BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

Internal distribution code:
(A) [ ] Publication in OJ
(B) [ ] To Chairmen and Members
(C) [X] To Chairmen

DECISION
of 17 March 1999

Case Number: T 0729/95 - 3.3.5

Application Number: 88102079.6

Publication Number: 0279374

IPC: B01J 23/88

Language of the proceedings: EN

Title of invention:
Catalyst for oxidation of olefin or tertiary alcohol and process for production thereof

Patentee:
Nippon Shokubai Kagaku Kogyo Co., Ltd.

Opponent:
BASF Aktiengesellschaft, Ludwigshafen

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56

Keyword:
"Inventive step (no) - obvious to try"

Decisions cited:
-

Catchword:
-

Case Number: T 0729/95 - 3.3.5

DECISION
of the Technical Board of Appeal 3.3.5
of 17 March 1999

Appellant: (Proprietor of the patent)	Nippon Shokubai Kagaku Kogyo Co., Ltd 1, 5-chome, Koraibashi Higashi-ku Osaka-shi Osaka-fu 541 (JP)
Representative:	Kraus, Walter, Dr. Patentanwälte Kraus, Weisert & Partner Thomas-Wimmer-Ring 15 80539 München (DE)
Respondent: (Opponent)	BASF Aktiengesellschaft, Ludwigshafen -Patentabteilung - C6- Carl-Bosch-Strasse 38 67056 Ludwigshafen (DE)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted 3 July 1995 revoking European patent No. 0 279 374 pursuant to Article 102(1) EPC.**

Composition of the Board:

Chairman: R. K. Spangenberg
Members: M. M. Eberhard
J. H. van Moer

Summary of Facts and Submissions

I. European patent No. 0 279 374 based on application No. 88 102 079.6 was granted on the basis of two claims.

II. The Respondent (Opponent) filed a notice of opposition requesting revocation of the patent on the grounds of lack of novelty, lack of inventive step and insufficiency of disclosure. He relied inter alia on EP-B-0 015 565 (EH1) and EP-B-0 293 859 (GEH1). The latter document (published after the filing date of the patent application) was cited as evidence in support of the alleged insufficiency of disclosure.

III. The Opposition Division revoked the patent on the grounds of lack of novelty and lack of inventive step. It was held that the catalyst according to claim 1 as granted was not novel over the catalyst of example 1c of EH1. The process according to claim 2 was considered to lack an inventive step in view of the teachings of EH1 and GEH1.

IV. The Appellant (Proprietor of the patent) lodged an appeal against this decision. He submitted an amended claim, as a main request, together with the statement of grounds of appeal on 8 November 1995. In reply thereto, the Respondent filed an English translation (EH14) of JP-A-52/117 292. The sole claim of the main request reads as follows:

"A process for preparing a catalyst for producing, by catalytic gas phase oxidation of propylene, isobutylene or tertiary butanol, the corresponding unsaturated aldehyde and unsaturated carboxylic acid, said catalyst comprising the oxides of molybdenum, iron and bismuth and having a specific surface area in the range from 1 to 20 m^2/g, a pore volume in the range from 0.1 to 1.0 cc/g and a pore diameter distribution in which the pore diameters are collectively distributed in the range of each from 1 to 10 µm and from 0.1 to 1 (exclusive) µm, and having a composition represented by formula

$$Mo_aW_bBi_cFe_dA_eB_fC_gD_hO_x$$

wherein Mo denotes molybdenum, W denotes tungsten, Bi denotes bismuth, Fe denotes iron, A denotes at least one element selected from the group consisting of nickel and cobalt, B denotes at least one element selected from the group consisting of alkali metal, alkaline earth metal and thallium, C denotes at least one element selected from the group consisting of phosphorus, tellurium, antimony, tin, cerium, lead, niobium, boron, arsenic, manganese and zinc, D denotes at least one element selected from the group consisting of silicon, aluminum, titanium and zirconium, and O denotes oxygen; and further a, b, c, d, e, f, g, h and x denote atomic ratios respectively; and when the material to be oxidized is propylene, then a = 2 to 12, b = 0 to 10 and a + b = 12, C = 0,1 to 10, d = 0,1 to 10,0, e = 2 to 20, f= 0,005 to 3,0, g = 0 to 4,0, h = 0,5 to 15 and x is a numerical value determined depending upon the atomic values of the elements other than oxygen, and when the material to be oxidized is isobutylene or tertiary butanol, then a = 12, then b = 0 to 10, c = 0,1 to 10, d = 0,1 to 20, e = 2 to 20, f = 0 to 10, g = 0 to 4, h = 0 to 30 and x is a numerical value determined depending upon the atomic values of the elements other than oxygen,

comprising charging an unfired material powder into a centrifugal flow coating device to form particles having the average diameter of 2 to 10 mm and then firing the particles thereby to obtain the catalyst which has a specific surface area in the range from 1 to 20 m^2/g, a pore volume in the range from 0.1 to 1.0 cc/g and a pore diameter distribution in which the pore diameters are collectively distributed in the range of each from 1 to 10 µm and from 0.1 to 1 (exclusive) µm."

In a communication the Board informed the parties that the teaching of EH14 seemed to be relevant to the claimed process. The question was raised whether or not the use of an "unfired material powder" as the starting powder was a distinguishing feature with respect to the prior art.

V. Oral proceedings were held on 16 and 17 March 1999. During the oral proceedings, the Appellant filed several amended claims as auxiliary requests and finally maintained three auxiliary requests, each containing a single claim. The claim of auxiliary request I differs from the claim of the main request in that (i) the word "comprising" before the phrase "charging an unfired material powder" is replaced by the expression "characterized by" and (ii) the last line of the claim contains the additional features "with the proviso that the catalyst is not supported on an inert carrier". The claim of auxiliary request II is identical to the claim of auxiliary request I except for the additional proviso in the last line, which reads "with the proviso that the catalyst is not supported on an inert carrier or a core consisting of said catalyst". The claim of auxiliary request III differs from the claim of the main request in that the phrase "comprising charging an unfired material powder" is replaced by the phrase "characterized by charging a powder of an unfired oxide composition not shaped or pre-stage catalyst particle material composition not converted to oxide".

VI. The Appellant's arguments can be summarised as follows:

The claimed process involved the use of an unfired oxide composition or a composition which had not yet been converted to the oxide form. The Respondent's experiments showed that the use of oxides which had been calcined at a high temperature (e.g. 500°C) led to a catalyst having physical properties lying far outside the claimed ranges. Calcined oxides were used as starting powder for the forming step in the process of EH1 contrary to the claimed process. Thus, the skilled person could not have easily achieved the present invention on the basis of this teaching. Furthermore, EH1 did not mention the physical properties of the catalyst. As neither the ranges of physical properties leading to an excellent catalytic performance nor methods for obtaining them with good reproducibility were known before the priority date, the physical properties measured by the Respondent on the catalysts of example 1c were obtained only incidentally. Therefore, the novelty of the catalysts was not destroyed. The advantages of the claimed process with respect to the process of EH1 were that a catalyst exhibiting an excellent performance could be produced with good reproducibility.

EH14 was not a bar to the patentability of the claimed process since it did not disclose that a catalyst having a surface area, pore volume and pore diameter distribution lying within a certain range exhibited an excellent catalytic performance. In the process of EH14 a coating of catalytic material was deposited onto a granular core or carrier whereas, in the claimed process, the particles were formed without using such a core or carrier. EH14 disclosed on page 9 that a granular core of an active substance could be used; however, the claim according to auxiliary request II explicitly excluded the use of an active material as a core, and the process as claimed in auxiliary request III did not involve deposition onto a core or carrier. As the word "comprising" had been replaced by "characterized by", the addition of a carrier or core was excluded. The skilled person would not have used the centrifugal flow coating method of EH14 for **forming** particles of the catalyst since the said method was a **coating** method. Neither EH1 nor EH14 pointed towards the production of a carrier-free catalyst.

In reply to the Board's question as to how the expression "unfired material powder" had to be understood in the claim, the Appellant indicated at the beginning of the oral proceedings that an unfired material powder or a powder of an unfired oxide composition was a powder which had been treated at a temperature lower than the minimum calcination temperature of 450°C stated in the examples of the patent in suit. The Appellant also mentioned later on that an unfired powder might be a powder which had been heated at a temperature not above 200°C, ie the maximum temperature of the drying step.

VII. The Respondent argued inter alia as follows:

It was questionable whether the process as claimed in the main request was novel over the process of EH1. The Respondent's experiments of 21 August 1992 and 1 February 1999 proved that the physical properties measured on the catalyst of example 1c fell within the claimed ranges. On the basis of the Appellant's definition of the expression "unfired powder" the powder used as coating material in example 1c was to be regarded as an unfired powder. Furthermore, the claimed process was not limited to the use of the centrifugal flow coating device disclosed in EH14. In the absence of a definition for the "centrifugal flow coating method" in the claim, the question arose whether or not this method encompassed the coating method using a rotating pan as disclosed in EH1. If the claimed process were considered to be new, it would lack an inventive step. The advantages of the centrifugal flow coating method over conventional methods were disclosed in EH14. This document further recommended use of the centrifugal flow coating method for preparing supported catalysts suitable for the vapour-phase oxidation. The application of this method to the preparation of known supported catalysts such as those of EH1 could not involve an inventive step in view of the teaching of EH14.

The process as claimed in auxiliary request I was also obvious to the skilled person since EH14 further disclosed forming catalyst spheres by using the catalytic substance as cores. If the skilled person had accepted that the selectivity be somewhat lower than that obtained with a supported catalyst but still very good, he would have used the catalytic substance as cores instead of an inert carrier. The additional features in the claim according to auxiliary request II did not meet the requirements of Article 123(2) EPC. Furthermore, this claim still included the use of an unfired catalyst as a core and this embodiment was disclosed in EH14. Therefore, the claimed process also lacked an inventive step. The reasons given before in connection with the inventive step issue applied likewise to the process according to auxiliary request III.

VIII. The Appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of the claim submitted on 8 November 1995 or on the basis of any of the auxiliary requests I, II or III submitted during the oral proceedings. The Respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

2. The amended claim of the main request and of the auxiliary requests I and III meet the requirements of Article 123(2) and (3) EPC.

3. The translation (EH14) of JP-A-117 292/1977 was submitted for the first time in reply to the statement

of the grounds of appeal. The Japanese document was already cited in the search report and was briefly analysed on page 4 of the patent in suit. In the Board's judgement, this document is clearly relevant to the claimed process and, therefore, EH14 had to be introduced into the proceedings.

Main request

4. At the oral proceedings, the Respondent raised the question of the novelty of the claimed process with respect to the disclosure of example 1 of EH1.

EH1 discloses a process for preparing a supported catalyst which is used for the catalytic gas phase oxidation of propylene or isobutylene to acrolein or methacrolein. The supported catalyst of example 1 contains a commercial magnesium silicate sphere having a diameter of 1.5 to 2.5 mm and a porosity of about 1%, as a support, and a catalytic active coating having for example the composition $Mo_{12}Ni_{6.5}Zn_2Fe_2Bi_1P_{0.065}K_{0.06}O_x + 10 SiO_2$ (see page 5, line 36). According to claim 1, the thickness of the coating is from 150 to 1500 µm. The process for preparing this catalyst comprises spray-drying a suspension comprising the precursors of the catalytic material in the form of salts, calcining rods of the spray-dried material at a temperature of 360°C for 2 hours, milling the calcined material and adding methylcellulose to the powder. The support is coated with the resulting powder by charging the latter at a specific rate in a rotating pan containing the support while continuously spraying water. The coated support is then dried and finally calcined at a temperature of 600°C for 1.5 hours (see page 4, line 54 to page 5, line 42 and claim 1).

The catalyst composition indicated above falls within the formula stated in the claim of the main request and the sizes of the formed particles overlap. The wording of the claim does not exclude charging a carrier into the centrifugal flow coating device before charging the unfired material powder and, thus, preparing a supported catalyst as illustrated in a great number of examples. Therefore, the novelty of the claimed process depends in particular on (i) whether or not the powder used in example 1 of EH1 is an "unfired material powder" within the meaning of the patent in suit and (ii) whether or not the coating step in the rotating pan can be considered as a coating in a "centrifugal flow coating device".

4.1 The expression "unfired material powder" is not defined in the claim. According to page 4 of the patent in suit, the powder which is charged into the centrifugal flow coating device can be an "unfired oxide composition" or a "pre-stage catalyst particle material composition not converted to oxide". In examples XI to XXI the solid material was dried at a temperature of 200°C for 5 hours before milling and the firing (final calcination) of the shaped catalyst was carried out at 500°C for 6 hours. Therefore, at least in these examples the "unfired material powder" seems to be a pre-stage catalyst powder which is either only partially converted to the oxide form or not converted thereto. The patent in suit contains no explanation about the alternative where an "unfired oxide composition" is used. As pointed out by the Appellant himself, most of the oxides concerned are subjected to a high temperature treatment (e.g. 500°C) at the stage of their production. At the oral proceedings the Appellant argued that in the examples of the patent in suit a "fired" catalyst was a catalyst which had been treated at a temperature of 450°C or 500°C, whereas a "dried" powder was a powder which had been treated at a temperature of only 120°C or 200°C. Therefore, so the Appellant, it was derivable from the patent in suit that an "unfired oxide composition" was an oxide composition which had been treated at a temperature lower than the minimum firing temperature stated in the examples, ie lower than 450°C. The Appellant further argued in the course of the oral proceedings that the expression "unfired material powder" might also be construed as meaning a powder which had been treated at 200°C or less. This argument is, however, not consistent with the fact that most of the concerned oxides are usually subjected to a treatment at a high temperature at the stage of their production and with the construction previously suggested for the expression "unfired oxide composition". In these circumstances and in the absence of any limitation in the claim, the expression "unfired material powder" has to be construed in the broadest possible meaning suggested by the Appellant on the basis of the data in the patent in suit, ie a material powder or a powder of an oxide composition which has been treated at a temperature lower than 450°C.

The powder of catalytic active material used for the coating step in example 1 of EH1 was calcined at 360°C for 2 hours in the form of rods, ie a temperature **well below the temperature of 450°C** indicated above, and the coated support was finally calcined at 600°C for 1.5

hours. As argued by the Respondent and not disputed by the Appellant, the precursors used in example 1 were not completely converted into oxides under the operating conditions of the first calcination. In these circumstances, the use of an "unfired material powder" as indicated in the present claim cannot be considered as a distinguishing feature over the disclosure in example 1 of EH1.

4.2 Concerning the Respondent's argument that the coating method in a rotating pan as disclosed in EH1 might fall within the claimed centrifugal flow coating method, the Board observes the following. Although the material travels in spiral rings under centrifugal action in a rotating pan, the latter is usually not called a "centrifugal flow coating device". In a rotating drum or pan the material is subjected to a rolling motion. In the comparative examples of the patent in suit the agglomeration or coating method in a rotating drum or pan is not expressly mentioned; however, the skilled person would understand that the conventional "rolling particle-forming method" referred to in the comparative examples (see examples I-5, XI-5 etc.) is in fact the well-known method performed in a rotating drum or pan. For these reasons, the Board holds that the step of forming particles "in a centrifugal flow coating device" indicated in the claim cannot be considered to encompass the conventional agglomeration or coating method in a rotating pan. Therefore, the claimed process is novel over the disclosure of EH1.

5. Both parties agreed that of the documents cited during the opposition procedure, EH1 was the closest prior art, in particular example 1 thereof. The Board can

follow this approach. The supported catalyst prepared by the process disclosed in example 1 of EH1 (see point 4 above) exhibits a very good performance in the gas phase oxidation of propylene to acrolein under the operating conditions of reaction reported in example 1b (see page 5, lines 40 to 42). According to the patent in suit, catalyst pellets formed by the conventional forming methods such as the tablet-forming method, extrusion, pill-forming method or rolling particle-forming method show in most cases a poor reproducibility of the catalyst performance.

5.1 Starting from this prior art, the technical problem underlying the claimed process can be seen in the provision of a process for preparing catalysts having a very good performance in the oxidation of propylene, isobutylene or tertiary butanol to the corresponding unsaturated aldehyde and carboxylic acid, which process makes it possible to produce the catalysts with good reproducibility.

It is proposed that this problem be solved by the process as defined in the claim of the main request. The examples of the patent in suit show that forming the catalyst particles by the centrifugal flow coating method makes it possible to obtain relatively small variations of the physical properties compared to the conventional rolling method and also leads to catalysts having a high activity and selectivity. In view of these examples, the Board is satisfied that the problem stated above has actually been solved by the claimed process, at least when the inert carrier is not porous or has a very low porosity, or in the absence of inert carrier. This was not disputed by the Respondent for these particular alternatives. The question whether or not this problem has been solved in the case where the inert carrier has a high porosity need not be decided taking into account the outcome of the present decision on the main request.

5.2 EH1 itself discloses coating a non-porous support or a support having a porosity lower than 5%, for example 1%, in a particle forming device such as a rotating drum or pan, wherein the powder of catalytic active material is brought into contact with the support in rolling motion. The support is preferably in the form of spheres having a diameter of 1-5 mm, for example 1.5-2.5 mm (example 1) or 3mm (example 2), and the thickness of the coating is of 150 to 1500 µm, preferably 300 to 1200 µm. Although EH1 mentions only the rotating drum and the rotating pan as coating devices, they are cited as examples and other coating devices in which the support is subjected to a strong motion are not excluded (see page 3, lines 36 to 65).

5.3 EH14 discloses a process for the manufacture of granular catalysts comprising coating a granular core with a catalytic active substance in a centrifugal flow coating device. The granular core, for example a catalyst carrier, is charged into the said device and a powder of catalytic substance having a particle size of generally about 50 mesh (0.297 mm) or less is applied onto the surface of the core granules while blowing air and spraying a binder. The resulting catalyst particles, which have a diameter of for example 5 mm, are dried and activated by thermal treatment. The teaching of EH14 is not restricted to the preparation of catalysts suitable for a specific reaction or having a specific composition but is very broad and general. Catalyst carriers and supported or unsupported catalysts of very different compositions can be manufactured by the described method. This method is said to be particularly advantageous for the manufacture of catalysts used for reactions performed at a high space velocity such as exhaust gas treatment or oxidation reactions, which require a minimum pressure loss and a catalytic effectiveness factor close to 1 (see claim; Figures 1 to 4; page 2, 1st paragraph; pages 7 and 8; page 9, first and third paragraphs).

EH14 further teaches that the use of the centrifugal flow coating method solves the problems encountered with the conventional granulation methods which lead to uneven effectiveness factors, poor activity and selectivity due to uneven particle sizes, distorted shape and poor mechanical strength of the catalyst thus obtained. According to EH14, the centrifugal flow coating method also overcomes the drawbacks of the known coating methods in which a catalytic active substance is applied onto the surface of separately shaped cores, in particular the coating method in a rotating pan. With the latter method it is difficult to regulate the layer thickness so that an uneven thickness is obtained. The resulting catalyst is, likewise, not satisfying as regards the catalytic activity, selectivity and mechanical strength. Impregnation of a fluid on a carrier is said to lack reproducibility (see pages 2 and 3). On the contrary, the thickness of the coating layer can be easily regulated with the centrifugal flow coating method and there is no loss of effectiveness factor or selectivity. This method leads to catalysts having a uniform particle size, an excellent mechanical strength, and a coating layer with uniform thickness (see pages 2 and 3; page 8, third paragraph).

In view of this teaching about the advantages of the centrifugal flow coating method, in particular the possibility of better controlling the coating thickness than with a rotating pan, and considering that the method of EH14 is recommended for the manufacture of catalysts used for gas phase oxidation reactions carried out at a high space velocity and requiring a minimum pressure loss, the skilled person faced with the problem stated above would have been encouraged to try the said method for the production of oxidation catalysts having the composition disclosed in EH1. He would have expected that the better control of the layer thickness would make it possible to achieve the desired good reproducibility as compared to the conventional forming methods. If, as argued by the Appellant, it was not known before the priority date that the specific surface area, the pore volume or the pore size distribution have to lie within specific ranges in order for the catalyst to have a very good performance, then the fact that these physical properties are not mentioned in EH1 or EH14 would not have deterred the skilled person from trying the promising centrifugal flow coating method for preparing catalysts of the compositions disclosed in EH1.

5.4 The powder used for the coating step in example 1 of EH1 can be considered as an "unfired powder material" within the meaning of the patent in suit (see point 4 above). At the oral proceedings, the Appellant confirmed that by using the catalytic powder composition and the low-porosity support of example 1 of EH1 (or a non-porous support also disclosed in EH1) and coating the support by the centrifugal flow coating method of EH14 instead of the coating method in a rotating pan, the skilled person would have obtained a catalyst which exhibits physical properties lying within the claimed ranges after calcination. No additional measures would be necessary to achieve the said properties. In the absence of evidence to the contrary, this is credible at least in the cases where the support has a low porosity or is non-porous. However, the mere measurement of the specific surface area, pore volume and pore size distribution of the resulting catalysts is obvious to the skilled person. As these parameters represent three of the most usual parameters in the field of catalysis and are further known to have an influence on the catalyst performance, the skilled person would have considered measuring them, without exercising an inventive skill.

It follows from the above that the claim according to the main request does not meet the requirements of inventive step set out in Article 52(1) and 56 EPC and is not allowable.

Auxiliary request I

6. The parties both considered that EH1 also represented the closest prior art with respect to the process as claimed in this request. Starting from EH1, the technical problem underlying the claimed process is also to provide a process for manufacturing catalysts having a very good performance in the oxidation of

propylene, isobutylene or tertiary butanol to the corresponding aldehyde and carboxylic acid, which process makes it possible to produce the said catalysts with good reproducibility.

It is credible in view of the examples I to X of the patent in suit that the catalysts obtained by the claimed process exhibit a very good performance at least for the oxidation of propylene to acrolein and acrylic acid and that they can be produced with good reproducibility (see table 2 of the description). Therefore, in the absence of evidence to the contrary, the Board is satisfied that the said problem has actually been solved by the claimed process.

6.1 According to EH14 the centrifugal flow coating method can also be used for producing a catalyst sphere which is exclusively made of catalytic active substance, ie an unsupported catalyst. In this case, **the catalytic active substance is used as the core** on which the coating is deposited (see page 9, third paragraph). Therefore, in view of the teaching of EH14, the skilled person was aware that the advantages of the centrifugal flow coating method, in particular the uniformity of the particle size or coating thickness, are also obtained with a core of a material other than an inert carrier, for example a core of catalytic substance. Furthermore, although the catalyst of example 1 of EH1 contains an inert carrier, it was well-known before the priority date that unsupported catalysts can be used for the catalytic gas phase oxidation of propylene into acrolein and acrylic acid. As pointed out by the Respondent, it was known that an unsupported catalyst might lead to a somewhat lower selectivity compared to a supported catalyst. However, in the Board's judgement this knowledge would not have discouraged the skilled person faced with the problem stated above from trying the centrifugal flow coating method for the manufacture of an unsupported catalyst. Firstly, the skilled person would, in view of the teaching of EH14, have expected this coating method to produce catalysts with a good reproducibility as a result of the more uniform particle size and coating thickness compared to the conventional methods. Secondly, the problem to be solved was to provide a process for producing a catalyst having a very good performance with good reproducibility. This objective does not imply that the resulting catalysts exhibit an improved or a similar selectivity and the file contains no evidence that such a problem would have been solved. A catalyst having a somewhat lower selectivity can still exhibit a very good performance for the catalytic reaction considered. Furthermore, EH14 suggests that the centrifugal flow coating method leads to an improved catalytic activity and selectivity in comparison with the conventional rolling granulation used to produce spherical unsupported catalysts (see page 2, lines 17 to 23 in combination with page 3, lines 26 to 29). Therefore, the skilled person would have expected that the lower selectivity resulting from the omission of the inert carrier might be partially compensated by the improvement resulting from the better uniformity of particle sizes. In these circumstances, the skilled person would have tried the method of EH14 for preparing unsupported catalysts having the composition disclosed in example 1 of EH14, using the catalytic substance as cores instead of the inert carrier. As confirmed by the Appellant, he would thereby have arrived at a product having physical characteristics falling within the ranges indicated in the claim of auxiliary request I. As already pointed out above, the measurement of these properties does not in itself involve an inventive step.

6.2 The Appellant's arguments that the skilled person would not have used the method of EH14 to produce unsupported catalysts since it involves coating a core or carrier with a catalytic substance contrary to the claimed process (see point VI above) cannot be accepted by the Board for the following reasons. First, the wording of the claim does not exclude charging a core of catalytic substance into the centrifugal flow coating device before charging the unfired material powder since the expression "characterized by" is not equivalent to "consisting of". It is observed in this context that in claim 2 as granted the expression "characterized by" was used to define a process in which an inert carrier may be previously charged into the centrifugal flow coating device as illustrated by a great number of examples. Secondly, it is expressly indicated in EH14 that the catalytic substance is deposited onto the surface of **nuclei or** cores, and these nuclei may have a particle size as low as 500 µm (see page 2, lines 1 to 4 and page 7, lines 20 to 21). In the claimed process, the powder introduced into the centrifugal flow device at the beginning of the forming process also constitutes nuclei or cores for the powder which is further charged into the said device within a certain period of time, and the claim does not contain any limitation as to the particle size of the powder. Further, according to page 4, lines 1 to 6, of the patent in suit, EH14 "discloses the coating of particulate cores with a catalyst or carrier by a centrifugal flow coating device" and "the present invention applies **this method** to the preparation of an oxide catalyst comprising Mo, Fe and Bi elements as essential components" (the number 17292/1977 should in fact read 117292/1977 as correctly indicated on page 1 of the patent in suit).

It follows from the above that the process according to the claim of auxiliary request I does not involve an inventive step.

Auxiliary request II

7. The claim of this request differs from the claim of auxiliary request I in that it contains the additional proviso that the catalyst is not supported on "a core consisting of said catalyst", the "said catalyst" being the catalyst obtained after the firing step. In view of the statement on page 4, lines 17 to 20, of the patent in suit, it is clear that this proviso in fact means that a core of the said catalyst (the fired catalyst) is not used for forming the catalyst particles.

The Appellant argued at the oral proceedings that this proviso did not meet the requirements of Article 123(2) EPC, since it was not directly derivable from the sentence on page 4, lines 17 to 20, of the patent in suit that the "catalyst per se" was the fired catalyst. The Board considers that the question of the allowability of this amendment can remain open since even if it were assumed in favour of the Appellant that the provisions of Article 123(2) EPC were complied with, then the claim would still not be allowable because of lack of inventive step, as set out below.

7.1 Although the claimed process excludes the use of a core of inert carrier or fired catalyst for forming the catalyst particles, it still encompasses the use of the "unfired material powder" or "unfired oxide composition" as nuclei or cores.

Starting from EH1, the technical problem underlying the claimed process is the same as that indicated above for auxiliary request I. It is also credible that the said problem has actually been solved by the claimed process.

As already indicated above, EH14 discloses the use of "catalytic active substance" as the cores or nuclei to produce a sphere made exclusively of the catalytic active substance by the centrifugal flow coating method (see page 9, third paragraph and page 2, lines 1 to 8). The Appellant argued at the oral proceedings that the teaching of EH14 was limited to the use of a fired catalytic substance as cores, since the expression "catalytic active substance" implied that the catalytic substance was active and, thus, had been subjected to the firing step. These arguments are not convincing for the following reasons. On page 2, lines 1 to 8, the expression "catalyst active substance" is employed to designate the starting powder which is deposited onto the nuclei or cores. On page 4, line 7; page 5, line 22 and page 7, line 12, the terms "catalyst" or "catalyst powder" designate the same starting powder whereas in the last paragraph of page 7, this powder is called "the per se known catalyst". On page 8, lines 7 to 10, it is disclosed that the "catalyst" shaped in the centrifugal flow coating device is dried and activated by thermal treatment or the like before use, or may be directly used after the forming step. The next sentence indicates the precautions to be taken during the thermal treatment if such a treatment is effected. Therefore, it clearly derives from the whole disclosure in EH14 that the expression "catalyst active substance" used on page 2 to designate the starting powder does not necessarily mean a substance which has been activated by heat treatment since such an activation can be effected after the coating step. In these circumstances, the skilled person reading the expression "catalytic active substance" on page 9 would have no reasons to conclude that only a fired catalytic substance can be used as nuclei, this expression or similar terms being also used in EH14 to designate a non-activated catalytic substance. The skilled person would thus have inferred from EH14 that either a fired or an unfired catalytic substance can be used as nuclei. Furthermore, the reasons given above in points 6.1 and 6.2 apply analogously to the claim of auxiliary request II. Therefore, the process according to the claim of this request lacks an inventive step and this request must also fail.

Auxiliary request III

8. The claim of this request differs from the claim of the main request in that the phrase "comprising charging an unfired material powder" has been replaced by the phrase "characterized by charging a powder of an unfired oxide composition not shaped or pre-stage catalyst particle material composition not converted to oxide". The substitution of the expression

"characterized by" for the word "comprising" in the claim does not exclude charging an inert carrier into the centrifugal flow coating device before charging a powder of an unfired oxide composition for the reasons already indicated above in point 6.2.

Furthermore, the claim of this request still contains the alternative where a powder of an "unfired oxide composition" is used for forming the particles. For the reasons given above in point 4.1, the expression "an unfired oxide composition" is to be construed as an oxide composition which has been treated at a temperature lower than 450°C. Thus, the powder of catalytic active material used in example 1 of EH1, which has been treated at 360°C for 2 hours, is considered to be a powder of an "unfired oxide composition not shaped".

It follows from the preceding considerations that the reasons indicated above in points 5 to 5.4 in connection with the inventive step issue of the main request apply analogously to the claim of auxiliary request III. Therefore, the process according to the claim of this request does not meet the requirements of inventive step set out in Article 52(1) and 56 EPC and is not allowable.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:	The Chairman:
S. Hue	R. Spangenberg