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D E C I S I O N
of 1 June 2006

Case Number: T 0175/04 - 3.3.10

Application Number: 97944611.9

Publication Number: 0920404

IPC: C07C 15/073

Language of the proceedings: EN

Title of invention:

A zeolite-based ethylbenzene process adaptable to an aluminium chloride-based ethylbenzene plant

Patentee:

THE DOW CHEMICAL COMPANY

Opponent:

ExxonMobil Chemical Patents Inc.
UOP LLC

Headword:

Zeolite-based ethylbenzene process/DOW

Relevant legal provisions:

EPC Art. 56

Keyword:

"Inventive step (yes) - non-obvious solution"

Decisions cited:

-

Catchword:

-



Case Number: T 0175/04 - 3.3.10

D E C I S I O N
of the Technical Board of Appeal 3.3.10
of 1 June 2006

Appellant: ExxonMobil Chemical Patents Inc.
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 4 December 2003
rejecting the opposition filed against European
patent No. 0920404 pursuant to Article 102(2)
EPC.

Composition of the Board:

Chairman: R. Freimuth
Members: J. Mercey
P. Schmitz

Summary of Facts and Submissions

I. The Appellant (Opponent I) lodged an appeal on 26 January 2004 against the decision of the Opposition Division dated 4 December 2003 rejecting the opposition against European patent No. 920 404 which was granted on the basis of nineteen claims, and on 6 April 2004 filed a written statement setting out the grounds of appeal. Claim 1 of the granted patent read as follows:

"1. A zeolite-based process for producing ethylbenzene which is suitable for retrofitting an aluminum chloride-based ethylbenzene plant comprising:

(A) contacting benzene with ethylene in at least a partial liquid phase in an alkylation reactor at a total benzene/ethylene molar ratio between 1.5:1 and 3.0:1 in the presence of a catalytic amount of an alkylation catalyst selected from acidic zeolites beta, Y, MCM-22, MCM-36, MCM-49, and MCM-56, the alkylation process conditions being sufficient to prepare an alkylation product mixture containing benzene, ethylbenzene, polyethylbenzenes including diethylbenzenes, and higher molecular weight residues, the diethylbenzenes/ethylbenzene weight ratio ranging from 1:2.5 to 1:8.0;

(B) passing the alkylation product mixture into a distillation train designed for an aluminum-chloride based ethylbenzene plant wherein benzene is recovered in a first distillation column; thereafter passing the bottoms from the first distillation column comprising ethylbenzene, polyethylbenzenes, and higher molecular weight residues into a second distillation column to recover ethylbenzene; and thereafter passing the bottoms from the second distillation column comprising

polyethylbenzenes and higher molecular weight residues into a third distillation column to recover polyethylbenzenes including diethylbenzenes;

(C) passing at least a portion of the polyethylbenzenes recovered from the third distillation column into a transalkylator wherein the polyethylbenzenes are contacted with benzene in the liquid phase at a molar ratio of total moles of benzenes in the benzene and polyethylbenzenes to total moles of ethyl groups on the polyethylbenzenes ranging from 1.5:1 to 3.0:1 in the presence of a catalytic amount of a transalkylation catalyst selected from acid zeolites mordenite, beta, and Y, the contacting being conducted under reaction conditions sufficient to produce a transalkylation product mixture containing benzene, ethylbenzene, and polyethylbenzenes including diethylbenzenes, the diethylbenzenes/ethylbenzene weight ratio ranging from 1:2.5 to 1:8.0; and

(D) passing the transalkylation product mixture into the aforementioned distillation train wherein the transalkylation product mixture is separated in the three distillation columns mentioned hereinbefore to recover benzene, ethylbenzene, and polyethylbenzenes."

II. Notice of Opposition had been filed by the Appellant and the Party as of right (Opponent II) requesting revocation of the patent as granted in its entirety on the grounds of lack of novelty and lack of inventive step (Article 100(a) EPC) and insufficient disclosure (Article 100(b) EPC). *Inter alia* the following documents were submitted in opposition proceedings:

(3) EP-A-485 683 and

- (5) B. Maerz et al., "EBMaxSM: Leading edge ethylbenzene technology from Mobil/Badger", Presented to the 21st Annual 1996 Dewitt Petrochemical Review, Houston, Texas, March 19-21, 1996, pages A-1 to A-10.

- III. The Opposition Division held that the invention was sufficiently disclosed, was novel and involved an inventive step. The claimed subject-matter was novel, since the prior art did not disclose the claimed benzene group to ethyl group molar ratio or the diethylbenzenes/ethylbenzene weight ratio for the transalkylation step, or the particular claimed catalyst for that step. Thus there was no direct and unambiguous disclosure in the state of the art of all the features of the claimed process in combination. With regard to inventive step, document (5), which was concerned with revamping existing ethylbenzene plants with the zeolite catalyst MCM-22, was considered to represent the closest prior art. There was, however, no reference in this, or in any other cited prior art document, to adapting the reaction conditions in such a way that the product streams of the alkylation and transalkylation steps should be close to the product streams of the aluminium chloride catalysed process.
- IV. The Appellant argued that the claimed process was not inventive. With respect to document (3), the Appellant argued that the skilled person, faced with the problem of adapting a zeolite-based ethylbenzene process to an aluminium chloride-based ethylbenzene plant, would know that he could not use the reactor of the aluminium chloride plant, since the zeolite process was heterogeneous and not homogeneous. He would also know

that the neutralisation and catalyst disposal section of the aluminium chloride plant was not necessary. Thus the only part of the aluminium chloride plant which could possibly be used was the distillation train, since the zeolite-based process produced similar products to the aluminium chloride-based process. The Appellant argued that it was well within the skilled person's common general knowledge that in order to modify the zeolite-based process of document (3) in such a way that the distillation train of an aluminium chloride-based ethylbenzene plant could be used to separate the products thereof, the product streams from the alkylation/transalkylation steps should be matched with the product streams obtained in an aluminium chloride-based process. Thus the claimed process consisted merely of choosing process conditions from within those disclosed in document (3) in order to produce product streams matching those produced by an aluminium chloride-based process, this being a trivial exercise for the skilled person, not requiring inventive ingenuity.

The Appellant also argued that the claimed process was not inventive over the disclosure of document (3) in combination with that of document (27):

- (27) SRI International, Process Economics Program, Report No. 33B, Styrene, Supplement B, H. W. Scheeline et al., December 1977, pages 27 to 49 and Figure 4.1.

This latter document disclosed an aluminium chloride-based process for the production of ethylbenzene from benzene and ethylene by a two step

alkylation/transalkylation reaction, wherein the product mixtures from each step were separated in a common distillation train comprising three distillation columns (cf. Figure 4.1). It was argued that since document (27) taught the compositions of the feed streams (cf. Table 4.2), and the compositions of the product stream (cf. Table 4.4, Stream 24) which were separated by said distillation train, the skilled person, wishing to adapt the zeolite-based process of document (3) to an aluminium chloride-based ethylbenzene plant would know that in order to use the distillation train thereof, feed and product streams having compositions as disclosed in document (27) should be used.

The same conclusion in respect of obviousness arose when starting from document (27), the process of which had the most features in common with the claimed process. In view of this prior art, the problem to be solved by the invention was to retrofit an aluminium chloride-based ethylbenzene plant with a zeolite catalyst. It was argued that the only difference between the claimed process and a conventional process for producing ethylbenzene using an aluminium chloride catalyst was in fact the use of the zeolite catalyst as specified in claim 1, no other modifications having to be made to the pre-existing aluminium chloride-based process, including the liquid phase conditions, the benzene to ethylene mole ratio and the diethylbenzene to ethylbenzene weight ratio.

- V. The Respondent (Proprietor of the patent) submitted that the process was inventive, and that starting from document (3), the problem to be solved comprised

adapting a zeolite-based process to an aluminium chloride-based ethylbenzene plant. It was argued that neither document (3), nor any of the other cited prior art documents, taught the skilled person to match the product streams of the zeolite-based process to those of the aluminium chloride-based process in order to be able to use the distillation train of an aluminium chloride-based ethylbenzene plant.

With regard to document (27), although this document taught feed and product streams used in an aluminium chloride-based plant, the compositions of which fell within those defined in claim 1, this document did not address the technical problem underlying the patent in suit of adapting a zeolite-based ethylbenzene process to an aluminium chloride-based ethylbenzene plant, such that the skilled person would not have turned to this document when looking for a solution to the problem underlying the invention. Hence the Appellant's arguments based on document (27), when taken either alone or in combination with document (3), were based on an *ex post facto* analysis. Document (5) was not concerned with revamping aluminium chloride-based plants but rather zeolite-based plants, such that said document also did not address the problem underlying the invention and thus could not be combined with document (3).

VI. The Party as of right made no submissions as to the substance of the appeal, nor did it file any requests.

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

The Respondent requested that the appeal be dismissed.

VIII. Oral proceedings were held on 1 June 2006 in the absence of the Party as of right, who, after having been duly summoned, informed the Board with a letter dated 3 March 2006 that it would not attend. At the end of the oral proceedings, the decision of the Board was announced.

Reasons for the Decision

1. The appeal is admissible.
2. *Sufficiency of Disclosure and Novelty*

The appealed decision found the invention to be sufficiently disclosed and the subject-matter of the claims to be novel (cf. point III supra). Sufficiency of disclosure and novelty were no longer contested during the appeal proceedings, nor does the Board see any reason to take a different view to the Opposition Division. Hence, it is unnecessary to go into more detail in this respect.

3. *Inventive step*

Thus, the sole issue arising from this appeal consists in deciding whether or not the subject-matter of the claims of the patent in suit involves an inventive step.

- 3.1 According to the established jurisprudence of the Boards of Appeal it is necessary, in order to assess inventive step, to establish the closest state of the

art, to determine in the light thereof the technical problem which the invention addresses and successfully solves, and to examine the obviousness of the claimed solution to this problem in view of the state of the art. This "problem-solution approach" ensures assessing inventive step on an objective basis and avoids an *ex post facto* analysis.

- 3.2 The patent in suit is directed to a zeolite-based process for producing ethylbenzene which is suitable for retrofitting an aluminium chloride-based ethylbenzene plant.

A similar process, namely one which prepares the same products employing the same starting materials and catalysts by a two step alkylation/transalkylation reaction, wherein the product mixtures from each step are separated in a common distillation train comprising three distillation columns, already belongs to the state of the art, namely to the disclosure of document (3). More particularly, this document discloses (cf. Figure 7 and page 11, line 50 to page 12, line 30) a process wherein benzene is contacted with ethylene in an alkylation reactor 56 under sufficient pressure to maintain the benzene in the liquid phase and at least 2 mole percent of the ethylene solubilised in the benzene, in the presence of a catalyst which is either zeolite beta, zeolite Y or zeolite omega, to produce an alkylation product mixture containing benzene, ethylbenzene and polyethylbenzenes. The alkylation product mixture is passed into a distillation train comprising a benzene distillation column 61 wherein benzene is recovered via line 62; the bottoms fraction from this column containing ethylbenzene and

polyethylbenzenes and heavier aromatics is passed to an ethylbenzene distillation column 65 wherein ethylbenzene is recovered. The bottoms fraction from the ethylbenzene column containing polyethylbenzene and heavier aromatics is passed into a polyethylbenzene distillation column 68 wherein *inter alia* polyethylbenzenes are recovered *via* line 70. The polyethylbenzenes recovered from the polyethylbenzene column are supplied to the transalkylation reactor 72 *via* line 70 where they are contacted with benzene (*via* line 73) at a molar ratio of benzene to polyethylbenzene preferably within the ratio of 1:1 to 4:1 in the presence of a catalyst which is either zeolite beta, zeolite Y or zeolite omega to produce a transalkylation product mixture containing benzene, ethylbenzene and polyethylbenzenes. The transalkylation product mixture is passed *via* line 75 into the aforementioned distillation train wherein said product is separated in the three distillation columns to recover benzene, ethylbenzene and polyethylbenzenes. A benzene to ethylene mole ratio of 2:1 is disclosed at page 20, line 50, zeolite beta is specifically described as the alkylation catalyst at page 6, lines 46 to 47 and page 16, lines 53 to 55, and diethylbenzenes:ethylbenzene weight ratios in the product mixture of the alkylation step of from 3.64 to 6.32 are disclosed in Table IV on pages 18 and 19 (cf. Samples 1 to 16 and 18 to 24). That the transalkylation is carried out in the liquid phase is disclosed at page 6, lines 9, 19 and 30-32, and zeolite Y is described as the preferred transalkylation catalyst at page 6, lines 52 to 53.

Thus, the Board considers that the disclosure of document (3) specified above represents the closest state of the art, and, hence, the starting point in the assessment of inventive step.

- 3.3 The Appellant, while not disputing the above finding, addressed also document (27) as representing the closest prior art, said document being directed to an aluminium chloride-based process for preparing ethylbenzene. However, although document (27) describes the preparation of the same products from the same starting materials as the patent in suit, employing a two step alkylation/transalkylation reaction, wherein the product mixtures from each step are separated in a common distillation train comprising three distillation columns, the reaction is of a different type, namely it is a homogeneous Friedel-Crafts alkylation using an aluminium chloride catalyst, and not a heterogeneous catalysed alkylation using an acidic zeolite. For this reason the process disclosed in document (27) is further away from the claimed invention than document (3) addressed in point 3.2 above.

The Respondent, while also not disputing the finding that document (3) represents the closest prior art, addressed additionally document (5) as representing the closest prior art, said document being directed to a zeolite-based process for preparing ethylbenzene. However, in the process of document (5), the transalkylation step is carried out in the vapour phase (cf. page A-5, first line under equation) and the transalkylation catalyst is a ZSM-5 variant (cf. page A-7, lines 4 to 5), whereas in the claimed invention said step is carried out in the liquid phase

and the transalkylation catalysts is a different acid zeolite. For these reasons the process disclosed in document (5) is also further away from the claimed invention than document (3).

- 3.4 The drawbacks of the process of D3 *vis-à-vis* the aluminium chloride-based ethylbenzene process lie in the necessity to construct a grass-roots plant which customises the benzene alkylation, transalkylation, and product recovery stages to the activity of the zeolite employed (cf. patent specification, page 2, lines 49 to 52).

Thus, the technical problem underlying the patent in suit consists in providing a zeolite-based ethylbenzene process adapted for an aluminium chloride-based ethylbenzene plant (cf. patent specification, page 2, lines 53 to 57).

- 3.5 As the solution to this problem, the patent in suit proposes the process according to claim 1 which is characterised by the particular benzene group to ethyl group molar ratio in the feed to, and the particular diethylbenzenes/ethylbenzene weight ratio in the product of, the transalkylation step, and by the combination of the other particular process features indicated in that claim.

- 3.6 The evidence in the patent in suit (cf. Tables I and III) convincingly demonstrates that this proposed solution successfully solves the problem underlying the invention. More particularly, the Examples and Comparative Experiments show that by using feed streams as defined in claim 1, it is possible using zeolite

catalysts to produce product streams which are similar to those produced by an aluminium chloride-based process, most particularly with respect to the diethylbenzenes/ethylbenzene weight ratio. The Appellant never disputed that the claimed process was successfully adapted for an aluminium chloride-based ethylbenzene plant and the Board is not aware of any reason for challenging this finding.

3.7 Finally, it remains to decide whether or not the proposed solution to that objective problem underlying the patent in suit is obvious in view of the state of the art.

3.7.1 The closest prior art document (3) teaches a zeolite based ethylbenzene process. It does not give any incentive to modify that process such that the product streams from both the alkylation and transalkylation steps match those of an aluminium chloride-based process. However, the similarity of the product streams of the zeolite-based process to those of the aluminium chloride-based process is critical in order to solve the problem, since only if particular feed stream compositions are used, and product streams from the alkylation and transalkylation reactors having particular diethylbenzenes/ethylbenzene weight ratios as defined in claim 1 are produced, can the distillation train of an aluminium chloride-based plant be operated successfully. This fact was not disputed by the Appellant. Thus selecting in combination the particular process features as defined in claim 1 in order to arrive at similar product streams as produced by an aluminium chloride-based process, is a purposive selection, since only such product streams may be

optimally separated by the distillation train of an aluminium chloride-based plant. Hence, document (3) on its own does not render obvious the solution proposed by the claimed invention.

- 3.7.2 None of the documents cited in the appeal proceedings addresses the technical problem underlying the patent in suit of adapting a zeolite-based ethylbenzene process to an aluminium chloride-based ethylbenzene plant, such that the skilled person would not have taken the teaching of any of these documents into consideration when looking for a solution to the problem underlying the invention.
- 3.7.3 Though document (5) is directed to revamping "existing Mobil/Badger ethylbenzene units" (cf. page A-9, penultimate paragraph), these Mobil/Badger units already used zeolite technology (cf. page A-2, fourth paragraph). Thus document (5) is concerned with revamping existing **zeolite**-based plants with newer zeolite catalysts and does not address the technical problem underlying the patent in suit of adapting a zeolite-based ethylbenzene process to an aluminium chloride-based ethylbenzene plant. For this simple reason, document (5) cannot give any hint as to how to solve that technical problem, since a skilled person would not take the teaching of that document into consideration when looking for a solution to the problem underlying the invention. In any case, the skilled person would not arrive at the process according to claim 1 when combining the teaching of document (5) with that of document (3), since document (5) contains no teaching regarding the matching of product streams when revamping ethylbenzene plants.

3.7.4 The Board is not convinced by the Appellant's objection of obviousness based on a combination of documents (3) and (27), since document (27) also does not address the technical problem underlying the patent in suit, such that it also cannot give any hint as to how to solve that technical problem. Indeed, document (27) does not even mention zeolite catalysts. Hence the skilled person would ignore document (27) when seeking a solution to the problem underlying the invention.

3.7.5 The Appellant further alleged that when seeking to modify the zeolite-based process of document (3) in such a way that it would be adapted for use in an aluminium chloride-based ethylbenzene plant, the skilled person, knowing that the distillation train was the only part of such an aluminium chloride-based ethylbenzene plant at all suitable for use in a zeolite-based process, would be motivated by economic considerations to operate said distillation train optimally. It was within the skilled person's common general knowledge that in order to achieve this aim, the product streams should be matched with those obtained in an aluminium chloride-based process. Thus the claimed process consisted merely of choosing process conditions from within those disclosed in document (3) in order to produce product streams matching those produced by an aluminium chloride-based process. That the product stream from the transalkylation step in document (3) was similar to the product stream from the alkylation step was apparent from the fact that both streams are fed to the same distillation column 61.

However, document (3) contains no teaching as to how to adapt the process described therein to an aluminium chloride-based plant and is completely silent in this respect. The Appellant did not indicate any further prior art which teaches that the product streams of the zeolite-based process should be matched with those obtained in an aluminium chloride-based process in order to be able to use any part of the aluminium chloride plant, let alone that this matching should involve the selection of the particular combination of feed stream compositions and product stream compositions from the alkylation and transalkylation reactors having the particular diethylbenzenes/ethylbenzene weight ratios claimed, such that the distillation train of such an aluminium chloride plant may be used for the separation of the products of the zeolite-based process. In the absence of any corroborating evidence, the Appellant's argument that the matching of product streams was within the skilled person's common general knowledge represents merely an unsubstantiated allegation. It rather appears that the Appellant's view is based on hindsight with knowledge of the present invention, such that the Appellant's obviousness objection based on document (3) alone, or in combination with common general knowledge, is devoid of merit.

3.8 To summarise, in the Board's judgement, none of the documents cited above renders the claimed invention obvious, either taken alone or in combination.

3.9 For these reasons, the Board concludes that the subject-matter of claim 1, and by the same token that of dependent claims 2 to 18, and of independent

claim 19, which includes all the features of claim 1, involves an inventive step within the meaning of Articles 52(1) and 56 EPC.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

G. Nachtigall

R. Freimuth