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**Datasheet for the decision
of 27 November 2012**

Case Number: T 1507/09 - 3.3.07

Application Number: 00310977.4

Publication Number: 1106246

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C07C 51/25, C07C 57/04

Language of the proceedings: EN

Title of invention:
Modified carrier, complex oxide catalyst and process for
preparation of acrylic acid

Patentee:
NIPPON SHOKUBAI CO., LTD

Opponent:
CeramTec GmbH

Headword:
-

Relevant legal provisions:
EPC Art. 56
RPBA Art. 13

Keyword:
"Amendments to parties' cases - new arguments and objections
against inventive step (admissible) - new claims request
(admissible) - new objections against novelty (not dealt with)"
"Inventive step (no) - obvious solution - all requests"

Decisions cited:
-

Catchword:
-



Case Number: T 1507/09 - 3.3.07

DECISION
of the Technical Board of Appeal 3.3.07
of 27 November 2012

Appellants:
(Opponents) CeramTec GmbH
CeramTec-Platz 1-9
D-73207 Plochingen (DE)

Representative: Uppena, Franz
Chemetall GmbH
Patente, Marken & Lizenzen
Trakehner Strasse 3
D-60487 Frankfurt/Main (DE)

Respondents:
(Patent Proprietors) NIPPON SHOKUBAI CO., LTD.
1-1, Koraibashi 4-chome
Chuo-ku
Osaka-shi (JP)

Representative: Benson, John Everett
J.A. Kemp & Co.
14 South Square
Gray's Inn
London WC1R 5JJ (GB)

Decision under appeal: Interlocutory decision of the Opposition
Division of the European Patent Office posted
14 May 2009 concerning maintenance of European
patent No. 1106246 in amended form.

Composition of the Board:

Chairman: J. Riolo
Members: G. Santavicca
M.-B. Tardo-Dino

Summary of Facts and Submissions

I. The appeal by the opponents lies from the interlocutory decision of the Opposition Division, according to which, account being taken of Claims 1 to 4 of the First Auxiliary Request filed during oral proceedings held on 23 April 2009, and of a description adapted thereto, European patent 1 106 246 (application N° 00 310 977.4) and the invention to which it relates met the requirements of the EPC. The decision also gave the reasons for refusing the Main Request as well as for apportioning the costs of oral proceedings held on 15 July 2008.

II. Claims 1 and 4 of the First Auxiliary Request underlying the decision under appeal read as follows (compared to the claims as granted, additional features are indicated in bold, deletions in strike-through):

"1. A **complex oxide catalyst which comprises a complex oxide containing molybdenum and vanadium supported on a modified** carrier which is characterized by carrying on at least a part of an inert carrier surface an oxide of formula (1):



Wherein X is one or more element selected from alkaline earth metals;

Y is one or more element selected from silicon, aluminum, titanium and zirconium;

Z is one or more element selected from Group IA elements and Group IIIB elements of the periodic table, boron, iron, bismuth, cobalt, nickel and manganese; and O is oxygen;

a, b, c and d denote the atomic ratios of X, Y, Z and O, respectively, such that when $a=1$, $0 < b \leq 100$ and $0 \leq c \leq 10$, and d is a numerical value determined by the degree of oxidation of the other elements, and wherein said **modified** carrier has been heat treated at a temperature of 1200 to 1700°C."

"4. A process comprising preparing acrylic acid through vapour phase oxidation of acrolein with molecular oxygen or molecular oxygen-containing gas in the presence of a catalyst, which process is characterized in that the catalyst is a complex oxide catalyst according to any one of claims ~~3 or 4~~ 1 to 3."

III. The patent in suit had been opposed in its entirety on the grounds of lack of novelty and inventive step (Article 100(a) EPC), having regard to *inter alia* documents D1 (EP-B1-0 714 700) and D4 (DE-C3-2 135 620). A further document (D7: H. Salmang & H. Scholze, *Keramik, Teil 2: Keramische Werkstoffe*, 6th edition, 1983, pages 111-112), allegedly representing common general knowledge, submitted during the oral proceedings held on 15 July 2008 and held to be highly relevant, was admitted into the proceedings. Comparative Example 2 (D8) was enclosed in the letter of 23 February 2009 by the patent proprietors.

IV. In the decision under appeal, it was *inter alia* held that:

- (a) As regards the Main Request, D4 disclosed an inert carrier coated with a mixed oxide, which was steatite, i.e. a natural raw material obtained from the calcination of soapstone $(Mg_3(Si_4O_{10})(OH)_2)$, a natural magnesium silicate, hence an oxide as

defined in Claim 1. D4 disclosed that the carrier with the mixed oxide was ceramically burnt, without however indicating any temperature therefor. In particular, D4 illustrated a mixture of soapstone, plastic clay and feldspar for making the steatite. According to D7, similar mixtures of soapstone, plastic clay and feldspar were burnt at a temperature of 1200 to 1400°C. Thus, D4 confirmed what was implicitly disclosed by D4, i.e. the temperature feature of Claim 1. Consequently the modified support of Claim 1 of the Main Request was not novel over D4.

- (b) As regards the First Auxiliary Request, its Claim 1 was restricted to a complex oxide catalyst. Since D1, the only document invoked against novelty, neither disclosed an inert carrier comprising an oxide of Formula (1) on at least part of its surface, nor that the carrier was treated at a temperature of from 1200 to 1700°C, the catalyst of Claim 1 of the First Auxiliary Request was novel.
- (c) The closest prior art was disclosed by D1, which addressed the problem of providing a complex oxide and a process for producing acrylic acid through vapour phase catalytic oxidation of acrolein, as the patent in suit. In particular, D1 disclosed ring-shaped coated catalysts attaining an improved lifespan, over prior art spherical catalyst particles, in exothermic gas phase oxidation reactions in fixed bed reactors such as in the production of acrylic acid through catalytic vapour phase oxidation. D4 instead addressed a process for making coated catalyst carriers and so would not have been considered as the closest prior art.

- (d) As regards the problem solved over D1, the patent in suit comprised examples illustrating spherical carriers of silica-alumina having an average particle diameter of 5 mm, with different oxide layers, on which the same catalyst layer ($\text{Mo}_{12}\text{V}_5\text{W}_1\text{Cu}_{2.2}\text{Sb}_{0.5}\text{O}_n$) was provided. The only comparative example in the patent in suit did not contain an oxide layer. Both acrolein conversion and acrylic acid selectivity were higher for the catalysts of the examples than for that of the comparative example. However, since D1 disclosed a catalyst having a carrier with the composition of the oxide layer of Example 1 of the patent in suit and with a catalytic layer, Comparative Example 1 of the patent in suit was not representative of D1. Comparative Example 2 (D8) dealt with spherical particles made of steatite and carrying the same catalytic layer used in the examples of the patent in suit. Hence, the catalyst of Comparative Example 2 (D8) represented catalysts of D1. A comparison between the catalyst of Example 1 and that of Comparative Example 2 credibly showed that the claimed catalysts improved selectivity and yield of acrylic acid in the vapour phase catalytic oxidation of acrolein over those of D1.
- (e) Since D4 concerned oxidation catalysts and had a totally different objective than the patent in suit and D1, the skilled person would not have combined D4 with D1 in order to attain an improvement in selectivity and yield. So the claimed process was not obvious.
- (f) Therefore, the claimed subject-matter of the First Auxiliary Request met the requirements of the EPC.

- (g) Since the late filing of D7 had caused the postponement of the first oral proceedings, the opponents were to bear the relevant costs thereof.
- V. In their statement setting out the grounds of appeal, the appellants attacked the decision under appeal to the extent that it acknowledged that the subject-matter of Claim 1 of the then First Auxiliary Request involved an inventive step as well as because an apportionment of costs had been ordered. Then, in their letter of 7 May 2012, the appellants argued against Comparative Example 2. Finally, in their letter of 5 November 2012, the appellants objected against the late filing of the second Auxiliary Request and attacked the novelty of the subject-matter of Claim 1 of all requests over D1.
- VI. In their letter of 17 February 2010, the respondents (patent proprietors) enclosed three sets of amended claims as their Main Request and their First and Second Auxiliary Requests. Then, in their letter of 27 September 2012, the respondents enclosed a new Second Auxiliary Request, to replace the previous Second Auxiliary Request. The latter became the Third Auxiliary Request.
- VII. Claim 1 of each of the Main Request and First, Second or Third Auxiliary Request contains the following amendments, if any:

Main Request

Claim 1 of the Main Request identically correspond to Claim 1 upheld in the decision under appeal (Point II, *supra*).

First Auxiliary Request

Claim 1 of the First Auxiliary Request, compared to Claim 1 according to the Main Request, additionally contains the features "wherein the inert carrier is selected from silica, alumina, silica-alumina, silicon carbide, silicon nitride, titanium dioxide and zirconium dioxide".

Second Auxiliary Request

Claim 1 of the Second Auxiliary Request, compared to Claim 1 according to the Main Request, additionally contains the features "and wherein the inert carrier supports thereon the oxide of Formula (1) at a carriage ratio of 1-50%, said ratio being calculated by the following equation: carriage ratio(%)=[1-(weight of inert carrier/weight of modified carrier)]x100".

Third Auxiliary Request

Claim 1 according to the Third Auxiliary Request identically corresponds to Claim 4 of the First Auxiliary Request underlying the decision under appeal (Point II, *supra*) limited to the use of the catalyst as defined in Claim 1 upheld in the decision under appeal.

- VIII. In a communication in preparation for oral proceedings, the Board drew attention to the issues needing debate and decision, in particular inventive step over D1.
- IX. Oral proceedings were held on 27 November 2012. The appellants withdrew their request to set aside the

decision as to an apportionment of costs. At the end of the oral proceedings, the decision was announced.

- X. As regards the issues dealt with in the present decision, the appellants essentially argued as follows:

Procedural questions

Amendments to appellants' case

- (a) (letter of 7 May 2012) Comparative Example 2 had no probative character. Also, in Claim 1 there was a lack of definition for a coating of oxide of Formula (1) which modified the inert carrier. So it was not clear which technical feature related to the sought improvement.
- (b) (letter of 5 November 2012) The subject-matter of Claim 1 of each of the Main Request and First to Third Auxiliary Requests lacked novelty over D1.

New claims requests by the respondents

- (c) The Second Auxiliary Request enclosed in the letter of 27 September 2012 had been filed late and so was not admissible.

Main Request

- (d) D1 described the closest prior art. The claimed subject-matter was distinguished from D1 by a modified carrier, i.e. an inert carrier coated with a multi metal oxide and calcined. The comparative examples on file did not prove any improvement over

D1, let alone over the whole breadth of Claim 1. In particular, the tested catalysts did not correspond to those of D1, which were obtained by spray drying, kneading and size reduction. Furthermore, only one catalyst had been exemplified in the patent in suit and Claim 1 was not even restricted to the carriage ratio described, which could instead be less than 1%, i.e. disadvantageous according to the patent in suit itself. The patent in suit did not mention what effect was attained by a coating as claimed. Hence, the problem stated in the patent in suit, i.e. the provision of a complex oxide catalyst with improved properties that were maintained over long time, had not been solved over D1, let alone over the whole breadth of Claim 1. Thus, the problem was to be reformulated, such as to provide further complex oxide catalysts having good properties, e.g. long lasting yield. Whilst D1 only described a catalyst as claimed, D4, the relevance of which had been ascertained in the decision under appeal, described a modified carrier as claimed. D4 aimed at a uniform coating of catalyst, which adhered well and lasted longer. Hence, D4 disclosed the features which distinguished the claimed subject-matter from D1. As regards the motivation of the skilled person to combine D1 with D4, starting from D1, there were only two ways for solving the reformulated problem, modifying the catalyst or modifying the carrier. Concerning the possible modification of the carrier, D1 concerned not only ring shaped but spherical catalysts too. D1 mentioned the problems relating to the fluctuations in the adhesion of the catalyst on porous carriers and suggested to use dense carriers having rougher

surfaces. Also D4 addressed similar problems and suggested to coat the dense carriers with an oxide in order to improve adhesion, hence the catalytic properties. D4 too concerned modified carriers for oxidation catalysts for saturated hydrocarbons, and acrolein was an unsaturated hydrocarbon. Therefore, the combination of D1 and D4 was obvious for the skilled person aiming at solving the reformulated problem.

First Auxiliary Request

- (e) The carrier materials defined in Claim 1 of First the Auxiliary Request being usual and no particular effect deriving from their use being disclosed, the claimed subject-matter was obvious.

Second Auxiliary Request

- (f) The examples provided in support of inventive step concerned carriage ratios of from 7 to 25%. The results of these examples could not be extrapolated, e.g. to back up the range of 1 to 50%. For instance a carriage ratio of 50% meant that the amount of inert carrier and the amount of coating material were the same, i.e. that the inert carrier might entirely be coated with the oxide of Formula (1), which became the actual carrier. In any case, a carriage ratio of 22% could be calculated for the coated carrier of D4. Hence, the claimed subject-matter was obvious over D1 and D4.

Third Auxiliary Request

- (g) Compared to the previous requests, Claim 1 now concerned a process for producing acrylic acid. In the patent in suit, that process was acknowledged as being not critical. Since the catalyst used was obvious, the process too was obvious.

XI. As regards the issues dealt with in the present decision, the respondents essentially argued as follows:

Procedural questions

Amendments to appellants' case

- (a) The objections by the appellants that Comparative Example 2 was not probative, that the extent of the coating (the carriage ratio) was not specified in Claim 1 as well as that the subject-matter of Claim 1 of all requests lacked novelty over D1, were not raised in the statement setting out the grounds of appeal. So these belated objections were not admissible.

New claims request

- (b) The Second Auxiliary Request was submitted in reaction to the belated objection by the appellants that the carriage ratio was not specified in Claim 1. So the Second Auxiliary Request was admissible.

Main Request

- (c) The patent in suit concerned catalysts useful for the production of acrylic acid from acrolein, which attained excellent results. The closest prior art according to the problem solution approach was the document concerning the same effect or result attained by the patent in suit. Only D1, which concerned molybdenum vanadium catalysts for the production of acrylic acid from acrolein, fulfilled these requirements. D4 instead concerned the production of ceramic catalysts carriers for various reactions, such as production of methanol and formaldehyde. Hence, D1 rather than D4 described the closest prior art. The claimed subject-matter was distinguished over the catalyst of D1 by a complex oxide catalyst based on three components (inert carrier/oxide coating/catalyst coating), rather than on two components (inert carrier/catalyst oxide) as in D1, hence in the nature of the carrier (an inert carrier modified by an oxide coating applied onto it). The modified carrier as claimed led to improvement, as apparent from Comparative Example 2 (D8). Thus, the formulation of the problem solved had to take into account the attained improvement. Hence, the problem solved was the provision of complex oxide catalysts having improved properties such as improved yield lasting longer. This problem had been solved over the whole breadth of Claim 1, as mentioned in the patent in suit in relation to the carriage ratio. Having regard to the improvement attained, the combination of D1 and D4 was not obvious. D1 mentioned spherical carriers but

concerned ring shaped carriers, which were not mentioned by D4, which preferred ellipsoidal or spherical carriers. Hence, D4 was not suitable for modifying a ring carrier of D1. This was also apparent from the fact that although D4 was a document of 1971 and D1 of 1995, the applicants of D1 albeit being aware of D4 did not use the modified carrier of D4. Even if the skilled person combined D1 and D4 in order to merely provide further catalysts, the subject-matter of Claim 1 would still not be obvious, in particular because D4 contains a single example.

First Auxiliary Request

- (d) Claim 1 of the First Auxiliary Request was a combination of previous Claims 1 and 2, in order to indicate the carrier materials. The arguments on inventive step over D1 and D4 were the same as those for the Main Request.

Second Auxiliary Request

- (e) Claim 1 of the Second Auxiliary Request was based on the previous requests but contained a limitation to the carriage ratio in order to support inventive step over the whole breadth of Claim 1. The further arguments on inventive step remained the same.

Third Auxiliary Request

- (f) Since Claim 1 of the Third Auxiliary Request concerned a process for producing acrylic acid, not mentioned in D4, there was no reason to consider D4.

So the claimed subject-matter was not obvious over D1.

- XII. The appellants (opponents) requested that the decision under appeal be set aside and the patent be revoked.
- XIII. The respondents (patent proprietors) requested that the appeal be dismissed or, as an alternative, that the patent be maintained on the basis of the claims of the First Auxiliary Request filed with the letter of 17 February 2010, or on the basis of the Second or Third Auxiliary Request filed with letter of 27 September 2012.

Reasons for the Decision

1. The appeal is admissible.

Amendment to appellants' case - New objections

2. In their statement setting out the grounds of appeal, the appellants attacked the decision under appeal to the extent that an inventive step was acknowledged. The appellants *inter alia* argued (e.g. page 3, fifth and sixth full paragraphs; page 4, second full paragraph) that application of the problem solution approach led to a formulation of the problem solved which was different from that stated in the application as filed and accepted by the Opposition Division.
 - 2.1 In their letter of 7 May 2012, for the first time in appeal proceedings, the appellants extensively attacked the probative nature of Comparative Example 2 (D8) and

the lack of definition of the extent of the coating of oxide of Formula (1) which modified the inert carrier.

- 2.2 This amendment to the appellants' case has been made after oral proceedings had been arranged by the Board. Hence, its admissibility and consideration is at the Board's discretion, as established in Article 13 of the Rules of Procedure of the Boards of Appeal of the EPO (RPBA). Some criteria for exercising the discretion are established in Paragraphs (1) and (3) of Article 13 RPBA, *inter alia* the complexity of the new subject-matter submitted, the current state of the proceedings, the need for procedural economy and whether new issues are raised thereby which cannot be dealt with without adjournment of the oral proceedings.
- 2.3 The new arguments are not complex and in fact merely expands on facts (which problem is effectively solved) already generally dealt with, which constitute a step of the application of the problem solution approach, i.e. which anyhow would have required discussion.
- 2.4 The respondents dealt with the new arguments in their letter of 27 September 2012, in which they enclosed a new Second Auxiliary Request. The new arguments could also be dealt with during the oral proceedings.
- 2.5 Therefore, this amendment to appellants' case is admissible.
3. In their letter of 5 November 2012, also for the first time ever in the appeal proceedings, the appellants attacked the novelty of the subject-matter of Claim 1 of all requests on file over D1.

- 3.1 Since the patent has to be revoked for lack of an inventive step (*infra*), the Board need not address this amendment to appellants' case in the present decision.

Amendment to respondents' case - New claims request

4. The Second Auxiliary Request was submitted with letter of 27 September 2012, i.e. after oral proceedings had been arranged. Hence, admissibility and consideration of the Second Auxiliary Request is at the Board's discretion as established in Article 13 RPBA.
- 4.1 The submission of the new Second Auxiliary Request was in reaction to the arguments submitted by the appellants in their letter of 7 May 2012, in particular the argument (page 4, second paragraph) that Claim 1 did not contain any limitation as regards the extent of the coating, if any. In fact, Claim 1 of the Second Auxiliary Request sought to overcome that objection by defining the carriage ratio which is a measure of the extent of coating the inert support with the Oxide of Formula (1). This restriction implies that the content of the Second Auxiliary Request converges with and expands on facts under discussion. Already for that reason, the submission of the Second Auxiliary Request fulfils the general principles set out in the case of the Boards of Appeal of the EPO (6th edition 2010, VII.E.16.1.1).
- 4.2 The appellants, who reacted to the submission of the Second Auxiliary Request in their letter of 5 November 2012 as well as during the oral proceedings, have had the opportunity to argue their case in respect of the

Second Auxiliary Request. Hence, the submission of the Second Auxiliary Request did not raise issues which could not be dealt with during the oral proceedings.

4.3 Therefore, the Second Auxiliary Request is admissible.

Amendments- All claims requests

5. The claims of the Main Request are identical to the claims of the First Auxiliary Request underlying the decision under appeal, i.e. the claims request upheld by the Opposition Division. Apart from the necessary renumbering and adaptation, and apart from the inclusion of the term "modified" in the last feature of Claim 1, the amendments consisted in the deletion of Claim 1 as granted, wherein Claim 3 as granted based on Claim 1 as granted became new Claim 1.

5.1 The First and Second Auxiliary Requests correspond to the Second and Third Auxiliary Requests before the Opposition Division. Claim 1 of the First Auxiliary Request, compared to Claim 1 of the Main Request, contains the additional features of Claim 2 as granted. Claim 1 of the Third Auxiliary Request is instead based on the process of Claim 4 as granted.

5.2 As regards Claim 1 of the Second Auxiliary Request, compared to Claim 1 of the Main Request, it contains the additional features relative to the carriage ratio, which as such are disclosed in the application as filed (page 4, line 24 to page 5 line 11).

5.3 The amended claims have neither been objected to by the then opponents nor by the present appellants. The

present Main Request was found to be allowable by the Opposition Division.

- 5.4 Since the patent has to be revoked for lack of an inventive step (*infra*), the Board need not treat in further detail why all amendments are allowable (Article 123(2) EPC).

Main Request

Novelty

6. In the decision under appeal, it was *inter alia* held that the subject-matter of Claim 1 of the Main Request was novel, as D1 did not disclose a modified carrier comprising an oxide layer of Formula (1) on an inert carrier, as defined in Claim 1.
- 6.1 This decision was not contested until the last letter of the appellants of 5 November 2012.
- 6.2 Since, having regard to the distinguishing features acknowledged in the decision under appeal, the patent has to be revoked for lack of an inventive step (*infra*), the Board need not address these belated objections against novelty raised by the appellants.

Inventive step

The patent in suit

7. The patent in suit concerns a complex oxide catalyst formed by supporting a complex oxide catalyst on a modified carrier and a process for the preparation of acrylic acid from acrolein by vapour phase catalytic oxidation reaction (Title and Paragraph [0001]).
 - 7.1 In particular, the patent in suit addresses the drawbacks of the known catalysts, which are not satisfactory for industrial working, as the yield of acrylic acid is insufficient, the deterioration rate of their activity is high and the catalyst life is short.
 - 7.2 According to the patent in suit, these problems are solved by a complex catalyst oxide as defined in Claim 1 and by a process for making acrylic acid as defined in Claim 4.

The closest prior art

8. There is agreement between the parties that D1, which concerns complex oxide catalysts for the production of acrylic acid from acrolein by vapour phase catalytic oxidation, describes the closest prior art. The Board has no reason to take a different position.

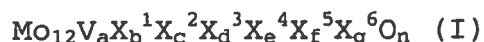
The disclosure of D1

9. D1 concerns a process for the preparation of a catalyst which consists of a carrier and a catalytically active oxide material applied to the surface of the carrier,

in which the carrier is first moistened with a liquid binder, a layer of active oxide material is then bound to the surface of the moistened carrier by bringing it into contact with dry, finely divided, active oxide material, and the liquid binder is then removed from the moistened carrier coated with active oxide material, wherein the liquid binder is a solution consisting of from 20 to 90% by weight of water and from 10 to 80% by weight of an organic compound having a boiling point or sublimation temperature at atmospheric pressure of 100°C (Claim 1).

- 9.1 As to the carrier used in the process of D1, it can:
- (a) *inter alia* consist of alumina, silica, steatite, aluminum silicate (Claim 7; Page 6, lines 15-19);
 - (b) have a surface roughness of from 40 to 100 μm (Claim 8; Page 6, lines 20-23);
 - (c) have a total volume of pores of less than or equal to 1% by volume, based on the volume of the carrier (Claim 9; page 6, lines 23-35), i.e. can be dense; and,
 - (d) comprise spherical particles or hollow cylinders (Claims 11 and 12; page 6, lines 25-28).

- 9.2 The catalytically active oxide material to be applied as a coat can be a multimetal oxide containing Mo and V (Claim 19), e.g. having the general stoichiometry (Claim 21; page 8, lines 34-55)



where

X^1 is W, Nb, Ta, Cr and/or Ce,

X^2 is Cu, Ni, Co, Fe, Mn and/or Zn,

X^3 is Sb and/or Bi,

X^4 is at least one or more alkali metals,

x^5 is at least one or more alkaline earth metals,
 x^6 is Si, Al, Ti and/or Zr,
a is from 1 to 6,
b is from 0.2 to 4,
c is from 0.5 to 18,
d is from 0 to 40,
e is from 0 to 2,
f is from 0 to 4,
g is from 0 to 40 and
n is a number which is determined by the valency and frequency of the elements differing from oxygen in (I).

9.3 The Examples of D1 concerning catalysts VS1, S1, S2, S3, S4, S5, S6 and VS2 (page 10, line 40, to page 13) *inter alia* illustrate:

- (a) the preparation of a catalytically active oxide material (A) having the stoichiometry $MO_{12}V_3W_{1.2}Cu_{2.4}O_n$ by a process including the steps of:
- (i) preparing an aqueous solution comprising copper(II) acetate monohydrate, heptamolybdate tetrahydrate, ammonium metavanadate and ammonium paratungstate heptahydrate;
 - (ii) spray-drying the solution at an outlet temperature of 110°C;
 - (iii) kneading the spray powder with water;
 - (iv) calcining the kneaded material in a through-circulation oven fed with an oxygen/nitrogen mixture, by first heating to 300°C at a rate of 10 K/min, then keeping at this temperature for 6 hours, then heating to 400°C at a rate of 10 K/min, and maintain this temperature for a further hour;

(v) milling the calcined catalytically active material to a finely divided powder.

(b) The preparation of coated catalysts by the steps of:

- (i) introducing into a rotating coating pan ring carriers (7 mm external diameter, 3 mm length, 4 mm internal diameter) of steatite, having a surface roughness of 45 μ m and a total pore volume of less than 1% by volume, based on the volume of the carriers;
- (ii) spraying water (VS1) or an aqueous solution of glycerine (S1), propionic acid (S2), formamide (S3), ethylene glycol (S4), 1,4-butanediol (S5), 1,6-hexanediol (S6), or ethylene glycol (VS2), onto the carriers;
- (iii) continuously and simultaneously metering the catalytically active oxide powders (A) by means of an oscillating conveyor outside the spray cone of the water atomizer nozzle, so that the powder fed in is completely absorbed onto the surface of the carriers, without agglomeration of the finely divided oxidic active material;
- (iv) after the end of the addition of powder and water, blowing hot air at 110°C into the coating pan for 20 minutes;
- (v) then drying for 2 hours at 250°C under air.

In the resulting ring shaped coated catalysts, the proportion of oxidic active material was 27% by weight, based on the total weight (i.e. the active material was in a supported ratio as described in the patent in suit, Paragraph [0019]).

(c) The preparation of acrylic acid from acrolein by gas-phase catalytic oxidation, in which coated catalysts VS1 and S1 were tested, in a model contact tube surrounded by a salt bath. The reaction mixture has a starting composition of 5% by volume of acrolein, 7% by volume of oxygen, 10% by volume of steam and 78% by volume of nitrogen, and was loaded at 3600 l/h (S.T.P.) of starting reaction gas mixture and the temperature of the salt bath was adjusted so that an acrolein conversion of 99 mol% resulted after a single pass.

The salt bath temperature T required for an acrolein conversion of 99 mol% and the selectivity S of the acrylic acid formation were as follows (Table 2 of D1):

TABLE 2

Coated catalyst used	T [°C]	S [mol %]
VS1	267	95.2
S1	263	95.3

9.4 Summing up, the process of D1 comprises the application of dry, finely divided oxide material as active catalyst mass on an inert support as such. The process of D1 does not contain a step of modifying the carrier before application of the active catalytic mass, let alone a step of calcining the so modified carrier at a temperature of 1200 to 1700°C.

9.5 Since the complex oxide catalyst of D1 is not supported on a modified carrier as defined in Claim 1 of the Main Request, the claimed catalyst is novel.

The problem solved

10. If the closest prior art were correctly acknowledged in the application as filed, the problem addressed in the application as filed would be taken as the starting point (Case Law of the Boards of Appeal of the EPO, 6th edition 2010, *supra*, I.D.4.3.2), in the present case as mentioned on page 1, lines 27-30 of the application as filed (Paragraph [0003] of the published specification), i.e. the development of catalysts which excel in stability and enable acrylic acid production at high yield over prolonged periods. Such is not the case in the present appeal however.

11. D1, considered by all parties, the Opposition Division and the Board as the closest prior art, was not acknowledged in the patent application as filed, and on which the patent in suit was granted. Hence, the problems and objectives mentioned in the application as filed, and on which the patent in suit was granted, were not formulated on the basis of the prior art D1.
 - 11.1 The problem effectively solved over D1 has thus to be established on the basis of the actual results provided by the claimed subject-matter over the known catalysts (Case Law, *supra*, I.D.4.4), having regard to the requirements that the problem should be derivable from the original application (Case Law, *supra*, I.D.4.4), that it should neither be artificial nor retrospective, and that it should not contain any pointer to the solution (Case Law, *supra*, I.D.4.3.1). Also, alleged advantages to which it is referred (e.g. by statements or examples) without offering sufficient evidence to support the comparison with the closest prior art D1, cannot be taken into consideration in formulating the problem solved by the alleged invention **over the whole**

breadth of the claims (see Case Law, *supra*, I.D.4.2 and VI.H.5.1.2(b)).

11.2 Thus, the question arises whether the attainment of an improvement by the claimed subject-matter over D1 has been demonstrated or is plausible, in particular over the whole breadth of Claim 1, more particularly when considering all conditions encompassed by Claim 1 but not exemplified, such as material, shape and size of the carrier other than those specified, carriage ratio beyond the limits given in the description and amounts of oxide/complex oxide catalyst other than illustrated.

11.3 The appellants have referred to alleged advantages illustrated by the examples and the comparative example of the patent in suit, and to the statements in Paragraph [0012] thereof as regards the role played by the carriage ratio. Having regard to D1, they have relied on Comparative Example D8.

11.4 The examples of the patent in suit illustrate carriers modified by the following oxides:

Example 1: $Mg_1Si_{1.5}Al_{0.1}$ (carriage ratio 18.3%);

Example 2: $(Ca_{0.5}Ba_{0.5})_1Si_{27}Na_{0.25}$ (carriage ration 5.4%);

Example 3: $Mg_1Si_{1.5}Al_{0.1}K_{0.01}Fe_{0.05}$ (carriage ratio 27%);

Example 4: $Sr_1Si_{2.4}Al_2$ (carriage ratio 12.6%);

Example 5: $Mg_1Si_{1.5}$ (carriage ratio 9.7%).

Comparative Example 1 used an inert carrier as such.

11.4.1 All the modified carriers and the inert carrier illustrated in the patent in suit are coated with the following, same, complex oxide catalyst:

$MO_{12}V_5W_1Cu_{2.2}Sb_{0.5}$ (supported ratio 22%)

11.4.2 Still according to the patent in suit (paragraph [0031], the catalytic performance was tested by charging 1000 ml of each of the above catalysts in a reaction tube, into which a gaseous mixture made up of 5% volume of acrolein, 5.5% volume of oxygen, 25% volume of steam and 64.5% volume of an inert gas comprising nitrogen was then introduced.

11.4.3 The catalyst performance is illustrated by Table 1 of the patent in suit, which is reproduced herebelow:

TABLE 1

	Catalyst No.		Reaction Temp. (°C)	Acrolein conversion (%)	Acrylic Acid Selectivity (%)	Acrylic Acid Yield (%)
Example 1	(1)	Initial stage of reaction	260	99.2	96.0	96.2
		After 8,000 hrs.	270	99.2	95.8	95.0
Comparative Example 1	(2)	Initial stage of reaction	260	98.4	94.4	92.0
		After 8,000 hrs.	287	98.6	93.9	92.6
Example 2	(3)	Initial stage of reaction	260	99.0	95.4	94.4
		After 8,000 hrs.	271	99.0	95.3	94.3
Example 3	(4)	Initial stage of reaction	260	99.3	96.9	96.2
		After 8,000 hrs.	269	99.2	96.7	94.9
Example 4	(5)	Initial stage of reaction	260	99.1	96.6	94.7
		After 8,000 hrs.	270	99.0	96.6	94.6
Example 5	(6)	Initial stage of reaction	260	99.0	96.1	94.1
		After 8,000 hrs.	273	99.1	96.0	94.1

11.5 These data show that the illustrated catalysts provide a high acrylic acid yield which is substantially maintained after 8000 hours. This fact is not contested, neither by the appellants nor by the Board.

11.6 The dispute rather concerns, on the one hand, the fact that the examples illustrated in the patent in suit concern a limited number of modified carriers, with a carriage ratio ranging from 5.4 to 27% and only one complex oxide catalyst, so that it is not apparent that these results can be attained with all complex oxides falling under Claim 1. On the other hand, Comparative example 1 does not represent D1. Hence, the examples of the patent in suit do not demonstrate that the good properties attained also represent an improvement over those attained by the catalysts disclosed in D1.

11.7 In particular, the statements given in Paragraph [0012] of the specification and referred to by the respondents read as follows:

"Where the carriage ratio is less than 1%, the effect of the modified carrier cannot be sufficiently obtained. Whereas, when it exceeds 50%, the surface properties of the inert carrier itself such as coarseness and porosity are impaired by the supported oxide, to reduce adhesion between the modified carrier and the catalytic component supported thereon, giving rise to such a problem as peel-off of the catalyst component".

The respondents have argued that an improvement is nevertheless attained also outside the given range for the carriage ratio, albeit not sufficiently.

11.8 The Board, in agreement with the appellants, instead considers that this statement does not credibly back up an attainment of the sought-for effect outside the disclosed carriage ratio. In particular, the question arises whether a carriage ratio (much) lower than 1% or (much) greater than 50% leads to a modified carrier. In fact, in the first case the amount of modification could be very small and in the latter case the amount of oxide of Formula (1) could be much greater than the amount of the inert carrier. Hence, either the inert carrier remains essentially unmodified, i.e. with little or no coating, or a new carrier is obtained by entirely coating the inert support with an amount of oxide of Formula (1) greater than that of the inert carrier. In the latter case, the catalytic mass would then be coated on the new carrier made of oxide of Formula (1). Both situations could not provide any effect linked to a partial coating of the inert carrier.

11.9 Hence, these statements referred to by the respondents cannot support the attainment of an improvement over the whole breadth of Claim 1.

11.10 D8 concerns a comparison over D1, which takes place in the context of the preparation of the complex oxide as illustrated in Paragraph [0029] of the patent in suit, which context is used for all the modified carriers illustrated in the patent in suit. In that context, the respondents have replaced the modified carrier of e.g. Example 1 of the patent in suit with a spherical carrier of steatite, which is one of the material of the carrier disclosed by D1 (and by D4 as well). Compared to Example 1 of the application as filed, the following results were obtained:

	Catalyst		Reaction temperature (°C)	Acrolein conversion	Acrylic acid selectivity	Acrylic acid yield
Example 1 of the present application	(I)	Initial stage of reaction	260	99.1	96.0	95.1
Comparative Example 2	(A)	Initial stage of reaction	260	96.7	93.7	90.6

11.11 However, apart from the choice of steatite for the inert carrier, the catalysts of D8 have not been prepared according to D1, let alone according to the most preferred embodiments of D1 (page 5, lines 22-37, and examples), in particular catalytic mass A and catalyst S1 of D1 have not been reproduced.

11.11.1 That the catalyst of Comparative Example 2 (D8) is not according to D1 is also confirmed by the results in Table 2 of D1, according to which Catalyst S1 of D1 attains acrylic acid selectivity of 95.3 at 263°C, which selectivity is higher than that illustrated by Comparative Example 2 (D8) and comparable with the values given in Table 1 of the patent in suit.

11.11.2 So D8 cannot be considered as reproduction of the closest catalysts of D1.

11.11.3 In any case, even if D8 were considered to represent D1, it would deal with only one catalyst within the whole breadth of Claim 1, i.e. would be insufficient.

11.11.4 Therefore, Comparative Example 2 of D8 does not back up the argument that an improvement over D1 in terms of catalytic properties is effectively attained over the whole breadth of Claim 1. As regards the deterioration of catalytic properties with time or stability, no comparison with D1 is available.

11.12 Since the problem solved cannot be formulated in terms of an improvement over the closest prior art D1, it has to be redefined, based upon the information present in the application as filed, as to provide further catalysts suitable for the vapour phase catalytic oxidation of acrolein to acrylic acid having good activity, selectivity and yield, which do not deteriorate too much with time.

Obviousness

12. D1 does not suggest an at least partial coating of an inert carrier with an oxide of Formula (1) according to the patent in suit, in order to obtain a modified carrier on which the catalytic active mass is coated. So D1 itself cannot lead the skilled person to a further complex oxide catalyst as defined in Claim 1.

12.1 D4 discloses (Claim 1) a process of production of ceramic catalyst carriers consisting of a dense carrier on which a coating of a desired thickness (e.g. 0.3 to 0.6 mm) is formed by coating the carrier with a ceramic material and a binder, then drying and calcining.

12.1.1 As regards the material of carrier and granular coating, D4 not only mentions steatite for the carrier and aluminium oxide for the granular coating (column 3, lines 10-11) but also illustrates specific compositions for carrier and granular coating, such as the one comprising 82-84% of soapstone, 8-9% plastic clay and 8-9% feldspar (column 3, lines 38-42).

12.1.2 It has been established in the decision under appeal that D7 (page 111, last full paragraph, penultimate sentence) confirms that it was known that calcination of the above composition at 1300-1400°C leads to steatite, whereby steatite indisputably falls under the definition of the oxide Formula (1) of Claim 1. These facts established in the decision under appeal are not in dispute.

12.2 Hence, the disclosure of D4 essentially corresponds to the features which distinguish the claimed subject-matter from the disclosure of D1.

13. The question which remains to be answered is whether the skilled person would have combined D1 and D4, e.g. in order to solve the reformulated problem.

13.1 The catalysts of D1 are suitable for oxidative chemical reactions carried out in the gas phase (page 2, lines 12-24), *inter alia* the oxidation of propylene to

acrolein, which is then oxidized to acrylic acid. The coated carriers of D4 are suitable for the production of oxidation catalysts (Claim 2), such as for making carboxylic acids from unsaturated aliphatic hydrocarbons (Column 4, lines 6-8). Hence, both D1 and D4 *inter alia* concern oxidation catalysts which are suitable for the production of carboxylic acids from unsaturated aliphatic hydrocarbons. Consequently, D1 and D8 also pertain to a common technical field.

13.2 According to D1 (page 3, lines 9-25), a general problem posed by coated catalysts was their production on an industrial scale, in such a way that:

- (a) they have the layer thickness required with regard to the catalyst activity;
- (b) the catalytically active coat satisfactorily adheres in the required thickness to the surface of the carrier;
- (c) the coat thickness shows very slight fluctuations over the surface of a carrier;
- (d) the coat thickness shows very slight fluctuations over the surface of different carriers;
- (e) the size of the specific, catalytically active surface area based on the mass unit of the active material is satisfactory; and,
- (f) the output of the production process is satisfactory.

13.3 The process for the preparation of catalysts disclosed by D1, which allegedly does not have the disadvantages of the prior art processes, *inter alia* comprises the following features (page 6, lines 20-28):

- (a) the surface of the carrier is rough, as increased surface roughness results in greater adhesion of the applied coat to the oxide active material;
- (b) the surface roughness of the carrier can range from 40 to 200 μm ;
- (c) the carrier material can be nonporous (i.e. having a total volume of pores of less than 1% by volume, based on the volume of the carrier) (this requirement implies that the carrier is dense);
- (d) the dense carrier can be spherical or annular.

13.3.2 Also D4 (Column 1, lines 28-42 and 44-50) addresses the preparation drawbacks and discloses that:

- (a) dense carrier are preferable over porous carriers, as they enable an easy and consistent production;
- (b) the spherical dense carriers also possess an improved resistance to attrition, i.e. better mechanical stability;
- (c) however, dense carrier generally have a smooth surface, which makes it difficult to coat with a porous catalytic layer in order to attain high activity and adhesion of the catalyst layer.
- (d) So the catalysts require replacement in short time, i.e. are not stable.

13.4 According to D4 (Column 2, lines 24-28; Column 3, line 55 - in particular "*Durch die rauhe Oberfläche des ursprünglichen glatten ...*", to Column 4, line 25), the modification of the dense carrier by the granular coating attains the following effects:

- (a) the catalytic oxide does not penetrate in the dense carrier;
- (b) the adhesion of the catalytic coating is good;
- (c) a desired thickness of the coating can be obtained;

- (d) the yield loss during the reaction can be reduced.
- 13.5 Hence, D1 and D4 also share a number of objectives and elements of solution.
14. The fact that D4 is older than D1 and nevertheless was not considered by the applicants of D1 is not decisive. In fact, the solution provided by D4 remains plainly suitable for making further catalysts starting from D1.
15. Therefore, the skilled person would have combined D1 with D4 in order to solve the reformulated problem and come to the claimed solution without inventive activity.

First and Second Auxiliary Requests

16. It is not in dispute that the amendments contained in the First and Second Auxiliary Requests do not change the closest prior art (D1), nor the problem effectively solved over the whole breadth of Claim 1 (as reformulated for the Main Request). Also not in dispute is the fact that e.g. steatite is mentioned in both D1, as possible material for the carrier, and D4, as possible material for the carrier or the coating. Finally, the appellants (letter of 5 November 2012, Point 3) have calculated, for a modified carrier according to D4, a carriage ratio within the range as claimed. Therefore, the modifications in Claim 1 of the First and Second Auxiliary Requests do not overcome the lack of inventive step decided for the Main Request. Hence, the First and Second Auxiliary Requests do not fulfil the requirements of the EPC and are rejected.

Third Auxiliary Request

17. In spite of the change in category, Claim 1 of the Third Auxiliary Request still relates exactly to the same catalyst composition as the Main Request, in particular its use for the specific purpose for which it has been acknowledged in the reformulation of the technical problem for the Main Request (Point 10.13, *supra*).
- 17.1 The appellants have decided in view of this not to present any different arguments for Claim 1 of the Third Auxiliary Request, but to refer to the arguments already presented for the Main Request.
- 17.2 The respondents have argued that D4 should no longer be considered, as it did not address the production of acrylic acid.
- 17.3 The Board considers, as also acknowledged in Paragraph [0022] of the patent in suit, that apart from the modified catalyst the process per se follows generally practiced methods. The modified catalyst is however still the catalyst of the Main Request, which has been found to be obvious.
- 17.4 Under such circumstances, the Board does not see any reason to come to a different conclusion as regards the appreciation of inventive step with respect to the same prior art and does not see any need to analyse the issue in any further detail.
- 17.5 The subject-matter of Claim 1 of the Third Auxiliary Request does therefore not involve an inventive step.

Conclusions

18. The patent in suit in the amended form of the Main Request or of any of the First to Third Auxiliary Requests does not fulfil the requirements of the EPC.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The patent is revoked.

The Registrar:



S. Fabiani



The Chairman:



J. Riolo

