**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 0430/91 - 3.3.1
Application No.:	84 114 576.6
Publication No.:	0 144 936
Classification:	C07C 51/12
Title of invention:	Production of carboxylic acids from alcohols

DECISION
of 15 June 1993

Applicant:	
Proprietor of the patent:	UNION CARBIDE CORPORATION
Opponent:	OI) HOECHST AKTIENGESELLSCHAFT, Frankfurt OII) BP Chemicals Limited

Headword:	Carbonylation/UNION CARBIDE
EPC:	Art. 56
Keyword:	"Inventive step (confirmed)" - "Non-obvious combination of co-catalysts, known *per se*"

**Headnote
Catchwords**

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Case Number: T 0430/91 - 3.3.1

DECISION
of the Technical Board of Appeal 3.3.1
of 15 June 1993

Appellant:
(Opponent)

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Decision under appeal: **Interlocutory decision of the Opposition Division of the European Patent Office of 5 April 1990, posted on 19 April 1991 concerning maintenance of European patent No. 0 144 936 in amended form.**

Composition of the Board:

Chairman: A. Jahn
Members: P. Krasa
J.A. Stephens-Ofner

Summary of Facts and Submissions

I. The mention of the grant of the European patent No. 0 144 936 in respect of European patent application No. 84 114 576.6 filed on 30 November 1984, was published on 24 June 1987 (cf. Bulletin 87/26) on the basis of nine claims.

II. In two notices of opposition which were duly filed by HOECHST AKTIENGESELLSCHAFT (Opponent I and later Appellant) and by BP INTERNATIONAL LIMITED (Opponent II) the revocation of the patent, on the grounds of lack of novelty and of inventive step was requested.

The oppositions were supported by a series of documents of which the following ones finally remained relevant:

(1) US-A-3 769 329
(2) DD-A- 113 521
(4) DE-A-2 450 965
(5) EP-A-0 055 618 and
(8) US-A-4 212 989.

After expiry of the opposition period the Appellant referred also to

(3A) DE-A-3 024 353, corresponding to DE-C-3 024 353, cited in the Appellant's notice of opposition (page 3, last line).

III. The Opposition Division, with its interlocutory decision of 5 April 1990, posted 19 April 1991, maintained the patent in amended form on the basis of six claims which were submitted to it during oral proceedings. The only independent Claim 1 reads

"A process for the production of acetic acid which comprises the catalytic reaction of methanol and carbon monoxide in contact with methyl acetate and a homogeneous catalyst system consisting of rhodium or a rhodium compound and a promoter characterized in that the promoter is a mixture of lithium iodide and methyl iodide in a mole ratio of Rh:LiI from 1:1 to 1:1,000 and a mole ratio of LiI:CH_3I from 1.1,000 to 1,000:1 and further characterized in that methyl acetate, but no water, is added to the reaction mixture."

The Opposition Division held that the claimed subject-matter was new and considered document (4) as the closest prior art. This document disclosed a process for the simultaneous production of acetic anhydride and acetic acid by carbonylation of a mixture of methyl acetate and methanol in the presence of a catalyst system comprising rhodium and methyl iodide. While the presence of minor amounts of water was not excluded, the reaction was carried out without the intentional addition of water.

The technical problem to be solved in respect to citation (4) was defined by the Opposition Division to be the provision of a process with improved efficiency, selectivity and conversion rate. In view of the available experimental evidence it was acknowledged that this problem was solved by the claimed process.

The suggested solution was also held to result from an inventive step as there was no incentive in document (4) to replace the co-catalyst methyl iodide by the particular combination of methyl iodide with lithium iodide. Document (2), which disclosed this co-catalyst combination was held to be irrelevant as relating to a different process, i.e. the manufacture of acetic anhydride.

The argument that the patent in suit lacked inventive step in view of the combined disclosure of documents (1) to (8) was rejected by the Opposition Division, as resulting from an *ex post facto* analysis.

IV. An appeal was lodged against this decision on 8 June 1991 with the payment of the prescribed fee. In his Statement of Grounds of Appeal, filed 20 August 1991, the Appellant disagreed that document (4) represented the closest state of the art, and regarded document (1) to be the more relevant one. While conceding that citation (1) did not disclose the incorporation of lithium iodide into the catalyst system, he argued that such incorporation was obvious in view of documents (2), (3A), (4), (5), and (8).

In particular, so he argued, a comparison of Examples 22 and 23 of document (2) would have shown that the combination of methyl iodide and lithium iodide was much more effective as a co-catalyst in carbonylation reactions than lithium iodide alone (document (2), page 22). Hence, so the Appellant concluded, it would have been obvious for the skilled person, who wished to solve the technical problem of improving the process known from citation (1), to try the incorporation of lithium iodide into the catalyst system disclosed therein. This was all the more true since the beneficial effect of lithium on rhodium catalysts was confirmed by documents (3A), (5) and (8) (document (3A), page 8, last paragraph and page 9; document (5), claim 14; document (8), column 2, lines 54 to 57).

In the Appellant's view, the absence of water could not render the claimed subject-matter inventive since from citation (4) it was known that the reaction proceeds rapidly also in the absence of appreciable amounts of water.

V. The Respondent maintained that there was no hint in the state of the art to use a combination of lithium iodide and methyl iodide in the present reaction and that the Appellant's arguments result from an *ex post facto* analysis.

VI. The Appellant requested that the decision under appeal be set aside and that the European patent No. 0 144 936 be revoked. The Respondent requested that the appeal be dismissed and that the patent be maintained as amended in the course of oral proceedings before the Opposition Division but subject to the further amendment of the last line of Claim 1 by the substitution of "1:1,000" for "1.1,000".

At the end of the oral proceedings, which took place on 15 June 1993, the chairman announced the Board's decision to allow the Respondent's request.

Reasons for the Decision

1. The appeal is admissible.

2. No objections arise against the amended claims under Article 123 EPC and as this is not in dispute, no further comments are required by the Board.

3. The subject-matter of Claim 1 is not disclosed in any citation and, thus, is novel. This not being contested, no further comments are necessary either by the Board.

4. The patent in suit relates to the manufacture of acetic acid by carbonylation of methanol in the presence of added methyl acetate and a homogeneous catalyst system consisting of rhodium or a rhodium compound and a

mixture of lithium iodide and methyl iodide as promoter in particular ratios whereby no water is added to the reaction mixture.

The sole issue in dispute is whether or not the process of the patent in suit results from an inventive step.

5. The Board accepts the Appellant's view that document (1) discloses, *inter alia*, a process for the manufacture of acetic acid, which differs from that of the patent in suit only in that lithium iodide is not used as an additional promoter and takes this citation as the starting point for evaluating inventive step. The reason for this view is that this citation is concerned with the production of acetic acid in high yield and selectivity (the methanol conversion is substantially 100% and the selectivity of acetic acid formation is greater than 95%; column 2, lines 61 to 69 in combination with column 12, lines 21 to 23). In contrast, document (4), which the Opposition division considered as the closest prior art, relates primarily to the production of acetic acid anhydride and yields acetic acid only as a by-product and always in admixture with the anhydride (see the paragraph bridging pages 1 and 2 and page 2, lines 18 to 20 [original numbering]).

6. According to the patent in suit, the invention is based on the finding that the rhodium - lithium iodide - methyl iodide catalyst system leads to a process with an unexpected high selectivity and catalyst efficiency/conversion rate (page 3, lines 25 to 26 and 51 to 55). On page 3, lines 42 to 45 it is explained that "activity or conversion rate" relates to the amounts of reactants the catalyst converts to product per unit of time, generally expressed in g mole/l.h. Thus, the Board sees the technical problem underlying the patent in suit as being the improvement of the

space-time yield of acetic acid and the catalyst selectivity as compared with the process disclosed in document (1).

7. According to the disputed patent, this technical problem is solved by the process of Claim 1 which differs, as already indicated, from that disclosed in citation (1) only in that a combination of lithium iodide and methyl iodide is used as a promoter.

In view of the experimental evidence available from the patent in suit (see Control Experiment B, which uncontestedly is representative for the state of the art as disclosed in document (1), and Example 1; page 6, line 63 to page 7, line 12) the Board accepts that the above problem is indeed solved by the claimed process. For example, the calculated rate to acetic acid was increased from 0.86 to 2.2 g mole/l.h at a selectivity of 100%.

8. The Board cannot accept the Appellant's argument that document (2), and in particular its Examples 22 and 23 rendered the claimed process obvious. It is true that the said Example 23 discloses a mixture of lithium iodide and methyl iodide as a promoter in a rhodium catalysed carbonylation, and that the catalyst system of this example is remarkably more active as that of Example 22, where only lithium iodide is used. However, this document and these examples do not relate to the manufacture of acetic acid - but to the production of monocarboxylic acid anhydrides (page 3, lines 16 to 18) and in particular of acetic anhydride (page 22, lines 14 to 31).

8.1 The skilled person in the field of catalytic reactions is aware that knowledge gained in respect of a particular process cannot, in general, be transferred to a different process. This is true in the case where, as here, the catalyst-system remains unchanged but one starting material is replaced by a different one. In the light of this fact the skilled person would not have transferred the knowledge obtained in connection with the carbonylation of methyl acetate to the carbonylation of methanol. Whilst the Appellant conceded this, he submitted that it was obvious at least for the skilled person to seek confirmation of this experimentally. Even if the Board were to accept this, which is not the case, in the current situation, the skilled person would have been deterred from carrying out such experiments.

8.2 Document (2) shows on page 5 a reaction scheme explaining the reaction sequences involved in the formation of the acid anhydride. The first two partial reaction are quoted (the third reaction relates to dialkyl ethers as starting material which is irrelevant in the present context):

$$RX + CO \rightarrow RCOX \qquad [1]$$
$$RCOX + RCOOR \rightarrow RX + (RCO)_2O \qquad [2]$$

On the one hand, this reaction scheme, where R is a hydrocarbon radical and X is I or Br, teaches the skilled person that, in the carbonylation of methyl acetate, the actual carbonylating agent is the acetyl iodide formed from the promoter methyl iodide, and not carbon monoxide, and that the catalytic function of the methyl iodide relies on it being set free in the second reaction step. On the other hand, the skilled person is aware that the highly reactive acetyl iodide will react spontaneously with methanol to form methyl acetate.

8.3 The skilled person, who is concerned with the question whether the improvement in the activity of the catalyst by changing from methyl iodide to the combination of methyl iodide and lithium iodide, which is deducible from Examples 22 and 23 of document (2), is transferable to the carbonylation of methanol, would immediately realise that the question must be answered in the negative, since the contemplated starting material, methanol, would immediately scavenge the actual carbonylating agent, acetyl iodide, by ester formation, and so render it unavailable for the second reaction and, hence, its reformation from methyl iodide, which is indispensable for its catalytic function, impossible.

Thus, the Board concludes that this disclosure would have deterred the skilled person from using the reaction system of document (2) in a carbonylation process for the manufacture of acetic acid from methanol.

8.4 The Respondents counter-argument that under the prevailing reaction conditions the acetic anhydride formed in equation [2] would react with the methanol to form acetic acid and methyl acetate is likewise not convincing. The skilled person would not have considered this possibility since, as explained above, he would have expected the loss of promoter activity of the methyl iodide by changing the starting material from methyl acetate to methanol. In other words, with the loss of acetyl iodide by the scavenging effect of methanol, acetic anhydride would no longer be formed.

8.5 Furthermore, document (2) is silent on the issue of product selectivity, and on whether the already high value known for this feature from document (1) (>95% for acetic acid, see column 12, lines 21 to 22), could be further increased by the use of the combination of lithium iodide and methyl iodide as a promoter. Thus, even if one were to accept, for the sake of argument, that the skilled person might have gained from citation (2) useful information regarding the catalyst system's activity in a process for the production of acetic acid, this would not have been sufficient for him to expect that such a catalyst combination would solve the present technical problem as defined, in which the product selectivity is of paramount importance. It has to be noted in the present context that the product yields in the Examples 22 and 23 of document (2) are by far lower than the theoretical ones when calculated on the basis of the starting material methyl acetate which, at least, casts some doubt on the selectivity of this catalyst system.

9. Document (3A) is concerned with the production of acetic anhydride and relates to the problem of improving carbonylation processes by avoiding catalyst pollution and loss of the valuable rhodium or iridium catalysts in the reaction work up (page 5, lines 9 to 15, in combination with page 3 line 25 to page 4, line 25). This problem is unrelated to that of the disputed patent so that the skilled person would not have considered this document helpful in solving of the present technical problem. In addition, the disclosure of this document is confined to the manufacture of acid anhydrides and the indication that promoters, like lithium and chromium (page 9, lines 10 to 11), can improve the "activity of rhodium- or iridium catalysts" (page 8, lines 29 to 32) is too vague to rouse expectations that a more sophisticated problem (selectivity, space-time yield) might be solved by this suggestion in a different carbonylation reaction (production of acetic acid). Therefore, the Board cannot see any reason, why a skilled person should have combined the disclosure of citation (3A) with that of citation (1).

10. Document (4), which also discloses a process for the manufacture of acetic anhydride from methyl acetate,

whereby acetic acid may be produced simultaneously with acetic anhydride, if methanol is added to starting material (page 2, paragraph 4) makes no mention of lithium iodide. Hence, it could not have induced the skilled person to incorporate this compound into the catalyst system of document (1) for solving the present technical problem.

11. Document (5) is concerned with reducing the catalyst precipitation problems encountered in the reaction work up of rhodium catalysed carbonylations (page 5, lines 19 to 22). Lithium acetate is disclosed, *inter alia*, as a catalyst stabiliser (example 18, page 22, lines 33 to 35 and Claim 14). Although the carbonylation of methanol is also disclosed as a possible reaction, no information can be found in this citation relating to product selectivity and space-time yield, which is the problem of the patent in suit. Thus, in the Board's judgment, there is no indication that the promoter combination of the patent in suit would, in combination with the other process features as claimed, solve the present technical problem.

12. Document (8) refers to the production of carboxylic acids and esters from alcohol and carbon monoxide without the formation of water and hydrogen halide (column 1, lines 5 to 8, and lines 41 to 45). Methyl iodide and lithium iodide are disclosed in column 2, lines 55 to 58 as examples of alternative iodine comprising catalyst promoters. However, as the Appellant conceded, the combination of these compounds as promoters is not disclosed in this citation, let alone that a beneficial effect of such a combination on the selectivity of the acetic acid formation and catalyst activity in a process according to document (1) would be expected.

The fact that this document, which was published in 1980, i.e. long after citations (1) and (2) published 1973 and 1975 respectively, does not even mention the combination of methyl iodide and lithium iodide as a possible promoter is a further indication that such combination was not obvious to the skilled person as a solution of the present technical problem and that the Appellant's opinion results from hindsight.

13. For all these reasons, in the Board's judgment, the proposed solution to the technical problem underlying the patent in suit is inventive, and in consequence independent Claim 1 is allowable. Dependent Claims 2 to 6, which relate to preferred embodiments of the process claimed in Claim 1, derive their patentability from that of Claim 1 and are likewise allowable.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Opposition Division with the order to maintain the patent in accordance with the Respondent's request made in the course of oral proceedings.

The Registrar:

Martorana

P. Martorana

The Chairman:

A. Jahn