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Datasheet for the decision of 5 June 2007

Case Number:	T 0964/02 - 3.3.07
Application Number:	99303655.7
Publication Number:	0962253
IPC:	B01J 37/02
Language of the proceedings:	EN

Title of invention:
A process for preparing a multi-metal oxide catalyst

Applicant:
ROHM AND HAAS COMPANY

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56

Keyword:
"Novelty (yes)"
"Inventive step (yes)"

Decisions cited:
-

Catchword:
-

Case Number: T 0964/02 - 3.3.07

DECISION

of the Technical Board of Appeal 3.3.07
of 5 June 2007

Appellant:
ROHM AND HAAS COMPANY
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Representative:
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 15 May 2002 refusing European application No. 99303655.7 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: S. Perryman
Members: B. ter Laan
F. Rousseau

Summary of Facts and Submissions

I. European patent application No. 99 303 655.7, filed on 11 May 1999 and published on 8 December 1999 under No. 0 962 253, was refused by a decision of the Examining Division of the European Patent Office announced on 29 April 2002 at the end of oral proceedings and issued on 15 May 2002.

Claims 1, 10 and 14 as originally filed read:

"1. A process for preparing a catalyst comprising:
(A) admixing metal compounds, at least one of which is an oxygen containing compound, and at least one solvent to form a solution;
(B) removing the solvent from the solution to obtain a catalyst precursor; and
(C) calcining the catalyst precursor at a temperature from 350°C to 850°C under an inert atmosphere, to form a catalyst having the formula

$$A_aM_mN_nX_xO_o$$

wherein $0.25<a<0.98$, $0.003<m<0.5$, $0.003<n<0.5$, $0.003<x<0.5$ and o is dependent on the oxidation state of the other elements, and A is selected from Mo, W, Fe, Nb, Ta, Zr, Ru, and mixtures thereof; M is selected from V, Ce, Cr, and mixtures thereof; N is selected from Te, Bi, Sb, Se, and mixtures thereof; and X is selected from Nb, Ta, W, Ti, Al, Zr, Cr, Mn, Fe, Ru, Co, Rh, Ni, Pd, Pt, Sb, Bi, B, In, Ce and mixtures thereof."

"10. A catalyst prepared according to the process of claim 1."

"14. A process for preparing an unsaturated aldehyde or carboxylic acid comprising subjecting an alkane to catalytic oxidation in the presence of a catalyst prepared by the process of claim 1."

II. According to the decision, page 2, the main request was based on claims 1 to 7 filed on 25 March 2002, which, according to the minutes, had been amended during the oral proceedings before the opposition division. Also according to the decision, page 2, when read in combination with page 6 (point III.1), the auxiliary request had the same claims as the main request but with claim 1 restricted to a process for preparing an unsaturated aldehyde or carboxylic acid comprising subjecting an alkane to catalytic oxidation in the presence of a catalyst prepared by the process according to claim 1 of the main request. No sets of claims in writing appear to be on file to give explicit confirmation of what the main request and the auxiliary request decided on were.

Claim 1 of the main request of 25 March 2002 read:

"1. A process for preparing a catalyst comprising:
(A) admixing metal compounds, at least one of which is an oxygen containing compound, and at least one solvent to form a solution;
(B) removing the solvent from the solution to obtain a catalyst precursor; and
(C) calcining the catalyst precursor at a temperature from 350°C to 850°C under an inert atmosphere, to form a catalyst having the formula

$$A_aM_mN_nX_xO_o$$

wherein $0.25<a<0.98$, $0.003<m<0.5$, $0.003<n<0.5$, $0.003<x<0.5$ and o is dependent on the oxidation state of the other elements, and A is selected from Mo and mixtures thereof; M is selected from V and mixtures thereof; N is selected from Te, Se, and mixtures thereof; and
X is selected from Nb, Ta, W, Ti, Al, Zr, Cr, Mn, Fe, Ru, Co, Rh, Ni, Pd, Pt, Sb, Bi, B, In, Ce and mixtures thereof; characterized in that the inert atmosphere is not flowing over the surface of the catalyst precursor, and wherein greater than 95% of metal solid added to the solvent in step (A) is dissolved."

III. The Examining Division held that the subject-matter of both the main and the auxiliary request satisfied the requirements of Article 54 EPC, but that they lacked an inventive step (Article 56 EPC) in view, *inter alia*, of the following documents:

D1: US-A-5 380 933
D2: EP-A-0 529 853

In particular, it was found that D1 was the closest prior art document and the technical problem to be solved was to find a process for preparing a catalyst with improved selectivity, conversion and yield in a process for preparing an aldehyde or carboxylic acid by subjecting an alkane to oxidation. Although catalysts when containing Mo, V, Te and Nb were considered to be inventive, the claim was not restricted to such catalysts, since Nb could also replaced by numerous other elements listed as X in claim 1, having widely different and not necessarily comparable or predictable properties. The effect obtained with a catalyst containing Nb was not necessarily also obtained with a catalyst containing another element than Nb. In the absence of additional evidence that compounds not containing Nb were even effective catalysts, it could not be accepted that the problem had been solved over the whole scope of the claim. The same reasoning was valid for the auxiliary request. Therefore, the claimed subject-matter lacked an inventive step.

The decision also refers to objections under Article 123(2) EPC, but fails to identify those. In particular, it is not clear whether the objections concern the main or the auxiliary request. The minutes of the oral proceedings state that an arbitrary selection of metals was not supported by the description and from the context it would appear that that was the case with both requests on file at that time.

IV. On 16 July 2002 a Notice of Appeal was lodged against that decision, together with payment of the prescribed fee. With the Statement of Grounds of Appeal filed on 7 September 2002, the Appellant submitted a set of eleven claims as the main request and a set of eight claims as an auxiliary request.

After a communication from the Board in which several problems under Articles 123(2), 84, 83, 54, 56 and 82 EPC were addressed, the appellant, with a letter dated 14 May 2007, filed two new sets of claims of five and four claims respectively, replacing the claims then on file. Attached to the letter were also a declaration by Scott Han, PhD and copies concerning the Periodic Table of the Elements and the effective ionic radii of various elements.

V. At the oral proceedings before the Board, held on 5 June 2007, after discussion of the claimed subject-matter, those claims were again replaced by a new set of three claims as the sole request.

Claim 1 of that request, the only independent claim, reads as follows:

"A process for preparing a catalyst comprising:

(A) admixing metal compound, at least one of which is an oxygen containing compound, and at least one solvent;

(B) removing the solvent from the solution to obtain a catalyst precursor; and

(C) calcining the catalyst precursor at a temperature from 350°C to 850°C under an inert atmosphere to form a catalyst having the formula

$$A_aM_mN_nX_xO_o$$

wherein $0.25<a<0.98$, $0.003<m<0.05$, $0.003<n<0.05$, $0.003<x<0.05$, and o is dependent on the oxidation state of the other elements, and A is selected from Mo, W and mixtures thereof; M is selected from V, Ce, Cr and mixtures thereof; N is selected form Te, Bi, Sb and mixtures thereof; and X is selected from Nb, Ta, Zr and mixtures thereof, and having X-ray diffraction peaks at a diffraction angle of 2θ at 22.1°, 28.2°, 36.2° 45.2°, and 50.0°;

characterized in that:

in step A), the metal compounds and solvent form a solution wherein greater than 95% of metal solid added to the solvent is dissolved;
in step B), the solvent is removed by a process selected from rotary evaporation, vacuum drying, air drying and freeze-drying; and
in step C), the inert atmosphere is not flowing over the surface of the catalyst precursor."

VI. The Appellant's arguments submitted in writing and during the oral proceedings can be summarised as follows:

(i) Regarding Article 123(2) EPC, the appellant indicated the passages in the application as originally filed that provided the basis for the amendments.

(ii) As to Articles 82, 84, 83 and 54 EPC, the amendments to the claims resolved the problems indicated by the Board in its communication and during the oral proceedings.

(iv) As regards inventive step, the closest document was D1 or possibly D2. The problem solved was to find an improved process for preparing a catalyst for catalyzing an alkane into an unsaturated aldehyde or carboxylic acid. That problem had been effectively solved, as could be seen by the examples in the description. The present process differed from the process described in D1 in the amount of metal that should be dissolved, in the drying process and in the no-flow requirement for the calcination step. Nothing in D1 suggested to use those requirements in order to improve the catalyst preparation process. It had not been possible to repeat the examples of D1, but some of the examples of the present application could be used for comparison purposes, showing an improvement. Also D2 did not suggest to change the process of D1 in such a way as to arrive at the claimed subject-matter. Therefore, the claimed subject-mater was inventive.

VII. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of claims 1 to 3 submitted as the sole request at the oral proceedings held on 5 June 2007.

Reasons for the Decision

1. The appeal is admissible.

Amendments

2. Present claim 1 differs from its original version in
- the restriction of the possibilities for A, M, N and X, for which the basis can be found in the paragraph bridging pages 4 and 5 as originally filed (paragraph [0017], penultimate sentence, as published),
- the obligatory presence of X-ray diffraction peaks at an angle of 2θ, the basis for which can be found on original page 21, lines 15 to 17 (paragraph [0053] as published),
- the requirement that greater than 95% of the metal should be dissolved (original page 4, last full paragraph; paragraph [0016] as published),
- that certain methods for removing the solvent are required (original claim 8) and

- that the inert atmosphere should not flow over the surface of the catalyst (original claim 7).

The wording of the cited passages also provides sufficient basis for the combination of the amended features.

The requirements of Article 123(2) EPC are therefore fulfilled.

3. The Board has no objections regarding clarity (Article 84 EPC), unity (Article 82 EPC) or disclosure (Article 83 EPC).

Novelty

4. D1 discloses a method for producing an unsaturated carboxylic acid, which comprises subjecting an alkane to a vapour phase catalytic oxidation reaction in the presence of a catalyst containing a mixed metal oxide comprising, as essential components, Mo, V, Te, O and X wherein X is at least one element selected from the group consisting of niobium, tantalum, tungsten, titanium, aluminium, zirconium, chromium, manganese, iron, ruthenium, cobalt, rhodium, nickel, palladium, platinum, antimony, bismuth, boron, indium and cerium, wherein the proportions of the respective essential components, based on the total amount of the essential components exclusive of oxygen, satisfy the following formulas:

$0.25<r_{Mo}<0.98$

$0.003<r_V<0.5$

$0.003<r_{Te}<0.5$

$0.003<r_X<0.5$

wherein r_{Mo}, r_V, r_{Te} and r_X are molar fractions of Mo, V, Te and X, respectively, based on the total amount of the essential components exclusive of oxygen (claim 1).

The mixed metal oxide may exhibit X-ray diffraction (Cu - $K\alpha$) peaks at a diffraction angle of 2θ at 22.1°, 28.2°, 36.2° 45.2°, and 50.0° (claim 7).

The catalyst can be prepared by adding aqueous solutions of e.g. telluric acid, ammonium niobium oxalate and a solution or slurry of ammonium paramolybdate to an aqueous solution containing a predetermined amount of ammonium metavanadate, so as to arrive at the prescribed atomic ratio of the respective metal elements. The mixture so obtained is described in all the examples as being a slurry. It is then dried, and the dried product is calcined, usually at 350 to 700°C, usually during 0.5 to 30 hours. The calcination can be carried out in an oxygen atmosphere, but the absence of oxygen, specifically the presence of an inert gas or vacuum, is preferred (D1, column 3, lines 27 to 48).

Although D1 mentions that the use of an inert atmosphere during calcination is preferred, the use of an oxygen atmosphere is also possible. D1 does not disclose that, when an inert atmosphere is used, it should not flow over the surface of the catalyst during calcination. On the contrary, in the catalyst preparation example calcination takes place in a nitrogen stream.

Therefore, in order to arrive at the claimed subject-matter, the skilled person would have to select the metal solutions such that when mixed a solution is obtained, as well as non-oxygen and non-flow conditions for calcination. That combination of features is not clearly and unambiguously disclosed in D1.

4.1 D2 discloses a similar type of catalyst as in D1, which is however used for the production of a nitrile from an alkane. It is prepared by drying an aqueous solution containing the necessary metal compounds and then calcining in the absence of oxygen (Claim 5). According to example 1, upon mixing the metal compound solutions, as in D1, a slurry is obtained, but in example 4 a solution is mentioned. D2 describes the general conditions during calcining (page 4, lines 7 to 17), but it is silent regarding flow or non-flow conditions. However, in the examples a nitrogen stream is used.

Therefore, D2 does not disclose the present combination of features.

4.2 In view of the above, it is concluded that the process now being claimed is novel.

Inventive step

5. The application in suit concerns a process for preparing a multi-metal oxide catalyst. Processes for the preparation of multi-metal oxide catalysts are known from D1 and D2. Since D1 concerns catalysts for the production of unsaturated carboxylic acids, as does the application in suit, whereas D2 refers to catalysts for the production of nitriles, the Board agrees with the examining division and with the appellant that D1

is the most suitable starting point for assessing the presence of an inventive step.

5.1 According to the description of the present application (page 2, lines 32 to 34), the aim is to provide a process for the preparation of a catalyst for catalyzing an alkane into an unsaturated aldehyde or carboxylic acid wherein phase segregation is minimized and improvement in selectivity, conversion and yield are achieved.

5.2 From the examples it can be seen that the highest selectivity, conversion and yield are obtained with catalysts prepared from a solution and calcined under non-flow conditions (Table 1, examples 1, 2, 4, 10 and 12). In Table 1 no exact details regarding the composition of those catalysts, in particular the percentage of metal, are given, but from Table 2 it can be seen that catalysts prepared under non-flow conditions, starting form the same amount of Te, retain a higher percentage of it. Thus, Table 2 supports the view given in the description (paragraph [0047] as published) that the non-flow conditions during calcination prevent the loss of Te from the catalytic material. It is however not clear if the improved catalyst performance in terms of conversion, selectivity and yield (Table 1, examples 10 and 11) would not be the consequence of the presence of a higher amount of Te in the catalyst and if the same results as in example 10 would not also be obtained in example 11 if a higher starting amount of Te were used.

5.3 Therefore, it can be concluded that the non-flow measure leads to a more reliable process for obtaining the catalyst composition sought, but not that the catalyst itself is improved.

5.4 Therefore, the problem that has been effectively solved by the claimed subject-matter can be seen as to provide an improved process for preparing a catalyst for catalyzing an alkane into an unsaturated aldehyde or carboxylic acid.

6. It remains to be decided whether the claimed subject-matter is obvious having regard to the documents on file.

6.1 The process described in D1 to prepare a catalyst for catalyzing an alkane into an unsaturated carboxylic acid entails the preparation of a solution or slurry and calcination, drying and calcining (see point 5 above). D1 does not in general state the nature of the mixture obtained when the mixing of all the necessary ingredients has been completed (column 3, lines 27 to 36), but in all the examples a slurry is mentioned. Likewise, there is no general statement that calcination should take place under flow conditions (column 3, lines 43 to 49), but in all the examples a nitrogen stream is used. Therefore, D1 does not contain any indication that the drying of a solution and calcining it under non-flow instead of flow conditions would have the advantages described in the present application. D1 alone does not render the claimed subject-matter obvious.

6.2 D2 concerns multi-metal oxide catalysts for the production of a nitrile from an alkane. Hence the skilled person would not look to D2 for suggestions to prepare a catalyst used in a different process, the production of unsaturated aldehydes or carboxylic acids.

Moreover, in example 1 of D2 a slurry is obtained upon mixing all the catalyst ingredients and in example 4 a solution. The other examples merely refer to example 1 (examples 2, 3, 7 to 23) or 4 (examples 5 and 6). The general conditions for calcination (page 4, lines 7 to 17) do not indicate whether flow or non-flow conditions should be used, let alone indicate any effect due to their use. In the examples, a nitrogen stream is used. Therefore, D2 contains no pointer to changing the process described in D1 in a way so as to arrive at the process now being claimed.

6.3 For the reasons given above, the subject-matter of claim 1 as well is its dependent claims 2 and 3 cannot in an obvious manner be derived from the cited art.

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 grant a patent on the basis of the three claims of the main request submitted at the oral proceedings on 5 June 2007 and a description yet to be adapted thereto.

Registrar	Chairman
K. Götz	S. Perryman