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D E C I S I O N
of 14 June 1995

Case Number: T 0688/93 - 3.3.1

Application Number: 88202870.7

Publication Number: 0321054

IPC: C07C 69/24

Language of the proceedings: EN

Title of invention:
Process for the production of propionate esters

Applicant:
SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V.

Opponent:
-

Headword:
Propionate esters/SHELL

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (yes) - non-obvious combination of process steps"

Decisions cited:
-

Catchword:
-

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Case Number: T 0688/93 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 14 June 1995

Appellant: SHELL INTERNATIONALE RESEARCH
MAATSCHAPPIJ B.V.
Carel van Bylandtlaan 30
NL-2596 HR Den Haag (NL)

Representative: -

Decision under appeal: **Decision of the Examining Division of the European Patent Office dated 8 March 1993 refusing European patent application No. 88 202 870.7 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: A. J. Nuss
Members: P. P. Bracke
W. Moser

Summary of Facts and Submissions

I. European patent application No. 88 202 870.7, filed on 13 December 1988 (publication No. 0 321 054), was refused by a decision of the examining division dated 8 March 1993.

II. The decision was based on the application as filed, containing ten process claims with the only independent claim reading:

"1. A process for the production of propionate esters which process comprises the simultaneous production of methyl propionate and the propionate ester of an alcohol other than methanol by means of the following steps:

step 1: carbonylating ethylene with carbon monoxide in the presence of methanol and a carbonylation catalyst with formation of methyl propionate;

step 2: isolating methyl propionate from the reaction mixture obtained in step 1;

step 3: transesterifying methyl propionate by reaction with the alcohol other than methanol in the presence of a transesterification catalyst;

step 4: isolating the propionate ester of said alcohol other than methanol and an azeotrope comprising methanol and methyl propionate by means of distillation from the reaction mixture obtained in step 3; and

step 5: recirculating the azeotrope isolated in step 4 to the carbonylation in step 1."

III. The ground of refusal was that the subject-matter of Claim 1 lacked inventive step (Article 56 EPC) in view of the contents of documents

(1) EP-A-0 235 864 and
(2) US-A-3 098 093.

In the Examining Division's view document (1), describing the preparation of methyl propionate by carbonylating ethene in the presence of, for example, methanol, was the closest state of the art.

The Examining Division came to the conclusion that, starting from the content of document (1), the claimed process was obviously derivable from the state of the art, because

- it was general knowledge that methyl propionate may be converted into another propionic acid ester by a transesterification reaction;

- the recirculation of the azeotrope isolated from the reaction equilibrium of a transesterification reaction

  reactant ester + reactant alcohol $\rightleftharpoons$ product alcohol + product ester

was proposed in document (2);

- a skilled person would expect that the recirculation of the azeotrope isolated from the transesterification step would, in the present case, negatively influence the transesterification equilibrium; and

- consequently, it was found obvious to recirculate such azeotrope not into the transesterification mixture but into the carbonylation mixture.

IV. An appeal was lodged against this decision.

V. The Appellant (Applicant) agreed that steps 1 and 2 of the claimed process were known from document (1). However, he contested that it was obvious to combine the teachings of documents (1) and (2). More particularly, he asserted that a skilled person would not consider document (2), since in the claimed process

- the azeotrope being recycled is formed by the reactant ester and the product alcohol, whereas in the process according to document (2) the azeotrope being recycled is formed by the product ester and the reactant alcohol, and

- the azeotrope is recirculated to the carbonylation step, whereas in the process described in document (2) the azeotrope is recycled to the transesterification reaction from which it was obtained.

Finally, he stated that interpreting and modifying the teaching of document (2), as done by the examining division, to construe a process as in Claim 1 was a reasoning based on hindsight.

VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted with the published Claims 1 to 10.

Reasons for the Decision

1. The appeal is admissible.

2. *Novelty*

After examination of the cited prior art, the board has reached the conclusion that the claimed process is not described in any of those documents. Since the Examining Division acknowledged the novelty of the claimed process, it is not necessary to give detailed reasons for this finding.

3. *Inventive step*

3.1 The Board considers document (1) to be the closest state of the art. This has also been accepted by the Examining Division and by the Appellant.

3.2 Document (1), which is illustrative of the state of the art mentioned on page 2, lines 3 to 7 of the application in suit, is concerned with a process for carbonylating an olefinically unsaturated compound in the presence of a specific homogeneous catalytic system (page 2, lines 13 to 29). More particularly, in this document a process of preparing methyl propionate, starting from ethylene, carbon monoxide and methanol is described (page 3, lines 18 and 19, and Examples 1 to 4).

3.3 According to the application in suit, by using the prior art processes, such as those described in document (1), the simultaneous production of methyl propionate and a propionate ester of an alcohol other than methanol may

only take place by carrying out two such carbonylation reactions, one starting from methanol and the other starting from the alcohol other than methanol (page 2, lines 7 to 9).

In view of this, the problem underlying the invention must be seen in the development of a process enabling the production of methyl propionate and a propionate ester of an alcohol other than methanol in a simpler way than by the total of two such carbonylation processes (page 2, lines 10 to 12).

According to the application in suit, this problem is solved by the claimed process of simultaneously producing methyl propionate and a propionate ester of an alcohol other than methanol, more particularly, by isolating the methanol-methyl propionate azeotrope in step 4 and recirculating the said azeotrope in the carbonylation step 1, irrespective of which alcohol other than methanol is used in step 3, thus effecting the separation of the azeotrope into its components, the methanol being used for the production of further quantities of methyl propionate in step 1 and the separated methyl propionate finally becoming available in step 2 (page 2, lines 26 to 29, and page 3, lines 11 to 15).

The simplicity of the claimed process has been made credible in the example describing the simultaneous preparation of methyl propionate and 2-butyl propionate, both of more than 99.9 % purity (page 4, lines 48 and 49; page 5, lines 33 to 36; and the output of lines 29 and 63 in the material balances on page 5), and the positive effect of the recirculation of the azeotrope obtained in step 4 in the carbonylation step has been made credible by a comparison between the material balances provided in the example and in the comparative

experiment, more particularly, by a comparison between the amounts of methyl propionate obtained via line 29 with and without recirculating the azeotrope obtained in step 4.

3.4 It remains to be decided whether, in the light of the teachings of documents (1) and (2), a skilled person would have chosen the process features according to Claim 1 with a view to simultaneously produce methyl propionate and a propionate ester of an alcohol other than methanol in high purity.

3.5 The Examining Division was of the opinion that a skilled person would have done so (see III. above).

3.6 Document (1) is concerned with a process for the carbonylation of an olefinically unsaturated compound with carbon monoxide in the presence of, for example, an alcohol and a specific homogeneous catalyst (page 2, lines 13 to 29), more particularly, with the preparation of methyl propionate starting from ethylene, carbon monoxide and methanol (page 3, lines 18 and 19, and Examples 1 to 4), which corresponds with steps 1 and 2 of the claimed process.

Although it is said in document (1), page 3, lines 20 to 28, that the products formed may be further reacted if desired, it is only taught there that esters obtained according to the carbonylation reaction may be further hydrolysed, without mentioning the possibility of converting such esters into other esters by a transesterification reaction, let alone to partially convert methyl propionate into a propionate ester of an alcohol other than methanol.

Therefore, a skilled person, looking for a process of simultanedusly preparing methyl propionate and a propionate ester of an alcohol other than methanol, both in relatively pure form, could not find any suggestion in document (1) leading to the claimed process.

3.7 Document (2) describes a continuous ester interchange process, resulting in nearly complete conversion of the alcohol to product ester (column 1, lines 35 to 38). This document furthermore indicates that the equilibrium mixture obtained in the interchange process is difficult to separate since the reactant alcohol and the product ester form an azeotrope and the separation is thereby complicated (column 1, lines 29 to 34) and, in order to obviate these drawbacks, it proposes recirculating said azeotrope to the transesterification reaction (column 5, lines 29 to 32).

Document (2) is therefore only concerned with a method of influencing the reaction equilibrium obtained during a transesterification reaction in such a way that the reactant alcohol is nearly completely converted to product ester (column 1, lines 35 to 38) and, in the meantime, recirculating the azeotrope to the reaction vessel, thus avoiding the complicated separation of the components of the azeotrope. Hence, this document in no way proposes to re-use the azeotrope in a reaction other than a transesterification reaction, let alone in a carbonylation reaction as in step 1 of the claimed process.

A skilled person, looking for a process for simultaneously preparing methyl propionate and a propionate ester of an alcohol other than methanol, both in relatively pure form, would therefore not have a pointer in document (2) for recirculating an azeotrope to a carbonylation step for preparing methyl propionate.

3.8 Since the possibility of transesterifying methyl propionate is not mentioned in document (1) and the recirculation of the azeotrope to a reaction other than a transesterification reaction is not mentioned in document (2), the claimed process is neither obviously derivable from document (1) or (2) nor from a combination of both documents.

3.9 The Board therefore concludes that the process claimed in Claim 1 is not obvious in the light of the cited state of the art.

The process Claims 2 to 10, which depend on Claim 1 and represent preferred embodiments thereof, are, consequently, also not obvious in the light of the cited state of the art.

4. Since these claims also comply with the other requirements of the EPC, a European patent may be granted on the basis of Claims 1 to 10 as originally filed.

Order

For these reasons it is decided that:

1. The examining division's decision 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 10 as originally filed.

The Registrar: The Chairman:

E. Görgmaier A. Nuss