BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

A		B		C	X

File No.: T 0346/92 - 3.3.1

Application No.: 84 114 575.8

Publication No.: 0 144 935

Classification: C07C 51/12

Title of invention: Production of carboxylic acids from alcohols

D E C I S I O N
of 29 July 1993

Applicant:

Proprietor of the patent: UNION CARBIDE CORPORATION

Opponent: 01) HOECHST AKTIENGESELLSCHAFT, Frankfurt
02) BP Chemicals Limited

Headword: Acetic Acid/UNION CARBIDE

EPC: Art. 56

Keyword: "Inventive step (affirmed); non-obvious alternative" - "Long delay in issuing written reasons after oral decision not acceptable"

Headnote
Catchwords

Case Number: T 0346/92 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 29 July 1993

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Decision under appeal: **Decision of the Opposition Division of the European Patent Office of 5 April 1990, with written reasons posted on 12 March 1992, concerning maintenance of European patent No. 0 144 935 in amended form.**

Composition of the Board:

Chairman: K.J.A. Jahn
Members: R.W. Andrews
S.C. Perryman

Summary of Facts and Submissions

I. European patent No. 0 144 935 in respect of European patent application No 84 114 575.8, which was filed on 30 November 1984, was granted on 16 June 1987 (cf. Bulletin 87/25).

II. Notices of Opposition which were filed on 4 December 1987 and 16 March 1988, requested the revocation of the patent on the grounds that its subject-matter lacked novelty and did not involve an inventive step. The oppositions were supported, *inter alia*, by the following documents:

(1) US-A-3 769 329 and
(4) EP-A-O 055 618.

III. By a decision delivered orally on 5 April 1990, with written reasons being issued on 12 March 1992, the Opposition Division maintained the patent in amended form on the basis of Claims 1 to 5 submitted during oral proceedings. The Opposition Division held that the subject-matter of the amended Claim 1 was novel. With respect to inventive step, the Opposition Division found it was not obvious that the use of lithium iodide in the presence of added methyl acetate and in the absence of any added water would solve the problem of improving the process of document (1) with respect to its efficiency, selectivity and conversion rate.

IV. An appeal was lodged against the decision by Opponent OI on 13 April 1992 with payment of the prescribed fee. In his Statement of Grounds of Appeal filed on 12 May 1992 and during the oral proceedings held on 29 July

1993, the Appellant contended that document (1) disclosed a process for the production of acetic acid by carbonylating a mixture of methanol and methyl acetate in a molar ratio of 0.001:1 to 2:1 using a homogenous catalyst system comprising rhodium and a metal halide. The Appellant argued that in view of results obtained in the examples of the disputed patent and those of document (1), in particular Example 4, with respect to conversion rate and selectivity, the only problem underlying the disputed patent must lie in improving the stability of the catalyst system of document (1).

However, Example 18 of document (4) disclosed that lithium iodide, prepared *in situ* from lithium acetate and hydrogen iodide, improved the stability of rhodium based carbonylation catalysts. Therefore, the claimed subject-matter was obvious in the light of the combined teaching of documents (1) and (4).

V. The Respondent alleged that the invention lay in the combined selection of the use of lithium iodide as promoter, the deliberate addition of methyl acetate and the absence of any added water. Document (1) was completely silent with respect to the use of lithium iodide and there was no incentive for the skilled person to combine the teaching of document (1) with that of document (4) which was concerned with the stability of rhodium carbonylation catalysts under substantially lower pressures than those used in the carbonylation reaction itself. Furthermore, both these documents emphasised that the addition of water to the reaction mixture exerted a beneficial effect on

reaction rate. In contrast to this the present process specifically excluded the addition of water.

The Respondent also argued that the results of the comparative tests submitted during the examination proceedings on 18 June 1986 demonstrated an improvement in conversion rate *vis à vis* document (1).

VI. The Appellant requested that the decision under appeal be set aside and that the patent be revoked.

The Respondent requested that the decision under appeal be set aside and that the patent be maintained in the amended form submitted during the oral proceedings.

Claim 1 in this amended form reads as follows.

"A process for the production of acetic acid by the catalytic reaction of methanol and carbon monoxide or synthesis gas in contact with a catalyst system consisting of rhodium metal or rhodium compound and lithium iodide, characterised in that the catalyst system is a homogeneous catalyst system and methyl acetate but no water is added to the reaction mixture".

The other party to the proceedings took no part in the appeal and did not attend the oral proceedings.

VII. At the conclusion of the oral proceedings the Board's decision to maintain the patent in amended form was announced.

Reasons for the Decision

1. The appeal is admissible.

2. There are no objections under Article 123 EPC to the present claim. n particular, Claim 1 is based on Claims 1 and 5 as granted and page 4, line 6 of the printed patent specification (cf. also Claims 1 and 5 as filed and page 10, lines 2 and 3 of the published patent application). Claims 2 to 5 correspond to Claims 2, 3, 6 and 7 as granted and Claims 2, 3, 6, and 8 as filed.

3. The disputed patent relates to a process for the production of acetic acid by the catalytic reaction of methanol and carbon monoxide or synthesis gas in the presence of added methyl acetate. Document (1), which represents the closest state of the art, discloses a process for the preparation of acetic acid by reacting methanol in with carbon monoxide in the presence of methyl acetate, a rhodium compound and a halogen compound. The examples of this document demonstrate that selectivities of greater than 95% with 100% conversion can be achieved.

In the light of this closest prior art, the technical problem underlying the disputed patent is to provide an alternative process for the efficient and selective production of acetic acid.

According to the disputed patent, this technical problem is essentially solved by carrying out the carbonylation reaction in the presence of a homogeneous catalyst consisting of rhodium metal or a rhodium compound and lithium iodide in the absence of any added water.

In the light of the examples in the disputed patent, the Board is satisfied that the technical problem as defined above has been solved.

The Respondent maintained that the comparative tests, submitted on 18 June 1986 during the examination proceedings, demonstrated that the conversion rate (as measured by the consumption of carbon monoxide) of the present process was higher than that of the process of document (1). Therefore, he contended that the technical problem underlying the patent in suit should be seen in providing an improved process for the production of acetic acid wherein the improvement lies in the higher conversion rate as compared to that obtained in this prior art process.

However, it can be seen from the Table summarising the results of these experiments that the reaction using methyl iodide as the promoter was carried out at a lower pressure than the one using lithium iodide; 55.2 bar or 800 psig as compared to 69 bar or 1000 psig. Since it is well known that the pressure is an important parameter for carbonylation reactions, the Board considers that these two experiments do not represent a fair comparison between the present process and the process of document (1). The Respondent alleged that the increase in the consumption of carbon monoxide using lithium iodide as the promoter by a factor of 3 as compared to that obtained using methyl iodide, nevertheless, should be taken as indicative of the improved conversion rates achieved with the claimed process. To counter this submission the Appellant relied on the non-linear relationship between the partial pressure of the carbon monoxide and the

conversion rate. In these circumstances, these experimental results cannot be taken into account in establishing the technical problem underlying the disputed patent.

4. After examination of the cited documents, the Board has concluded that the claimed subject-matter is novel. Since novelty is no longer in dispute, it is not necessary to give detailed reason for this finding.

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

5.1 As previously mentioned document (1) discloses a process for the production of acetic acid by the carbonylation of methanol, optionally in the presence of, *inter alia*, methyl acetate using a catalyst containing a rhodium component and a bromine or iodine component (cf. column 3, lines 14 to 30, column 8, lines 8 to 14 and Examples 4 and 20).

This document also discloses that it is generally preferred to have an excess of halogen present in the catalyst system as a promoting component. This promoting component consists of a halogen and/or halogen compound such as, for example, a metal halide (cf. column 4, lines 46 to 62). However, there is no mention of lithium iodide in this document.

Moreover, document (1) also teaches that when an ester is present in the feedstock it is normally charged with equimolar amounts of water, although more or less water may be used (cf. column 8, lines 42 to 44). In fact, it was found that water may exert a beneficial effect on

the rate of reaction and that an amount of water in excess of the equimolar quantity of water to ester promotes the production of the carboxylic acid (cf. column 8, lines 48 to 53 and Claims 1 and 16). Therefore, a skilled person would conclude from the disclosure of document (1) that, to obtain the optimum results from the carbonylation of a feedstock consisting of methanol and methyl acetate, it is also necessary to include water in the feed stream to the reactor.

In contrast thereto, although it is absolutely essential for methyl acetate to be present in the feedstock of the process of the disputed patent using lithium iodide as the promoter (cf. page 5, lines 62 to 65 and page 6, lines 41 to 43), the reaction is carried out in the absence of any added water.

In the Board's judgment, document (1) would not provide the skilled person with any indication that an alternative to the process disclosed therein lies in the use of a feed stock consisting of methanol and methyl acetate but no added water, and a catalyst system consisting of rhodium metal or a rhodium compound and lithium iodide.

5.2 Document (4) discloses a process for carbonylating an alcohol, or an ester, halide or ether derivative of said alcohol in the liquid phase using a catalyst system containing a rhodium component and an iodine or bromine component wherein the catalyst system is stabilised against decomposition under conditions of reduced carbon monoxide partial pressure by the presence of a stabilising component (cf. Claim 1 in

combination with page 4, lines 11 to 16). Suitable stabilisers include compounds of alkali metals (cf. group (4) in Claim 1). In Example 18, lithium iodide, formed *in situ* from lithium acetate and hydrogen iodide, is used as the stabilising component. This document also discloses that water present in the reaction mixture exerts a beneficial effect upon the reaction rate (cf. page 8, lines 23 to 25).

According to the disputed patent, one of the criteria required of the catalyst in the catalytic reaction of synthesis gas or carbon monoxide in processes to produce oxygenated organic compounds is that it must be as stable as possible (cf. page 3, lines 42 to 44). Therefore, it could be argued that, in order to satisfy this criterion, the skilled person would contemplate combining the teaching of documents (1) and (4).

However, in order to arrive at the proposed solution to the above-defined technical problem, the skilled person would have to take the following steps:

(a) He would have to select lithium iodide from the large number of stabilisers disclosed and taught by document (4).

(b) He would have to realise that the presence of both methanol and methyl acetate in the feedstock was absolutely essential for the success of the process, and

(c) He would have to ignore the clear teaching of both documents (1) and (4) regarding the beneficial effect of water in the reaction mixture.

In the Board's judgment, there is nothing in the disclosure of documents (1) and (4) which would have led the skilled person to take these necessary steps to arrive at the proposed solution to the technical problem of providing an alternative process for the preparation of acetic acid by the carbonylation of methanol to the one described in document (1).

6. Therefore, the subject-matter of Claim 1 involves an inventive step. Dependent Claims 2 to 5, which relate to preferred embodiments of the process as claimed in Claim 1, are also allowable.

7. The Board has dealt with this appeal as quickly as possible in view of the delay of more than two years between the delivery of the decision at the oral proceedings before the Opposition Division and the issuing of the written decision. It is emphasised that such a delay cannot be regarded as acceptable because of the considerable risk of all kinds of error which it is likely to engender (cf. T 243/87 of 30 August 1989, published in EPOR [1990] 136).

Order

For these reason, 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 the amended form submitted during the oral proceedings.

The Registrar: The Chairman:

E. Görgmaier K.J.A. Jahn