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**Datasheet for the decision
of 20 March 2018**

Case Number: T 1284/16 - 3.3.03

Application Number: 07757312.9

Publication Number: 1989238

IPC: C08F10/00, C08F4/659, C07F15/00

Language of the proceedings: EN

Title of invention:
DUAL METALLOCENE CATALYSTS FOR POLYMERIZATION OF BIMODAL
POLYMERS

Applicant:
CHEVRON PHILLIPS CHEMICAL COMPANY LP

Relevant legal provisions:
EPC Art. 56

Keyword:
Inventive step - (no) - all requests



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Case Number: T 1284/16 - 3.3.03

D E C I S I O N
of Technical Board of Appeal 3.3.03
of 20 March 2018

Appellant: CHEVRON PHILLIPS CHEMICAL COMPANY LP
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted on 15 February
2016 refusing European patent application No.
07757312.9 pursuant to Article 97(2) EPC.

Composition of the Board:

Chairman D. Semino
Members: M. C. Gordon
C. Brandt

Summary of Facts and Submissions

I. The appeal of the applicant lies from the decision of the examination division posted on 15 February 2016 refusing European patent application number 07 757 312.9.

II. The application as filed had 42 claims whereby claim 1 read as follows:

"1. A composition comprising the contact product of: 1) at least one first metallocene; 2) at least one second metallocene; and 3) at least one activator, wherein:
a) the at least one first metallocene comprises an *ansa*-metallocene having the formula:

$(X^1)(X^2)(X^3)(X^4)M^1$ (M1-A), wherein

M^1 is titanium, zirconium, or hafnium;

(X^1) and (X^2) are independently a substituted cyclopentadienyl, a substituted indenyl, or a substituted fluorenyl;

one substituent on (X^1) and (X^2) is a bridging group having the formula ER^1R^2 , wherein E is a carbon atom, a silicon atom, a germanium atom, or a tin atom, and E is bonded to both (X^1) and (X^2) , and wherein R^1 and R^2 are independently an alkyl group or an aryl group, either of which having up to 12 carbon atoms, or hydrogen, wherein at least one of R^1 and R^2 is an aryl group; at least one substituent on (X^1) or (X^2) is a substituted or an unsubstituted alkenyl group having up to 12 carbon atoms;

(X^3) and (X^4) are independently: 1) F, Cl, Br, or I; 2) a hydrocarbonyl group having up to 20 carbon atoms, H, or BH_4 ; 3) a hydrocarbyloxy group, a hydrocarbylamino group, or a trihydrocarbylsilyl group, any of which having up to 20 carbon atoms; or 4) OBR^A_2 or SO_3R^A , wherein R^A is an alkyl group or an aryl group, any of

which having up to 12 carbon atoms; and any additional substituent on the substituted cyclopentadienyl, substituted indenyl, substituted fluorenyl, or substituted alkenyl group is independently an aliphatic group, an aromatic group, a cyclic group, a combination of aliphatic and cyclic groups, an oxygen group, a sulfur group, a nitrogen group, a phosphorus group, an arsenic group, a carbon group, a silicon group, or a boron group, any of which having from 1 to 20 carbon atoms; a halide; or hydrogen;

b) the at least one second metallocene comprises an unbridged metallocene having the formula:

i) $(X^5)(X^6)(X^7)(X^8)M^2$ (M2-A), wherein

M^2 is titanium, zirconium, or hafnium;

(X^5) and (X^6) are independently a cyclopentadienyl, an indenyl, a substituted cyclopentadienyl, or a substituted indenyl;

(X^7) and (X^8) are independently: 1) F, Cl, Br, or I; 2) a hydrocarbyl group having up to 20 carbon atoms, H, or BH_4 ; 3) a hydrocarbyloxy group, a hydrocarbylamino group, or a trihydrocarbylsilyl group, any of which having up to 20 carbon atoms; or 4) OBR^B_2 or SO_3R^B , wherein R^B is an alkyl group or an aryl group, any of which having up to 12 carbon atoms; and

any substituent on the substituted cyclopentadienyl or substituted indenyl is independently an aliphatic group, an aromatic group, a cyclic group, a combination of aliphatic and cyclic groups, an oxygen group, a sulfur group, a nitrogen group, a phosphorus group, an arsenic group, a carbon group, a silicon group, or a boron group, any of which having from 1 to 20 carbon atoms; a halide; or hydrogen;

ii) $(X^9)(X^{10})(X^{11})(X^{12})M^3$ (M3-A), wherein

M^3 is titanium, zirconium, or hafnium;

(X^9) is a substituted cyclopentadienyl group, wherein

one substituent is an aliphatic group, an aromatic group, or a combination of aliphatic and cyclic groups, any of which having up to 20 carbon atoms;

(X¹⁰) is a substituted indenyl group, wherein one substituent is an aliphatic group, an aromatic group, or a combination of aliphatic and cyclic groups, any of which having up to 20 carbon atoms;

(X¹¹) and (X¹²) are independently: 1) F, Cl, Br, or I; 2) a hydrocarbyl group having up to 20 carbon atoms, H, or BH₄; 3) a hydrocarbyloxy group, a hydrocarbylamino group, or a trihydrocarbylsilyl group, any of which having up to 20 carbon atoms; or 4) OBR^C₂ or SO₃R^C, wherein R^C is an alkyl group or an aryl group, any of which having up to 12 carbon atoms; and

any additional substituent on the substituted cyclopentadienyl or substituted indenyl is independently an aliphatic group, an aromatic group, a cyclic group, a combination of aliphatic and cyclic groups, an oxygen group, a sulfur group, a nitrogen group, a phosphorus group, an arsenic group, a carbon group, a silicon group, or a boron group, any of which having from 1 to 20 carbon atoms; a halide; or hydrogen;

or

iii) any combination thereof;

and

c) the at least one activator is selected independently from:

i) an activator-support selected from a solid oxide treated with an electron-withdrawing anion, a layered mineral, an ion-exchangeable activator-support, or any combination thereof;

ii) an organoaluminum compound;

iii) an organoboron compound or an organoborate compound; or

iv) any combination thereof."

III. The decision of the examining division was based on amended sets of claims forming a main request and first to third auxiliary requests, filed with letter dated 21 October 2015 and fourth and fifth auxiliary requests filed with letter dated 12 November 2015.

Claim 1 of the main request was identical to claim 1 of the application as filed.

It is not necessary for the purposes of this decision to give further details of the wording of the auxiliary requests beyond the indication that starting with the first auxiliary request indenyl had been excluded from the definition of the first metallocene in claim 1.

According to the decision the closest prior art was US-2005/159300 (D1), in particular the examples of Tables 4A and 4B.

The subject-matter claimed was held not to involve an inventive step over this disclosure either alone or in combination with the teachings of, *inter alia*:

D2: US-A-5 886 202.

The same conclusions applied to the auxiliary requests in which the definition of the catalyst architecture had been progressively restricted.

Consequently the application was refused.

IV. The applicant lodged an appeal against the decision. Experimental reports - E1 and E2 - were submitted with the statement setting out the grounds of appeal,

whereby E1 had previously been submitted in proceedings before the examining division.

The sets of claims on which the decision was based were maintained.

Central to the appellant's arguments was that the disclosure within D1 identified as the closest prior art was a comparative example. A different part of D1 was considered to be more appropriate.

- V. The Board issued a summons to oral proceedings and a communication, in which *inter alia* matters relating to Article 123(2) and Article 84 EPC were identified.

With respect to inventive step, it was held that there were no data to demonstrate the existence of a technical effect with respect to that aspect of D1 identified by the appellant as forming the closest prior art.

- VI. In response, the appellant with letter dated 5 March 2018 provided a new set of claims as main request, amended in view of the observations with respect to Articles 84 and 123(2) EPC and a further experimental report (E3).

- VII. In a further communication the Board noted that E3 did not appear to correspond either to the teaching of D1 or to the subject-matter as claimed since in all examples only a single bridged metallocene, but no unbridged metallocene had been employed.

- VIII. The appellant made a further written submission with letter of 7 March 2018 addressing these aspects, including filing a complete set of amended first to

fifth auxiliary requests.

IX. Oral proceedings were held before the Board on 20 March 2018.

X. The arguments of the appellant can be summarised as follows:

The problem underlying the application was to provide a catalyst composition suitable for producing a resin having a good balance of properties such as physical strength, processability and low level of long chain branching.

The closest prior art was represented by D1, Table 6B, notwithstanding that D1 was directed to resins for film applications whereas the application in suit was concerned with pipe resins. This example of D1 related to catalyst systems based on a loosely bridged metallocene (i.e. more than a single atom in the bridge) and an unbridged metallocene.

The following distinguishing features could be identified:

- the position of the alkenyl group on X^1 and X^2
- at least one of R^1 and R^2 on ER^1R^2 was independently aryl or alkyl or if one was H the other was mandatorily aryl.
- X^1 and X^2 were both substituted
- a single atom in the bridge.

The experimental report E3 demonstrated that the effect of the tightly bridged metallocenes compared to the loosely bridged metallocenes of D1 was to provide polymers of higher molecular weight as well as higher

catalyst activity. The higher molecular weight resulted in increased physical strength of the polymer.

The same effect was evident from a comparison of example E6 of the application with example 9.3 of D1.

Although report E3 concerned catalyst systems employing only a single - bridged - metallocene, the results would apply also to the subject-matter of claim 1 incorporating additionally the unbridged metallocene. The data of E3 isolated and emphasised the technical effect arising from the distinguishing features of the bridged metallocene, i.e. the bridge being a single atom and the position of the alkenyl substituent, the significance of which was furthermore explained in the application.

It was acknowledged that there were a number of differences between the data of the application and those of report E3 on the one side and D1 on the other in terms of the specific metallocenes employed as representative of the subject-matter claimed and those representative of the prior art which went beyond the distinguishing features. However these differences would not materially influence the activity of the catalyst and hence would not detract from the suitability of these data to demonstrate the technical effect deriving from the distinguishing features.

This evidence established that the objective technical problem solved with respect to D1 was to provide a catalyst system which resulted in a resin with higher molecular weight and hence increased physical strength wherein the catalyst had higher activity.

There was no teaching in D1 to employ the claimed

metallocene structure to solve said problem or to make the necessary modifications to the metallocenes of D1. On the contrary, in D1 structures corresponding broadly to those now claimed in that they had a tightly bridged metallocene structure were presented as comparative examples, providing inferior results, notwithstanding that the structures differed in other respects from those claimed. D1 was concerned with different polymers and different catalysts. Nor did reference to other documents, for example D2 provide any indication that the particular combination of structural features would give rise to the observed effects.

For the auxiliary requests the argument remained essentially the same, notwithstanding that these requests were directed to emphasising the difference over D1. In particular the restrictions made to the organic ring structures would modify the electronic environment of the metal and hence influence the activity of the catalyst and the properties of the product obtained.

- XI. The appellant requested that the decision under appeal be set aside and a patent be granted on the basis of the set of claims filed as main request with letter dated 5 March 2018, or alternatively on the basis of one of the sets of claims according to first to fifth auxiliary requests filed with letter dated 7 March 2018.

Reasons for the Decision

1. Main request - inventive step

1.1 The application - technical field

The application is directed in general to organometal compositions, olefin polymerisation catalyst compositions and methods of polymerisation (first paragraph of the application). In particular the problem which the application addresses is being able to tailor the properties of the resin (page 1, line 18). According to the passage bridging pages 1 and 2 there existed a need for a resin and a pipe prepared therefrom that exhibited improved physical properties. However with resins prepared using conventional metallocene catalyst systems there was a trade-off between high stiffness and high environmental stress cracking resistance.

This problem is solved according to the application by a dual metallocene catalyst system, which can provide polyolefin having bimodal or multimodal molecular weight distribution and having a good balance of the required properties (page 2 lines 6-18, in particular lines 12-14).

1.2 Closest prior art

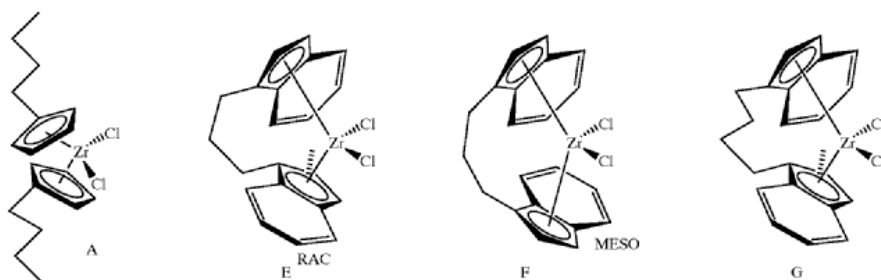
It is the position of the appellant that the closest prior art is represented by D1 which is directed not to resins for pipes but to resins for film applications (D1, title, paragraphs [0001], [0005] and [0056]). Nevertheless, in common with the application, D1 also addresses the question of tailoring the properties of the resins to meet a particular profile (paragraphs

[0002]-[0004]).

The Board is satisfied that, notwithstanding the different end uses of the polymer resins, D1 can be taken as representing the closest prior art since, like the application, it addresses the problem of tailoring the properties of polymers to meet specific needs.

According to paragraphs [0005]-[0007] and [0056] of D1 it was found that a dual catalyst system containing a combination of metallocene catalysts resulted in a useful combination of polymer properties.

Within D1 the most relevant disclosure is considered - in agreement with the appellant - to be the catalyst systems of Table 6A:



wherein A is an unbridged metallocene and E, F and G are the bridged metallocenes and the dual catalyst systems A+E, A+F and A+G were tested.

As activator, these examples of D1 employ triethyl aluminium and fluorided CTSO (passage bridging page 27 and 28 of D1) i.e. a solid oxide treated with an electron withdrawing anion (fluoride), as required according to feature c)i) of operative claim 1.

1.3 Distinguishing features

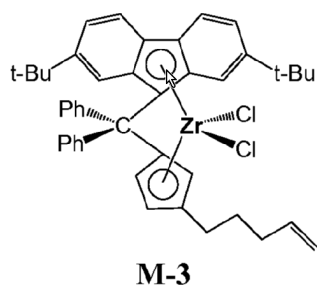
The definition of the unbridged metallocene of operative claim 1 encompasses that of D1, i.e. this

feature of the claim does not represent a distinction over D1 feature. This was stated in the decision under appeal (section 11.1 of the reasons) and was acknowledged by the appellant in the discussion of the identity of the prior art in the statement setting out the grounds of appeal.

The subject-matter of claim 1 is thus distinguished from the disclosure of Table 6A of D1 by the following features of the bridged metallocene:

- E in the bridging group is a single atom;
- the bridging group is substituted by an alkyl group, an aryl group, either of which having up to 12 carbon atoms, or hydrogen whereby at least one of the groups is aryl;
- at least one of the substituent groups on the one of the aromatic systems (X^1 and X^2 in the claim) is a substituted or unsubstituted alkenyl having up to 12 carbon atoms.

Representative and illustrative of the bridged metallocene as required by the claim is, for example that identified as M-3 in the application (Figure 1, page 97) which was also employed in the experimental reports E1 and E3:



1.4 Technical effect

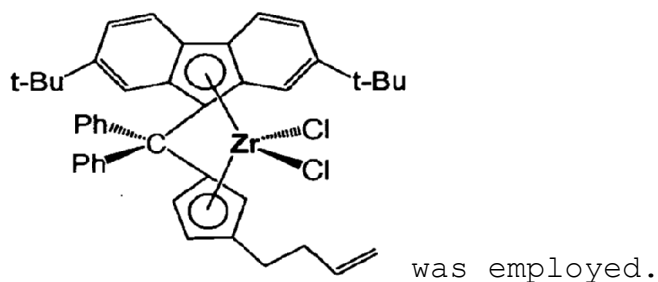
The examples of the application (pages 90-105, in

particular 100-105) do not provide any comparison with the subject-matter of D1 since the nature of the bridged metallocene is not varied.

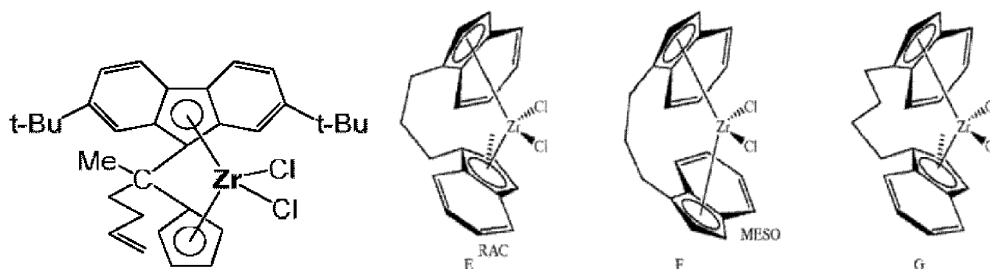
With experimental report E1 the appellant provided a (comparative) example comparing a metallocene in which the - methylene, i.e. single carbon atom - bridging group was substituted by a methyl group and an alkylene group. However this example is structurally different from the bridged catalysts of D1 having an unsubstituted bridging group comprising 3 to 5 contiguous ansa carbon atoms. This comparison cannot be seen as representative of D1 and consequently cannot demonstrate whether any technical effect arises as a result of the distinguishing feature of the operative claims.

With experimental report E2 comparisons were provided with bridged metallocenes in which either no alkylene group was present on the metallocene ring, or in which the bridging group was substituted with two alkyl groups. This evidence also did not serve to demonstrate whether a technical effect is present over the systems of D1.

Finally, in response to observations by the Board about the unsuitability of the existing evidence to demonstrate an effect with respect to D1, the appellant filed experimental report E3 in which, as representative of the subject-matter claimed, in addition to the above-mentioned metallocene M-3 the metallocene identified as M-4 in the application (figure 1, pages 97-98):



These were compared with the following bridged metallocenes:



The first of these metallocene structures resembles that of Tables 4A and 4B of D1, which was in the nature of a comparative example but - nevertheless - considered in the decision under appeal as representing the closest prior art. The structures of metallocenes E-G represent the teaching of D1.

Common to all these experimental reports was that only a single - bridged - metallocene was employed. There was no unbridged metallocene present, meaning that these systems correspond neither to the subject-matter claimed nor to the subject-matter of D1, so that they did not permit any conclusion to be reached with respect to the claimed system.

Regarding report E3, the three metallocenes E, F and G which were employed by the appellant to represent the teaching of D1 differed from those employed as representative of the subject-matter of the application (M3, M4) not only in the above noted distinguishing

features but also in the nature of the metallocene ring structure.

There is accordingly no basis for concluding to which extent, if at all, the demonstrated differences in the performance of the catalysts were due to the above identified distinguishing features. The evidence provided does not allow to exclude that the observed differences in performance were - either in part or in total - a consequence of the differing structures of the organic framework. Indeed, the appellant explicitly acknowledged - albeit with respect to the auxiliary requests - that modification to the ring structure would influence the electronic environment of the metal atom, which is the active centre in these catalysts.

The first of the above indicated "comparative" catalysts is the same as employed in report E1 which as discussed above also does not permit any conclusion to be drawn with respect to the distinguishing features with respect to D1.

Regarding the comparison between example E6 of the application and example 9.3 of D1, referred to by the appellant, it is observed that the metallocenes employed in these examples likewise differed in a plurality of features. Thus the bridged metallocene employed in example E6 of the application - designated M-1 - corresponds to the above referenced M-3 with the difference that the metal atom is hafnium (see figure 1 of the application) and the bridged metallocene employed in example 9.3 of D1 is the above reported metallocene F. Thus this juxtaposition of data from D1 and the application suffers from the same defects as the data provided in E3 in terms of unsuitability to provide any unambiguous evidence for an effect

associated with the distinguishing features.

Consequently none of these data provide evidence for a technical effect associated with any of the distinguishing features with respect to D1.

1.5 Objective technical problem/the solution

In view of the absence of evidence for a technical effect associated with the distinguishing features the only technical problem that can be objectively formulated with respect to D1 is the provision of further dual metallocene catalyst systems.

This problem was solved by modifying - in the light of the available evidence arbitrarily - the structure of the bridged metallocene, as represented by the above identified distinguishing features.

1.6 Obviousness

The arbitrary modification of an existing teaching is inherently an obvious solution to the problem of providing a further composition.

Furthermore the structural features of the metallocene which represent distinctions with respect to D1 are known in the prior art.

D2 relates to bridged metallocenes consisting of an indenyl and a fluorenyl system whereby the bridge can be a single atom (claim 2, Table 7). D2 investigates the effect of the substituent pattern on bridged metallocenes and proposes that the substitution of methyl by phenyl in a single atom bridge can be advantageous (D2, column 9 lines 48, 49). Also the

placement of an alkenyl substituent on the 3-position of the indenyl group is proposed and it is postulated that this will give rise to higher molecular weight (column 9, lines 60-63).

D2 thus establishes that the various substitution patterns as defined in the application were already known in the art and at least in part associated - if only as postulates - with beneficial properties of the catalyst.

Thus D2 provides a pointer to the restrictions made, further underlining the obviousness of said subject-matter as a solution to the formulated technical problem.

1.7 The subject-matter of the main request does not meet therefore the requirements of inventive step.

2. Auxiliary requests

These requests differ from the main request by making progressive restriction of the nature of the metallocene structures. *Inter alia* indenyl groups are excluded as from the first auxiliary request. However there is likewise no evidence for a technical effect associated with any of the restricted groups of metallocenes. Nor were any additional arguments in respect of these requests advanced by the appellant beyond the above mentioned observation that modification of the ring structure affects the electronic environment of the metal.

Accordingly the Board has no reason to come to a different conclusion in respect of inventive step for any of the auxiliary requests to that reached for the

main request.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:



B. ter Heijden

D. Semino

Decision electronically authenticated