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File Number: T 619/89 - 3.3.1

Application No.: 80 108 181.1

Publication No.: 0 031 590

Title of invention: Rhodium catalyzed hydroformylation process

Classification: C07C 45/50

D E C I S I O N
of 7 March 1991

Proprietor of the patent: Union Carbide Corporation

Opponent: BASF Aktiengesellschaft, Ludwigshafen

Headword: Hydroformylation/Union Carbide

EPC Articles 54 and 56

Keyword: "Novelty (confirmed) - proper construction of claim"
"Inventive step (confirmed) - unexpected result in the light
of the prior art"

Headnote



Case Number : T 619/89 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 7 March 1991

Appellant :
(Proprietor of the patent)

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Respondent :
(Opponent)

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Representative :

-

Decision under appeal :

Decision of the Opposition Division of the
European Patent Office of 21 February 1989,
posted on 18 July 1989, revoking European patent
No. 0 031 590 pursuant to Article 102(1) EPC.

Composition of the Board :

Chairman : Jahn, K.J.A.
Members : Andrews, R.W.
Holtz, E.M.C.

Summary of Facts and Submissions

- I. European patent No. 0 031 590 in respect of European patent application No. 80 108 181.1, which was filed on 23 December 1980, was granted on 14 December 1983 (cf. Bulletin 83/50) on the basis of five claims.

- II. On 24 August 1984 a notice of opposition was filed in which the revocation of the patent was requested on the grounds that its subject-matter lacked novelty and did not involve an inventive step having regard to the disclosure in the document

(1) DE-A-2 354 217

- III. By a decision delivered orally on 21 February 1989, with written reasons posted on 18 July 1989, the Opposition Division revoked the European patent. The Opposition Division held that only the subject-matter of the single claim filed during the oral proceedings was novel in the light of the disclosure of document (1). However, the subject-matter of this claim did not involve an inventive step in the light of the teaching of GB-A-1 120 277 (document (2)) since it had not been shown that the claimed process overcame the drawback of this prior art process with regard to the amount of ethylene that may be present in the olefinic feed or that an unexpected effect compared to the process of document (2) had been achieved insofar as the catalyst efficiency was concerned.

- IV. An appeal was lodged against this decision on 15 September 1989 with payment of the prescribed fee. In his Statement of Grounds of Appeal filed on

28 November 1989 and during the oral proceedings held on 7 March 1991, the Appellant maintained that document (1) disclosed the hydroformylation of ethylene by itself or of a mixture comprising ethylene, propylene and butylene.

With respect to inventive step, the Appellant argued that, unexpectedly, the hydroformylation of propylene to a product having a high ratio of normal to iso-butyraldehyde could be carried out simultaneously with that of ethylene without reducing the production rate of butyraldehyde below that achieved when propylene is hydroformylated in the absence of ethylene. The Appellant contended that there was no support in the prior art for the allegation that the hydroformylation of two different olefins would obviously proceed independently and lead to an increase in catalyst efficiency. On the contrary, document (2) shows that the presence of ethylene during the hydroformylation of ethylene and propylene using a cobalt catalyst negatively influences the rate of production of butyraldehyde.

- V. The Respondent argued that the expression "up to 5% by volume of butene" in document (1) has to be construed as meaning that the percentage of butene can be zero.

The Respondent also contended that the unexpected effect relied upon by the Appellant to support inventive step had not been shown to be present and even if it were, could not be used in view of the Decision T 192/82. The Respondent also argued that from a comparison of Run 3 of Example 2 with Run 4 of Example 3 of document (2) it cannot be concluded that ethylene has a negative influence on the production rate of the butyraldehydes. The Respondent further alleged that the Appellant's calculation purporting to show this negative influence was incorrect. In any case, it was impossible to compare the results of Tables 1 and 2 of document (2) in view of differences in reaction

conditions. Similarly, a comparison could not be drawn between the results obtained in document (2) and those of the disputed patent.

The Respondent supported his arguments that the results obtained by the claimed process were to be expected by means of various diagrams and calculations.

- VI. The Appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of three sets of claims filed during oral proceedings.

The only independent claim in accordance with the Appellant's main request reads as follows:

"A process for the production of butyraldehyde products rich in normal aldehydes which comprises

- (1) forming a reaction mixture containing (a) propylene; (b) carbon monoxide; (c) hydrogen; (d) a catalytic quantity of a complex catalyst consisting essentially of rhodium in complex combination with (i) carbon monoxide and (ii) a triorgano phosphorus ligand of the group consisting of trialkylphosphites, tricycloalkylphosphites, triarylphosphites and triarylphosphines; each organic moiety thereof having up to 18 carbon atoms; and said triorgano phosphorus ligand possessing a Δ HNP value of at least 425; and (e) at least 2 moles of free ligand as defined above per mole of rhodium;
- (2) maintaining the reaction mixture at a temperature in the range of from 50°C to 145°C; at a total pressure of carbon monoxide and hydrogen of less than 31 bar (450 psia), and a partial pressure attributable to carbon monoxide no greater than 75 percent of said total pressure,; thereby

(3) reacting said propylene with said carbon monoxide and hydrogen with the formation of said butyraldehyde products, characterized by including in said reaction mixture ethylene and additional carbon monoxide and hydrogen in amounts at least sufficient to react with said ethylene and reacting said ethylene with said carbon monoxide and hydrogen with the formation of propionaldehyde in addition to and concurrently with said butyraldehyde products."

The main claim in accordance with the alternative request 1 additionally specifies that the ethylene and propylene are the only olefins present and that the mole ratio of ethylene to propylene is 0.1:1 to 10:1. The only claim according to the Appellant's alternative request 2 also requires that the process be carried out in high boiling liquid aldehyde condensation products formed during the reaction.

The Respondent requested that the appeal be dismissed.

VII. At the conclusion of the oral proceedings the Chairman announced the Board's decision to allow the appeal according to the main request.

Reasons for the Decision

1. The appeal is admissible.
2. There are no formal objections under Article 123 EPC to any of the claims in accordance with the Appellant's three requests. Thus, Claim 1 of the main request represents a combination of Claims 1 and 3 as filed and granted. Claims 2 and 3 of this request correspond to Claims 2 and 5

as filed and granted. Claim 1 of the first alternative request is a combination of Claims 1 to 3 as filed and granted. Claim 2 of this request corresponds to filed and granted Claim 5. The only claim according to the second alternative request represents a combination of Claims 1, 2, 3 and 5 as filed and granted.

3. The disputed patent relates to a process for the production of aldehydes by reacting olefins with carbon monoxide and hydrogen in the presence of a catalytic quantity of a complex catalyst consisting of rhodium in complex combination with carbon monoxide and a triorganophosphorus ligand having a Δ HNP value of at least 425 selected from trialkyl phosphites, tricycloalkyl phosphites, triaryl phosphites and triarylphosphines and at least 2 moles of free ligand per mole of rhodium.

3.1 In the Board's judgement, GB-A-1 338 237 (document B) represents the closest prior art. This document discloses the low pressure rhodium catalysed hydroformylation of an alpha-olefin having 2 to 20 carbon atoms, such as ethylene or propylene (cf. Claim 1 in combination with page 7, lines 55 to 68). However, in this document there is no indication that a mixture of olefins could be hydroformulated.

In the light of this closest state of the art, the technical problem underlying the patent in suit is to be seen in providing an improved hydroformylation process, wherein the improvement lies in increasing the catalyst efficiency, i.e. increasing the unit weight of aldehyde produced in a given time per unit weight of catalyst (cf. printed patent specification page 6, lines 28 to 32).

- 3.2 According to the disputed patent, this technical problem is essentially solved by the hydroformylation of a mixture of ethylene and propylene.
- 3.3 Having regard to Example II of the disputed patent, the Board is satisfied that the above-defined technical problem is effectively solved. According to Part A of this Example, which corresponds to the process disclosed in document B, the hydroformylation of propylene (partial pressure 1.65 bar) resulted in the formation in the steady state of 1.1 gram moles per litre-hour of butyraldehydes, which equates to a catalyst efficiency of 7,845 kg of aldehyde/(day)/(kg of contained rhodium). In Part B, a mixture of ethylene (partial pressure 0.034 bar) and propylene (partial pressure 1.65 bar) hydroformylated under the same reaction conditions resulted in the formation of 1.2 gram moles per litre-hour of propionaldehyde and 1.1 gram moles per litre-hour of butyraldehydes. This is equivalent to a catalyst efficiency of 14,700 kg of aldehydes/(day)/(kg of contained rhodium).
4. In view of the fact that both parties disagreed in respect of the proper construction of Claim 1 in accordance with the Appellant's main request, it is necessary first of all to clarify the point. According to the Respondent the above-mentioned claim covered the hydroformylation of a mixture of ethylene, propylene and other olefins. However, the Appellant insisted that the claim was restricted to the hydroformylation of a mixture in which ethylene and propylene are the only olefins present. The Appellant stressed that the auxiliary requests in which the presence of olefins other than ethylene and propylene are specifically excluded were only filed to safeguard the Appellant's position in the event of the rejection of his main request. Therefore, these requests are not to be

interpreted as meaning that it was intended that the claim of the main request should include within its ambit the hydroformylation of mixtures containing olefins in addition to ethylene and propylene.

From an examination of the whole disclosure of the disputed patent, it is clear that the process is confined to the hydroformylation of mixtures containing ethylene and propylene as the only olefins. Since the whole tenor of the disputed patent excludes the hydroformylation of other olefinic mixtures, Claim 1 in accordance with the Appellant's main request has to be construed as claiming the hydroformylation of reaction mixtures in which ethylene and propylene are the only olefins present. During the oral proceedings, the Appellant's Representative agreed that this was the proper and only possible construction of Claim 1 in accordance with the main request.

- 4.1 Document (1) discloses the hydroformylation of a so-called lean gas in the presence of a complex rhodium catalyst. The lean gas, which is the gaseous mixture remaining after the separation of valuable constituents from the gaseous mixture resulting from the autothermal cracking of oil, contains olefins having 2 to 4 carbon atoms, carbon monoxide and hydrogen (cf. first two paragraphs on page 1, original numbering).

According to the first paragraph on page 2 of this document, the lean gas contains, for instance, 10 to 60% by vol. carbon monoxide, 10 to 60% by vol. hydrogen, 5 to 50% by vol. methane, ethane, propane and butane, 5 to 30% by vol. ethylene, 1 to 10% by vol. propylene, up to 5% by vol. butylene, up to 2% by vol. acetylene and propyne, up to 2% by vol. allene and butadiene, up to 1% by vol. carbon dioxide and very small amounts of oxygen, carbonyl sulphide and hydrogen sulphide. This paragraph also discloses that a

typical mixture contains, for example, 5.4% by vol. methane, 42.8% by vol. carbon monoxide, 30.6% by vol. hydrogen, 20.6% by vol. ethylene, 0.6% by vol. air and 13 mg hydrogen sulphide per cubic metre.

In a passage referring to the advantages obtained by the presence of an excess of triphenylphosphine in the hydroformylation reaction medium, it is stated that this measure is particularly advantageous if propylene is also present in the lean gas (cf. page 3, lines 8 and 9). Finally, the Examples describe the hydroformylation of a lean gas containing ethylene as the only olefin.

- 4.2 Thus, from the above, it is clear that this document contains a number of inconsistencies which render it difficult to reach a conclusion on the actual technical teaching a skilled person would derive from it. Nevertheless, in the Board's judgement, the expression "such lean gases contain ... up to 5% by volume of butylene" (solche Armabgase enthalten ... bis zu 5 Volumenprozent Buten) has to be construed as meaning that butylene is always present in these lean gases.
- 4.3 Furthermore, since these particular lean gases also contain 1 to 10% by volume propylene, the above-mentioned reference on page 3, lines 8 and 9 could apply to these gases or to the more generally defined lean gases referred to in the second paragraph on page 1. However, in the latter case, the implication that propylene may not be present in the lean gas would disclose a gas containing ethylene and butylene. Moreover, in view of this implication of the absence of propylene, the skilled person would not combine the passage on page 3, lines 8 and 9 with any disclosure relating to specific lean gases in which ethylene is the only olefin present.

- 4.4 Therefore, the disclosure of this document cannot be considered as having made available to the public the hydroformylation of a gaseous mixture containing, as the only olefins, ethylene and propylene. Thus, the subject-matter of Claim 1 in accordance with the Appellant's main request is novel.
- 4.5 After examination of the other cited and more remote prior art, the Board finds that this claimed subject-matter is also novel with respect to the disclosure of these documents.
5. It still remains to be examined whether the requirement of inventive step is met by claimed subject-matter.
- 5.1 Document (2) discloses a process for producing aldehydes by reacting olefins, carbon monoxide and hydrogen in the presence of a cobalt oxo catalyst which is insoluble in the reaction product. The concentration by volume of olefins in the reaction mixture is 5 to 20%, provided that the concentration of ethylene, if any, in the mixture when this contains an olefin having 3 or 4 carbon atoms, is less than 5% (cf. Claim 1).
- 5.2 Examples 3 and 4 of this document illustrate the simultaneous production of propionaldehyde and n- and isobutyraldehydes by hydroformylating a mixture of ethylene and propylene. In particular, Example 3 illustrates the effect of the concentration of ethylene and propylene on the reaction rate, yield and catalyst efficiency using the reactor and reaction conditions described in Example 2 in which the effect of propylene concentration on the same parameters was investigated.
- 5.3 The Respondent argued that the results of Tables 1 and 2 were not comparable in view of the lack of details in

Table 2 concerning the amount of carbon monoxide, hydrogen and inerts in the feed gas and the variations in the amounts of these constituents in the feed gases of Table 1. However, in the light of the statement that the same reactor and reaction conditions were used in both Examples (cf. page 3, lines 62 to 64) and, in the absence of any evidence to the contrary, the Board considers that it is possible and permissible to compare the results of these two Tables.

According to the third run of Table 1, the hydroformylation of a feed gas containing 5% by volume propylene resulted in a catalyst efficiency of 5430 lbs aldehyde per lb cobalt used in the reaction. However, the catalyst efficiency obtained in the hydroformylation of a feed gas containing 5% propylene and 2% ethylene was 5440 lbs aldehydes (propionaldehyde and butyraldehydes) per lb cobalt used (cf. fourth run in Table 2).

From the very small increase in catalyst efficiency, the skilled person would conclude that the production rate of butyraldehydes is negatively influenced by the presence of ethylene in the feed and that the two hydroformylation reactions are not independent of each other.

In the absence of any evidence to the contrary, the Board considers that the skilled person would expect that the same type of negative influence would also occur if mixtures of ethylene and propylene were hydroformylated using a rhodium catalyst. Consequently, it is surprising and unobvious that, in the presence of a rhodium hydroformylation catalyst under the specified reaction conditions, the two reactions are independent of each other. This unexpected independence of the two hydroformylation reactions enables the technical problem

underlying the patent in suit of increasing the catalyst efficiency to be solved.

- 5.4 In view of the technical teaching of document (2), the Respondent's assumption that the hydroformylation of ethylene and propylene would proceed independently of each other is unjustified. Moreover, the diagrams and calculations presented by the Respondent to support his contention that the claimed process is obvious are either based on this unwarranted assumption or in fact demonstrate the independence of the ethylene and propylene hydroformylation reactions.
- 5.5 The Respondent argued that a comparison could not be drawn between the results obtained by the process of document (2) and those achieved by the process of the disputed patent. However, by defining the technical problem underlying the patent in suit in the above manner, such a comparison is rendered unnecessary.
- 5.6 The Board cannot accept the Respondent's arguments that in the present case the skilled person was in a "one-way street" situation (cf. Decision T 192/82, OJ EPO 1984, 415). Faced with the problem of improving the prior art process disclosed in document B in the manner specified above, there are several possibilities the skilled person could have tried, such as, for example, modification of the catalyst or varying the reaction conditions. Moreover, in view of the disclosure of document (2), which is the only cited prior art to specifically describe the hydroformylation of mixtures of ethylene and propylene, the skilled person would be positively discouraged from hydroformylating mixtures of ethylene and propylene with any expectation of solving his technical problem.

- 5.7 The disclosure of document (1) as outlined above would not provide the skilled person with any indication that the solution to the technical problem underlying the disputed patent lies in the hydroformylation of mixtures of ethylene and propylene.
6. In the Board's judgement, the proposed solution to the technical problem underlying the patent in suit is inventive. Therefore, the subject-matter of Claim 1 in accordance with the Appellant's main request involves an inventive step. This claim and dependent Claims 2 and 3, which relate to preferred embodiments of the process of Claim 1, are allowable.
7. In view of the above, it is not necessary to consider the Appellant's other requests.

Order

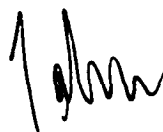
For these reasons, it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the first instance with the order to maintain the patent in amended form on the basis of the main request submitted during oral proceedings.

The Registrar:


E. Gorgmaier

The Chairman:


K.J.A. Jahn

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LWA
C. M. J.