BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

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File No.: T 0558/92 - 3.3.1

Application No.: 85 308 635.3

Publication No.: 0 183 545

Classification: C07C 29/16

Title of invention: Hydroformylation of olefins

D E C I S I O N
of 13 September 1993

Applicant:

Proprietor of the patent: EXXON RESEARCH AND ENGINEERING COMPANY

Opponent: HOECHST AKTIENGESELLSCHAFT

Headword: Hydroformylation/EXXON

EPC: Art. 56

Keyword: "Inventive step (denied) allowable combination of two documents"

Headnote
Catchwords

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0558/92 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 13 September 1993

Appellant:
(Proprietor of the patent) EXXON RESEARCH AND ENGINEERING COMPANY
P.O. Box 390
180 Park Avenue
New Jersey 07932 (US)

Representative: Bawden, Peter Charles
Exxon Chemical Ltd
P.O Box 1
Abindon, Oxforshire OX13 6BB (GB)

Respondent:
(Opponent) HOECHST AKTIENGESELLSCHAFT
Werk Ruhrchemie
Postfach 13 01 60
D-46128 Oberhausen (DE)

Representative:

Decision under appeal: **Decision of the Opposition Division of the European Patent Office dated 11 March 1992, with written reason posted on 21 April 1992, revoking European patent No. 0 183 545 pursuant to Article 102(1) EPC.**

Composition of the Board:

Chairman: K.J.A. Jahn
Members: R.W. Andrews
J.A. Stephens-Ofner

Summary of Facts and Submissions

I. European patent No. 0 183 545, in respect of European patent application No. 85 308 635.3, which was filed on 27 November 1985, was granted on 27 December 1989(cf. 89/52) on the basis of ten claims. Independent Claim 1, 9 and 10 read as follows:

"1. A process for producing a higher alcohol from an olefinic feedstock which comprises hydroformylating the feedstock with synthesis gas in the presence of a hydroformylation catalyst to form a product mixture containing higher aldehyde, alcohol, unreacted feed and secondary products; removing catalyst therefrom; hydrogenating the substantially catalyst free mixture to convert higher aldehyde to the desired higher alcohol; distilling the higher alcohol-containing product mixture to separate (i) a lower boiling Light Oxo Fraction (LOF) and (ii) desired higher alcohol from (iii) a higher boiling Heavy Oxo Fraction (HOF); subjecting the HOF to catalytic steam cracking at a temperature of from 260°C to 380°C using as catalyst an active metal oxide or pseudo-metal oxide, to form HOF residue and a cracked HOF mixture comprising a major proportion of higher alcohol and higher aldehyde, and a minor proportion of olefin and saturated hydrocarbon; and recycling the cracked HOF mixture to the hydroformylation or hydrogenation stage of the process.

9. A process for producing a higher alcohol from a C6 to C12 olefinic feedstock which comprises hydroformylating the feedstock with

synthesis gas at a temperature of from 125-175°C and a pressure of from 150-300 atm in the presence of a hydroformylation catalyst to form a product mixture containing higher aldehyde, alcohol, unreacted feed and secondary product; removing catalyst therefrom; hydrogenating the substantially catalyst free mixture to convert higher aldehyde to the desired higher alcohol having a carbon number which is one carbon atom greater than the olefinic feedstock; distilling the higher alcohol-containing product mixture to separate (i) a lower boiling Light Oxo Fraction (LOF) and (ii) desired higher alcohol from (iii) a higher boiling Heavy Oxo Fraction (HOF); subjecting the HOF to catalytic steam cracking at a temperature of from 260 to 380°C and a pressure of from 1-10 atm. abs. employing steam and HOF in a weight ratio of from 0.1:1-2:1 and using alumina as catalyst, to form HOF residue comprising dimeric and heavier components based on said higher alcohol carbon number and no more than 10 wt% of monomeric components, and a cracked HOF mixture comprising monomeric components based on said higher alcohol carbon number and no more than 20 wt% of dimeric and heavier components and containing a major proportion of higher alcohol and higher aldehyde, and a minor proportion of olefin and saturated hydrocarbon; and recycling the cracked HOF mixture to the hydrogenation stage of the process.

10. HOF residue comprising a mixture of 0-10 wt% alcohol/aldehyde. 45-75 wt% ethers, 20-35 wt%

ether-alcohols, 1-6 wt% acetals and 0-7 wt% esters, which mixture contains dimeric, trimeric and heavier compounds, the dimeric compounds being of carbon number C_{14}-C_{26} and the trimeric compounds of carbon number C_{21}-C_{39} and which contains no more than 10 wt% of monomeric compounds."

II. A notice of opposition, which was filed on the 17 September 1990, requested the revocation of the patent on the grounds of that its subject-matter did not involve an inventive step. The opposition was supported, *inter alia*, by the following documents:

(3) DE-A-2 443 995 equivalent to (3c) GB-A-1 411 073 and

(4) US-A-2 595 096.

In a letter filed on 21 October 1991, the Opponent also contended that the claimed subject-matter lacked novelty having regard to the disclosure of document (4).

III. By a decision delivered orally on 11 March 1992, with written reasons being issued on 21 April 1992, the Opposition Division revoked the patent. The Opposition Division held that the subject-matter of the disputed patent was novel. With respect to inventive step, the Opposition Division found that the proposed solution to the problem of selecting the conditions for the hydrolysis of the Heavy Oxo fraction (HOF) in order to increase the overall yield of desired alcohol over that obtained in document (4) was obvious in the light of the disclosure of this document combined with that of document (3).

IV. An appeal was lodged against this decision of 15 June 1992, with payment of the prescribed fee. In his

statement of grounds of appeal filed on 11 August 1992, the Appellant contended that three key statements in the decision under appeal were unfounded. Thus, the Opposition Division concluded that document (4) disclosed not only the hydrolysis of acetals but also other by-products in the HOF which are hydrolysable under the conditions used. Furthermore, since the cracking of the bottoms did not appear to be so dependent on the fact that said bottoms, had previously been hydrogenated, it was possible to combine the teaching of documents (3) and (4).

The Appellant argued that his experimental data demonstrated that the conditions used in documents (4) only result in the hydrolysis of the acetals and not the entire bottoms.

The Appellant also maintained that there was no reason for the skilled person to combine the teachings of documents (3) and (4) since the former was concerned with the production of aldehydes and the latter with alcohols. In any case, the combination did not bring the skilled reader to the present invention. Finally, the Appellant argued that neither document recognised the presence of ether aldehydes in the heavy fraction, let alone suggested that by applying the claimed cracking conditions to a process for the production of alcohols employing a hydrogenation step to convert aldehydes to alcohol, the uncrackable ether aldehydes present in the heavy fraction would be converted to ether alcohol which can be cracked under the conditions employed.

V. The Respondent argued that the disclosure of document (4) was not restricted to the hydrolysis of acetals with water or dilute acids at 350°F. With respect to document (3), the Respondent contended that the analysis of the crude oxo product of Example 2

indicated that the mixture hydrolysed in this example corresponds to that hydrolysed according to the process claimed in the disputed patent.

The Respondent maintained that the teachings of documents (3) and (4) may be combined since both documents are concerned with high boiling residues of the same origin and with the same object of cracking, the high molecular weight compounds to monomeric ones, which are economically useful.

According to the Respondent the Appellant's experiments are not comparable since they were carried out under completely different temperature conditions and, in any case, document (4) does not specify an upper temperature limit.

VI. The Appellant requests that the decision under appeal be set aside and the patent be maintained in the form as granted.

The Respondent requests that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible

2. The disputed patent relates to a hydroformylation process for the production of higher alcohols from the corresponding olefins in which the heavy oxo fraction remaining after the removal of a light oxo fraction and the desired alcohol is treated to improve the overall alcohol yield of the process.

Document (4), which represents the closest state of the art, describes such a process in which the said heavy

oxo fraction is treated with dilute mineral acid or steam or by other catalytic means and, after separation, the hydrolysed material is advantageously recycled to the hydrogenation stage of the process (cf. Claim 1 in combination with column 3, lines, 39 to 49). If 50% of the HOF can be converted to alcohols, the overall yield may be increased by about 10% (cf. column 3, lines 49 to 52).

Although the prior art process was satisfactorily in improving the overall alcohol yield of the process in which the distillation residues, after the removal of the higher alcohol, contain a relatively large proportion of acetals, there was still a need to provide a method for improving the overall alcohol yield even in cases where the acetal content of the said distillation residues (HOF) is relatively small (cf. page 2, lines 41 to 48 of the disputed patent).

Therefore, in the light of this closest prior art the technical problem underlying the patent in suit is to provide a method of improving the overall yield of higher alcohols of an oxo process which is less dependent on the acetal content of the heavy oxo fraction.

According to the disputed patent this technical problem is solved by subjecting the heavy oxo fraction to catalytic steam cracking at a temperature of 260° to 380°C, using as catalyst an active metal oxide or pseudo metal oxide and recycling the resulting cracked heavy oxo fraction to the hydrogenation or hydroformylation stage of the oxo process.

In the light of the results obtained in Examples 1, 2 and Comparative Example 5 of the disputed patent and in the Tests 2, 3 and 4 filed on 11 August 1992 (the

corresponding chromatograms being submitted on 17 July 1993), it is plausible that the above-defined technical problem has been solved.

3. After examination of the cited prior art the Board has reached the conclusion that the claimed subject-matter is novel. Since novelty is no longer in dispute, it is not necessary to give detailed reasons for this finding.

4. It still remains to be decided whether the claimed subject-matter involves an inventive step.

4.1 As previously mentioned, document (4) discloses an oxo process for the production of alcohols in which, after hydrogenation of the crude product mixture and removal of the desired alcohols by distillation, the thus obtained distillation residue is hydrolysed in a neutral medium at a temperature above about 300°F (148°C) and at least a proportion of the products of the hydrolysis is hydrogenated (cf. Claim 1). Preferably, the hydrolysed products are separated from the non-hydrolysed material by vacuum or steam distillation and the separated alcohols and aldehydes recycled to the hydrogenation stage of the process (cf. column 3, lines 45 to 49).

According to column 3, lines 39 to 42, column 5, line 68 to column 6, line 2 and column 6, lines 23 to 27, the hydrolysis may be carried out using dilute mineral acid, steam at about 300°C to about 400°F (149° to 204°C) and catalytic agents such as alumina, silica and metals, or metal oxides of the eighth group of the Periodic Table.

From the disclosure of this document it is clear that the primary aim of this prior art process was to increase the overall alcohol yield by regenerating alcohols and aldehydes from the acetals contained in the distillation residues. In fact, the process is based on

the unexpected discovery that the distillation residues comprise a substantial portion , up to 50% by volume of acetals (cf. column 3, lines 17 to 20). Previously, it had been considered that the distillation residues were a mixture of polymeric material, such as polymerised aldehydes and ketones, high molecular weight ethers and secondary alcohols, and polymerised hydrocarbons (cf. the sentence bridging columns 2 and 3).

Therefore, the skilled person would immediately realise that the conditions used for the treatment of the distillation residues in this prior art process were especially and primarily suitable for the conversion of the acetals to aldehydes and alcohols. Thus, faced with the technical problem underlying the disputed patent, the skilled person would consider changing the conditions to which the heavy oxo fraction (distillation residues) are subjected with the aim of converting not only the acetals present therein to alcohols and aldehydes but also some of the other components of the mixture.

4.2 Document (3c) discloses a process for the production of aldehydes by the oxo synthesis in which the heavy oxo fraction remaining after the removal of the desired aldehydes and the alcohols and formates produced as by-products is hydrolysed with steam in the presence of known hydrolysation catalysts at atmospheric pressure and at a temperature between 250° and 400°C to form a mixture of alcohols, aldehydes and residual heavy products, the alcohols and aldehydes of which are separated and recycled to the output from the hydroformylation reactor (cf. Claim 1). The dehydration of the alchohols and formates can be neglected in the present context. A suitable hydrolysation catalyst is a metallic oxide, preferably in the active state, such as,

for example, silica, alumina and titanium oxide (cf. page 2, lines 57 to 62).

The skilled person is aware that, due to the fact that the hydroformylation catalyst are also hydrogenation catalysts and the presence of hydrogen in the synthesis gas, hydrogenation of the initially produced aldehydes occurs. This is clearly demonstrated by the analysis of the crude oxo product given in Example 2 of document (3c). As soon as both aldehydes and alcohols are present in the reaction mixture other side reactions take place. Clearly any by-products that are susceptible to hydrogenation will, at least to some extent, be hydrogenated. The same side reactions occur irrespective of whether the object of the oxo process is the synthesis of aldehydes or alcohols, since they take place at the hydroformylation stage of the process.

Therefore, the skilled person, faced with the present technical problem , would take the teaching of document (3) into consideration despite the fact that, since the aim of the process described therein is the production of aldehydes, the catalyst free reaction product is not hydrogenated. Thus, in the Board's judgment, the skilled person would have no hesitation in combining the disclosure of document (3) with that of document (4).

According to page 2, lines 53 to 74, catalytic steam cracking (hydrolysis) of the heavy oxo fraction, which, as pointed out above is at least partially hydrogenated in the hydroformylation reactor, covers at least two thirds of this heavy fraction into aldehydes and alcohols.

Therefore, the disclosure of document (3) provides the skilled person with the incentive to replace the mild

hydrolysis conditions used in document (4) to hydrolyse the heavy oxo fraction by the more severe ones described in document (3) in the expectation of solving the technical problem of providing a method of improving the overall yield of alcohols in an oxo process which is less dependent on the acetal content of the heavy oxo fraction.

4.3 Therefore, in the Board's judgement, the proposed solution to the technical problem underlying the patent in suit is obvious. Hence, Claim 1 is not allowable. Since the Appellant's only request is that the decision under appeal be set aside and the patent be reinstated as granted, dependent Claims 2 to 8 and independent Claims 9 and 10 must share the fate of Claim 1.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

E. Görgmaier K.J.A. Jahn