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File Number: T 496/89 - 3.3.1
Application No.: 83 302 995.2
Publication No.: 0 096 987
Title of invention: Hydroformylation process

Classification: C07C 45/50

D E C I S I O N
of 21 February 1991

Proprietor of the patent: Davy McKee (London) Limited
Opponent: BASF AG

Headword: Hydroformylation/DAVY

EPC Art. 56

Keyword: "Inventive step (denied)"

Headnote



Case Number : T 496/89 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 21 February 1991

Appellant :
(Proprietor of the patent)

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(Opponent)

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Decision under appeal :

Decision of Opposition Division of the European
Patent Office of 26 April 1989, posted on
7 June 1989 revoking European patent
No. 0 096 987 pursuant to Article 102(1) EPC.

Composition of the Board :

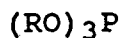
Chairman : K.J.A. Jahn
Members : R.W. Andrews
J.A. Stephens-Ofner

Summary of Facts and Submissions

- I. European patent No. 0 096 987 in respect of European patent application No. 83 302 995.2 which was filed on 24 May 1983, was granted on the basis of ten claims; the only independent claim reading as follows:

"A process for the production of an aldehyde by hydroformylation of an olefin which comprises:

providing a hydroformylation zone containing a charge of a liquid reaction medium having dissolved therein a complex rhodium hydroformylation catalyst comprising rhodium in complex combination with carbon monoxide and with an organic phosphite ligand of the general formula:



in which each R represents an optionally substituted hydrocarbyl radical;

supplying said olefin to the hydroformylation zone;

maintaining temperature and pressure conditions in the hydroformylation zone conducive to hydroformylation of the olefin;

supplying make-up hydrogen and carbon monoxide to the hydroformylation zone;

recovering from the liquid hydroformylation medium a hydroformylation product comprising at least one aldehyde;
and

supplying make-up phosphite ligand to the hydroformylation zone at a rate sufficient to maintain a predetermined

level of free phosphite ligand in the hydroformylation medium."

- II. A notice of opposition was filed on 15 July 1986 in which the revocation of the patent was requested on the grounds set out in Article 100(a) and (b) EPC.
- III. By a decision delivered orally on 26 April 1989, with written reasons posted on 7 June 1989, the Opposition Division revoked the patent. The Opposition Division considered that the process of the disputed patent differed from that disclosed in US-A-3 527 809 (document (1)) in that it operated continuously and that free ligand was added to the reaction zone during hydroformylation. However, since it was known that phosphite ligands are less stable and that hydroformylation processes may be operated on a continuous basis, the Opposition Division held that the claimed process in accordance with the main and two auxiliary requests was obvious.
- IV. A notice of appeal was lodged on 1 August 1989 with payment of the prescribed fee. In his statement of grounds of appeal filed on 17 October 1989 and during the oral proceedings held on 21 February 1991, with respect to his main and first auxiliary requests the Appellant contended that there was no recognition in the prior art that any problem would arise in practice in operating a rhodium catalysed hydroformylation plant continuously using a triorganophosphite as the sole ligand. Thus, for example, document (1) describes the use of a batch reaction technique to successfully hydroformylate a range of alpha-olefins employing a rhodium catalyst with a variety of triorganophorus ligands including triphenyl phosphite. Document (2) (GB-A-1 338 237) claims a continuous rhodium catalysed hydroformylation process in which a

triorganophosphite may act as the ligand, the continuous operation of this process as described in Examples 12 to 14 utilise triphenylphosphine as the ligand.

The Appellant argued that, in the light of this prior art, it was surprising that using a triorganophosphite as the ligand there was a large decrease in catalyst activity upon extended continuous operation and, having regard to the plethora of possible explanations for this loss of catalyst activity, it was not obvious that it could be prevented by the controlled addition of the ligand. Particularly since the addition of triphenylphosphine does not restore catalyst activity loss due to rhodium cluster formation which may occur, under unfavourable circumstances, despite the presence of a plentiful amount of free triphenylphosphine ligand during the hydroformylation utilising triphenylphosphine as the ligand.

The Appellant also maintained that, faced with this previously unrecognised problem, the skilled person may not have even persisted with his efforts to find a solution, but rather would have turned his attention to other ligands. If the skilled person had continued, he would have discovered that the loss of catalyst activity was caused by the disappearance of triorganophosphite ligand from the reaction medium. However, the Appellant argued that the skilled person would consider that the addition of ligand would make matters worse if the disappearing ligand was converted into a catalyst poison or deactivator, such as an acidic substance formed by hydrolysis. Only in the event that the disappearing ligand produced an inert substance would its addition give the desired result. In the Appellant's view the arrival at this insight is inventive, even if its technical application is obvious.

With respect to his second auxiliary request, the Appellant argued that the specified ligand to rhodium molar ratios were considerably lower than those disclosed in the Examples 12 to 14 of document (2) describing continuous operation of this known hydroformylation process.

- V. The Respondent contended that the conversion of the batch process disclosed in document (1) to a continuous process required no inventive activity on behalf of the skilled person. The Respondent also argued that, if a decrease in catalyst activity occurs as a result of chemical or physical changes in the catalyst components in the course of time, it is obvious to replace the used catalyst components by fresh ones. It is clear that if the activity of a multicomponent catalyst system decreases, this may be traced back to all of the components or, for example, to only one of them. If it is found that only one of the components has been noticeably consumed, it is obvious to add only this one.

The Respondent maintained that, although it can be seen from Table A in documents (1) and (2) that triorganophosphites are better ligands than triorganophosphines, because of the known chemical sensitivity of phosphites, phosphines are generally the preferred ligands.

The Respondent did not agree with the Appellant that cluster formation cannot be reversed, but, in any case, the reason for the loss of catalyst activity was irrelevant to the consideration of inventive step.

Finally, with respect to the Appellant's second auxiliary request the Respondent referred to Table VI of document (1).

VI. The Appellant requested that the decision under appeal be set aside and that the patent be maintained in unamended form. Alternatively, the Appellant requested that the patent be maintained on the basis of the amendments submitted in his letter dated 3 February 1987 (1st auxiliary request) or on the basis of the amendments formally resubmitted during oral proceedings (2nd auxiliary request). The claims in accordance with the Appellant's first auxiliary request differs from those of the granted patent only in that in Claim 1 the words "continuous" and "continuously" have been inserted between "A" and "process" in the first line thereof and before the expression "supplying said olefin to the hydroformylation zone" respectively. In accordance with the second auxiliary request, Claim 1 has additionally been amended by specifying that the ligand to rhodium molar ratio in the hydroformylation medium should be from about 3:1 to about 8:1. In addition granted Claim 6 has been deleted with consequential amendments to granted Claims 7 to 10.

The Respondent requested that the appeal be dismissed.

VII. At the conclusion of the oral proceedings, the Board's decision to dismiss the appeal was announced.

Reasons for the Decision

1. The appeal is admissible.
2. The Respondent (Opponent) sought to introduce a fresh document in the course of the oral proceedings. His representative, when questioned by the Board about the reason for such tardiness, freely admitted that the search

for the document, which had been published for over ten years, only began one week before the date set for the oral proceedings. Although the Board has decided not to admit this document into the proceedings on the ground of its irrelevance, it wishes to reiterate that the late introduction of documents and of other matter into the appeal proceedings is inimical and contrary to the public interest, quite apart from being unfair to the other party to the proceedings.

Attempts by either party to spring a surprise on the other by deliberate late-filing, as well as inadvertent omissions to present arguments and the evidence needed to support them, run counter to the spirit and intent of the EPC, as expounded in the note on "Opposition Procedure in the EPO" OJ EPO 1989, 457, and laid down in cases such as T 117/86, OJ EPO 1989, 401, T 182/89 (to be published) and T 326/87 (to be published) that arguments, facts, evidence and requests must be submitted at the earliest possible opportunity, i.e. within the ample nine-month period provided for oppositions (Article 99(1) EPC).

3. There are no formal objections under Article 123 EPC to the claims in accordance with the Appellant's first and second auxiliary requests. In the Board's view, the amendments made by way of clarification to the granted Claim 1 in accordance with the first auxiliary request are totally unnecessary. From a proper construction of the claim, even without reference to the description, it is clear that the claim could only embrace a continuous hydroformylation process. In particular, the reference to supplying make-up hydrogen and carbon monoxide to the hydroformylation zone and the fact that a predetermined level of free phosphite ligand in the hydroformylation zone is maintained by the addition of phosphite ligand at an appropriate rate would only make sense in connection

with a process operating on a continuous basis. In view of this, the Appellant's main and first auxiliary request will not be treated separately in the following.

- 3.1 Support for the ligand to rhodium molar ratios specified in Claim 1 in accordance with the Appellant's second auxiliary request is to be found on page 12, lines 24 to 29 of the published patent application (cf. also page 5, lines 55 and 56 of the printed patent specification).
4. The disputed patent concerns a continuous hydroformylation process for the production of an aldehyde by reacting in a liquid medium an olefin with carbon monoxide and hydrogen in the presence of a complex rhodium hydroformylation catalyst comprising rhodium in complex combination with carbon monoxide and with a triorganophosphite and free phosphite ligand.
 - 4.1 Document (2), which is considered to represent the closest prior art, also discloses a continuous hydroformylation process in the presence of a complex rhodium catalyst wherein the triorganophosphorus ligand has a Δ HNP (a measure of the basicity of the ligand) of at least 425, such as, for example, triaryl or trialkyl phosphites and triarylphosphines (cf. Claims 1, 2 and 3, Table A bridging pages 6 and 7 and page 7, lines 20 to 36).
 - 4.2 However, it was found that, although this prior art process could be successfully operated on a continuous basis with a triarylphosphine as the phosphorus-containing ligand, if a triorganophosphite was utilised as the ligand continuous operation could not be achieved due to the tendency of the catalyst to deactivate moderately rapidly (cf. disputed patent, page 2, lines 44 to 49).

- 4.3 Therefore, in the light of this closest state of the art, the technical problem underlying the patent in suit is to be seen in providing a continuous rhodium catalysed hydroformylation process in which a triorganophosphite is employed as the ligand.
- 4.4 In the Board's judgment, no contribution to inventive step can be seen in the recognition of the above-defined problem. Although, to the Board's knowledge, the Appellant was the first to realise the existence of this problem and it is conceivable that the skilled person, in the light of the prior art, may have been surprised by his failure to operate this known process using, for example, triphenyl phosphite as the ligand, nevertheless the problem would be easily recognised by the skilled person in the course of his routine investigation of this prior art process. Particularly, since the advantages of operating chemical processes on a continuous basis are well-known and any attempt at commercialisation of this prior art process would routinely involve experiments of a reasonably long duration (cf. for example, Example 13 and 14 of document (2) in which the experiments lasted for 720 and 250 hours respectively).
- 4.5 According to the disputed patent (main and first auxiliary request), the above-mentioned technical problem is essentially solved by supplying make-up phosphite ligand to the hydroformylation zone at a rate sufficient to maintain a predetermined level of phosphite ligand in the hydroformylation medium.

In view of the results set forth in Tables I and II of the disputed patent, the Board is satisfied that this technical problem is effectively solved.

5. After examination of the cited documents, the Board has reached the conclusion that the claimed subject-matter is

novel. Since novelty is not in dispute, it is not necessary to give detailed reasons for this finding.

6. It still remains to be examined whether the requirement of inventive step is met by the process claimed in accordance with the main and first auxiliary requests.
- 6.1 According to document (2) an aldehyde may be obtained by the continuous hydroformylation of an olefin in the presence of a catalyst comprising rhodium in complex combination with carbon monoxide and a trialkyl or triaryl phosphite and at least 2 moles of free phosphite ligand per mole of rhodium (cf. Claim 1 in combination with page 7, lines 20 to 24). However, it is clear from the Comparative Example and Examples 2 and 3 of the disputed patent that, in fact, this prior art process cannot be operated on a continuous basis.
- 6.2 The Board cannot accept the Appellant's contention that the skilled person faced with this initial failure would not persist in his efforts to develop a continuous hydroformylation process using a phosphite as the ligand. Particularly since he is aware that the ΔH_{NP} value of, for example, triphenyl phosphite, suggests that this compound would be an extremely good ligand.
- 6.3 It is true that there could be numerous reasons for the loss of catalyst activity, however, in the Board's opinion, after establishing the purity of the feedstock, the skilled person would, as a matter of course, analyse the reaction mixture at various time intervals in an attempt to discover why the reaction rate decreased with time. In the course of this routine investigation the skilled person would find that the free phosphite ligand was disappearing from the reaction mixture.

Since it is well-known in this field that it is essential to carry out the hydroformylation reaction in the presence of free ligand, i.e. ligand that is not complexed with the rhodium atom in the active complex catalyst, the skilled person's first thought would be to add free phosphite to compensate for the disappearance of the free ligand.

- 6.4 In the Board's judgment, the skilled person would adopt this course of action before investigating the reason for the free ligand's disappearance. Therefore, it is irrelevant whether the disappearing ligand reacts to produce a catalyst poison or deactivator or whether it is converted into an inert substance. In the former case, the addition of phosphite would not solve the problem and the skilled person would be forced to continue his search for a solution.

Furthermore, it is immaterial to the decision with respect to inventive step whether or not it is possible to reverse a rhodium cluster formation by the addition of triphenylphosphine. Therefore, the difference of opinion on this point between the Appellant and Respondent can remain unresolved.

Thus, the skilled person's obvious response to the discovery of the loss of phosphite ligand from the hydroformylation medium solves the technical problem underlying the disputed patent. The reason why this measure solves the problem is of no relevance in deciding the question of obviousness.

- 6.5 Therefore, the subject-matter of Claim 1 in accordance with both the Appellant's main and first auxiliary request does not involve an inventive step.

6.6 Dependent Claims 2 to 10 in accordance with both these requests relate to preferred embodiments of the process of their respective main claims. It was not argued that these claims contain any independent features and, therefore, in the absence of such features, they are unallowable in the absence of an acceptable main claim.

6.7 Claim 1 in accordance with the Appellant's second auxiliary request also includes the feature that the ligand to rhodium molar ratio in the hydroformylation medium is from about 3:1 to about 8:1.

However, from document (1) it is known to carry out hydroformylation reactions using triphenyl phosphite as the ligand in which the ligand to rhodium molar ratio falls within the above-mentioned range. Thus, Examples 9 to 11, 13, 17 and 25 disclosed the use of a ligand to rhodium molar ratio of 6.5:1.

With this known molar ratio in mind, it would be a matter of routine experimentation to determine the optimum ligand to rhodium molar ratio to be employed in the present process. Therefore, the subject-matter of Claim 1 in accordance with the Appellant's second auxiliary request does not involve an inventive step.

For the reason given above in paragraph 5.6, dependent Claims 2 to 9 in accordance with this request are also unallowable.

Order

For these reasons, it is decided that:

The appeal is dismissed.

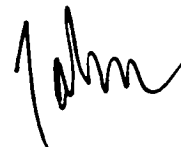
The Registrar:

The Chairman:

E. Görgmaier



K.J.A. Jahn



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