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**Datasheet for the decision
of 28 June 2011**

Case Number: T 0334/07 - 3.3.07

Application Number: 94308612.4

Publication Number: 0654302

IPC: B01J 27/26

Language of the proceedings: EN

Title of invention:

Improved double metal cyanide complex catalysts

Patent proprietors:

BAYER ANTWERPEN N.V.

Opponents:

Shell Internationale Research Maatschappij B.V.
THE DOW CHEMICAL COMPANY

Headword:

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Relevant legal provisions:

EPC Art. 54

Relevant legal provisions (EPC 1973):

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Keyword:

"Novelty (no)"

Decisions cited:

-

Catchword:

-



Case Number: T 0334/07 - 3.3.07

D E C I S I O N
of the Technical Board of Appeal 3.3.07
of 28 June 2011

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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 21 December 2006
revoking European patent No. 0654302 pursuant
to Article 102(1) EPC.

Composition of the Board:

Chairman: J. Riolo
Members: D. Semino
M.-B. Tardo-Dino

Summary of Facts and Submissions

I. The appeal of the patent proprietors (appellants) lies against the decision of the opposition division announced at the oral proceedings on 16 November 2006 to revoke European Patent 0 654 302. The granted patent comprised 6 claims, independent claim 1 reading as follows:

"1. A double metal cyanide (DMC) complex comprising i) up to 10 wt.% of a crystalline DMC component and ii) at least 90 wt.% of a DMC component which is amorphous to X-rays and which comprises a complexing agent which is a water-soluble aliphatic alcohol."

II. Three notices of opposition were filed against the granted patent requesting revocation of the patent in its entirety on the grounds of lack of novelty, lack of inventive step and insufficiency of disclosure as set out in Article 100(a) and (b) EPC. The opposition of opponents 01 was withdrawn before the opposition division reached a decision.

III. In the decision the following documents were cited *inter alia*:

D1: JP-A-4 145123

D1': English translation of D1 filed by opponents 01

D2: Experimental report conducted by Gao Guohua filed by opponents 01 with the notice of opposition

D4: Experimental report conducted by Shigeru Ikai filed by opponents 01 with the notice of opposition

D9: Experimental report filed by opponents 02 with the notice of opposition

D16: US-A-5 158 922

D18: US-A-5 712 216

D20: Experimental report filed by opponents 03 with the notice of opposition

D21: Experimental report filed by the patent proprietors with letter of 6 December 2004

IV. The decision of the opposition division can be summarised as follows:

Sufficiency of disclosure

(a) The alternative definitions of the expression "amorphous to X-rays" in the description used terms ("well-defined crystal structure" and "sharp lines") which had no clear meaning. In the context of the description of the patent in suit the meaning of "amorphous to X-rays" could only be understood as lacking the peaks that corresponded to the crystalline DMC prepared in absence of an organic complexing agent.

The measurements of the degree of crystallinity had to be carried out by spiking experiments, which were known by the person skilled in the art, but had a low level of accuracy.

The experiments submitted by the parties during opposition showed that the skilled person was able to prepare substantially amorphous zinc hexacyanocobaltate t-butanol complexes and no evidence had been filed that other substantially amorphous DMC complexes could not be obtained.

Therefore the patent in suit enabled the skilled person to carry out the invention as claimed over the whole range, although the claimed subject-matter lacked clarity in view of the alternative, but not equivalent definitions of the term "amorphous to X-rays".

Novelty

- (b) D1 disclosed a zinc hexacyanocobaltate catalyst complexed with t-butanol and its method of preparation. All the reproductions of D1 carried out by the parties (D2, D4, D9, D20, D21) lead to a substantially amorphous zinc hexacyanocobaltate t-butanol complex except when the mixing was done with a stirrer at 200 rpm at room temperature for a time or 10 minutes or less. Since a rework of D1 which carried out the maturation step at room temperature for 10 minutes or less did not correspond to what was commonly understood in the art as adequate conditions for maturing an organic ligand containing DMC suspension, novelty of the subject-matter of claims 1 and 6 over D1 could not be acknowledged.

The subject-matter of claims 1 and 6 was also not novel in view of D16, since the preparation methods disclosed therein lead to a complex falling under claim 1 by taking into account technical information given in D18, a late published document by the same inventor as the patent in suit.

V. The patent proprietors appealed that decision. With the statement setting out the grounds of appeal they submitted some further experiments meant at reproducing the examples of D1 and D16 and a first auxiliary request, in which claim 1 had been amended as follows:

"1. A double metal cyanide (DMC) complex comprising i) up to 1 wt.% of a crystalline DMC component and ii) at least 99 wt.% of a DMC component which is amorphous to X-rays and which comprises a complexing agent which is a water-soluble aliphatic alcohol."

VI. In a communication sent in preparation of oral proceedings the Board expressed its concerns over the meaning of the expression "amorphous to X-rays" due to the presence of relative terms in its definition, to the lack of a method of measurement of the percentage of the complex which is amorphous to X-rays and to the equivalence of that expression with the expression "substantially amorphous" used in the original application which indicated that the part of the complex which was defined as "amorphous" was itself not totally amorphous and might contain a (quantitatively not specified) crystalline part.

VII. Oral proceedings were held on 28 June 2011 in the announced absence of the opponents 02 and 03.

VIII. The arguments of the appellants (patent proprietors), as far as relevant to the present decision, can be summarised as follows:

(a) In reference example 1 of D1 there was no disclosure of the conditions of the maturation

step, namely the severity of agitation, the amount of time and the temperature. The reproduction of that example in D21 in which reaction and maturation were carried out by stirring at 200 rpm for 5 minutes showed that the obtained complex contained approximately 50% of a crystalline component. Since those conditions had been considered as not adequate by the opposition division, further experiments had been provided with the statement of grounds in which maturation had been conducted under more severe mixing conditions und for a longer period of time. The choice of mixing the solutions containing the two salts for 30 to 60 minutes before addition of the solution of the complexing agent followed by maturation instead of mixing the three solutions followed by reaction and maturation had an impact on the structure of the product, but was in agreement with the teaching in the description of D1 and therefore also in accordance with its example. Since those experiments resulted in a highly crystalline product, it could not be concluded that the reproduction of reference example 1 of D1 inherently resulted in a product comprising up to 10 wt.% of a crystalline DMC component and at least 90 wt.% of a DMC component which is amorphous to X-rays. The information in the patent and also later advancements in the field had shown that such a result could be obtained only with specific methods of preparation which had not been disclosed in D1.

- (b) The measurement of the quantity of the amorphous component in the complex had to be done by means

of spiking experiments as mentioned in the patent, which were well-known to the skilled person. The use of the expression "substantially amorphous" in the original application (which was then amended to "amorphous to X-rays") was due to the fact that the complex could not be properly indicated as "amorphous", since in amorphous solids the atoms are completely disordered, while in the present case a certain degree of order at short distance was present due to the ionic structure of the salt. In any case it was clear from the patent that no crystalline component was present in the amorphous part within the tolerance of the measurements which was around 1%. Claim 1 according to the first auxiliary request expressed clearly that no crystalline component was present within the tolerance of the method of measurement.

- (c) For those reasons, novelty of the product of granted claim 1 with respect to the disclosure of D1 had to be acknowledged. With even stronger reasons that conclusion was valid for the first auxiliary request.

IX. The arguments of the respondents (opponents 02 and 03), as far as relevant to the present decision, can be summarised as follows:

- (a) The term "amorphous to X-rays", which related to the only feature alleged to distinguish the claimed material from the prior art, was not present in the original text and was fundamentally unclear. The only possible definition in the patent ("lacking a well-defined crystal structure

or characterised by the substantial absence of sharp lines in the X-ray diffraction pattern of the component") left unclear whether it was a single requirement or two alternative requirements and there was no guidance as to what was meant by "sharp lines" or "substantial absence". The presence of certain "broad" lines in the X-ray diffraction spectrum of the amorphous component rendered the expression even more unclear, since it was apparently contradictory with the definition and the term "broad" was not defined.

(b) The fundamental lack of clarity of the term "amorphous to X-rays" resulted in insufficiency of disclosure. Moreover, the feature related to it was incapable of distinguishing the invention as claimed in the granted patent and in the first auxiliary request from the disclosure in D1 and D16, so as to result in lack of novelty.

X. The appellants (patent proprietors) requested that the decision under appeal be set aside and the patent be maintained as granted or, alternatively, on the basis of the set of claims filed as first auxiliary request with the statement setting out the grounds of appeal.

XI. The respondents (opponents 02 and 03) had requested in writing that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

Novelty of granted claim 1

2. Reference example 1 of D1 (see pages 10 and 11 of its translation into English D1') discloses the production of a double metal cyanide complex according to the following procedure: 15 cc of an aqueous solution containing 10 g of zinc chloride, 75 cc of an aqueous solution containing 4 g of potassium cyanocobaltate and 100 cc of a 50 wt.% t-butanol aqueous solution were mixed together at room temperature, and reaction and aging were conducted while stirring it to obtain a solution in a slurry state. Then, this solution was subjected to filtration by suction filtration to obtain a white product. This filter cake was washed with a 30 wt.% t-butanol aqueous solution, and filtered to obtain a filter cake, and further washed with t-butanol and then filtered to obtain a filter cake. This filter cake was dried at 40°C under reduced pressure and then milled to obtain a double metal cyanide complex.
 - 2.1 There can be no doubt (and it was not contested by the parties) that by means of this procedure a double metal cyanide complex is obtained which comprises a complexing agent which is a water-soluble aliphatic alcohol (t-butanol). It needs therefore to be determined whether this complex can be considered as comprising at least 90 wt.% of a DMC component which is amorphous to X-rays and up to 10 wt.% of a crystalline DMC component.

2.2 The critical issue, as acknowledged by the parties in their submission, lies in the interpretation of the term "amorphous to X-rays". The patent defines amorphous to X-rays as "lacking a well-defined crystal structure or characterized by the substantial absence of sharp lines in the X-ray diffraction pattern of the component" (paragraph [0019]). The critical term was not present in the original application and replaced in the granted patent the expression "substantially amorphous" in the application as filed, which was defined in exactly the same way (page 3, lines 46-48 of the A publication and claims) with no change in meaning (otherwise a problem under Article 123(2) EPC would have arisen).

2.3 It is not specified in the patent whether the two definitions ("lacking a well-defined crystal structure" and "characterized by the substantial absence of sharp lines in the X-ray diffraction pattern of the component") are equivalent or alternative. Moreover, no further information is given about what is meant by "well-defined", "substantial absence", "sharp lines", which are all relative terms lacking a defined boundary, and a method of measurement of the percentage of the complex which is amorphous to X-rays is neither given, nor referred to. Both the expression "substantially amorphous" and the relative terms in the definition indicate therefore that the part of the complex which is defined as "amorphous" is itself not totally amorphous and may contain a (quantitatively not specified) crystalline part.

2.4 The appellants indicated the final part of paragraph [0021] in the patent and the data in Table 1 as a

source of further information which should make it clear what is meant by "amorphous to X-ray" and how the weight percent of the component which is "amorphous to X-rays" should be measured.

2.4.1 The sentence in paragraph [0021] cited by the appellants reads "Spiking experiments demonstrate that DMC catalysts prepared by the method of the invention typically contain less than about 1 wt.% of highly crystalline DMC compound". While this passage makes a clear reference to the possibility of running spiking experiments, it does not add anything as to how these experiments should be used to determine the amount of the component which is "amorphous to X-rays", nor does it give any indication of when it should be concluded that a sharp line present in the X-ray diffraction pattern is substantially absent. Moreover, there is no reference in claim 1 to the method of preparation of the product so that the statement related to catalysts prepared by the method of the invention has no weight on the product of claim 1.

2.4.2 Table 1 lists for a number of catalysts according to the examples which X-ray diffraction lines are present and which are absent. However, the X-ray diffraction patterns are not shown. Neither this table, nor the corresponding information in the description of the examples give any information on which criteria have been used to determine when a line is considered to be absent (or substantially absent according to the definition in paragraph [0019]).

2.4.3 Moreover, these results are apparently contradictory with the definition of "amorphous to X-rays", since

also the catalysts of the invention (catalysts D and E in the table) are characterised by the presence of certain diffraction lines. This is partially explained in paragraph [0021] of the patent in suit (second sentence), where it is said that a zinc hexacyanocobaltate catalyst prepared according to the method of the invention shows essentially no lines for crystalline zinc hexacyanocobaltate, but instead has only two major lines, both relatively broad, at specific d-spacings. This is understood to indicate that, while "sharp lines" should be substantially absent, "relatively broad lines" can be present. However, as is the case for the term "sharp lines", also the relative term "relatively broad lines" is not explained, so that the skilled person is left without clear criteria to determine where the boundaries are.

2.5 Also the arguments of the proprietor that the term "substantially amorphous" had been used in the original application in view of the presence of a certain degree of order at short distance in the complex due to the ionic structure of the salt cannot change the fact that no clear boundaries are given to distinguish what is "amorphous to X-rays" according to the patent in suit from what is not. Moreover, there is no information in the original application related to the issue of the presence of a certain degree of order at short distance, which could help in the interpretation of the term.

2.6 In view of this only a very broad interpretation of the term "amorphous to X-rays" is possible, which does not put clear limits to the quantity of crystalline material which is present in the amorphous component. Taking this broad interpretation into consideration,

the products of D1 could be distinguishable from the claimed complex only if the former were totally or predominantly crystalline.

- 2.7 A large number of tests aimed at reproducing reference example 1 of D1 had been filed by the parties during opposition proceedings and had been taken into account in the appealed decision. Since the tests of the appellants had been considered as employing inadequate conditions for maturing an organic ligand containing DMC suspension, they filed three further experiments with the statement setting out the grounds of appeal meant at reproducing reference example 1 of D1.
- 2.7.1 In these experiments an aqueous solution of zinc chloride was added to an aqueous solution of potassium hexocyanocobaltate within 30 or 60 minutes at room temperature while mixing with a conventional laboratory stirrer at 200 or 600 rpm, followed by addition of the t-butanol aqueous solution and then stirring at 200 or 600 rpm for further 60 minutes.
- 2.7.2 These experiments cannot be considered as reproducing the method of reference example 1 of D1 in which the three solutions were mixed and then reaction and maturation were carried out. Indeed, in the experiments filed with the statement of grounds reaction between the salts is carried out in the absence of the complexing agent which is only added afterwards before maturation takes place.
- 2.7.3 While it is true that the description of D1 discloses that it is preferred to conduct reaction between the salts first and then add the complexing agent dropwise

(page 7 of D1', last full paragraph), this is not the situation in reference example 1 and there is no reason to presume that in reference example 1 something different is meant from what is clearly written. Moreover, the appellants themselves have confirmed that the order of addition and the presence of the complexing agent during reaction have a strong impact on the properties of the obtained product. On this basis the experiments filed with the statement setting out the grounds of appeal are not relevant to assess the characteristics of the product of reference example 1 of D1.

- 2.7.4 All the other tests filed by the parties during opposition proceedings in order to reproduce reference example 1 of D1 (D2, D4, D9, D20, D21) lead to a predominantly amorphous zinc hexacyanocobaltate t-butanol complex except when the reaction and maturation were done with a stirrer at 200 rpm for a time of 10 minutes (experiment A of D9) or 5 minutes (single test in D21). The weight percent of the crystalline DMC component was 26 wt.% with 10 minutes mixing and approximately 50 wt.% with 5 minutes mixing.
- 2.7.5 The Board concurs with the opposition division that the mixing conditions of experiment A of D9 and of the single test of D21 (200 rpm for a time of 10 minutes or less) are not adequate for proper reaction and maturation of the DMC complex, since in all the available prior art reaction and maturation take place over much longer times. However, since also these experiments do not result in a complex which is totally or predominantly crystalline and the produced products may even fall under the granted claim in its broad

interpretation, which does not set clear boundaries for the crystalline part, there is no need to discuss this issue in any more detail.

- 2.8 In view of the evidence on file and of the broad interpretation of the term amorphous to X-rays, which does not exclude the presence of a (not quantified) crystalline part in the amorphous component as long as the component is "substantially" amorphous, the Board can only conclude that the feature that the complex comprises at least 90 wt.% of a DMC component which is amorphous to X-rays and up to 10 wt.% of a crystalline DMC component is not able to distinguish the complex according to granted claim 1 with respect to the product of reference example 1 of D1. The product of claim 1 as granted is therefore not novel with respect to the disclosure in D1.

First auxiliary request

3. Claim 1 according to the first auxiliary request has been amended with respect to granted claim 1 in that the weight percent of the DMC component which is amorphous to X-rays is changed from at least 90 to at least 99 and correspondingly the weight percent of the crystalline DMC component from up to 10 to up to 1.
- 3.1 In view of the broad interpretation of the term "amorphous to X-rays" this amendment does not result in a relevant change of the subject-matter of claim 1, as the presence of a (not quantified) crystalline part in the amorphous component is not excluded (see point 2.6 above).

3.2 For the same reasons as detailed under point 2 above, the product of claim 1 according to the first auxiliary request is therefore not novel with respect to the disclosure of D1.

4. Since all the requests of the appellants fall for lack of novelty with respect to the disclosure in D1, there is no need to discuss any other issue.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar

The Chairman

S. Fabiani

J. Riolo