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**Datasheet for the decision
of 16 June 2020**

Case Number: T 2406/18 - 3.3.06

Application Number: 04773122.9

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C07C51/235, C07C57/055,
C07B61/00, B01J23/887,
B01J23/888

Language of the proceedings: EN

Title of invention:

Process for producing composite oxide and composite oxide
catalyst

Applicant:

Mitsubishi Chemical Corporation

Headword:

Composite oxide catalyst / MITSUBISHI CHEMICAL

Relevant legal provisions:

EPC Art. 56

Keyword:

Inventive step (all requests) : no - no effect convincingly
shown over the closest prior art

Decisions cited:

T 0181/82, T 0197/86, T 1397/08

Catchword:



Beschwerdekammern

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Chambres de recours

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Case Number: T 2406/18 - 3.3.06

D E C I S I O N
of Technical Board of Appeal 3.3.06
of 16 June 2020

Appellant:
(Applicant)

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Decision under appeal:

**Decision of the Examining Division of the
European Patent Office posted on 4 May 2018
refusing European patent application No.
04773122.9 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman J.-M. Schwaller
Members: L. Li Voti
R. Cramer

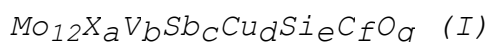
Summary of Facts and Submissions

- I. The appeal lies from the decision of the examining division to refuse European patent application n° 04773122.9 on the ground that the claimed subject-matter lacked inventive step over the combined teachings of documents D1 (JP 2003 210991 A) and D2 (DE 43 29 907 A1).
- II. With its statement of grounds the appellant maintained the requests presented before the examining division with some amendments to the first and second auxiliary requests.
- III. In the communication expressing its preliminary opinion, the board held the claimed subject-matter to lack inventive step over the disclosure of D1 taken in combination with the teaching of either of D2, D3 (DE 196 38 249 A1) or D5 (JP 2003 251 184 A).
- IV. With its reply dated 13 May 2020 the appellant filed two amended sets of claims as first and second auxiliary requests.
- V. During the oral proceedings held on 16 June 2020 the question focused on the question whether or not the subject-matter of claim 1 of all the requests then on file met the requirements of Article 56 EPC starting from document D1 as representing the closest prior art.
- VI. After closure of the debate, the appellant's final requests were that the decision under appeal be set aside and a patent be granted on the basis of the claims of the main request filed before the first instance on 23 August 2012, or alternatively of the

claims of one of the first or second auxiliary request filed with the letter of 13 May 2020, or of the third auxiliary request filed during the oral proceedings before the first instance on 11 April 2018.

VII. Claim 1 according to the **main request** reads as follows:

"1. A method for producing a composite oxide catalyst, which is a method for producing a composite oxide catalyst having the following formula (I), to be used at the time of gas phase catalytic oxidation of acrolein with a molecular oxygen-containing gas to produce the corresponding acrylic acid, characterized in that Sb₂O₃ of isometric system is used as at least a part of an antimony-supplying source compound:



(wherein the respective components and variables have the following meanings:

*X is at least one element selected from the group consisting of Nb and W;
a, b, c, d, e, f and g represent atomic ratios of the respective elements, and against 12 of molybdenum atom, $0 < a \leq 10$, $0 < b \leq 10$, $0 < c \leq 5$, $0 < d \leq 5$, $0 \leq e \leq 1,000$, $0 \leq f \leq 1,000$, and g is a number determined by the degrees of oxidation of the above respective components),
wherein Sb₂O₃ is used as the antimony-supplying source compound, and the ratio (a_1/a_2) of the intensity (a_1) at $2\theta =$ about 28.3 to the intensity (a_2) at $2\theta =$ about 27.6, of its X-ray diffraction (anticathode Cu-K α), is at most 0.2."*

Claim 1 of the **first auxiliary request** is further defined by the features "*and wherein an aqueous dispersion of supply source compounds containing the respective catalyst component elements shown by the formula (I), is prepared, and such an aqueous dispersion is dried to a powder, which is molded, and the molded product is calcined, wherein the calcination is carried out in an atmosphere containing molecular oxygen in an amount of at most 10 vol%.*"

Claim 1 of the **second auxiliary request** further requires that the powder is molded "***by using at least one binder selected from the group consisting of silica, graphite and cellulose***".

Claim 1 of the **third auxiliary request** further requires that the molded product is calcined "***at a calcining temperature of from 250°C to 600°C over a calcining time of from 1 to 50 hours***".

Reasons for the Decision

1. *Main request - Inventive step (Article 56 EPC)*
- 1.1 The present application (page 1, lines 5-9 and claim 1) concerns a method for producing a catalyst to be used for gas phase catalytic oxidation of acrolein with a molecular oxygen-containing gas to produce the corresponding acrylic acid.
- 1.2 As stated in the description (page 3, lines 21-25) conventional composite oxide catalysts for this type of reaction exhibit excellent properties but it is desirable to achieve a still higher conversion of the starting unsaturated aldehyde material or a still

higher selectivity in the produced unsaturated carboxylic acid.

According to the application (page 4, lines 5 to 12 read in combination with the passage bridging pages 6 and 7) the purpose of the invention is thus to provide a method for producing a composite oxide catalyst which allows a high conversion of acrolein and a high selectivity for acrylic acid when used in a gas phase catalytic oxidation with a molecular oxygen containing gas for producing acrylic acid from acrolein.

- 1.2.1 Document D1, which deals with the same purpose (see abstract and paragraphs [0011] and [0012]), is the most suitable starting point for the evaluation of inventive step and it is undisputed that its example 1 represents the closest prior art.

It is also not in dispute that the catalyst produced in this prior art example already allows high conversion of acrolein and high selectivity to acrylic acid in the same reaction as in the claimed subject-matter.

As D1 does not disclose the source of antimony used, the preparation disclosed in its example 1 thus differs from the subject-matter of claim 1 at issue only in that D1 is silent as to whether or not the Sb_2O_3 used as the antimony source provides under X-ray diffraction (anticathode Cu-K α) a ratio (a_1/a_2) of the intensity (a_1) at $2\theta =$ about 28.3 to the intensity (a_2) at $2\theta =$ about 27.6 of at most 0.2. It is undisputed that the intensities (a_1) and (a_2) correlate with the amounts of rhombic and isometric crystalline phases present in the Sb_2O_3 ; this means that the isometric crystalline phase (also called senarmontite) must be present in an amount

of at least 83% (see also point 11.5 of the decision under appeal).

- 1.3 The appellant formulated the underlying technical problem as the provision of a method for producing a composite oxide catalyst to be used in a gas phase catalytic oxidation with molecular oxygen for producing acrylic acid from acrolein, resulting in a catalyst exhibiting **improved** activity, i.e. conversion rate (of acrolein), and **improved** selectivity (to acrylic acid) or, in the alternative, **superior** activity while maintaining **high** selectivity.

The board notes in this respect that the catalyst of the closest prior art (example 1 of D1) provides an acrolein conversion rate of 99.8%, an acrylic acid selectivity of 98.6% and an acrylic acid yield (conversion rate multiplied by the selectivity) of 98.4%, i.e. values which are almost identical to and in any case not worse than those achieved by the catalyst used in example 1 of the present application, namely 99.7%, 98.7% and 98.4%, respectively. So an improvement over the closest prior art is not identifiable.

As correctly argued by the appellant, the catalyst of example 1 of the present application contains less niobium (Nb) and vanadium (V) and more silicon (Si) and carbon (C) as the one from example 1 of D1, but as acknowledged by the appellant himself - when referring to the selectivity of certain catalysts listed in the table on page 7 of D2 (M4 vs M7 and M5 vs M9) - even small variations in the catalyst formula may affect its performance. There is however no data on file showing how such variations may affect the claimed catalyst. The board furthermore notes that there are even further differences between example 1 of the application and

the closest prior art, namely in the reaction conditions, in particular the space velocity, the composition of the reacting gas and the reaction temperature, which may all also affect the catalytic performance, but also here there is no data showing how these differences may affect the results.

It follows that in the absolute it cannot be concluded that the closest prior art catalyst is worse than the one of example 1 of the application, and also the appellant's argument that the conversion rate in example 1 of the application would be allegedly better than that of example 1 of D1 - because of the much higher space velocity - is speculative in the absence of evidence and can thus not be taken into consideration.

It follows that it cannot be established whether or not the catalyst of example 1 performs better than the closest prior art catalyst and whether or not the technical problem proposed by the appellant has been convincingly solved by the subject-matter of claim 1 at issue.

- 1.3.1 In such a situation, according to established jurisprudence, the only factors of importance for the determination of the objective technical problem are results actually achieved in relation to the closest prior art (T 1397/08, catchword and point 3.3, third full paragraph, of the reasons). Furthermore, if comparative tests are chosen to demonstrate an inventive step on the basis of an improved effect, a comparison should be made with the closest state of the art and should convincingly show to have its origin in the distinguishing feature of the invention (Case Law

of the Boards of Appeal of the EPO, 9th edition 2019, page 271, I.D.10.9, first paragraph).

1.3.2 The appellant referred to decisions T 181/82 (OJ EPO 1984, 401) and T 197/86 (OJ EPO 1989, 371) and maintained that it was not necessary to provide evidence of an improvement over the closest prior art since the application already showed an improvement by comparing examples 1 and 2, which both made use of the same process conditions, but example 2 used a catalyst which differed from that of example 1 in that its ratio (a_1/a_2) in the antimony-supplying source was more than 0.2, thus containing less isometric crystalline phase than required in claim 1 at issue.

1.3.3 The board notes that in case **T 181/82** it was decided (Headnotes I and II) that where comparative tests are submitted as evidence of an effect, there must be the closest possible structural approximation - in a comparable type of use - to the subject-matter of the invention and that only known substances - not notionally described ones - qualify for use when comparing compounds.

Therefore, according to this decision only subject-matter belonging to the state of the art qualifies as suitable comparison. Example 2 of the present application is thus not an appropriate comparison. Moreover, it is not disputed that example 1 of D1 has the closest possible structural approximation to the subject-matter of claim 1 at issue, since it differs therefrom only in that it does not explicitly disclose that the antimony-supplying Sb_2O_3 has a ratio (a_1/a_2) of at most 0.2, and so if at all, it differs from the claimed subject-matter in only one feature, like example 2 of the present application. Hence by

following T 1818/82 a comparison should have been made with example 1 of D1.

In decision **T 197/86** the board applied the same criteria as T 181/82 in order to establish an effect over the closest prior art (point 4 of the reasons). Further the board decided that additional comparisons not belonging to the prior art and differing from the claimed subject-matter only in the distinguishing feature allegedly responsible for the effect and not in several characteristics as the closest prior art considered in that case, were acceptable in order to establish that the same effect existed across the entire scope of the claims (see Headnote and points 6.1.2 and 6.1.3).

Thus, also this decision confirmed that the criteria established in T 181/82 has to be applied in order to ascertain that an effect exists over the closest prior art and that additional comparisons not belonging to the prior art, like example 2 of the present application in the present case, are allowable in order to establish the existence of an effect across the entire scope of the claims in case the closest prior art differs in terms of several technical features from the claimed subject-matter.

The tenure of this decision is thus not applicable to the present case wherein already the closest prior art differs from the claimed subject-matter only in one feature, like in example 2 of the application, and wherein the existence of an effect over the entire scope of the claim does not need to be discussed since no evidence of an improvement over the closest prior art was made credible.

- 1.4 It follows that the technical problem underlying the invention, starting from Example 1 of D1 as representing the closest prior art, is to be reformulated as the provision of a further method for producing a composite oxide catalyst which presents high conversion of acrolein and high selectivity for acrylic acid when used in a gas phase catalytic oxidation of an unsaturated aldehyde to produce the corresponding unsaturated carboxylic acid.
- 1.5 Since the subject-matter of claim 1 manifestly solves this problem, it remains to be decided whether it was obvious for the skilled person to use in the method of example 1 of D1 a catalyst prepared by using as antimony-supplying source an Sb_2O_3 containing at least 83% of the isometric crystalline phase (senarmontite).
- 1.5.1 The appellant submitted that, starting from example 1 of D1, several other modifications were indeed possible for the skilled person in order to provide a further method for producing a composite oxide catalyst able to provide high conversion of acrolein and a high selectivity for acrylic acid.
- 1.5.2 The board does not deny this fact but in the absence of indication in D1, the skilled person has **in any case** to select an antimony-supplying source among those known from the prior art. In this respect, he would inevitably have noted that senarmontite, i.e. Sb_2O_3 containing at least 83% of the isometric crystalline phase, had already been used in the prior art (see e.g. D2, page 3, lines 1-23 and page 5, lines 23-29); D3 (page 3, lines 6-28; page 4, lines 40-46; page 5, line 21) and D5 (paragraphs [0014]-[0017])) for preparing catalysts containing all the elements of the catalyst of example 1 of D1 and that all these catalysts were

suitable for use in the gas phase oxidation of unsaturated aldehydes with molecular oxygen-containing gas to produce the corresponding unsaturated carboxylic acids, including the oxidation of acrolein to acrylic acid.

Moreover, he would have noted that both D2 (page 2, lines 30-37) and D3 (page 3, lines 21-24) teach that the use of senarmontite can provide better catalytic performance at least in the oxidation of methacrolein to methacrylic acid.

- 1.5.3 Even though - as submitted by the appellant - the reaction of methacrolein to methacrylic acid is not necessarily comparable to that of acrolein to acrylic acid, the board notes that initially the application described the claimed catalyst as plainly suitable for both reactions, so that the above information from D2 and D3 clearly also qualifies senarmontite as a promising antimony-supplying source not affecting negatively the catalytic performance in the reaction of acrolein to acrylic acid.

Therefore, the board is convinced that in the light of the teaching of the prior art, it would have been directly apparent to the skilled person that an Sb_2O_3 containing at least 83% of the isometric crystalline phase (senarmontite) was a very suitable antimony source for the preparation of the catalyst disclosed in example 1 of D1.

- 1.5.4 As regards the further argument of the appellant concerning an alleged non obvious combination in view of the different microstructure of the catalyst used in D2 (point 5.4 of the statement of grounds), the board notes that claim 1 at issue does not contain any

limitation as to the retention of the antimony oxide having isometric structure in the final catalyst nor to the microstructure of the catalyst used, so that this argument is irrelevant for supporting inventive step.

- 1.5.5 The board thus concludes that it would have been obvious for the skilled person, faced with the underlying technical problem and looking for a suitable source of antimony for the catalyst of D1/example 1, to use senarmontite, i.e. a Sb_2O_3 containing at least 83% of the isometric crystalline phase, as an antimony source for the catalyst of example 1 of D1.

The subject-matter of claim 1 of the main request was thus obvious from the prior art and thus lacks inventive step (Article 56 EPC).

2. *Auxiliary requests 1 to 3 - Inventive step*

- 2.1 The method of preparation disclosed in example 1 of D1 (paragraphs [0041]-[0043]), which represents the closest prior art, differs from the subject-matter of each claim 1 of these auxiliary requests only in that the calcination step of the catalyst is carried out in a nitrogen atmosphere instead of an atmosphere containing molecular oxygen in an amount of at most 10%. All the other additional features of claim 1 of these requests are in fact already disclosed in example 1 of D1. This is not in dispute.

- 2.2 The board notes however that the claimed method does not require any lower limit for the amount of molecular oxygen and thus it includes methods wherein the calcination atmosphere contains extremely small amounts of oxygen, with the rest being for example nitrogen.

The application moreover does not contain any example showing that the use of an atmosphere containing molecular oxygen in an amount of at most 10% gives rise to any particular or unexpected effect. To the contrary, like in the closest prior art, example 1 of the application makes use of a nitrogen atmosphere during calcination.

2.3 Therefore, considering that the technical problem remains as formulated in point 1.4 above and that D1 (paragraph [0036]) itself teaches that the calcination step may be carried out in the presence of an inert gas (like nitrogen as in example 1) or molecular oxygen, and a similar teaching being also present in D2 (page 4, lines 25-27), it would have been obvious for the skilled person to try, as an alternative to the calcination under nitrogen atmosphere of example 1 of D1, a calcination step wherein a small amount of molecular oxygen is added to nitrogen. For the board the choice of a suitable amount of molecular oxygen is nothing else a mere optimisation of the process that the skilled person would obviously carry out and so arrive without inventive skill at the claimed subject-matter.

2.4 Therefore, the board concludes that the subject-matter of claim 1 of all auxiliary requests is obvious from the prior art and thus lacks inventive step as well (Article 56 EPC).

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:



A. Pinna

J.-M. Schwaller

Decision electronically authenticated