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File Number: T 565/90 - 3.3.1

Application No.: 82 105 793.2

Publication No.: 0 071 032

Title of invention: Process for the preparation of methyl tert-butyl ether and ethyl tert-butyl ether

Classification: C07C 43/04

D E C I S I O N
of 15 September 1992

Proprietor of the patent: EUTECO IMPIANTI S.p.A., et al

Opponent: Hüls Aktiengesellschaft

Headword: Ethyl tert.-butylether/EUTECO

EPC Articles 54 and 56

Keyword: "Novelty (no) - overlapping numerical ranges"
"Novelty (yes - auxiliary request)"
"Inventive step for a chemical process (yes) - homolgy of reactants"



Case Number : T 565/90 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 15 September 1992

Appellant :
(Proprietor of the patent)

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Representative :

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Respondent :
(Opponent)

Hüls Aktiengesellschaft
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Decision under appeal :

Decision of Opposition Division of the European
Patent Office of 8 November 1988 posted on 9 May
1990 revoking European patent No. 0 071 032
pursuant to Article 102(1) EPC.

Composition of the Board :

Chairman : K.J.A. Jahn
Members : P. Krasa
C. Holtz

Summary of Facts and Submissions

- I. The mention of the grant of patent No. 0 071 032 in respect of European patent application No. 82 105 793.2, filed on 29 June 1982, was published on 2 May 1985 (cf. Bulletin 85/18) on the basis of three claims. The only independent Claim 1 reads as follows:

"Process for the preparation of methyl tert-butyl ether or ethyl tert-butyl ether by the selective reaction of methanol or ethanol respectively with isobutylene contained in a C₄-hydrocarbon fraction, in the liquid phase, in the presence of acid catalysts, with a molar ratio of the alcohol to the isobutylene of from 1:1 to 1.25:1, at a temperature of from 40° to 100°C and at a pressure of from 15 to 40 bars, the reaction medium being diluted by an aliphatic hydrocarbon or mixtures of aliphatic hydrocarbons which are liquid under the operating conditions and inert to the other constituents of the reaction medium and which are the recycle product obtained from the reaction mixture after separation of at least the formed ether, characterised in that the recycle product is fed to the reaction medium in an amount to maintain, at the equilibrium, a molar ratio of aliphatic hydrocarbon with the formed ether higher than 7:1."

- II. A notice of opposition, which was filed on 9 January 1986, requested revocation of the patent on the ground of lack of novelty and of inventive step. The opposition was based, inter alia, on the following document:

(1) DE-A-3 005 013.

In the course of the opposition proceedings the following document was additionally cited:

(1A) GB-A-2 043 065 (corresponding to citation (1)).

- III. By a decision delivered orally on 8 November 1988, with written reasons posted on 9 May 1990, the Opposition Division revoked the patent for lack of inventive step. It held that the claimed subject-matter was novel but did not involve an inventive step, as far as the manufacture of methyl tert.-butyl ether (MTBE) was concerned.

The Opposition Division stated that citation (1A) disclosed the beneficial effect of increasing the recycle rate in respect to both the increased MTBE yield and the reduced isobutylene content in the reaction mixture. It found it highly plausible that a skilled person would have carried out experiments with increasing recycle rates, which means with increasing dilution, and, thus, would have arrived at the technical teaching of the patent in suit. The Opposition Division also indicated, that the claimed subject-matter involved an inventive step, insofar as the manufacture of ethyl tert.-butyl ether (ETBE) was concerned.

- IV. An appeal was filed against this decision on 6 July 1990, the prescribed fee having been paid on 2 July 1990. A Statement of Grounds of Appeal was filed on 14 September 1990.

- V. The Appellant (Patentee) submitted that citation (1A) generically disclosed a recycle rate of 0.1:1 to 10:1, preferably of 0.3:1 to 3:1. He argued that nothing in this citation suggested that a recycle rate higher than 1:1 should be preferred over a recycle rate below 1:1.

Regarding inventive step, he submitted that reference (1A) only showed an increase of isobutylene conversion when the methanol/isobutylene ratio in the total feed is increased

together with an increase of the recycle rate from 0 to 1. In contrast to this the patent in suit discloses an increased isobutylene conversion at a fixed molar ratio between methanol and isobutylene with a very low methanol excess in the overall feed.

- VI. The Respondent argued that citation (1) anticipated the process of manufacturing MTBE according to the patent in suit. In particular, when examples 3 and 6 of document (1) are performed with a recycle rate of 10, molar ratios of inert C₄-hydrocarbons to formed ether are obtained which are within the range of Claim 1 of the present main request. To that end, he submitted calculations during oral proceedings, held on 15 September 1992, which were based on the mass balance of feed and effluent. Moreover, the molar ratio methanol/isobutylene of 1:1 to 1.1:1 was also said to be disclosed in reference (1).

The manufacture of ETBE was considered as being analogous to that of MTBE and to lack inventive step since the starting material ethanol was a homologue of methanol used in the process known from citation (1).

- VII. The Appellant requested that the impugned decision be set aside and that the patent be maintained as granted (main request) or, alternatively, on the basis of an amended Claim 1 submitted on 14 September 1990 and Claims 2 and 3 as granted (first auxiliary request) or on the basis of a single claim submitted during oral proceedings (second auxiliary request). Claim 1 of the first auxiliary request differs from Claim 1 as granted by replacing "from 1:1 to 1.25:1," by "from 1:1 to 1.1:1,". The claim of the second auxiliary request differs from Claim 1 as granted by being limited to the manufacture of ETBE and by specifying that the molar ratio of aliphatic hydrocarbon to the formed ether is from 15:1 to 30:1.

The Respondent requested that the appeal be dismissed.

VIII. At the end of the oral proceedings the Chairman announced the Board's decision to allow the Appellant's second auxiliary request.

Reasons for the Decision

1. The appeal is admissible.

2. Main request

2.1 Novelty

2.1.1 Claim 1 of the patent in suit relates to the manufacture of, inter alia, MTBE by reacting isobutylene, comprised in a C₄-hydrocarbon fraction, with methanol in the liquid phase and in the presence of an acidic catalyst, with a mandatory molar ratio of inert aliphatic hydrocarbon to the formed MTBE higher than 7:1.

Documents (1) and (1A) are corresponding patents with essentially identical disclosure. Therefore, it is sufficient to refer hereinafter only to document (1).

This citation discloses a process for the manufacture of MTBE in the liquid phase from isobutylene, comprised in a C₄-hydrocarbon fraction, and methanol in the presence of an acidic catalyst (Claim 1). The reaction temperature is 30 to 120°C, preferably 40 to 90°C at a pressure sufficient to maintain the reactants in the liquid phase (page 7, second paragraph). The molar ratio of methanol to isobutylene is usually 0.9:1 to 2:1, preferably 1.05:1 to 1.4:1 (page 7, last paragraph). No molar ratio of inert

hydrocarbon to formed MTBE is mentioned in this citation expressis verbis. However, the process of document (1) requires, after distillation of the reaction effluent, recycling of inert C₄-hydrocarbons, non-converted isobutylene, and methanol (Claim 1). The process may be performed preferably with a recycle rate R of 0.1:1 to 10:1, a recycle rate R of 0.3:1 to 3:1 being especially preferred (Claims 2, 3 and page 8, penultimate paragraph). R is defined as the ratio by weight of the recycle stream to the fraction which is discharged from the system (Claim 2).

In Examples 4 to 6 of citation (1) a C₄ cut containing 50% by weight isobutylene is reacted with methanol with varying recycle rates. In Example 6 the value of R is 1.

2.1.2 It is not in dispute between the parties that the recycle rates R may be linked to the molar ratios of inert hydrocarbon to MTBE once the compositions of the streams entering and leaving the reactor are known. Thus, the Respondent demonstrated convincingly on the basis of calculations, which were submitted during oral proceedings and which were not contested by the Appellant, that Example 6 of reference (1), if it were performed with R = 10:1, resulted in a molar ratio of inert hydrocarbons to formed MTBE of 12:1. Therefore, on the basis of these calculations, the molar ratio of the said inert compounds: formed MTBE disclosed in citation (1) and in the patent in suit overlap.

2.1.3 The Appellant submitted that the range of 0.1:1 to 10:1 given for R in reference (1) was a generic one, which could not anticipate the more specific teaching of the disputed patent. In his opinion only preferred ranges or examples amount to a novelty destroying technical disclosure. He referred to Decisions T 188/83 and T 17/85 which allegedly support this argument.

The Board does not share this opinion. According to its consistent jurisprudence a definition of an invention, which differs from the prior art only in its wording is insufficient to establish novelty; what has to be established in the examination as to novelty is whether or not the state of the art makes available the subject matter of the invention to the skilled person in the form of a technical teaching (see T 198/84, OJ EPO 1985, 209, paragraph 4, the English translation being corrected in T 124/87, OJ 1989, 491, paragraph 3.2, T 17/85, OJ EPO 1986, 406, paragraphs 7 to 7.3).

When evaluating the disclosure of a document in the course of examination as to novelty, it was set out in T 666/89 (of 10 September 1991, paragraph 5 of the Reasons for the Decision, to be published in the OJ EPO, summarised in supplement to OJ EPO 6/1992, pages 18 to 20) that "... the evaluation must therefore not be confined to a comparison of the claimed subject-matter with only the examples of the citation, but must extend to all information contained in the earlier document (T 332/87, paragraph 2.2 of 23 November 1990, not published in the OJ EPO, confirming T 424/86 paragraph 4.2 of 11 August 1988, not published in the OJ EPO)". In particular, the skilled person may combine the technical teaching of an example with the general information disclosed elsewhere in the same document, provided that the respective example is indeed representative for the general technical teaching of that document (following T 332/87, paragraph 2.2).

- 2.1.4 Applying these principles, citation (1) discloses to the skilled person, a liquid phase process for the manufacture of MTBE from methanol and isobutylene, preferably in a molar ratio of 1.05:1 to 1.4:1, in the presence of an

acidic catalyst and inert C₄-hydrocarbons at preferred temperatures of 40 to 90°C and a pressure sufficient to maintain the reaction mixture in the liquid phase. No reason can be seen which would prevent the skilled person from combining the specific embodiment of Example 6 with the teaching of Claim 2. On the contrary, this example demonstrates the feasibility of the technical teaching of document (1) throughout the range given in this claim and, therefore, this document makes available to the skilled person the technical teaching that the molar ratio of inert hydrocarbons to formed MTBE can be as high as at least 12:1. Thus, it follows that the process for manufacturing MTBE as claimed in Claim 1 of the main request is anticipated by document (1).

- 2.1.5 With respect to Decision T 188/83, the Appellant overlooks that the Board's decision with regard to lack of novelty in that case was based on the finding that processes corresponding in all essential technical features to those of the subject-matter claimed in the application were already state of the art. In particular, previously described examples gave specific numerical values for the molar ratio of acetic acid : oxygen which were scattered within the claimed range according to the application (paragraph 4 of the Reasons). It was explained in paragraph 5 that the anticipation resulted not only from the individual values, which could be calculated from the examples, but also from the fact that those values - as in the present case - had to be considered in the light of the broader technical teaching disclosed therein. Nothing can be gained from this in favour of the Appellant. The same holds true for the Appellant's reference to the Headnote I of that decision reading "If for the purpose of a chemical production process previously described a certain ratio of reactants, defined in terms of a range, is chosen, the said ratio being covered by the

conventional teaching but not mentioned in it, this may involve a new invention." The term "may" indicates that this question has to be decided on the merits of each individual case.

Overlapping numerical ranges were dealt with in Decision T 17/85 (OJ EPO 1986, 406). In that case the novelty of a claimed numerical range of 4 to 8 had to be considered in respect to a previously disclosed preferred numerical range of 6.67 to 31.6. The Board's conclusions were summarised in paragraph 7.4 of the Reasons as follows. "If the preferred numerical range in a citation in part anticipates a range claimed in an application, the said claimed range cannot be regarded as novel at least in cases where the values in the examples given in the citation lie just outside the claimed range and teach the skilled person that it is possible to use the whole of its range." This makes it quite clear that the anticipatory character of a known numerical range overlapping with a claimed numerical range is not necessarily confined to a situation where examples are disclosed in the state of the art which are "close" to the claimed range. The decisive question to be answered is, whether or not the anticipation discloses to the skilled person that he could indeed apply the known technical teaching in the range now claimed. In this context the existence of "close" examples may provide additional evidence to answer this question.

Document (1) teaches, as explained in the above paragraph 2.1.4, that the process disclosed there can also be performed with a molar ratio of inert hydrocarbons to formed MTBE of at least 12:1, i.e. higher than 7:1 and, thus, in the range now claimed, which, therefore, is not novel. Hence, the Board's finding in the present case is in agreement with Decisions T 188/83 and T 17/85.

3. First auxiliary request

3.1 Novelty

Claim 1 of the first auxiliary request differs from Claim 1 as granted only by requiring a molar ratio of the alcohol to isobutylene of from 1:1 to 1.1:1 instead of from 1:1 to 1.25:1. This restricted range overlaps with the respective preferred range disclosed in reference (1) - see the above paragraph 2.1.1 - the lower limit of which is exactly in the middle of the range now claimed. Therefore, this feature cannot distinguish the process of Claim 1 of the first auxiliary request from that of reference (1) and, hence, the subject-matter of this claim lacks novelty.

4. Second auxiliary request

4.1 Novelty

The only claim of this request relates to the manufacture of ETBE. None of the citations, which are before the Board, discloses a process for the manufacture of ETBE. Thus, the claimed subject-matter is novel.

4.2 Technical problem and solution

In this situation and having regard to the introduction to the present patent specification, according to which MTBE and ETBE are products of great commercial interest due to their anti-knock properties (cf. column 1, lines 9 to 15), the Board is satisfied that the embodiments performed in the Examples 5, 6, and 7 of the disputed patent are representative for the manufacture of ETBE according to the state of the art and accepts as the starting point for

evaluating inventive step the technical problem as it can be deduced from the patent in suit. According to column 2, lines 25 to 54 the problem was to provide a process for the preparation of, inter alia, ETBE with an improved isobutylene conversion resulting in an isobutylene content in the exhaust C₄-hydrocarbon fraction of about 1% by volume or less.

Examples 10, 13 and 14, which have molar ratios of inert hydrocarbon to the ETBE of about 26.2:1, 15.7:1, and 22.9:1, respectively, show an isobutylene content in the exhaust gas of about 1, 1, and 0.8% by volume and isobutylene conversion rates of 96, 98.8, and 99% respectively (the patent in suit, columns 14, 15, and 18 to 21). In contrast, Examples 5, 6, and 7, designated as comparative examples, which were performed at molar ratios of inert hydrocarbon to ETBE of 3.8:1, 4.3:1, and 5.6:1 (compare also Table I enclosed to the Appellants submission of 2 September 1990 to the Opposition Division) show higher isobutylene concentrations in the exhaust gas (5.6, 5, and 4% by volume) and lower isobutylene conversion rates (76, 79, and 83.3%; cf. the disputed patent columns 9 to 12). Hence, the Board is satisfied that the above problem is plausibly solved.

4.3 Inventive Step

The Respondent's sole argument, that it would have been obvious for the skilled person to apply the MTBE manufacturing process known from citation (1) to the manufacture of ETBE because these compounds and the respective reactants methanol and ethanol are (adjacent) members of homologous series of compounds fails for the following reason. An attack on inventive step of a chemical process based on the argument of "homology" of starting materials amounts in fact to an assertion that it

is known to the skilled person that there is no essential difference in the reactivities of these homologues in respect to the process concerned and that, consequently, the skilled person could have reasonably expected an essentially identical performance of the homologues in the said process. However, no document was cited and no common general knowledge was put forward by the Respondent to support this allegation.

On the contrary, the Appellant maintained that the chemical equilibrium of the ETBE formation from ethanol and isobutylene - which is obviously important for the claimed process - is less favourable than that of the MTBE formation (see the patent in suit, column 1, lines 59 to 62). This was contested by the Respondent. However, he did not offer any supporting evidence and, thus, the Board disregards this objection, applying the principles laid down in T 219/83, in particular paragraph 12 of the Reasons for the Decision (OJ EPO, 1986, 211).

Therefore, it was not established by the Respondent, who has the burden of proof, that in the present case "homology" means that the skilled person would have known or expected the homologue ethanol to behave analogously to methanol in the specific ether formation known from citation (1). Thus, the Respondent's argument that the claimed process does not result from an inventive step, simply because the starting material ethanol is a homologue of methanol used as starting material in the said known process is rejected by the Board.

Furthermore, the lower limit of the range of the molar ratio of inert compounds to formed ETBE according to the claim is well above the value of 12:1 as deduced for the MTBE-process of document (1); see the above paragraph 2.1.2. There was no indication in the state of

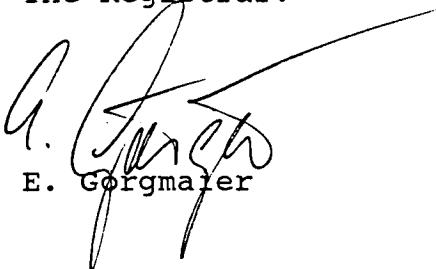
the art that the present technical problem would be solved by maintaining the molar ratio of inert compounds to formed ETBE between 15:1 and 30:1 during the ether formation. Hence, the Board concludes that the subject-matter of the present claim is inventive.

Order

For these reasons, it is decided that:

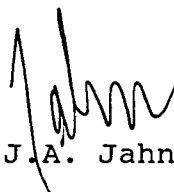
1. The decision under appeal is set aside.
2. The case is remitted to the first instance with the order to maintain the patent on the basis of the claim as submitted during the oral proceedings (second auxiliary request).

The Registrar:



E. Gorgmaier

The Chairman:



K.J.A. Jahn