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D E C I S I O N
of 17 December 1996

Case Number: T 0537/94 - 3.3.2

Application Number: 88108780.3

Publication Number: 0293859

IPC: B01J 23/28

Language of the proceedings: EN

Title of invention:
Catalyst for oxidation of acrolein and process for production thereof

Patentee:
NIPPON SHOKUBAI KAGAKU KOGYO KABUSHIKI KAISHA

Opponent:
BASF Aktiengesellschaft, Ludwigshafen

Headword:
Catalyst/NIPPON

Relevant legal provisions:
EPC Art. 54, 56 EPC

Keyword:
"Novelty - yes"
"Inventive step - no"

Decisions cited:
-

Catchword:
-

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Case Number: T 0537/94 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 17 December 1996

Appellant: (Proprietor of the patent)
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Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted 27 May 1994 revoking European patent No. 0 293 859 pursuant to Article 102(1) EPC.**

Composition of the Board:

Chairman: P. A. M. Lançon
Members: G. J. Wassenaar
S. C. Perryman

Summary of Facts and Submissions

I. The appeal lies from a decision of the Opposition Division to revoke European patent No. 0 293 859, which was granted with 6 claims in response to European patent application No. 88 108 780.3.

II. A Notice of Opposition was filed against the European patent by the Respondent (Opponent). Revocation of the patent was requested on the grounds of lack of novelty, lack of inventive step and insufficient disclosure (Articles 100(a) and (b) EPC).

The opposition was supported, inter alia, by the following documents:

US-A-4 035 262 (D1)
DE-A-2 909 671 (D2)

Later in the proceedings, inter alia, the following documents were cited:

DE-A-2 626 887 (D14)
DE-A-2 550 440 (D15)
DE-A-2 449 992 (D16)
EP-B-0 279 374 (D17)
Chemical Abstract 88: 79851b (D20)

III. The decision under appeal was based on the set of claims as granted (main request) and a set of claims 1 to 3 corresponding to granted process claims 4 to 6 (auxiliary request).

The Opposition Division held that the subject-matter of claims 1 to 3 of the main request lacked novelty over D1 (Example 15), D2 (Example 1), D14 (Examples 1 and 10), D15 (Example 11) or D16 (Example 2) and that

the subject-matter of claim 1 to 3 of the auxiliary request lacked an inventive step over D2 in combination with D20. With respect to inventive step the closest prior art was considered to be D2 from which the process of claim 1 of the auxiliary request differed only in the use of a centrifugal flow coating apparatus to form the catalyst particles. The use of such an apparatus was considered to be obvious in view of D20 disclosing such an apparatus for the preparation of granular coated catalysts. The objection of insufficient disclosure was rejected.

IV. With the Statement of Grounds the Appellant filed a test report in which a catalyst obtained according to D2 was compared with a catalyst obtained according to the process of the patent in suit. The Appellant contested the arguments of the Opposition Division with respect to novelty and inventive step. He submitted that none of the cited documents disclosed the pore size distribution as claimed. The new comparative examples showed that the catalyst of Example 1 of D2 did not have the required pore size distribution and was less active than the corresponding catalyst prepared according to the patent in suit.

V. The Respondent contested the arguments of the Appellant and filed an English translation of JP-A-52-117292 (D21), which formed the basis of D20. With respect to novelty it was argued that D2 was not the only novelty destroying document and that the new comparative examples provided by the Appellant did not prove that Example 1 of D2 falls outside the granted claims. With respect to inventive step it was submitted that D21 confirmed that centrifugal flow coating was a known method for improving the activity, selectivity and the mechanical properties of granular supported catalysts. With respect to insufficiency it was agreed that it was possible to reproduce the examples in the patent in

suit but it was submitted that there was no direct relationship between the claimed process features and the required physical specifications. Moreover, the invention could not be performed over the whole breadth of the process claims.

VI. On 13 December 1996 the Appellant filed new claims 1 and 2 corresponding to claims 3 and 6 as granted. In the oral proceedings, which were held on 17 December 1996, the Appellant filed new claims 1 and 2, corresponding to claims 3 and 6 as granted, as a main request for the Contracting States DE, GB, FR, IT and BE, and a single process claim, corresponding to claim 6 as granted, as main request for Contracting State ES. As auxiliary request the Appellant at the oral proceedings filed a single process claim, corresponding to claim 6 as granted, for all designated Contracting States.

The Respondent maintained the novelty objection against the product claim and the inventive step objection against the process claim.

With respect to novelty, the Appellant argued that the reworking of the prior art examples by the Respondent was somehow incorrect, and that correct reworking would only produce something falling outside the product claim. The processes for obtaining the catalysts in the examples according to D1, D14, D15 and D16 were essentially the same as the carrier added forming method disclosed in Example I-7 of the patent in suit in which an entirely different pore size distribution was obtained without pores in the range of 10 to 100 µm. D2 disclosed a more specific coating process. The comparative examples given in the Grounds of Appeal proved that the process of D2 also resulted in a pore size distribution outside the claimed range. With respect to inventive step it was argued that the

comparative examples on file proved that the centrifugal flow coating resulted in catalysts with high reproducibility, which increased the acrolein conversion and the yield of acrylic acid. Because D2 disclosed that the catalysts made by the coating method described therein were already of high quality, i.e. very uniform in size and of high abrasion resistance, there would be no reason for the skilled person to apply other coating methods such as disclosed in D21 to further improve the catalysts. D21 provided no indication that the yield of acrylic acid could be improved using the centrifugal flow coating method.

VII. The Appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of the main request, or respectively, the auxiliary request submitted at the oral proceedings on 17 December 1996.

The Respondent requested that the appeal be dismissed.

VIII. Claim 1 of the main request for the Contracting States DE, GB, FR, IT and BE reads as follows:

A catalyst for use in the production of acrylic acid by catalytic gas phase oxidation of acrolein, comprising active substances represented by general formula

$$Mo_{(a)}V_{(b)}A_{(c)}B_{(d)}C_{(e)}D_{(f)}O_{(x)}$$

wherein Mo represents molybdenum, V represents vanadium, A represents at least one element selected from the group consisting of iron, copper, bismuth, chromium, antimony and thallium, C represents at least one element selected from the group consisting of alkali metals and alkali earth metals, D represents at least one element selected from the group consisting of silicon, aluminum and titanium, and O represents

oxygen; and a, b, c, d, e f and x represents atomic ratios of Mo, V, A, B, C, D and O, respectively, and where a=12, b=2 to 14,c=0 to 12,d=0 to 6,e=0 to 6 and f=0 to 30 and x is a numerical value determined depending upon the oxidation states of the other elements, and characterized by having a specific surface area of 0.50 to 15.0 m^2/g , a pore volume of 0.10 to 0.90 cc/g and a pore diameter distribution in which the pore diameters are distributed concentratedly in the ranges of from 0.1 to less than 1.0 µm, from 1.0 to less than 10.0 µm and from 10.0 to 100 µm, such that the pore volume composed of pores having pore diameters in the range of from 0.1 to less than 1.0 µm, the pore volume composed of pores having pore diameters in the range of from 1.0 to less than 10.0 µm and the pore volume composed of pores having pore diameters in the range of from 10.0 to less than 100.0 µm are respectively 15 to 40%, 15 to 65% and 15 to 65% based on the entire pore volume.

Claim 2 of the main request for DE, GB, FR, IT and BE, Claim 1 of the main request for ES, and Claim 1 of the auxiliary request each reads as follows:

1. A process for preparation, with good reproducibility, of a catalyst which is used in the production of acrylic acid by catalytic gas phase oxidation of acrolein and which comprises active substances represented by general formula

$$Mo_{(a)}V_{(b)}A_{(c)}B_{(d)}C_{(e)}D_{(f)}O_{(x)}$$

wherein Mo represents molybdenum, V represents vanadium, A represents at least one element selected form the group consisting of tungsten and niobium, B represents at least one element selected form the group consisting of iron, copper, bismuth, chromium, antimony and thallium, C represents at least one element

selected from the group consisting of alkali metals and alkali earth metals, D represents at least one element selected form the group consisting of silicon, aluminum and titanium, and O represents oxygen; and a, b, c, d, e, f and x represent atomic ratios of Mo, V, A, B, C, D and O, respectively, and where a=12,b=2 to 14,c=0 to 12,d=0 to 6,e=0 to 6 and f=0 to 30 and x is a numerical value determined depending upon the oxidation states of the other elements; the process characterized by charging an unfired catalyst material powder composition into a centrifugal flow coating apparatus to form particles having an average diameter of 2 to 10mm, and then firing the particles thereby to obtain the catalyst having a specific surface area of 0.50 to 15.0 m^2/g, a pore volume of 0.10 to 0.90 cc/g and a pore diameter distribution in which the pore diameters are distributed concentratedly in the ranges of from 0.1 to less than 1.0 µm, from 1.0 to less than 10.0 µm and from 10.0 to 100 µm, such that the pore volume composed of pores having pore diameters in the range of from 0.1 to less than 1.0 µm, the pore volume composed of pores having pore diameters in the range of from 1.0 to less than 10.0 µm and the pore volume composed of pores having pore diameters in the range of from 10 to 100 µm are respectively 15 to 40%, 15 to 65% and 15 to 65% based on the entire pore volume.

Reasons for the Decision

1. The appeal is admissible.

2. The patent in suit has been amended by deleting granted claims. By deleting claims, neither the content of the application as filed nor the protection conferred can be extended. The amendments, therefore, are allowable under Article 123(2) and (3) EPC.

3. *Main request product claim*

Novelty

3.1 Novelty was contested on the basis of D1, D2, D14, D15 and D16 each taken individually. Each of these documents discloses at least one catalyst with a composition as required by the product claim. The pore structure of the catalysts is not disclosed in these documents but these documents contain detailed examples for producing the catalysts. The Respondent has prepared catalysts according to examples in each of these documents and has found that the preparation resulted in catalysts with a specific surface, a pore volume and a pore size distribution as required by present claim 1 (Notice of opposition and Respondent's letter filed 29 January 1994). In particular a catalyst was prepared according to Example 1 of D2 which had the composition $Mo_{12}V_3W_{1.2}Cu_{2.2}O_{49.3}$, a specific surface of 13.54 m^2/g, a pore volume of 0.45 cm^3/g and a pore size distribution such that 28% have a size in the range of 0.1 to 1.0 µm, 25% have a size in the range of 1.0 to 10.0 µm and 31% have a size in the range of 10.0 to 100 µm (pages 2 and 3 of the reasons filed with the notice of opposition).

3.2 The Appellant's argument that the working of the prior art examples by the Respondent is incorrect is not convincing, given the positive evidence relied on by the Respondent.

With respect to the examples according to D1, D14, D15 and D16 no specific counter evidence was filed. It was rather argued that since the coating method according to comparative Example I-7 of the patent in suit did not lead to the desired result the similar processes

according to said prior art documents would likewise not lead to the required pore size distribution. According to comparative Example I-7, porous alpha alumina carrier particles having diameters of 3 to 5 mm were mixed with a suspension of the catalyst mass, which mixture was evaporated to dry solidness with stirring, and then fired at 400°C for 5 hours. The processes of the examples provided by the Respondent differ, however, from said comparative Example I-7. The process of Example 15 of D1 differs at least in using a carrier having a larger diameter (¼ inch (6.3 mm) and a longer heat treatment (1½ hours at 200 to 400°C and 5 hours at 400°C). The process of Examples 1 and 10 of D14 differs at least in that the catalyst mass is calcined before coating at a temperature of 395°C for 3 hours. The process of Example 11 of D15 differs in that the catalyst mass is calcined before coating. The process of Example 2 of D16 differs at least in the use of a carrier of a different composition and of larger diameter (silicon dioxide-aluminium oxide spheres of 6.3 mm).

According to the Appellant's own submission heat-treatment conditions in the preparation of catalysts have a large influence on the performance and physical properties of the resulting catalyst (Appellant's letter filed 9 May 1994, page 4). By the comparative examples filed with the Appellant's letter of 9 May 1994, it was further demonstrated that a slight variation in particle size of the catalyst resulted in a substantial change in surface area; viz. by increasing the size of 4.5 to 5 mm, the specific surface increased from 14.0 to 18.5 m^2/g.

Thus, the failure of said comparative Example I-7 does not mean that with other coating methods such as disclosed in D1, D14, D15 and D16 the desired result cannot be obtained.

The Appellant's argument that the test report filed with the grounds of appeal proves that Example 1 of D2 does not fulfil the requirements of claim 1, likewise fails to convince, because in the preparation of catalyst 1 of said test report a carrier was used with a diameter of 5 mm instead of 6 mm as indicated in Example 1 of D2.

3.3 In the absence of convincing evidence to the contrary, the Board accepts that working of the examples according to the prior art, as presented by the Respondent, results in a product fulfilling all the requirements of claim 1, so that claim 1 lacks novelty.

4. *Main request and auxiliary request process claim*

4.1 *Novelty*

The claimed process is not disclosed in any of the cited documents, so that the process is new. In fact, the novelty of the process has never been disputed.

4.2 *Inventive step*

4.2.1 The Board agrees with the Parties and the Opposition Division that, with respect to the claimed process, D2 represents the closest prior art. It discloses a process for the preparation of a coated catalyst by charging simultaneously but separately from each other catalytically active material in powder form together with water into a coating apparatus containing carrier particles under such conditions as to form coated granules (claim 1). The carrier particles preferably have a diameter of between 1 to 9 mm and the loading of the particles is preferably from 5 to 200 wt.% (page 7, line 1 and page 9, lines 21 to 25). In Example 1 a process for the preparation of a catalyst for use in the production of acrylic acid by catalytic gas phase

oxidation of acrolein has been disclosed whereby 6 mm diameter non porous magnesium silicate spheres are coated with a catalytic mass of the composition $Mo_{12}V_3W_{1.2}Cu_{2.2}O_{49.3}$ by the above mentioned coating procedure in a rotating pan. The thickness of the coating is not disclosed but the loading is indicated to be 24 kg catalyst powder on 112 kg catalyst carrier, which means that the coating thickness cannot be more than a few mm. The catalytic mass in this example was calcined (fired) before the coating process. The firing before coating is, however, only an optional measure. According to the general disclosure of D2, it may be advantageous in some cases to calcine the catalytic mass after the coating (page 9, lines 5 to 8). The supported catalyst obtained according to Example 1 has been used in a process for the oxidation of acrolein to acrylic acid, whereby an acrolein conversion of 98 mol% and a yield of 95 mol% acrylic acid was obtained (page 15, lines 5 and 6).

4.2.2 According to the description of the patent in suit the catalyst for the oxidation of acrolein obtained with the claimed process has high activity, excellent durability and good reproducibility (page 1, lines 5 to 7). When used in a process for the production of acrylic acid by catalytic gas phase oxidation of acrolein, high yields of acrylic acid could be obtained as demonstrated by the examples in the patent specification. In the oral proceedings the Appellant maintained that one of the advantages of the claimed process was that the catalyst so produced makes it possible to produce acrylic acid in higher yields than with comparable prior art catalysts.

The technical problem underlying the claimed invention is therefore regarded as providing a process for preparing a supported catalyst for the preparation of acrylic acid by gas phase oxidation of acrolein with higher activity and/or selectivity in order to improve the acrylic acid yield.

According to the process claim, this problem is solved by charging an unfired catalyst material powder of specified composition into a centrifugal flow coating apparatus to form particles having an average diameter of 2 to 10 mm, and then firing the particles thereby to obtain a catalyst having the specified specific surface, pore volume and pore size distribution.

In the test report filed with the grounds of appeal, the performance of a catalyst made according to D2 (Catalyst 1) was compared with a catalyst of the same composition coated on the same carrier with the coating process of the patent in suit (Catalyst 2). By applying these catalysts under the same conditions in the acrolein oxidation a higher conversion and a higher yield was obtained with Catalyst 2 (98.5 mol% against 96.5 mol% and 92.0 mol% against 87.3 mol% respectively). Although Catalyst 1 was not exactly prepared according to Example 1 of D2 (see point 3.2) it can be regarded as a representative example of a process according to D2. The Respondent has not provided counter evidence which would invalidate the measured improvement.

The Board is, therefore, satisfied that the said problem is actually solved by the claimed process.

4.2.3 It remains therefore to be decided if, for solving the above stated problem, the claimed solution would have been obvious to a person skilled in the art.

Apart from the centrifugal flow coating apparatus and the explicit mention of the pore structure of the catalyst, all the features of the present process claim are known from D2. The use of a centrifugal flow coating apparatus for the preparation of granular supported catalyst has been disclosed in D20, which is an abstract of D21. Abstract D20 teaches the skilled person that with the centrifugal flow coating apparatus mixed oxide catalysts comprising Mo, V, Ag, P and Na with good mechanical strength and abrasion resistance for the oxidation of benzene to maleic acid could be obtained with a conversion of 97 mol%. In the introduction of D21 it is further indicated that with the known coating methods it is difficult to regulate the coating layer thickness, which results in catalysts which are problematic in respect of catalytic activity, selectivity and mechanical strength (page 3, lines 9 to 17). D21 teaches that this problem is solved by use of the centrifugal fluidisation coating apparatus disclosed therein. It is stated that "because of the simplicity for regulation of coating layer thickness, and owing that the optional selection for size, quality and so on of the core is possible, there is therefore no loss of effectiveness factor caused by vainly left catalytic active component, or there is no loss of selectivity" (page 8, last complete paragraph of translation). The process disclosed in D21 is not limited to specific catalysts but discloses in particular mixed oxide catalysts (Examples 1 to 6). D20 and D21 are publications in the same specialist field of catalysts as the problem to be solved by the patent in suit, and so would be considered by the skilled person seeking a solution to this problem of improving acrolein catalysts. They provide the person skilled in the art seeking to achieve higher activity and/or selectivity of the granular mixed oxide supported acrolein oxidation catalyst of D2 with a strong incentive to use the centrifugal flow coating apparatus

instead of the traditional rotating coating pan in the method of D2. The broad applicability of the method taught in D21, together with the plausible explanation given for why it achieves the advantages claimed for it, and the example showing that it works for a similar catalysis problem, give the skilled person a reasonable expectation that he can successfully achieve the desired improvement also for an acrolein oxidation catalyst. D20 and D21 thus render the use centrifugal flow coating apparatus for producing improved acrolein catalysts obvious.

4.2.4 The tests filed by the Appellant with the grounds of appeal show that the use of the centrifugal flow coating apparatus in a process according to D2 results in a specific surface, a pore volume and a pore size distribution as required by the process claim, without any special measures being needed to achieve this specific surface, pore volume and pore size. Thus these requirements of the process claim are fulfilled merely by following the suggestion of D20 and D21 of the use of centrifugal flow coating apparatus, so that D20 and D21 render the subject-matter of the process claim obvious.

4.2.5 The Board does not accept the Appellant's argument that because it is indicated in D2 that the coated catalysts obtained with the process of D2 have a very uniform distribution of the active mass and a high abrasion resistance (page 6, lines 6 to 13) the skilled person would have had no reason to use another coating method. According to the Appellant the skilled person **could** have used the known centrifugal flow coating apparatus but without incentive he **would** not use it.

In the Board's opinion, however, the skilled person will always try to improve existing processes even if they are already highly sophisticated and will always as far as possible make use of the latest developments in his technical field. Keeping an eye open for improvements is an immanent characteristic of the "person skilled in the art" of Article 56 EPC. Moreover, D21 tells the skilled person that the centrifugal flow coating method is superior to all existing coating methods for preparing a supported catalyst, and thus provides a clear incentive to use this teaching to improve the coating method of D2.

4.2.6 The Appellant's argument that D21 is not related to the patent in suit because in the examples thereof oxides were used as starting material cannot be accepted either. The fact that the process claim requires that an unfired catalyst material powder composition is used as starting material does not exclude an oxide composition. According to Example 1 of D21, the coated catalyst after drying is activated by a thermal treatment at 500°C, implying that the coating mixture was regarded as "unfired". Moreover, the fact that the composition of a coating mixture is indicated with reference to the oxides, does not necessarily means that the coating mixture was an oxide mixture. In chemistry and in particular in the field of catalysts, it is not uncommon to refer to the oxide composition to indicate any composition which, after calcination, has this oxide composition. Anyhow, D21 contains no indication that the process disclosed therein is restricted to oxide mixtures as starting material.

4.2.7 In view of the preceding considerations, the subject-matter of the process claim is considered to be the obvious result of the use of a known coating method in a known process for producing a catalyst of known composition and does not involve an inventive step in the meaning of Article 56 EPC.

4.2.8 It follows that neither the main request nor the auxiliary request meet the requirements of the European patent convention, and the appeal must be dismissed.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

P. Martorana

The Chairman:

P. A. M. Lançon

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