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Datasheet for the decision of 9 March 2010

Case Number: T 1404/06 - 3.3.05

Application Number: 97105316.0

Publication Number: 0799795

IPC: C01B 25/37

Language of the proceedings: EN

Title of invention:
Vanadium-phosphorus oxide, method for production thereof, catalyst for vapor phase oxidation formed of the oxide, and method for partial vapor phase oxidation of hydrocarbon

Patentee:
NIPPON SHOKUBAI CO., LTD.

Opponent:
BASF Aktiengesellschaft

Headword:
VPO Catalyst/NIPPON SHOKUBAI

Relevant legal provisions:
EPC Art. 123(2)

Relevant legal provisions (EPC 1973):
EPC Article 54 (1)(2)

Keyword:
"Main and 1st to 4th auxiliary request: Novelty (no)"
"5th auxiliary request: amended claims extend beyond the content of the application as filed"

Decisions cited:
-

Catchword:
-

Case Number: T 1404/06 **-** 3.3.05

DECISION
of the Technical Board of Appeal 3.3.05
of 9 March 2010

Appellant: (Patent Proprietor)	NIPPON SHOKUBAI CO., LTD. 1-1, Koraibashi 4-chome Chuo-ku Osaka-shi Osaka-fu 541 (JP)
Representative:	Luderschmidt, Schüler & Partner Patentanwälte Postfach 3929 D-65029 Wiesbaden (DE)
Respondent: (Opponent)	BASF Aktiengesellschaft -Patentabteilung - C6- Carl-Bosch-Strasse 38 D-67056 Ludwigshafen (DE)
Representative:	-
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted 4 July 2006 revoking European patent No. 0799795 pursuant to Article 102(1) EPC 1973.**

Composition of the Board:

Chairman:	G. Raths
Members:	J.-M. Schwaller S. Hoffmann

Summary of Facts and Submissions

I. This appeal was lodged by the proprietor (hereinafter "the appellant") against the decision of the opposition division revoking European patent 0 799 795.

II. During the opposition proceedings, the parties relied *inter alia* upon the documents:

D1: C. Busca et al., Journal of Catalysis, 99 (1986), pages 400 to 414;

D4: P. M. Michalakos et al., Ind. Eng. Chem. Res., 34 (1995), pages 1994 to 2000;

D11: WO 95/29006.

III. In the contested decision, the opposition division concluded that:

- claim 7 of the main request, claim 7 of the first auxiliary request as well as claim 3 of the second auxiliary request contravened Article 123(3) EPC;

- claim 1 of the sixth, seventh and eight auxiliary requests lacked novelty in particular over document D11.

The third, fourth and fifth auxiliary requests then on file had been withdrawn during the oral proceedings before the opposition division.

IV. Along with the grounds of appeal dated 13 November 2006, the appellant filed six sets of claims as the main and

1st to 5th auxiliary requests, respectively, with the first four requests corresponding respectively to the main, 6th, 7th and 8th auxiliary requests on which the contested decision was based.

Independent claim 4 of the main request reads as follows:

"4. A method for the production of a vanadium-phosphorus oxide possessing the nature of having an X-ray diffraction spectrum (Cu-K•) showing main peaks of the diffraction angle 2• (± 0.2°) at 18.5°, 23.0°, 28.4°, 29.9°, and 43.1° and having the intensity ratio of the peaks of the diffraction angle 2• (± 0.2°) at 23.0° and 28.4° in the following range

$$0.3 \bullet I(23.0)/I(28.4) \bullet 0.7$$

wherein I(23.0) and I(28.4) respectively represent the intensities of the peaks of the diffraction angle 2• (± 0.2°) at 23.0° and 28.4°, which method comprises reducing a pentavalent vanadium compound in an organic solvent, then causing the reduced compound to react with a phosphorus compound at a temperature in the range of 60°-150°C, and firing the resultant reaction product."

Claim 1 of the first auxiliary request reads as follows:

"1. A method for the partial vapor phase oxidation of a hydrocarbon with a molecular oxygen-containing gas by the use of a vanadium-phosphorus oxide having an X-ray diffraction spectrum (Cu-K•) showing main peaks of the diffraction angle 2• (± 0.2°) at 18.5°, 23.0°, 28.4°, 29.9°, and 43.1° and having the intensity ratio of the

peaks of the diffraction angle 2• (± 0.2°) at 23.0° and 28.4° in the following range

0.3 • I(23.0)/I(28.4) • 0.7

wherein I(23.0) and I(28.4) respectively represent the intensities of the peaks of the diffraction angle 2• (± 0.2°) at 23.0° and 28.4°, and being produced either by causing a tetravalent vanadium compound to react with a phosphorus compound in an organic solvent at a temperature in the range of 60°-150°C and firing the resulting reaction product or by reducing a pentavalent vanadium compound in an organic solvent, then causing the reduced compound to react with a phosphorus compound at a temperature in the range of 60°-150°C, and firing the resultant reaction product."

Claim 1 of the second auxiliary request corresponds to claim 1 of the first auxiliary request with the additional feature that "*the atomic ratio of vanadium/phosphorus is in the range of 1/0.9-1/1.2*".

Claim 1 of the third auxiliary request corresponds to claim 1 of the first auxiliary request with the additional features that "*the hydrocarbon is butane and the partial oxide is maleic anhydride*".

Independent claim 1 of the fourth auxiliary request corresponds to claim 1 of the first auxiliary request with the two temperature ranges (60° - 150°C) being each restricted to the range "*80° - 140°C*".

V. Under cover of a letter dated 13 February 2007, the respondent raised objections under Article 54(1) and (2) EPC to claim 4 of the main request as well as to claim 1 of the first to fourth auxiliary requests, respectively. It based its objections in particular on document D11.

It further held claims 1 to 3 of the fifth auxiliary request to contravene Article 123(3) EPC.

VI. With a letter dated 1 October 2007, the appellant filed an amended set of seven claims as a fifth auxiliary request, which set comprised three independent claims, of which claim 3 reads as follows:

"3. A vanadium-phosphorus oxide having an X-ray diffraction spectrum (Cu-K•) showing main peaks of the diffraction angle 2• (± 0.2°) at 18.5°, 23.0°, 28.4°, 29.9°, and 43.1° and having the intensity ratio of the peaks of the diffraction angle 2• (± 0.2°) at 23.0° and 28.4° of 0.6, wherein said vanadium-phosphorus oxide is obtainable by a method comprising the following steps:
a) suspending 400 g of vanadium pentoxide (V_2O_5) in 4000 ml of benzyl alcohol and stirring and meanwhile heating at 130°C and leaving reducing for 2 hours to effect thorough dissolution of vanadium pentoxide;
b) preparing a phosphoric acid solution by dissolving 477.4 g of 99% orthophosphoric acid in 1000 ml of benzyl alcohol and keeping at 80°C;
c) heating at 110°C for 10 hours the blackish blue solution of reduced vanadium and the phosphoric acid solution added thereto at 80°C, thereby producing a dark blue precipitate;
d) cooling the reaction solution slurry and separating the formed precipitate;
e) washing the precipitate with acetone and drying at 140°C for 12 hours;

f) forming the resultant dry mass into pellets, 5 mm in length and 5 mm in diameter;
g) calcining the pellets in a current of air at 500°C for 4 hours;
h) cooling to 400°C, sweeping with a current of a mixed gas consisting of n-butane and air and having a n-butane concentration of 1.5 % by volume, heating to 500°C at a temperature increasing rate of 1°C/minute, and activating at 500°C for 12 hours."

VII. Oral proceedings took place on 15 July 2009 in the absence of the appellant, as announced with its letter dated 11 March 2009. During the discussion, which in essence focused on the novelty and inventive step of the subject-matter claimed in the different requests on file, the respondent also objected under Article 123(2) EPC to independent claim 3 of the fifth auxiliary request. This objection having been raised for the first time at the oral proceedings, the board decided to continue the appeal proceedings in writing.

VIII. Following a communication dated 22 July 2009 wherein the board raised the question of the allowability under Article 123(2) EPC of claim 3 of the fifth auxiliary request, the appellant announced under cover of a letter dated 18 December 2009 that it did not intend to file any comment or amended claims.

IX. From the written submissions, the board establishes that the appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of the claims according to the main request dated 13 November 2006, or alternatively according to one of the first to fourth auxiliary requests dated 13 November 2006, or alternatively on the basis of the claims according to the fifth auxiliary request filed on 1 October 2007.

The respondent requested that the appeal be dismissed.

Reasons for the Decision

1. *Main request - Novelty*

1.1 The respondent argued that the subject-matter of claim 4 of this request lacked novelty over in particular catalyst 2-A-1 disclosed in document D11, which relates to a method of producing active vanadium-phosphorus mixed oxide catalysts for the commercial production of maleic anhydride by oxidation of aliphatic hydrocarbons in the vapor phase (D11, page 1, first paragraph).

1.2 The particular catalyst 2-A-1 of D11 is prepared as follows:

A 10-liter, four-neck, round-bottom flask, fitted with a mechanical stirrer with a 15 cm teflon paddle, a thermometer, a heating mantle, and a reflux condenser is charged with 6480 ml (5196 g) of isobutyl alcohol and 720 ml (750 g) of benzyl alcohol. After stirring is started (about 350 r.p.m.), 670 g (3.7 moles) of vanadium pentoxide (V_2O_5) is added. The mixture is heated to reflux - about 107°C - and maintained at reflux for 3 hours. After the initial reflux period the stirred mixture is cooled to about 20°C below the reflux temperature and 816 g (8.3 moles) of freshly prepared phosphoric acid (106% H_3PO_4) is added. The resultant mixture is again heated to reflux and maintained at reflux for 16 hours. This mixture is cooled to about 50°C and suction filtered to yield a bright blue cake. The blue solid is transferred to four open 2-liter dish trays and dried in a forced-draft oven at 150°C for 10 hours to yield about 1300 g of a grey-blue catalyst precursor powder. The resultant powder is passed with some pressing through a 65-mesh sieve, blended with approximately 4% by weight graphite, and 4 mm x 4 mm cylindrical tablets are formed in a Stokes-512 tabletting machine equipped with a die. 100 ml of tablets are then charged to a 50 mm diameter borosilicate tube and placed in a vertical Lindberg oven. Before starting the heating program, a 25% air/75% nitrogen gas mixture is passed (160 L/hr) through the catalyst bed. When the temperature of the tablets reaches 150°C, the gas mixture is replaced by a 25% air/25% nitrogen/50% steam gas atmosphere, using the same flow of 160 L/hr. The temperature is thereafter increased to 420°C at a programmed rate of 0.5°C/min. At the end of the heating program, the atmosphere is replaced by a flow of nitrogen and the calcined tablets cooled (D11, page 12, lines 13 to 33; page 13, lines 9 to 11 and 22 to 24; page 14, Table 1).

1.3 D11 does not disclose any X-ray diffraction pattern of the catalysts therein prepared, but the respondent reproduced the preparation protocol of the catalyst 2-A-1 as detailed on pages 12 to 14 of D11 and submitted the results of this experiment in Annex 1 to its letter dated 13 February 2007.

The appellant did not contest the validity of this experiment and the board does not see any reason either to cast doubt on it, as the reproduction of the catalyst 2-A-1 followed the preparation protocol as disclosed in D11, in particular the reaction temperature (called "reflux temperature" in D11) which was kept in the range of 87°C to 107°C (D11, page 12, line 19 and lines 21 to 22), i.e. according to the requirements of claim 4 under dispute.

The experimental results summarized in Annex 1 attached to the respondent's letter of 13 February 2007 show (see "Nachstellung 2" and Figure 2b) that the product reproduced as the catalyst 2-A-1 of D11 exhibits an X-ray diffraction pattern having main peaks at 18.5°, 22.9°, 28.5°, 30.0° and 43.3° with an intensity ratio of the peaks at 22.9° and 28.5° of 0.59. So, the catalyst 2-A-1 of D11 falls under the wording of claim 4 in dispute.

1.4 The appellant argued that claim 4 had to be understood as meaning that the vanadium compound reacted with the phosphorus compound under such conditions that, before mixing, **both** organic solutions were held at a temperature of 60 to 150°C, preferably 80 to 140°C (emphasis added by the board). In support of this interpretation of claim 4, it referred to Examples 1 to 3 of the patent in suit, which showed that both reactant organic solutions were heated before mixing. The appellant concluded that the preparation protocol of the catalyst 2-A-1 did not disclose the above characterising feature of claim 4.

The board disagrees with the way the appellant interprets claim 4 because apart from Examples 1 to 3, the patent in suit nowhere else refers to the temperature of the reagents before mixing, and in paragraphs [0035] or [0046] it clearly and unambiguously refers to the temperature of the **reaction mixture** (emphasis added by the board). Therefore, the claim in dispute should be understood merely as it reads, namely that it requires "the reduced compound to react with a phosphorus compound at a temperature in the range of 60 to 150°C".

1.5 For the above reasons, the board concludes that the method for producing the catalyst 2-A-1 of document D11 falls under the wording of claim 4 of the present request, which claim thus lacks novelty under Article 54(1) and (2) EPC 1973.

2. *First auxiliary request - Novelty*

2.1 Claim 1 of this request relates to **a method for the partial vapor phase oxidation of a hydrocarbon with a molecular oxygen-containing gas** using a vanadium-phosphorus oxide as defined in claim 4 of the main request and produced according to inter alia the method of claim 4 of the main request.

2.2 The board observes that the catalysts prepared according to the method disclosed in D11, such as the catalyst 2-A-1 described at pages 12 to 14, are described as being "suitable for commercial production of maleic anhydride by oxidation of aliphatic hydrocarbons in the vapor phase" (D11, page 1, paragraph "Field of the invention").

The catalyst 2-A-1 being - as indicated in items 1.2 to 1.4 supra - a vanadium-phosphorus oxide as defined in claim 4 of the main request and said catalyst having been moreover prepared according to the method as defined in claim 4 of the main request, document D11 thus also directly and unambiguously anticipates the subject-matter of claim 1 of the first auxiliary request, which is therefore not allowable under Article 54(1) and (2) EPC 1973.

3. *Second auxiliary request - Novelty*

3.1 Claim 1 of this request differs from that of the first auxiliary request in that the catalyst is further defined as having an **atomic ratio vanadium/phosphorus of 1/0.9-1/1.2.**

3.2 The experimental results - in particular the "Nachstellung 2" - referred to in items 1.3 and 1.4 above further establish that the product the respondent reproduced as the catalyst 2-A-1 exhibited a phosphorus/vanadium atomic ratio of 1.051, i.e. a vanadium/phosphorus atomic ratio of 0.951, which clearly and unambiguously falls under the wording of the claim 1 in dispute.

The other features of the process defined in the present claim 1 being - as indicated in items 2.1 and 2.2 above - also disclosed in combination in document D11, the board concludes that claim 1 of the second auxiliary request is not allowable under Article 54(1) and (2) EPC 1973 either.

4. *Third auxiliary request - Novelty*

4.1 Claim 1 of this request differs from that of the first auxiliary request in that the **hydrocarbon is butane and the partial oxide is maleic anhydride.**

4.2 As indicated in item 2.2 above, the catalysts prepared according to the method disclosed in D11 are suitable for the production of maleic anhydride by oxidation of aliphatic hydrocarbons in the vapor phase.

D11 (page 11, lines 5 to 12) further discloses that the performance of said catalysts, in particular the catalyst 2-A-1, has been evaluated in the reaction of n-butane with air.

The other features of the process defined in the present claim 1 being - as indicated in item 2. above - also disclosed in combination in document D11, the board concludes that the subject-matter of claim 1 of the second auxiliary request is also no longer novel and that claim 1 is not allowable under Article 54(1) and (2) EPC 1973 either.

5. *Fourth auxiliary request - Novelty*

5.1 Claim 1 of this request differs from that of the first auxiliary request in that the temperature range for the reaction of the vanadium compound with the phosphorus compound has been reduced from the range 60°-150°C to the range **80°-140°C** (fourth auxiliary request).

5.2 As indicated in item 1.4, the catalyst 2-A-1 has been reproduced according to the experimental protocol disclosed in D11, in particular with a reaction

<u>temperature</u> (called "reflux temperature" in D11) <u>kept in the range of 87°C to 107°C</u>.

Since this range of temperature falls entirely under the range defined in claim 1 of the present request, document D11 also directly and unambiguously anticipates the subject-matter of claim 1 of the fourth auxiliary request, which is therefore not allowable under Article 54(1) and (2) EPC 1973.

6. *Fifth auxiliary request - Allowability of the amended claims*

At the oral proceedings, the respondent argued that claim 3 of this request infringed the requirements of Article 123(2) EPC.

Having been questioned on this point by the board, the appellant did not comment on this issue, nor did it file amended claims. In its letters dated 13 November 2006 and 1 October 2007, the appellant argued that the subject-matter of independent claims 1, 2 and 3 had been limited to the vanadium-phosphorus oxides obtainable by the processes respectively described in Examples 1, 2 and 3.

The board observes that, while the subject-matter of independent claims 1 and 2 corresponds exactly and in all details to the respective disclosures of Examples 1 and 2 as filed, the subject-matter of independent claim 3 differs from the disclosure of Example 3 in one single detail, namely in that in step a) of claim 3 vanadium **pentoxide (V_2O_5)** is used as the starting vanadium component, whereas in Example 3 as filed, vanadium **dioxide (VO_2)** is disclosed as the starting component.

The application as filed does however make a marked distinction between the method of producing a vanadium-phosphorus oxide making use of a pentavalent vanadium compound (page 8, line 1 to page 10, line 16) and the method making use of a tetravalent vanadium compound as the starting material (page 10, line 17 to page 11, line 20). The claims as filed furthermore reflect this difference with, on the one hand, an independent claim 5 describing the method starting from a tetravalent vanadium compound, and on the other hand, an independent claim 8 describing the method starting from a pentavalent vanadium compound.

Since furthermore the vanadium-phosphorus oxide obtained in Examples 1 and 2 (produced from a **penta**valent vanadium compound) has an intensity ratio of the peaks of the diffraction angle 2• (± 0.2°) at 23.0° and 28.4° different from the vanadium-phosphorus oxide obtained in Example 3 (produced from a **tetra**valent vanadium compound), the board concludes that there is no basis in the application as filed for replacing the starting tetravalent vanadium dioxide (VO_2) material, originally described in Example 3, with the pentavalent vanadium pentoxide (V_2O_5).

It is therefore concluded that there is no basis in the application as filed for the amendment proposed in claim 3 of the request at issue. Claim 3 according to this request therefore contravenes Article 123(2) EPC.

7. Since each request on file includes at least one claim which does not meet the requirements of the EPC, none of the requests is allowable.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:	The Chairman:
C. Vodz	G. Raths