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**Datasheet for the decision  
of 10 September 2008**

**Case Number:** T 1022/06 - 3.3.07

**Application Number:** 01308130.2

**Publication Number:** 1192992

**IPC:** B01J 23/887

**Language of the proceedings:** EN

**Title of invention:**

Method for reactivating catalyst for methacrylic acid preparation

**Applicant:**

SUMITOMO CHEMICAL CO., LTD.

**Opponent:**

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**Headword:**

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**Relevant legal provisions:**

EPC Art. 56, 123(2)

**Relevant legal provisions (EPC 1973):**

-

**Keyword:**

"Inventive step (yes) - non obvious alternative"

**Decisions cited:**

-

**Catchword:**

-



Case Number: T 1022/06 - 3.3.07

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.07  
of 10 September 2008

**Appellant:** SUMITOMO CHEMICAL CO., LTD.  
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**Representative:** Cresswell, Thomas Anthony  
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**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office posted 8 February 2006  
refusing European application No. 01308130.2  
pursuant to Article 97(1) EPC.

**Composition of the Board:**

**Chairman:** S. Perryman  
**Members:** F. Rousseau  
B. ter Laan

## Summary of Facts and Submissions

I. The appeal, filed on 12 April 2006, lies from the decision of the Examining Division posted on 8 February 2006, to refuse European application No. 01 308 130.2.

II. The application has European publication No. 1 192 992, is entitled "Method for reactivating catalyst for methacrylic acid preparation", and was originally filed with eleven claims, of which claims 1, and 11 read as follows:

"1. A process of reactivating a deteriorated catalyst containing phosphorus and molybdenum suitable for use in the production of methacrylic acid by vapor-phase oxidation of methacrolein or vapor-phase oxidative dehydrogenation of isobutyric acid which process comprises treating the catalyst with a gas containing a nitrogen-containing heterocyclic compound."

"11. A process of producing methacrylic acid through catalytic vapor-phase oxidation of methacrolein or catalytic vapor-phase oxidative dehydrogenation of isobutyric acid, using a catalyst which has been reactivated by the process as defined in any one of claims 1 to 10."

III. The decision of the Examining Division referred to documents

D1 US-A-4 303 550,

D2 EP-A-0 268 201, and

D3 JP-A-60-232247 in the form of its abstracts in  
Patent Abstracts of Japan

The decision can be summarized as follows:

*Main Request*

- (a) Claim 1 of the main request before the Examining Division, filed on 21 November 2005, corresponded to claim 1 as originally filed with at its end the added words "at a temperature at which components in the gas do not liquefy". These added words were found to have no basis in the application as originally filed, contrary to the requirements of Article 123(2) EPC, and not to be clear contrary to the requirements of Article 84, since it was not clear which components should not liquefy.
- (b) Novelty was acknowledged since D1 did not explicitly disclose the use of a nitrogen-containing heterocyclic compounds in the regeneration process.
- (c) Inventive step of the subject-matter of this claim was denied, referring to the problem to be solved as being that stated on page 2, paragraph three of the application, to provide a method of efficiently reactivating a catalyst, containing P and Mo, which is used in methacrylic acid preparation and whose activity level has dropped.
- (d) D1 disclosed treatment of the deactivated catalyst with an aqueous vapour of a volatile base, which could be ammonia, amines tetraalkyl ammonium

hydroxides or other free bases having a pH of at least 8. Having regard to this disclosure a person skilled in the art would know of his own knowledge or on the basis of D2 or D3 that nitrogen-containing heterocyclic compounds could be used. This was unambiguously the case for a process taking place in liquid phase. However this applied also to the embodiments in D1 involving a gas phase for the reactant(s), the preferred embodiment disclosed in D1. *A priori* the skilled person expected in view of D1 that all "bases having a pH of at least 8", that were used in the liquid phase, could be used in the vapour phase as far as they could be vaporized at the operating conditions. Moreover, the applicant had not shown that the use of these nitrogen-containing heterocyclic led to a surprising effect or had an advantage over the regeneration method used in the prior art.

- (e) The applicant's arguments that D1 related to a liquid phase process could not be accepted, since it was clearly stated in D1 that regeneration was preferably carried out in the vapour phase. Additionally, the current application did not exclude a fluid phase in the pores of the catalyst during regeneration either.
- (f) Further the process of the present application was a process **comprising** said steps, thus leaving open the option for further process steps such as disclosed in the description page 6, paragraph one, effectively reducing the claimed process to the process of D1.

(g) The auxiliary request, with a claim 1 corresponding to claim 1 as originally filed with at its end the added words "at a temperature at which the nitrogen-containing heterocycle does not liquefy" while apparently overcoming the objection under Article 123(2) EPC raised against claim 1 of the main request, and being novel over D1, lacked inventive step for the same reasons as given for claim 1 of the main request.

IV. With their statement setting out the grounds of appeal received on 16 June 2006, the appellants (applicants) submitted two sets of claims, as their main and first auxiliary requests, respectively. In reply to a communication of the Board sent as an annex to the summons for oral proceedings, the appellants filed a main and four auxiliary requests, replacing the requests then on file.

V. Oral proceedings were held on 10 September 2008, in the course of which the appellants submitted a new main request of five claims. In the main request, independent claim 1 corresponded to claim as originally filed, and independent claim 5 read as follows (with additions compared to the claim 5 as originally filed being shown in bold and underlined and deletions shown struck out):

"5. A process of producing methacrylic acid which process comprises reactivating a deteriorated catalyst in accordance with any one of claims 1 to 4 and which process further comprises producing methacrylic acid through catalytic vapor-phase

oxidation of methacrolein or catalytic vapor-phase oxidative dehydrogenation of isobutyric acid, using a the catalyst which has been reactivated ~~by the process as defined in any one of claims 1 to 10.~~"

VI. The arguments of the appellants, submitted in writing and at the oral proceedings can be summarised as follows:

- The amendments were based on the original disclosure.
- Document D1 did not disclose the use of a nitrogen-containing heterocyclic compound and documents D2 and D3 did not disclose a gas phase treatment of the catalyst. Hence, the claimed subject-matter was novel.
- As regards inventive step, the problem solved over document D1, the closest state of the art, was to provide an improved process to obtain catalysts having high performance over a prolonged period of time compared to the catalysts reactivated in document D1 which were indicated in D1, column 7, lines 47 to 63 to last at most 1080 hours. In addition, the claimed process did not require the use of an acid and was therefore much simpler.
- Even if an improvement over document D1 could not be acknowledged, the problem solved over document D1 had then to be formulated as an alternative to the process disclosed in document D1. The skilled person starting from document D1 would not have

arrived at the process now being claimed, because he would have had to leave out the essential step in the process of document D1 of using an acid. Furthermore, the passage relating to the use of a base in D1, column 4, lines 51-57 did not suggest the use of a nitrogen-containing heterocyclic compound, as in particular the meaning of the wording "and other free bases having a pH of at least 8 and which are capable of decomposing any residual PMA structure in the deactivated catalyst" did not allow the skilled person to understand which free base was meant.

- The other documents suggested a regeneration process, according to which a fluid phase within the pores of the catalyst was necessary. In contrast, in all steps of the claimed process, the nitrogen-containing heterocyclic compound was gaseous, as illustrated by Example 1 of the present application, using the Antoine equation.
- The appellants, however, admitted at the oral proceedings before the Board that they had no evidence for the absence in the present process of liquefaction of the nitrogen-containing heterocyclic compound within the pores of the catalyst.
- The reformulation of claim 5 explicitly required the regeneration as a first process step, and so the process of claim 5 was inventive for the same reasons as the process of claim 1. The reformulation avoided the objection raised by the Board that it had not been shown that the catalyst



produced according to the process of claim 1 was necessarily different from the catalyst produced by the process of D1.

- VII. The appellants requested that the decision under appeal be set aside and that a patent be granted on the basis of the claims of the Main Request submitted at the oral proceedings on 10 September 2008.

## **Reasons for the Decision**

### *Amendments*

1. Claims 1 to 4 correspond to claims 1 to 4 as originally filed. Claim 5, which is directed to a process of producing methacrylic acid comprising the step of reactivating the deteriorated catalyst in accordance with the process of claims 1 to 4, is based on claim 11 as originally filed reformulated to explicitly include as a process step the process of one of claims 1 to 4. The Board is therefore satisfied that the claims according to the main request satisfy the requirements of Article 123(2) EPC.

### *Clarity*

2. The appellants returned to the wording of original claim 1 which, in contrast to the claims underlying the decision under appeal, does not give rise to clarity objections.

*Novelty*

3. The novelty of the claimed process was not objected to by the Examining Division and the Board sees no reason to take a different view as document D1 does not disclose the use of a nitrogen-containing heterocyclic compound and neither of documents D2 and D3 discloses the treatment of the deteriorated catalyst with a gas containing said nitrogen-containing heterocyclic compound.

*Inventive step*

4. *Closest prior art*

- 4.1 The present application aims at providing a method for efficiently reactivating a catalyst containing phosphorous and molybdenum suitable in the production of methacrylic acid by vapour-phase oxidation of methacrolein or vapour-phase oxidative dehydrogenation of isobutyric acid. The claimed solution comprises the treatment of the deteriorated catalyst with a gas containing a nitrogen-containing heterocyclic compound.
- 4.2 The Board, in agreement with the appellants and the examining division, considers document D1 to represent the closest state of the art. It discloses a method for the regeneration of deactivated phosphomolybdic acid based catalysts comprising the steps of: imbibing an aqueous volatile base within the pore structure of said deactivated catalyst so as to form a fluid phase within the pore structure; adding hydrochloric acid to said fluid phase so as to form a precipitate (column 3, lines 18 to 33 and example 2). The precipitate is

thereafter dried and calcined to yield the regenerated catalyst. The treatment is conducted in vapour phase and the pressure is selected so as to obtain condensation of the volatile base and volatile acid within the catalyst particle pores (column 5, lines 13-32 and 48-52, and column 3, lines 29-33). Useful bases for the process of document D1 are disclosed as including ammonia, amines such as trimethylamine, tetraalkyl ammonium hydroxides having from 4 to about 16 carbon atoms and as defined in this prior art "other free bases having a pH of at least 8 and which are capable of decomposing any residual PMA structure in the deactivated catalyst to form a molybdate salt of the base" (column 4, lines 51-57), wherein PMA means phosphomolybdic acid (column 1, lines 21-22). The preferred base is ammonia (column 5, lines 1-2).

5. *Problem solved*

5.1 In view of this state of the art, the appellants submitted that the technical problem underlying the application consisted in providing a process of reactivating the deteriorated catalyst which restores the activity level of the catalyst approximately to the original level, whereby the reactivated catalyst exhibits a longer activity in comparison to a catalyst reactivated with the method of document D1. The appellants in particular referred to D1, column 7, lines 47 to 63, alleging that the method described permitted 2 to 10 catalyst turn-overs per three month period, meaning that the reactivated catalyst of document D1 was used at most for 45 days, i.e. 1080 hours. In contrast the catalysts reactivated with the process employed in example 1 of the application could

be used during a longer period of time while maintaining the same level of activity.

5.2 The Board, however, notes that the process shown in example 1 of the present application does not use a reactivated catalyst for more than 1000 hours and therefore does not support the appellants' allegation. In addition, when comparing results of tests in support of the allegation that an improved technical effect has been achieved by claimed subject-matter, it is necessary to convincingly show that the improved effect has its origin in the features distinguishing the claimed invention from the closest prior art, i.e. in the present case in the use of a gas containing a nitrogen-containing heterocyclic compound. However, such comparison is not available, as the processes which have been compared by the appellants, have not been shown to employ the same catalytic composition and identical reaction parameters for the production of methacrylic acid. As alleged but unsupported advantages cannot be taken into consideration for the determination of the problem underlying the claimed invention, the technical problem defined by the appellants must be reformulated in a less ambitious way.

5.3 Thus, in view of the teaching of document D1, the objective problem underlying the present application can only be seen in providing a further process of effectively reactivating a deteriorated catalyst containing phosphorus and molybdenum which is suitable for use in the production of methacrylic acid by vapour-phase oxidation of methacrolein, and this problem can on the information provided in the present application be regarded as solved.

6. *Obviousness*

6.1 It remains to be decided whether or not the skilled person starting from document D1 and wishing to solve the above defined problem would have been guided by the available prior art to treat the deteriorated catalyst with a gas containing a nitrogen-containing heterocyclic compound.

*Document D1*

6.2 The appellants have argued that document D1, in contrast to the process presently claimed, relies on the liquefaction of the treating gas within the pores of the catalyst and thus, would not have taught the presently claimed process to the skilled person. In the absence of any evidence for the appellants' contention, that the claimed process does not lead to condensation of the nitrogen-containing heterocyclic compound in the pores of the catalyst, their argument based on this unsubstantiated allegation, however, must be rejected. The applicants' argument that it would not be obvious to arrive at the subject-matter of present claim 1 starting from document D1, as it would be necessary to suppress the essential step of an acid treatment in the process of document D1, also fails to convince, as the absence of an acid treatment is not a feature of present claim 1. The question to be answered in the present case is rather whether the skilled person would have found any motivation in the prior art to treat the deteriorated catalyst with a gas containing a nitrogen-containing heterocyclic compound (see point 6.1 above).

6.3 According to the broadest teaching of D1, the vapours used for regenerating the deactivated phosphomolybdic acid based catalyst are those of volatile and decomposable bases and acids (column 4, lines 49-51). The bases which have been found in document D1 to have utility are disclosed in column 4, lines 51-57. They are defined to be ammonia, amines such as trimethylamine, tetraalkyl ammonium hydroxides having from 4 to 16 carbon atoms and "other free bases having a pH of at least 8 and which are capable of decomposing any residual PMA structure in the deactivated catalyst to form a molybdate salt of the base". Therefore the bases for the regeneration of the deactivated phosphomolybdic acid based catalyst which are explicitly disclosed in document D1 are not nitrogen-containing heterocyclic compounds. Concerning the meaning of the expression "other free bases having a pH of at least 8 and which are capable of decomposing any residual PMA structure in the deactivated catalyst to form a molybdate salt of the base nitrogen-containing heterocyclic compound", it is not clear whether the other free bases mentioned in document D1 refer to the term "amines" used at the beginning of the sentence or to any kind of other bases in general. In addition, the characteristics of these free bases, i.e. a pH of at least 8 and the ability of decomposing any residual PMA structure in the deactivated catalyst to form a molybdate salt of the base, do not provide any clear disclosure of which bases are in fact contemplated. The indication of a pH value, rather than a dissociation constant, gives no clear indication of which bases are to be considered, as the pH of the base depends not only on the base itself but also on many other variables which are not defined in document D1.

Furthermore, the functional definition of the bases chosen in document D1, i.e. "which are capable of decomposing any residual PMA structure in the deactivated catalyst to form a molybdate salt of the base" has no clear meaning for the skilled reader. In particular, neither is the meaning of "residual PMA structure" in the expression "capable of decomposing any residual PMA structure" defined in document D1, nor can any clear meaning for it be deduced from the purpose of the treatment step with the free base, which is the regeneration of the phosphomolybdic acid based catalyst. Nor do the phenomena described in document D1 as being responsible for the deactivation of the catalyst, namely loss of acid sites via condensation cross-linking or decomposition of the acid structure (column 1, lines 29-31 and lines 42-51) serve to elucidate the meaning. Thus, the skilled person would be unable to extract any meaningful teaching from document D1 with respect to "the other free bases" to be used in the regeneration treatment of the deactivated catalyst. Hence, the skilled person could not find in document D1 the suggestion to use within the framework of the invention disclosed therein a nitrogen-containing heterocyclic compound, if he wanted to solve the problem mentioned above. Therefore, document D1 does not by itself render the now claimed subject-matter obvious.

*Documents D2 and D3*

- 6.4 Document D2 discloses the regeneration treatment of a phosphomolybdic acid based catalyst, wherein the deactivated catalyst is treated in a dispersed state with an aqueous solution to which is added a nitrogen-

containing heterocyclic compound (page 2, lines 46-53, examples 1, 4). After aging, the substance obtained is dried, reshaped and calcined to give the regenerated catalyst.

6.5 Document D3 in the form of its abstracts provides a similar teaching. It discloses the regeneration treatment of a phosphomolybdic acid based catalyst, wherein the deactivated catalyst is after dispersion in water treated with an aqueous solution comprising a nitrogen-containing heterocyclic compound. The material treated is then dried, reshaped and calcined, resulting in a regenerated catalyst.

6.6 Thus, documents D2 and D3 both describe regeneration treatments that, contrary to the process used in document D1, require as a preliminary step the dispersion of the catalyst in an aqueous solution before it is brought into contact with the treating agent and as additional necessary step drying and reshaping of the treated catalyst. Thus, the methods taught in document D1 on the one hand and in document D2 or D3 on the other hand are different in nature. There is no hint that the treating agent used in document D2 or D3 in a regeneration method requiring that the deteriorated catalyst must be first prepared in a dispersed state before the treating agent is introduced, would be considered by the skilled person to be suitable for effective regeneration of the catalyst in a treatment method, which does not require the deteriorated catalyst to be dispersed before being treated. Thus, documents D2 and D3 do not suggest, either by themselves or taken in combination with document D1, that a deteriorated phosphomolybdic acid



based catalyst should be treated, i.e. brought into contact, with a gas containing a nitrogen-containing heterocyclic compound, as required by the presently claimed process, in order to effectively regenerate said catalyst.

6.7 Therefore, there is no evidence that the skilled person in view of the prior teaching available would have arrived at the subject-matter of present claim 1 in an obvious manner. To arrive at a different conclusion would rather require hindsight based on the knowledge of the claimed invention.

7. Therefore, the subject-matter of present claim 1 and by the same token that of dependent claims 2 to 4 and that of independent claim 5, which include all the features of claim 1, meets the requirements of Article 56 EPC.

**Order**

**For these reasons it is decided that:**

1. The decision under appeal is set aside.
  
2. The case is remitted to the first instance with the order to grant a patent on the basis of claims 1 to 5 of the Main Request submitted at the oral proceedings on 10 September 2008 and a description to be adapted thereto if necessary.

The Registrar

The Chairman

C. Eickhoff

S. Perryman