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D E C I S I O N
of 10 October 1994

Case Number: T 0170/92 - 3.3.2

Application Number: 87401987.0

Publication Number: 0263005

IPC: - B01J 27/28

Language of the proceedings: EN

Title of invention:
A process for regenerating a catalyst and its application to an oxidative dehydrogenation process

Applicant:
ELF ATOCHEM S.A

Opponent:
-

Headword:
-

Relevant legal norms:
EPC Art. 56

Keyword:
"Inventive step (yes) after amendment of the claims"

Decisions cited:
-

Catchword:
-

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Case Number: T 0170/92 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 10 October 1994

Appellant: ELF ATOCHEM S.A.
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Decision under appeal: **Decision of the Examining Division of the European Patent Office dated 2 October 1991 refusing European patent application No. 87 401 987.0 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: P. A. M. Lançon
Members: M. M. Eberhard
C. Holtz

Summary of Facts and Submissions

I. European patent application No. 87 401 987.0 (publication No. 0 263 005) was refused by a decision of the Examining Division. The decision was based upon the amended set of claims filed on 4 February 1991, with the further amendments requested in the Applicant's letter of 17 July 1991.

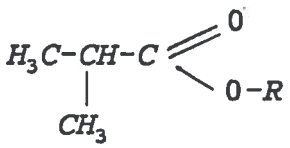
II. The ground for the refusal was that the process of the amended Claim 1 did not involve an inventive step in the light of the disclosure in document US-A-3 716 545 (hereinafter D1). According to the decision it was quite clear from the documents FR-A-2 497 795 (D4) and GB-A-2 097 690 (D5) that, for the oxidative dehydrogenation of isobutyric acid, the presence of one or more metals M as defined in claim 1 was optional. Thus, the catalysts to be regenerated in the claimed process were of the same type as those of D1. The Examining Division took the view that the claimed process was merely the application of the known regeneration process according to D1 to a similar situation since D1 also related to oxidative dehydrogenation reactions under comparable reaction conditions albeit with different feeds.

III. The Appellant lodged an appeal against this decision. In the Statement of Grounds of Appeal, the Appellant argued that D5 represented the closest prior art since it concerned the regeneration of a catalyst which was identical to that of the present invention and which was used in the same catalytic reaction. However, the regeneration process known from D5 was based on a very different theory.

It was further contended that D1 related to the oxidative conversion of olefins and/or diolefins including the oxidative dehydrogenation in the presence of an iron-phosphorus-oxygen catalyst, i.e. a catalyst which did not contain any metal M, and that the oxidative dehydrogenation of isobutyric acid to methacrylic acid was not mentioned in D1. It was pointed out that the latter reaction was very different from the oxidation of methacrolein to methacrylic acid cited in D1. Furthermore the sole example illustrating the regeneration of the catalyst by addition of a phosphorus-containing compound (Example XI) was directed to the oxidative dehydrogenation of 2-methyl-2-butene to isoprene and the catalyst used in this example did not contain any metal M. Therefore, the process according to the invention could not be considered as being merely the application of a known regeneration process to a similar situation.

IV. In reply to a communication of the Board pursuant to Article 110(2) EPC, the Appellant filed an amended set of claims on 26 September 1994. Claim 1 thereof reads as follows:

"A process for the oxidative dehydrogenation of a compound having the formula:



wherein R is selected from the group consisting of the hydrogen atom and linear or branched saturated aliphatic groups having from 1 to 8 carbon atoms,

in the presence of at least one modified iron phosphate catalyst having the gram-atom empirical formula $FeP_xM_yO_z$ wherein:

- M is at least one metal selected from the group consisting of lithium, sodium, rubidium, cesium, magnesium, calcium, strontium, baryum, cobalt, lanthane, tellurium and silver,
- x is not below 0.2 but not above 2,
- y is not below 0.01 but not above 2, and
- z is the amount of oxygen linked to the other elements and corresponding to their oxidation stage,

characterized in that the said catalyst is regenerated, intermittently or continuously, by the addition thereto of an effective amount of at least one phosphorous-containing compound."

The dependent Claims 2 to 8 relate to preferred embodiments of the process according to Claim 1.

V. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 8 submitted on 26 September 1994.

Reasons for the Decision

1. The appeal is admissible.

2. There are no formal objections under Article 123(2) EPC to the amended set of claims received on 26 September 1994. The subject-matter of claim 1 is supported by the process Claims 8, 9 and 1 as originally filed. The dependent Claims 2 to 8 find a support in the original Claims 10 and 2 to 7.

3. The amended Claim 1 states the starting organic compounds and the kind of catalytic reaction in which the catalyst was involved and deactivated. In these circumstances the expression "an effective amount of ..." used in Claim 1 to define the amount of the phosphorus containing compound(s) which is necessary to regenerate the deactivated catalyst is considered to meet the requirements of clarity set out in Article 84 EPC.

4. The process according to Claim 1 is novel with respect to the cited documents since none of them discloses a catalytic process as defined in Claim 1, which involves a regeneration of the catalyst by the addition of at least one phosphorus containing compound.

5. D5, which is analysed at page 1 of the application as originally filed, is considered by the Board to represent the closest prior art.

This document relates to the regeneration of a catalyst comprising iron, phosphorus, oxygen and possibly one or more other elements such as the alkali or alkaline earth metals, which is preferably used in the oxidative dehydrogenation of isobutyric acid to form methacrylic acid. The proportion of the elements in the catalyst can be expressed by the formula $FeP_xMe_yO_z$ in which Me is at least one element selected from Li, Na, K, Rb, Cs, Mg, Ca, Sr or Ba, x is a value from 0.2 to 2.0, y is o.o to 2 and z has a value sufficient to compensate the average valence of the elements in the oxidation states which exist in the catalyst (cf. page 1, lines 5 to 7 and 33 to 39). The example of D5 discloses the oxidative dehydrogenation of isobutyric acid into methacrylic acid in the presence of a catalyst having an iron/phosphorus/cesium ratio of 1 : 1.11 : 0.127. Thus, D5 describes a catalytic process according to the

preamble of Claim 1. The process of D5 further includes a regeneration of the deactivated catalyst by subjecting the latter to an extreme oxidizing atmosphere at a temperature of about 350°C or greater for at least 2 hours and then to an reducing atmosphere at about 350°C or greater (cf. page 4, Claim 1).

The regeneration method used in this process has two disadvantages, namely the catalyst can only be partially regenerated and, after regeneration, nearly fresh catalyst activity is attained but the rate of subsequent deactivation is greatly increased (cf. page 1, lines 22 to 30, of the present application).

5.1 Starting from this closest prior art, the technical problem underlying the present application can be seen in providing a process as defined in the preamble of Claim 1, in which complete regeneration of the catalyst can be achieved without increasing the rate of subsequent deactivation. It is proposed to solve this problem by intermittently or continuously regenerating the catalyst by addition thereto of an effective amount of at least one phosphorous containing compound. In view of the results reported in the examples of the present application the Board is satisfied that the technical problem has been plausibly solved by the claimed process.

5.2 As pointed out by the Appellant, the method of regeneration disclosed in D5 is based on a principle which is very different from that of the invention. Furthermore this document does not contain any information which might lead the person skilled in the art in the direction of the claimed solution.

5.3 D1 relates to the oxidative conversion including the oxidative dehydrogenation of organic compounds in the presence of an iron-phosphorus-oxygen catalyst. Contrary to the catalysts used in the process of the present application the catalysts of D1 do not contain a metal M of the group defined in Claim 1.

The iron-phosphorus-oxygen catalysts of D1 can be used in the oxidative conversion of olefins into diolefins and oxygenated compounds such as furans, furfural, acetaldehyde, pyrans, acetic acid, acrylic acid, methacrylic acid, acrolein, methacrolein, crotonaldehyde, crotonic acid, acetone, ethanol or in the oxidative conversion of diolefins into the listed oxygenated compounds. The catalysts are in particular suitable for converting alkenes, alkadienes, cycloalkenes, cycloalkadienes, alkylpyridines and alkylaromatics to less saturated and/or oxygen containing products. They can also be used to convert oxygen containing compounds to compounds having a greater oxygen content for example methacrolein into methacrylic acid (cf. column 2, lines 43 to 63; column 11, Claim 1). The oxidative conversions illustrated in the examples of D1 are the oxidative dehydrogenation of 2-methyl-2-butene to isoprene, the conversion of 1,3-butadiene to furan and acetaldehyde and the conversion of isoprene to furfural, acetic acid and acetaldehyde. Thus, this document does not concern the same catalytic reaction as the present application.

D1 further teaches that the catalysts disclosed therein can be reactivated by the continuous or intermittent addition thereto of phosphorus containing compounds, such as phosphoric acid, phosphorus pentoxide, organophosphorus compounds (cf. column 4, lines 31 to 46). Example XI shows that the iron-phosphorus-oxygen catalysts which were used for the oxidative conversion

of 2-methyl-2-butene primarily to isoprene can be regenerated by the addition of phosphoric acid or P_2O_5.

Taking into account that the catalysts involved in the catalytic reaction of D1 do not contain a metal M and that the oxidative dehydrogenation of isobutyric acid to methacrylic acid is not comparable to the oxidative dehydrogenation of 2-methyl-2-butene to isoprene in particular in view of the substantial differences between the starting products, the person skilled in the art could not expect that the drawbacks of the regeneration method of D5 might be overcome by applying the teaching of D1 to the modified iron phosphate catalysts of D5. Without such an expectation, in the Board's view, the skilled person faced with the problem stated above would not be encouraged to regenerate the modified iron phosphate catalysts of D5 by the addition of a phosphorus containing compound thereto (cf. T 2/83, OJ EPO 1984, 265).

5.4 The remaining documents cited in the Search Report are far more removed from the claimed process than D1 and D5. D4 does not disclose any method for regenerating the modified iron phosphate catalyst and the other documents relate to vanadium-phosphorus-oxygen catalysts used in the production of maleic anhydride by the vapour phase oxidation of n-butane. Therefore, their teaching would be of no assistance to the skilled person faced with the technical problem stated above.

6. It follows from the above that the process of the amended Claim 1 filed on 26 September 1994 meets the requirements of Article 52(1) EPC.

7. The dependent Claims 2 to 8 which relate to preferred embodiments of Claim 1 derive their patentability from that of Claim 1.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Examining Division with the order to grant a patent on the basis of Claims 1 to 8 filed on 26 September 1994 with a description to be adapted.

The Registrar: The Chairman:

P. Martorana P. A. M. Lançon