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D E C I S I O N
of 20 December 1999

Case Number: T 1062/96 - 3.3.1

Application Number: 89122983.3

Publication Number: 0379691

IPC: C07C 57/04

Language of the proceedings: EN

Title of invention:

Process for preparing unsaturated carboxylic acid or ester thereof

Patentee:

MITSUBISHI GAS CHEMICAL COMPANY, INC.

Opponent:

BASF Aktiengesellschaft

Headword:

Methacrylates/MITSUBISHI

Relevant legal provisions:

EPC Art. 56

Keyword:

"Main request - submission not allowed"

"Auxiliary request - inventive step (yes) - claimed method not obvious"

Decisions cited:

-

Catchword:

-



Case Number: T 1062/96 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 20 December 1999

Appellant:
(Proprietor of the patent)

MITSUBISHI GAS CHEMICAL COMPANY, INC.
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Representative:

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Respondent:
(Opponent)

BASF Aktiengesellschaft
-Patentabteilung - C6-
Carl-Bosch-Strasse 38
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Representative:

Decision under appeal:

Decision of the Opposition Division of the
European Patent Office posted 25 October 1996
revoking European patent No. 0 379 691 pursuant
to Article 102(1) EPC.

Composition of the Board:

Chairman: A. J. Nuss
Members: P. P. Bracke
R. E. Teschemacher

Summary of Facts and Submissions

- I. The appeal lies from the Opposition Division's decision to revoke European patent No. 0 379 691 due to lack of inventive step.

The set of claims underlying the decision consisted of 6 claims, with the only independent claim reading:

"1. A process for preparing an α,β -unsaturated carboxylic acid, an α,β -unsaturated carboxylic ester or a mixture thereof from at least one ester selected from the group consisting of α -hydroxycarboxylic acid ester, α -alkoxycarboxylic acid ester, and β -alkoxycarboxylic acid ester by a vapour-phase catalytic reaction, characterized in that the reaction temperature is 200-350°C, and the catalyst is X-type zeolite or Y-type zeolite wherein the use of X-type zeolite is excluded in case of preparing acrylic acid or acrylic acid ester."

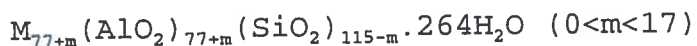
The Opposition Division was of the opinion that the dehydration of oxy-isobutyric acid esters using molecular sieve 13X at 200-350°C was inventive in view of the comparative data filed with letters dated 20 November 1992 and 26 April 1993, said data showing an unexpected effect for said catalyst over the activated alumina catalyst used in document (7), GB-A-584 607, in the dehydration of α -hydroxy-, α -alkoxy- and β -alkoxyisobutyric acids.

However, the Opposition Division did not accept an inventive step for Claim 1, because the unexpected effect had not been shown for all the catalysts and all the starting materials embraced by the scope of the claimed process.

II. With the statement setting out the grounds of appeal received on 5 March 1997 the Appellant (Proprietor) filed a set of claims as a main request and eleven sets of claims as auxiliary requests 1 to 11. He also provided arguments in support of the superiority of the process according to the main request over the prior art processes.

The two independent claims according to the main request read:

"1. A process for preparing methacrylic acid, methacrylic acid ester or a mixture thereof from at least one ester selected from the group consisting of α -hydroxyisobutyric acid ester, α -alkoxyisobutyric acid ester and β -alkoxyisobutyric acid ester by a vapor-phase catalytic reaction, characterized in that the catalyst is a X-type zeolite having the following structure



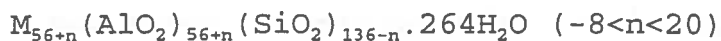
wherein M means alkali metal, or

a Y-type zeolite having the following structure,



wherein M means alkali metal."

"2. A process for preparing acrylic acid, acrylic acid ester or a mixture thereof from at least one ester selected from the group consisting of lactic acid ester, α -alkoxypropionic acid ester and β -alkoxypropionic acid ester by a vapor-phase catalytic reaction, characterized in that the catalyst is a Y-type zeolite having the following structure,



wherein M means alkali metal."

III. The former Opponent stated in a letter dated 10 February 1998 that it did not have any objection to the maintenance of a patent with a scope as defined in the claims according to the main request filed with the statement setting out the grounds of appeal.

By a letter dated 18 November 1999 it withdrew its opposition.

III. At the oral proceedings before the Board, which took place on 20 December 1999, the Appellant filed as a main request a set of 6 claims and as an auxiliary request a set of 4 claims. Those sets of claims corresponded to the sets of claims filed by telefax on 16 December 1999, which were amended at the oral proceedings before the Board.

The only independent claim of the main request read:

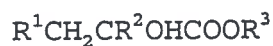
"1. A process for preparing an α, β -unsaturated carboxylic acid of the general formula



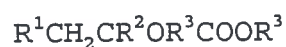
wherein R^1 and R^2 each represent a hydrogen or a C_1 - C_3 alkyl group, an α, β -unsaturated carboxylic ester of the general formula



wherein R¹ and R² are each defined as above and R³ represents a C₁-C₅ alkyl group, or a mixture thereof from at least one ester selected from the group consisting of α-hydroxycarboxylic acid ester of the general formula



wherein R¹ and R² are defined as above, α-alkoxycarboxylic acid ester of the general formula



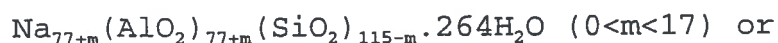
wherein R¹, R² and R³ are each as defined above, and β-alkoxycarboxylic acid ester of the general formula



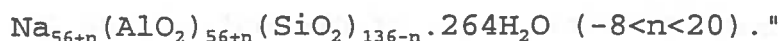
wherein R¹, R² and R³ are each as defined above, by a vapour-phase catalytic reaction, characterized in that the reaction temperature is 200-350°C, and the catalyst is X-type zeolite or Y-type zeolite."

The only independent claim of the auxiliary request read:

"1. A process for preparing methacrylic acid, methacrylic acid methylester or a mixture thereof from at least one ester selected from the group consisting of methyl α-hydroxyisobutyrate, methyl α-methoxyisobutyrate and methyl β-methoxyisobutyrate by a vapor-phase catalytic reaction, characterized in that the catalyst is a X-type zeolite having the following structure



a Y-type zeolite having the following structure



The Appellant essentially argued that Claim 1 according to the main request concerned an alternative process for preparing α, β -unsaturated carboxylic acids or esters thereof and that such process was not made obvious by the cited prior art.

As far as the auxiliary request was concerned, the Appellant confirmed the Opposition Division's opinion that, with the data of the comparative tests provided during the examination and opposition procedure, an unexpected effect had been shown for the whole scope claimed.

- IV. The Appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of the main request or the auxiliary request as submitted during the oral proceedings.

Reasons for the Decision

1. Although the statement of grounds of appeal was not filed within the time limit pursuant to Article 108, third sentence, EPC, the rights of the Appellant were re-established by the Board's decision of 11 December 1997.
2. *Main request*

Although in the written statement setting out the grounds of appeal the Appellant requested, as a main request, the maintenance of the patent with the claims as specified in point II above, he requested at the

oral proceedings before the Board of Appeal that the patent be maintained on the basis of a set of claims having a much broader scope of protection, as presented in point III above.

The introduction of this request into the proceedings would raise questions which neither formed the basis of the present appeal nor were a consequence of previous discussions. In particular, the Appellant submitted for the first time that the problem to be solved was to provide an alternative process for preparing an α,β -unsaturated carboxylic acid whereas previously he had taken the position that the presence of an inventive step should be accepted, taking into account an unexpected effect of the claimed process. Such substantive amendment could and should have been submitted early in the appeal proceedings. Since the amended claims are not clearly allowable and no justification for the late filing is apparent, in accordance with the well-established case-law of the Boards of Appeal, their submission is not allowed in the present case (see, for example, the decisions cited in EPO Board of Appeal Case Law in 1998, Special edition of the OJ EPO 1999, VI. D.10).

3. *Auxiliary request*

3.1 Amendments

Claim 1 is supported by Claim 1 as originally filed and by

- page 7, lines 11 and 12, of the application as filed, mentioning methyl α -hydroxyisobutyrate, methyl α -methoxyisobutyrate and methyl β -methoxyisobutyrate as starting compounds;

- the last paragraph of page 4 of the application as filed, describing a X-type or Y-type zeolite as catalyst; and
- page 7, lines 15 and 16, of the application as filed, mentioning methacrylic acid and methyl methacrylate as α, β -unsaturated carboxylic acid ester.

The additional features in Claims 2, 3 and 4 are supported by Claims 6, 7 and 8 as originally filed.

Consequently, the subject-matter of all Claims 1 to 4 meets the requirement of Article 123(2) EPC.

3.2 Novelty

Since the claimed process differs from the processes known from the prior art at least by the use of the specific X-type or Y-type zeolite, as defined in present Claim 1, the Board comes to the conclusion that Claim 1 and, consequently, also Claims 2 to 4 are novel over the cited prior art. Since the novelty of the claimed process has never been contested, it is not necessary to give detailed reasons for this finding.

3.3 Inventive step

- 3.3.1 The Board considers document (7), which is discussed on page 2, lines 39 to 41, of the patent in suit, to represent the closest state of the art, which was not contested.

- 3.3.2 Document (7) concerns a method of converting α -alkoxy-isobutyric acid into the corresponding methacrylate by treatment with a dehydrating agent (page 1, lines 39 to 46). Example 1 describes the conversion of methyl α -methoxy isobutyrate into methyl methacrylate in almost quantitative yield by passing the vaporised isobutyrate over activated alumina at about 300°C.
- 3.3.3 Starting from the disclosure of document (7), the problem underlying the invention must be seen as the provision of an industrial process for the preparation of acrylic acid or methyl methacrylate in more moderate conditions and with a high yield (see the patent in suit, page 2, lines 45 to 47, and page 4, line 36 to 38).
- 3.3.4 The patent in suit claims to solve this problem by the process defined in Claim 1, more particularly, by using in the vapour-phase catalytic reaction a catalyst as defined in Claim 1.
- 3.3.5 From the data presented in examples 1 and 3 of the patent in suit and example A, provided with letter of 26 April 1993 (see the table submitted on 16 December 1999), it follows that methanolic solutions of methyl- α -hydroxyisobutyrate, methyl β -methoxyisobutyrate and methyl α -methoxyisobutyrate are each converted in a vapour-phase catalytic reaction at 240°C in the presence of molecular sieve 13X in methyl methacrylate for 99% (α -hydroxy- and β -methoxyisobutyrate) or 98% (α -methoxyisobutyrate) with a selectivity for methylmethacrylate of 93%, 95% and 96% respectively and from the data presented in example 2 of the patent in suit it follows that aqueous solutions of methyl- α -hydroxyisobutyrate are converted under the same circumstances for 99% with a selectivity for methacrylic acid of 92%.

From the comparative example 1, filed with a letter dated 20 November 1992, and comparative examples A and B, provided with a letter dated 26 April 1993 (see table submitted on 16 December 1999), it follows that such conversions do not take place at 240°C when γ -alumina is used as catalyst and that the conversion of methyl- α -hydroxyisobutyrate in methyl methacrylate at 290°C is only 64% with a selectivity of 34% for methyl methacrylate, whereas such conversions of methyl α - or β -methoxyisobutyrate at 320°C with γ -alumina is only 81% or 72% with a selectivity for methyl methacrylate of 94% and 89%.

The Board therefore accepts that a credible case has been put forward that the problem underlying the invention, as defined in point 3.3.3, is effectively solved by the claimed method, a point which was not contested by the former Opponent.

- 3.3.6 It remains to be decided, whether, in the light of the teachings of the cited documents, a skilled person seeking to solve the above-mentioned problem, would have arrived at the claimed method in an obvious way.
- 3.3.7 Document (7) is completely silent about the use of a zeolite in converting α -alkoxy-isobutyric acid into methacrylic acid or methyl methacrylate and it has never been alleged that the use of a **X**-type or a **Y**-type zeolite in the preparation of methacrylic acid or methyl methacrylate has been suggested in any of the other cited documents.

Therefore, the Board comes to the conclusion that Claim 1 is not obvious in the light of the teachings of the cited prior art.

3.3.8 Claims 2 to 4, which represent preferred embodiments of Claim 1, derive their patentability from the same inventive concept.

Order

For these reasons it is decided that:

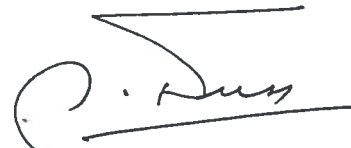
1. The decision under appeal is set aside.
2. The case is remitted to the first instance with the order to maintain the patent on the basis of Claims 1 to 4, submitted as auxiliary request during the oral proceedings, and a description yet to be adapted.

The Registrar:



E. Görgmaier

The Chairman:



A. Nuss