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**Datasheet for the decision
of 15 November 2012**

Case Number: T 0166/10 - 3.3.07

Application Number: 00309956.1

Publication Number: 1205246

IPC: B01J 37/02, C07C 67/055,
C07C 69/01, B01J 23/66,
B01J 23/58, B01J 23/52,
B01J 23/89

Language of the proceedings: EN

Title of invention:
Process for preparation of catalyst

Patentee:
Dairen Chemical Corporation

Opponent:
Celanese International Corporation

Headword:
-

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

Keyword:
"Late submitted material - document admitted (yes)"
"Novelty - implicit disclosure (no)"
"Inventive step - yes (main request)"

Decisions cited:
-

Catchword:
-



Case Number: T 0166/10 - 3.3.07

D E C I S I O N
of the Technical Board of Appeal 3.3.07
of 15 November 2012

Appellant: Celanese International Corporation
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Decision under appeal: **Decision of the Opposition Division of the
European Patent Office posted 20 November 2009
rejecting the opposition filed against European
patent No. 1205246 pursuant to Article 101(2)
EPC.**

Composition of the Board:

Chairman: D. Semino
Members: R. Hauss
M.-B. Tardo-Dino

Summary of Facts and Submissions

I. The appeal lies from the decision of the opposition division, announced at the oral proceedings on 1 October 2009, rejecting the opposition filed against European patent No. 1 205 246.

II. European patent No. 1 205 246 was granted on the basis of six claims.

Claim 1 of the granted patent, which is the only independent claim, reads as follows:

"1. A process for the preparation of a catalyst, comprising the steps of;

- (a) impregnation of a catalyst supporting material surface with a solution containing an oxidative state noble metal and an oxidative state metal for assisting catalysis, wherein said oxidative metal for assisting catalysis is selected from the group consisting of gold, copper, molybdenum, cadmium and magnesium;
- (b) reducing the metals from the oxidative state into a metallic state in gaseous phase with gaseous reducing agent while controlling the moisture content in the range of from 50 to 5000ppm by periodical purging of the reducing system with nitrogen to remove water vapour produced during reduction, wherein said gaseous reducing agent is hydrogen;
- (c) washing the treated carrier with deionized water to remove soluble ions;
- (d) drying the catalyst;
- (e) impregnating the reduced catalyst with a solution of alkali or alkaline earth metal compound; and
- (f) drying the catalyst at between 80 and 150°C until the water content is between 0 and 6%."

- III. An opposition was filed, opposing the patent in its entirety under Article 100(a) EPC for lack of novelty and inventive step.
- IV. The documents cited during the opposition proceedings included the following:
- D1: EP 0 976 713 A1
- D3: Farrauto, Bartholomew: Fundamentals of Industrial Catalytic Processes, Chapman & Hall, 1997, pp. 101-102
- D4: Richardson: Principles of Catalyst Development, Plenum Press, 1989, pp. 116-121
- D5: US 6 022 823 A
- D6: WO 98 18553 A1.
- V. In the decision to reject the opposition, the opposition division came to the conclusion that the process of claim 1 of the patent as granted was novel over the disclosure of D5, since said document did not disclose controlling the moisture content in the range of from 50 to 5000 ppm by periodical purging of the reducing system with nitrogen to remove water vapour produced during reduction. For the same reason, the claimed subject-matter was novel over D6, which belonged to the same patent family as D5 and had the same content. As far as the assessment of inventive step was concerned, the opposition division regarded D1 as the most relevant prior-art document. D1 described the same process steps for preparing a catalyst as the opposed patent, with the sole exception of the feature in step (b) concerning moisture control during reduction in a range of 50 to 5000 ppm by periodical purging with nitrogen, which was absent from D1. The opposition division acknowledged that the comparison between example 1 and comparative example 5 and between

example 5 and comparative example 10 of the opposed patent showed improved properties in terms of activity, selectivity and surface area for the catalysts of examples 1 and 5 which had been prepared according to the claimed process with moisture control during the reduction step. The technical problem was to improve the efficiency and active life of a catalyst of a noble metal on a carrier. The proposed solution consisted in controlling the moisture content during reduction by periodical purging with nitrogen as defined in step (b) of claim 1. The opposition division came to the conclusion that none of the prior-art documents cited in the opposition proceedings could lead the skilled person to this solution, which therefore involved an inventive step.

VI. The opponent (appellant) lodged an appeal against that decision. With the statement setting out the grounds of appeal, the appellant submitted document D7.

D7: Ullmann's Encyclopedia of Industrial Chemistry
Vol. A5, VCH Verlagsgesellschaft, Weinheim, Germany,
5th edition 1986, pp. 350-353

VII. With the reply to the statement of the grounds of appeal, dated 4 August 2010, the patentee (respondent) submitted a first auxiliary request.

VIII. The parties were summoned to oral proceedings. In a preparatory communication, the board observed that a comparison of the experimental data presented in D1 and in the patent in suit raised questions which might be relevant for the analysis of inventive step. The process features of examples 1 to 7 of D1 corresponded to those of examples 1 to 7 in the patent in suit, with the sole exception of the feature of moisture control

during reduction, which was not mentioned in D1. In spite of that difference, the results in terms of parametric properties of the catalysts obtained were identical in D1 and in the patent in suit. When the process was however repeated in the patent in suit while omitting the process step of moisture control during reduction (as in comparative example 5, which should accordingly be a repetition of example 1 as described in D1), a catalyst with less favourable properties was obtained. The board noted that in order to facilitate the assessment of the available experimental evidence, the respondent, who was also the applicant of D1, was expected to be able to explain the apparent discrepancy in the experimental results presented in D1 and in the patent in suit, and in particular to clarify whether the disclosure in D1 was incomplete.

- IX. Oral proceedings were held before the board on 15 November 2012. The appellant was not present, as announced by letter of 23 October 2012.
- X. The appellant's arguments can be summarised as follows:
The claimed process, comprising process steps (a) to (f), lacked novelty over the process disclosed in examples 1 to 7 of D1. The catalysts prepared according to examples 1 to 7 of the patent in suit were respectively identical, in their composition and properties (activity, surface area of metals and selectivity), to the catalysts prepared according to examples 1 to 7 of D1. The description of the process for preparing said catalysts was the same in D1 and in the patent in suit, except for the feature of controlling the moisture content during reduction step (b), which was foreseen in the patent in suit but not

explicitly mentioned in D1. That process feature was nevertheless implicitly disclosed in D1: since the same catalysts with identical properties were obtained in D1 and in the patent in suit, the process of preparation had to be identical in that it involved removal of moisture to the same extent.

Alternatively, if the feature of moisture control during reduction were to be regarded as a distinguishing feature over the closest prior art D1, then it merely represented the skilled person's common general knowledge that it was necessary to remove water vapour formed during the reduction of the oxidative state metals with hydrogen in order to shift the reversible reaction towards the product side of the equilibrium. In this context, the appellant referred to D7 (see paragraph bridging pages 351 and 352) which advised keeping the partial pressure of water low in order to increase the reduction potential and to minimise hydrothermal growth of the crystallites of oxidic catalyst precursors. The skilled person would in any case routinely take measures to reduce the moisture content during the reduction step. Claim 1 of the patent in suit differed from the corresponding process of D1 merely by defining means to remove water vapour (periodical purging with nitrogen) and by defining the extent of the moisture control (50 to 5000 ppm). No surprising advantage was obtained by those features, as evidenced by the fact that the catalysts prepared according to the examples of D1 were identical to those prepared according to the patent in suit. Periodical purging with nitrogen gas to keep the moisture content as low as possible was just one of several straightforward options the skilled person would have considered in such a case, as an alternative embodiment

to carrying the moisture away in the continuous stream of hydrogen gas used for reduction (as suggested in documents D3 and D4), or in a stream of hydrogen gas diluted with nitrogen (as foreseen in D7). In consequence, the proposed solution did not involve an inventive step.

XI. The respondent's arguments can be summarised as follows:

Document D7, cited by the appellant as a new reference, was not *prima facie* highly relevant, and not more relevant than any of the documents already cited in the opposition proceedings. Therefore D7 should not be admitted into the appeal proceedings.

As to novelty, D1 did not contain a direct and unambiguous disclosure of the feature "controlling the moisture content in the range of from 50 to 5000 ppm by periodical purging of the reducing system with nitrogen to remove water vapour produced during reduction". Nor could it be assumed that the results in D1 were down to controlling the moisture content to be in the range of 50 to 5000 ppm, when this feature was not recited in D1, which was silent on the issue of moisture control.

The disclosure of D1 was, in fact, incomplete: in the preparation of the catalysts according to the examples of D1, the moisture content had actually been controlled in a range of 50 to 5000 ppm by periodical purging with nitrogen during reduction of the metals with hydrogen in gaseous phase. This explained why the resulting catalyst data were the same as in the patent in suit. Although the step of moisture control by periodical purging did happen, it was however not disclosed in document D1 and thus it was not derivable from the information in D1. The test results presented in D1 should therefore be ignored. What was relevant

for the assessment of inventive step was the direct comparison presented in the patent in suit. D1, excluding the test results presented therein, was the closest prior art. The technical problem to be solved in view of D1 was to provide a catalyst which had improved efficiency and activity. The solution to this problem involved carrying out the reduction step of the process while controlling the moisture content of the reducing system in a range of 50 to 5000 ppm by periodical purging with nitrogen to remove water vapour produced during reduction. The comparison of example 1 with comparative example 5 and of example 5 with comparative example 10 of the patent in suit showed that metal catalysts with increased activity, selectivity and surface area were obtained when the moisture content of the reducing system was controlled in accordance with the claimed invention. The proposed solution was not obvious, as there was no teaching or suggestion of it in the prior art. Neither D1 nor any of D3, D4 or D7 disclosed or suggested periodical purging with nitrogen to remove water vapour, or a moisture content in the range of 50 to 5000 ppm.

- XII. The appellant (opponent) requested that the decision under appeal be set aside and that European patent No. 1 205 246 be revoked.
- XIII. The respondent (patentee) requested that the appeal be dismissed or, in the alternative, that the patent be maintained on the basis of the claims of the first auxiliary request submitted with letter dated 4 August 2010. The respondent also requested that D7 not be admitted into the proceedings.

Reasons for the Decision

1. The appeal is admissible.
2. Admission of document D7
 - 2.1 Document D7 was filed early in the appeal proceedings, together with the appellant's statement of the grounds of appeal, in conformity with Article 12(2)(a) of the Rules of Procedure of the Boards of Appeal of the EPO, in support of the applicant's argumentation about the skilled person's prior knowledge. Document D7 concerns the preparation of metal catalysts by reduction of oxidative state precursors (see page 351, paragraph 5.9). The filing of D7, which recommends dilution of hydrogen with nitrogen as an inert gas in the reduction step, was evidently a reaction to the decision of the opposition division, which had expressed the view that documents D3 or D4, both teaching the fast removal of reaction water due to a high hydrogen space velocity, did not suggest purging with an inert gas.
 - 2.2 In view of this, the board finds it appropriate to exercise its discretion under Article 114(2) EPC by admitting D7 into the proceedings.
3. Main request - novelty
 - 3.1 In the decision under appeal, novelty of the subject-matter of claim 1 was acknowledged with respect to documents D5 and D6. Since that part of the decision was not contested by the appellant, the board is not required to take a position on the issue.
 - 3.2 D1 is the only document invoked by the appellant in the appeal proceedings as being prejudicial to the novelty of the claimed process.

3.3 The process of preparing a catalyst as described in D1 involves process steps conforming to each of process steps (a) to (f) defined in claim 1 of the patent in suit (see D1: claim 1, paragraphs [0012] and [0018]), except that in the context of the reduction of the oxidative state metals with hydrogen (step (b)) document D1 neither mentions control of the moisture content of the reducing system, nor does it mention periodical purging with nitrogen (see D1: claim 1, example 1/step 4, paragraphs [0009]-[0011]). Since this evaluation of D1 was agreed by the parties it is not necessary to analyse it in further detail.

3.4 Although the respondent (who is also the applicant of D1) confirmed that the specific moisture control measures as defined in step (b) of the patent in suit were indeed also carried out when preparing the catalysts according to the examples of D1, this information could nevertheless not have been derived from the disclosure of D1. Since moisture control or variation of moisture in the reducing system are not discussed in D1 at all, the skilled person reading said document could not have inferred a mandatory step of controlling the moisture content in the specific range of from 50 to 5000 ppm by the specific method of periodical purging of the reducing system with nitrogen to remove water vapour produced during reduction.

3.5 The fact that the catalyst properties reported for some of the examples of D1 are the same as the properties reported for the corresponding examples of the patent in suit cannot lead to a different conclusion, for the following reasons:

Before the filing date of the patent in suit, the skilled person had no knowledge of the experimental data presented in the examples of the patent and could

not have effected a comparison with the examples of D1 for the purpose of interpreting the disclosure of D1. Nor could such a comparison lead to any conclusive result. Even if the catalysts obtained according to examples 1 to 7 of D1 appeared to be identical to the catalysts obtained according to the corresponding examples 1 to 7 of the patent in suit (cf. table 1 of the patent specification and table 1 in D1) this would not necessarily imply that their processes of preparation must have been identical. While identical processes will indeed yield identical products, identical products may be prepared by different processes. In consequence, the disclosure of the mere parametric properties of the catalysts obtained according to the examples of D1 cannot be regarded as constituting a direct and unambiguous disclosure in D1 of the required specific process feature of moisture control during reduction.

3.6 Hence, the missing feature concerning control of the moisture content of the reducing system in a range of 50 to 5000 ppm by periodical purging with nitrogen is neither explicitly nor implicitly disclosed in D1.

3.7 As a consequence, the subject-matter of the claims of the main request is novel over the disclosure of D1.

4. Main request - inventive step

4.1 The patent in suit concerns a process for the preparation of a supported catalyst comprising a noble metal as main component and a further specified metal as catalysis promoter in combination with an alkali or alkaline earth metal compound. The catalyst is suitable for producing alkenyl acetates, in particular vinyl acetate or allyl acetate, through reaction of olefins, acetic acid and oxygen in vapour phase.

4.2 According to the patent in suit (paragraphs [0006] and [0007]), by means of the claimed process a catalyst is obtained which has a high metal surface area and exhibits high catalytic activity; thus the catalytic efficiency and life of the heterogeneous catalyst are improved.

Closest prior art

4.3 Document D1 has been regarded as the closest prior art both in the decision under appeal and in the parties' submissions. The board does not see any reason to differ.

4.4 D1 seeks to prepare the same type of supported noble metal catalyst as the patent in suit and addresses the same technical problem. D1 states that the catalyst prepared therein has a high surface area and exhibits high catalytic activity, so that the catalytic ability and life of the heterophase catalyst are improved (see paragraphs [0006], [0007]).

4.5 D1 (see in particular example 1, paragraphs [0018] and [0012]) also discloses the same preparation process as defined in claim 1 of the patent in suit by process steps (a) to (f), except that D1 does not mention controlling the moisture content in the range of from 50 to 5000 ppm by periodical purging of the reducing system with nitrogen to remove water vapour produced during reduction. This feature is however mandatory according to claim 1, step (b), of the patent in suit.

Objective technical problem

4.6 In order to determine the objective technical problem, the question has to be answered whether it can be confirmed that the alleged advantage (i.e. more favourable catalyst properties) is obtained with the

claimed process involving the mandatory moisture control measures defined in step (b) of claim 1, in comparison with the process which is effectively disclosed in D1, viz. a process not defining such mandatory measures.

4.7 Example 1 and comparative example 5 of the patent in suit each describes the preparation of a supported palladium/gold catalyst and its use in preparing vinyl acetate. Example 5 and comparative example 10 of the patent in suit each describes the preparation of a supported palladium/copper catalyst and its use in preparing allyl acetate. In examples 1 and 5, the catalyst was prepared in accordance with claim 1 by controlling the moisture content in the reducing system at 800 + 200 ppm by periodical purging of the reducing system with nitrogen gas. In comparative examples 5 and 10, the moisture content of the reducing system was not controlled and was above 5000 ppm. Apart from this difference, the comparative examples were carried out in the same way as examples 1 and 5, respectively. The comparison is thus deemed to be an adequate comparison with the process of D1 (see D1: paragraph [0018]).

4.8 The catalysts obtained according to examples 1 and 5 of the patent in suit showed a higher surface area of metals and a higher activity, and to some extent also a higher selectivity, than the corresponding catalysts of comparative examples 5 and 10 (cf. table 1 of the patent in suit). Thus the integration, into the process disclosed in D1, of moisture control as defined in step (b) of the patent in suit allows the skilled worker to obtain catalysts with more favourable properties.

4.9 The objective technical problem can therefore be defined as the provision of a process for preparing supported noble metal catalysts with improved activity and efficiency.

4.10 That problem has been solved by the process defined in claim 1, as confirmed by the comparative test results presented in the patent in suit.

Obviousness of the solution

4.11 In order to decide whether the suggested solution was obvious in the light of the prior art, the question to be answered is whether starting from the process which is disclosed in the closest prior-art document D1, the skilled person would have introduced the specific moisture control measures defined in step (b) of claim 1 of the patent in suit, in order to solve the objective technical problem of providing a process for preparing supported noble metal catalysts with improved activity and efficiency.

4.12 Document D1 by itself gives no indication that, in connection with the process disclosed therein, specific measures for controlling the moisture content of the reducing system might be required, and effective, to obtain more favourable catalyst properties. Even if such moisture control was in fact carried out in the context of the process examples of D1, the skilled person could not have derived this information from the content of D1, where it is neither explicitly nor implicitly disclosed (see paragraphs 3.4 to 3.6 *supra*).

4.13 Documents D3 (see page 101) and D7 (see point 5.9 on pages 351 to 352) both address the preparation of general unspecified metal catalysts from oxidative state precursors by reduction with hydrogen, while the teaching of document D4 (see point 6.4.5 on pages 117

to 120) appears to be restricted to nickel catalysts. All three documents acknowledge the fact that water is produced during the reversible reduction of oxidative state metal with hydrogen, and that the partial pressure or content of water should be kept at a low level so as not to hinder the reduction, i.e. the activation of the catalyst. To that end, it is suggested to use low heating rates (D3), and to use a high space velocity of the gaseous reducing agent, hydrogen, to carry away the reaction water (D3, D4). None of documents D3, D4 or D7 specifies a particular range for the moisture content. Removal of the reaction water in the context of D3 and D4 is effected with the regular gas stream of the reducing agent hydrogen, while D7 does not indicate any specific means for reducing the water content. Nitrogen is mentioned in D7 for the purpose of diluting the reducing agent hydrogen in order to reduce the heat generated by the reaction, but not in connection with any additional purging steps. Thus the specific measures for controlling the moisture content which are defined in claim 1, step (b) of the patent in suit are not suggested by those documents.

- 4.14 Although the skilled person was thus in general aware that the removal of reaction water during the reduction of oxidative state metals might be of potential benefit, the prior art contains no indication that working in the specific concentration range of 50 to 5000 ppm and using periodical, i.e. intermittent, purging steps instead of a continuously high gas flow would be effective, in the case of supported noble metal catalysts of the type described in the patent in suit, to achieve the desired improvement of catalyst properties.

4.15 As a consequence, the proposed solution is not obvious when considering D1 either alone or in combination with D3, D4 and D7, and the subject-matter of the claims of the main request involves an inventive step.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

S. Fabiani

D. Semino