Case Number: T 0009/99 - 3.3.1
Application Number: 89202995.0
Publication Number: 0372617
IPC: C07F 5/06

Language of the proceedings: EN

Title of invention: Synthesis of methylaluminoxanes

Patentee: TEXAS ALKYLS, INC.

Opponent: Albemarle Corporation

Headword: Methylaluminoxanes/TEXAS ALKYLS

Relevant legal provisions: EPC Art. 123(2)(3), 54(1)(2), 56, 69(1)

Keyword: "Main request: support in the application as filed (no)"
"Auxiliary request: extention of scope of protection (no); novelty (yes) - combination of features not anticipated; inventive step (yes) - nonobvious solution"

Decisions cited:

Catchword:
Case Number: T 0009/99 - 3.3.1

DECISION
of the Technical Board of Appeal 3.3.1
of 26 March 2002

Appellant: Albemarle Corporation
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Composition of the Board:
Chairman: A. J. Nuss
Members: P. P. Bracke
R. T. Menapace
Summary of Facts and Submissions

I. The appeal lies from the Opposition Division's interlocutory decision, dispatched on 3 November 1998, to maintain European patent No. 0 372 617 with a set of 13 claims, of which the independent claims read:

"1. A process for the synthesis of methylaluminoxane which comprises reaction of a tetraalkyldialuminoxane containing C₂ or higher alkyl groups with an amount of trimethylaluminum that is not in stoichiometric excess on the basis of molar equivalents of aluminum."

"4. A process for the synthesis of methylaluminoxane which comprises the reaction of a tetraalkyldialuminoxane containing C₂ or higher alkyl groups with water to form a polyalkylaluminoxane which is then reacted with trimethylaluminum."

"5. A process for the synthesis of methylaluminoxane which comprises the reaction of a trialkylaluminum compound containing higher than C₂ alkyl groups with water to form a polyalkylaluminoxane which is then reacted with trimethylaluminum."

"8. A process for the synthesis of methylaluminoxane which comprises reaction of a polyalkylaluminoxane containing C₂ or higher alkyl groups with trimethylaluminum and then with water."

"11. A solution of a methylaluminoxane in an aliphatic hydrocarbon solvent which methylaluminoxane, when hydrolyzed, evolves hydrolysis products comprising methane and C₂ or higher alkanes."
"13. A method of polymerizing olefins which comprises polymerizing the olefin using a catalytically effective amount of a catalyst system comprising a metallocene component and a methylaluminoxane, characterized in that the methylaluminoxane is used in the form of a solution as defined in claim 11 or 12."

II. In particular, the Opposition Division found that the claimed subject-matter was novel and inventive over the teaching of *inter alia* documents

(1) Journal of Organometallic Chemistry, 234, pages 1 to 6 (1982),

(4) EP-A-0 279 586 and


III. With the statement setting out the grounds of appeal dated 11 March 1999 the Appellant (Opponent) for the first time introduced *inter alia* document

(15) DE-A-3 240 383.

IV. At the oral proceedings before the Board the Respondent (Proprietor of the patent) filed, as an auxiliary request, a set of 12 claims which were identical with Claims 1 to 12 as maintained by the Opposition Division.

V. During the written procedure and at the oral proceedings the Appellant objected that Claim 1 as maintained by the Opposition Division contravened Article 123(3) EPC and that Claims 4 and 11 were not novel over the teaching of document (1) or,
respectively, over the teaching of document (4). Moreover, he argued that the claimed subject-matter was obviously derivable from the combined teaching of documents (15) and (1) and the combined teachings of documents (15) and (8).

Moreover, at the oral proceedings it was questioned by the Board, on its own motion, whether Claim 13 as maintained by the Opposition Division met the requirement of Article 123(2) EPC.

VI. The Appellant requested that the decision under appeal be set aside and that the European patent No. 0 372 617 be revoked.

The Respondent requested that the appeal be dismissed (main request) or that the decision under appeal be set aside and the patent be maintained with Claims 1 to 12 as filed at the oral proceedings (auxiliary request).

VII. At the end of the oral proceedings held on 26 March 2002 the decision was given orally.

Reasons for the Decision

1. **Admissibility**

   The appeal is admissible.

2. **Main request**

2.1 **Article 123(2) EPC**

   In the application as filed Claim 16 was related to a method of polymerising olefins using a catalyst system comprising a methylaluminoxane which, when hydrolysed,
evolves hydrolysis products comprising methane and C2 or higher alkanes. Contrary thereto, present Claim 13 concerns a method of polymerising olefins by using a catalyst system comprising a solution of a methylaluminoxane in an aliphatic hydrocarbon solvent, which methylaluminoxane when hydrolysed, evolves hydrolysis products comprising methane and C2 or higher alkanes.

The Respondent submitted that support for the use of a solution of a methylaluminoxane in an aliphatic hydrocarbon solvent could be found in examples 76 and 77 of the application as filed.

However, the Respondent had to admit that those examples were the only ones describing the preparation of aluminoxanes in an aliphatic hydrocarbon, namely by reacting triisobutylaluminum with water in heptane and subsequently with trimethylaluminum, possibly followed by the addition of further water, and the use of the solutions thus obtained in the polymerisation of polyethylene. As both examples concern processes according to Claims 5 and 8 and nowhere in the application as filed can any indication be found that the methylaluminoxanes obtained according to Claims 1 and 4 would also be sufficiently soluble in an aliphatic hydrocarbon in order to be useful in the polymerisation of polyolefins, it cannot be directly and unambiguously derived from the application as filed that any solution of methylaluminoxanes in an aliphatic hydrocarbon solvent would be useful for polymerising olefins.

It has not been contested that in the first paragraph on page 8 of the application as filed it is mentioned that the disclosed methylaluminoxanes have a high solubility in aliphatic hydrocarbons, such as heptane,
hexane and cyclohexane. However, as it is nowhere taught in the application as filed that such solutions would be useful in the polymerisation of polyolefins, this teaching also cannot be considered as providing support for the subject matter of Claim 13.

2.2 As Claim 13 cannot be considered to meet the requirement of Article 123(2) EPC, the main request cannot be allowed.

3. Auxiliary request

3.1 Article 123(2) EPC

The Board has reached the conclusion that the claims meet the requirement of Article 123(2) EPC. Since this was not disputed, it is not necessary to give detailed reasons for this finding.

3.2 Article 123(3) EPC

According to the Appellant's submissions it cannot be derived from the patent as granted that the requirement in Claim 1 that the trimethylaluminum is not in stoichiometric excess in comparison with tetraalkyldialuminoxane is to be interpreted as a stoichiometric excess on the basis of molar equivalents of aluminum and that by this specification the scope was extended over the scope of granted Claim 1. In particular, he argued that granted Claim 1 was limited to a process wherein the molar ratio of trimethylaluminum to tetraalkyldialuminoxane was limited to 1:1, whereas in present Claim 1 such molar ratio is limited to 2:1.
However, according to Article 69(1) EPC, in determining the extent of the protection conferred by a European patent the description shall be used to interpret claims.

The only information obtainable on how the term "stoichiometric" is to be interpreted is the one given in examples 7-23, where on page 7, lines 49 and 50, it is said that the TMAL (trimethylaluminum)/IBAO (isobutylaluminoxane) ratios in Table II are calculated by dividing molar equivalents of Al in TMAL by the molar equivalents of Al in IBAO or DIBAL-O (tetraisobutylidialuminoxane) and in the heading TMAL/IBAO of the third column of Table II in combination with the explanation on page 10 "a Ratio of moles Al in TMAL to moles Al in IBAO".

The Appellant argued that this information could not be considered as a suitable support for interpreting the term "stoichiometric" in Claim 1, because TMAL/IBAO represents the ratio of trimethylaluminum to isobutylaluminoxane and not the ratio of trimethylaluminum to tetraisobutylidialuminoxane as in Claim 1.

However, since it follows from the fourth column in Table II that examples 7 to 10 and 17 are conducted under condition A, which is specified on page 10, line 9, as "Neat TMAL and neat commercial DIBAL-O"; those examples thus clearly describe the reaction of trimethylaluminum with tetraisobutylidialuminoxane and the ratio of trimethylaluminum to tetraisobutylidialuminoxane in Table II thus clearly is the ratio of equivalents Al in trimethylaluminum to the equivalents Al in tetraisobutylidialuminoxane.
In the absence of any other explanation in the description of the patent in suit as to how the term "stoichiometric" is to be interpreted, a skilled person could clearly and unambiguously derive from the patent in suit that the stoichiometric excess in Claim 1 was expressed on the basis of molar equivalents of aluminum.

Because the wording of present Claim 1 differs from the wording of Claim 1 as granted only by the feature "on the basis of molar equivalents of aluminum", the scope of present Claim 1 is not extended over the scope of Claim 1 as granted for those reasons given above.

Furthermore, Claims 2 to 12 are identical with Claims 2 to 12 of the set of claims as granted and the solution of a methylaluminoxane according to Claim 11 is incontestably restricted in comparison with Claim 12 as granted, relating to the methylaluminoxanes as such.

Therefore, the requirement of Article 123(3) EPC is met.

3.3 Novelty

3.3.1 The Appellant argued that all the parameters of Claim 4 were known from document (1), describing on page 2 the reaction of equimolar amounts of triethylaluminum and water according to the equation (2)

\[ n \text{Et}_3\text{Al} + n \text{H}_2\text{O} \rightarrow \text{Et}_2\text{Al} (\text{OAlEt})_{n-1} \text{OH} \quad (\text{II}) + \ (2n-1) \text{EtH} \]

and on page 3 the reaction of (II) with an equimolar quantity of trimethylaluminum according to the equation (6)

\[ \text{Et}_2\text{Al} (\text{OAlEt})_{n-1} \text{OH} \quad (\text{II}) + n \text{Me}_3\text{Al} \rightarrow \text{MeH} + [\text{Et}_2\text{Al} (\text{OAlEt})_{n-1} \text{OAlMe}_2 \cdot (n-1) \text{Me}_3\text{Al}] \quad (\text{VI}). \]
As it followed from page 1 of document (1) that by the reaction of trialkylaluminum with a 0.5 molar amount of water tetraalkyldialuminoxane was obtained, the Appellant alleged that in the reaction according to equation (2) tetraethylidialuminoxane was intermediately formed, which further reacted with water to form a polyethylaluminoxane. In support of this allegation, the Appellant referred to page 3, lines 51 to 53, of the patent in suit saying that the tetraalkyldialuminoxane and polyalkylaluminoxane can be present in non-isolated form when aluminum alkyls are initially reacted with appropriate amounts of water to form them. Since document (1) disclosed thus implicitly the reaction of tetraethylidialuminoxane with water to form a polyethylaluminoxane which is then further reacted with trimethylaluminum, all features of Claim 4 were known from document (1).

3.3.2 The Appellant did not provide any evidence that in the reaction of triethylaluminum and equimolar amounts of water tetraethylidialuminoxane was intermediately formed and the Respondent strongly contested this unsubstantiated allegation. Also, from the teaching on page 3, lines 51 to 53, of the patent in suit it does not follow that tetraethylidialuminoxane is intermediately formed when reacting equimolar amounts of triethylaluminum and water. From that passage it rather follows that a tetraalkyldialuminoxane is only formed when aluminum alkyls are reacted with appropriate amounts of water.

As a matter of principle, however, the burden of proof is upon the party making an allegation. Since, in the present case, the Appellant made an unsubstantiated allegation, which the Respondent contested, the Board does not have any reason to accept such allegation and
comes therefore to the conclusion, that not all process features according to Claim 4 could be derived directly and unambiguously from document (1).

Therefore, the subject matter disclosed in document (1) is not novelty-destroying for that of Claim 4.

3.3.3 The Appellant also argued that all the parameters of Claim 11 were known from document (4), describing in Claim 5 and on page 6, lines 43 to 54, aluminoxanes composed of mixed oxyaluminum units of formula -O-AlR- containing at least an oxymethylaluminum unit of formula -O-AlCH₃- and disclosing on page 7, lines 14 and 15, n-hexane solutions containing up to 40 mole% aluminoxanes.

3.3.4 However, in assessing novelty, the relevant question is whether the claimed combination of all the features was directly and unambiguously derivable from a document.

Since in document (4) none of the examples is concerned with aluminoxanes composed of mixed oxyaluminum units of formula -O-AlR- and the teaching on page 7, lines 14 and 15, only mentioning aluminoxane solutions in n-hexane, is completely silent about the nature of the aluminoxane, solutions of a methylaluminoxane in an aliphatic hydrocarbon solvent evolving, when hydrolysed, hydrolysis products comprising methane and C₂ or higher alkanes as now claimed, are not directly and unambiguously derivable therefrom.

The Appellant further argued that it was clear from the teaching on page 6, lines 23 to 42, that preferred R-groups in -O-AlR- were, besides the methyl group, also the ethyl- and the isobutyl group and that, consequently, from the aluminoxanes composed of mixed oxyaluminum units of formula -O-AlR- those containing methyl and ethyl or isobutyl groups were preferred.
In assessing novelty, however, the content of a document must not be considered as a reservoir from which features pertaining to separate embodiments could be combined in order to create artificially a particular embodiment. In order to be novelty destroying the teaching of a document must be such that a skilled person would consider combining the different features cited in that document.

This is not the case here, since, in order to come to the solutions of Claim 11, a skilled person had to select (i) from the aluminoxanes those composed of mixed oxyaluminum units of formula \(-O-\text{AlR}\) and (ii) from the mixed aluminoxanes those composed of \(-O-\text{AlR}\) units wherein \(R\) is partly methyl and partly an \(C_2\) or higher alkyl and, subsequently, to combine such an undisclosed selection with the teaching on page 7, lines 14 and 15, that (unspecified) aluminoxane is usually soluble in n-hexane in a ratio up to 40 mole%. Such a combination of features was not directly and unambiguously derivable from document (4).

Therefore, the Board comes to the conclusion that document (4) is not novelty-destroying for Claim 11.

3.4 Inventive step

In accordance with the "problem-solution approach" applied by the Boards of Appeal to assess inventive step on an objective basis, it is in particular necessary to establish the closest state of the art forming the starting point, to determine in the light thereof the technical problem which the invention addresses and solves, and to examine the obviousness of the claimed solution to this problem in view of the state of the art.
3.4.1 The "closest state of the art" is normally a prior art document disclosing subject-matter aiming at the same objective as the claimed invention and having the most relevant technical features in common.

According to the patent in suit, page 3, lines 7 to 9, and page 5, lines 37 to 39, the invention relates to the synthesis of methylaluminoxanes which are useful as cocatalysts in the homogeneous polymerisation and copolymerisation of olefins and/or dienes in conjunction with metallocene compounds and which are highly soluble in aliphatic hydrocarbons. Therefore, only a document relating to such objective could qualify as representing the closest state of the art.

As a method of preparing a polymethylaluminoxane in heptane is described in document (15), in particular in its example 3, and it is said there that such aluminoxane is suitable as a cocatalyst of Ziegler catalysts and since none of the other cited documents concerns aluminoxane solutions in an aliphatic hydrocarbon, only document (15) qualifies as the most suitable starting point for assessing inventive step. This was finally not contested any more by the Parties.

3.4.2 In view of example 3 of document (15), the technical problem underlying the patent in suit consists in providing further processes for preparing Ziegler cocatalysts soluble in aliphatic hydrocarbon solvents and also solutions of such cocatalysts in said solvents.

The patent in suit claims to solve this problem with the processes defined in Claims 1, 4, 5 and 8 and the solutions defined in Claim 11 (see point I above).
3.4.3 The first point to be considered in assessing inventive step is then whether it has been convincingly shown that by the processes according to Claims 1, 4, 5 and 8 and the solutions according to Claim 11 the problem underlying the patent in suit has effectively been solved.

It has never been contested that by the data presented in the examples 76 and 77 of the patent in suit, showing that a product obtained from triisobutylaluminum and water, subsequent addition of trimethylaluminum and, possibly, subsequent addition of water showed a solubility in heptane to be suitable to be used as heptane solution in the polymerisation of polyethylene. Moreover, the Appellant agreed that in view of the test report filed by the Respondent during the opposition proceedings with letter of 11 November 1997, aluminoxanes obtained by reacting tetraisobutylidialuminoxane with trimethylaluminum in hexanes showed suitable solubility in aliphatic hydrocarbons.

The Board has, thus, no reason to deny that a credible case has been put forward that the problem underlying the invention, as defined in point 3.4.2 above, is effectively solved by the claimed process.

3.4.4 Therefore, it remains to be decided whether the processes defined in Claims 1, 4, 5 and 8 and the solutions defined in Claim 11 were rendered obvious by the cited prior art.

3.4.5 The Appellant submitted that it was known from document (4) that mixed aluminoxanes were soluble in aliphatic hydrocarbon solvents and that it was disclosed in the second paragraph on page 5 of document (1) that the preliminary results revealed significant catalytic properties for the oligoaluminoxanes. Therefrom the
Appellant concluded, that the claimed processes and solutions were obviously derivable from the teaching of document (15) combined with the teaching of document (1) and/or (4).

However, document (4) only discloses on page 7, lines 14 and 15, that aluminoxanes are soluble in n-hexane, and not that mixed methylaluminoxanes, in general, let alone those evolving hydrolysis products specifically comprising methane and C₂ or higher alkanes upon hydrolysis, would be sufficiently soluble for being useful as Ziegler cocatalysts in solution. This passage should not be interpreted in isolation, but in combination with the teaching of the complete document, which is concerned with the preparation of finely divided aluminoxanes, by bringing a solution of an aluminoxane in contact with an aluminoxane-insoluble or aluminoxane-sparingly soluble solvent to precipitate a finely divided solid aluminoxane in suspension (see page 7, lines 33 to 51). Since as aluminoxane-insoluble or -sparingly soluble solvents saturated solvents such as hexane are mentioned on page 7, lines 54 to 57, document (4) does not give any hint that aluminoxanes according to a process such as claimed or solutions of them such as claimed, would be suitable Ziegler cocatalysts in an aliphatic hydrocarbon solvent.

Moreover, the passage referred to by the Appellant in the second paragraph on page 5 of document (1), stating that the preliminary results reveal significant catalytic properties for the oligoaluminoxanes, should be interpreted in the context of the complete paragraph, which states in the first sentence that the solubility of oligoaluminoxanes in Lewis acids allows the use of oligoaluminoxanes II and III as an active component or carrier of Ziegler-Natta type catalysts. As the oligoaluminoxanes II and III are both
ethylaluninoxanes, contrary to the mixed methylaluninoxanes according to the present claims, and as this paragraph is completely silent about the catalytic activity of mixed aluninoxanes, such as those of formula (VI), also the teaching of this paragraph does not lead to the claimed subject-matter.

Consequently, the subject-matter of none of the Claims 1, 4, 5, 8 or 11 is rendered obvious by the teaching of documents (15) in combination with the teaching of document (1) and/or (4).

3.4.6 The Appellant further argued that it was known from the first paragraph on page 2 of document (8) that methylaluminoxanes were Ziegler cocatalysts, but that a high excess of alunoxane to the Ziegler catalyst was necessary. As this document was concerned with the exchange of methyl groups by other alkyl groups, such as the exchanges described on pages 20 (point 6.3) and 40 (point 9.4.2), he concluded that it would have been obvious to replace the alunoxane described in example 3 of document (15) by the ones described in document (8).

From the second paragraph on page 8 of document (8) it follows that it was the sole objective of the experiments described therein to study the exchange of methyl groups in methylaluminoxanes by other alkyl- or aryl groups and from the third paragraph on page 32 it follows that it would be interesting to test whether the catalytic activity is maintained after the alkyl-exchange and that such testing is left to other studies. Document (8) does not provide any information about the catalytic activity of aluninoxanes obtained by exchanging methyl groups by alkyl- or aryl groups, let alone about the catalytic activity of mixed methylaluminoxanes, and document (8) also does not
provide any information about the solubility of mixed methylaluminoxanes in aliphatic hydrocarbons. Therefore, document (8) does not contain any suggestion to replace the methylaluminoxane described in example 3 of document (15) by mixed methylaluminoxanes according to the claimed claims.

3.4.7 Therefore, the Board comes to the conclusion that the processes according to Claims 1, 4, 5 and 8 and the solutions according to Claim 11 are not obvious over the cited prior art.

Claims 2, 3, 6, 7, 9, 10 and 12, which represent preferred embodiments of Claims 1, 4, 5 and 8, derive their patentability from the same inventive concept.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to maintain the patent with Claims 1 to 12 as filed at the oral proceedings on 26 March 2002, description pages 3, 7 to 17 as granted, pages 4 to 6 as filed on 22 September 1998.

The Registrar: 

N. Maslin

The Chairman:

A. Nuss