it does not bar the skilled person from adding the highly concentrated aluminoxane solution in several separate steps or directly to the reactor.



6.2.3 Accordingly, the process feature of Claim 1 not only encompasses the possibility of adding all of the metallocene to the aluminoxane solution before adding the combined catalyst components to the reactor, as described in all the examples of the patent in suit, but it includes also other possibilities, such as e.g. solving all of the metallocene in part of the aluminoxane solution, having it stand and adding it to the reactor separately from the remaining part of the aluminoxane solution which is added to the reactor directly, as described in all the worked examples of D1.

Although none of those examples actually mentions the use of a an aluminoxane solution having a concentration of at least 20 wt%, saturated solutions are mentioned and preferred concentrations of as high as 30 wt% are specifically mentioned in the description (page 6, lines 42 and 43). Therefore, the use of an aluminoxane solution having a concentration of at least 20 wt% is clearly and unambiguously derivable from D1.

6.2.4 From the above it is clear that the process to prepare a homogeneous catalyst system consisting of a metallocene and an aluminoxane solution as defined in product-by-process Claim 1 and in process Claim 5 of the main request includes processes described in D1. As a result, the product according to Claim 1 cannot differ from products prepared in D1.

6.2.5 Additionally, all the worked examples of D1 describe the preparation of a catalyst system consisting of a metallocene component and an aluminoxane solution and, as a consequence, also the resulting product.

According to Example 1, for instance, bisindenyl-dimethylsilyl zirconium dichloride is solved in a solution of methyl aluminoxane in toluene and preactivated by having it stand for 15 minutes. Using an aluminoxane concentration as preferred (page 6, line 43), one arrives at a system that is catalytically active also without any further aluminoxane addition (see e.g. the examples of the patent in suit). Since it is generally accepted as a principle underlying the EPC that a patent which claims a product as such, confers absolute protection upon such product including all its uses (see Decision G 2/88 (OJ 1990, 093, point 5), Claim 1 is not limited to any particular restrictions regarding the further polymerization conditions, such as the addition of a cocatalyst or electron donor or the like. Therefore, the catalytically active combination of metallocene and aluminoxane solution described in all the worked examples of D1, read in combination with page 6, lines 42 to 43 of D1, falls within the scope of present Claim 1.

6.3 In view of the above considerations, the Board comes to the conclusion that the catalyst system as defined in Claim 1 and the process as defined in Claim 5 of the main request are clearly and unambiguously derivable from D1 and hence not novel.

7. The main request is therefore refused.

*Auxiliary request*

8. Since Claim 1 of the auxiliary request corresponds to Claim 5 of the main request, the same considerations apply, so that its subject-matter is not novel

either.

9. The auxiliary request is therefore refused.

### **Order**

**For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

The Chairman:

E. Görgmaier

B. ter Laan