

A		B		C	X
---	--	---	--	---	---

File No.: T 146/92 - 3.3.1  
Application No.: 84 307 092.1  
Publication No.: 0 138 617  
Classification: C07C 4/18  
Title of invention: Processes for the hydrodealkylation and/or  
isomerization of alkylaromatic hydrocarbons

**D E C I S I O N**  
of 23 June 1993

Applicant: -  
Proprietor of the patent: Amoco Corporation  
Opponent: Mobil Oil Corporation

Headword: Hydrodealkylation/AMOCO  
EPC: Art. 54 and 56  
Keyword: "Novelty (yes) of second auxiliary request" - "Inventive step  
(yes)"

**Headnote**  
**Catchwords**



Case Number: T 146/92 - 3.3.1

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.1  
of 23 June 1993

**Appellant:**  
(Proprietor of the patent) Amoco Corporation  
200 East Randolph Drive  
Chicago  
Illinois 60601 (US)

**Representative:**  
Ritter, Stephen David  
Mathys & Squire  
10 Fleet Street  
London EC4Y 1AY (GB)

**Respondent:**  
(Opponent) Mobil Oil Corporation  
150 East 42nd Street  
New York, N.Y. 10017 (US)

**Representative:**  
Cooper, John Anthony  
Carpmaels & Ransford  
43 Bloomsbury Square  
London WC1A 2RA (GB)

**Decision under appeal:** Decision of the Opposition Division of the  
European Patent Office of 12 November 1991, with  
written reasons issued on 18 December 1991,  
revoking European patent No. 0 138 617 pursuant to  
Article 102(1) EPC.

**Composition of the Board:**

**Chairman:** K.J.A. Jahn  
**Members:** R.W. Andrews  
J.A. Stephens-Ofner

## Summary of Facts and Submissions

- I. European patent No. 0 138 617 in respect of European patent application No. 84 307 092.1, which was filed on 16 October 1984, was granted on 25 January 1989 (cf. Bulletin 89/04).
- II. A notice of opposition, which was filed on 6 October 1989, requested the revocation of the patent on the grounds set forth in Article 100(a) EPC. The opposition was supported, *inter alia*, by the following documents
- (1) EP-A-0 055 529
  - (3) US-A-4 152 363
  - (6) GB-A-2 033 358
  - (7) GB-A-1 444 481.
- III. By a decision delivered orally on 12 November 1991, with written reasons being issued on 18 December 1991, the Opposition Division revoked the patent on the basis that the claimed subject-matter lacked novelty in the light of the disclosure of document (3) when read in conjunction with that of document (6). The Opposition Division also observed that, even if the use of molybdenum or tungsten were regarded as being a selection from the metals listed in document (6), the claimed subject-matter would not involve an inventive step.
- IV. An appeal was lodged against this decision on 12 February 1992 with payment of the prescribed fee. In his statement of grounds of appeal filed on 24 April 1992 and during the oral proceedings held on 23 June 1993, the Appellant contended that the decision under appeal was erroneous insofar as the conclusion that the claimed subject-matter lacked novelty was based on the combined disclosures of documents (3) and (6). With

respect to documents (3) and (6) considered separately, the Appellant argued that there was no specific disclosure of a process with the three features called for in Claim 1 as granted. Moreover, from the formula and process of preparation it would appear that the entity  $M_{2x}O$  forms part of the lattice of the zeolite structure and, therefore, cannot be considered to have been placed on the zeolite. Furthermore, in view of the disclosure of column 10, lines 22 to 60 of document (3), the present process did not follow inevitably from this document.

With respect to inventive step, the Appellant contended that the finding that alkylaromatic compounds may be converted by hydrodealkylation at moderate temperatures ran quite counter to the teaching of document (3) and that there was nothing in the cited prior art which could possibly lead the skilled person to the idea of using the present catalyst.

- V. The Respondent maintained that the subject-matter of Claim 1 of the main and first auxiliary requests lacked novelty in the light of the disclosure of document (3). In his facsimile of 9 June 1993, the Respondent also alleged that the subject-matter of at least Claims 1 to 4, 7 to 10 and 12 lacked novelty in the light of US-A-4 400 573 (document (9)) when read in conjunction with US-A-4 269 813 (document (10)).

The Respondent maintained that, in view of the problem to be solved by the disputed patent, it was impossible to maintain Claims 1 to 7 of the main requests and that the main claims of the second and third auxiliary requests were unallowable in view of the passage on page 3, lines 27 to 38 of the printed patent specification.

The Respondent also contended that the subject-matter claimed in accordance with the second and third auxiliary requests did not involve an inventive step with respect to the disclosure of documents (3), (6) and (7).

VI. The Appellant requested that the decision under appeal be set aside and that the patent be maintained as granted or on the basis of Claims 1 to 11 submitted in the course of oral proceedings (auxiliary request No. 1) or on the basis of Claims 1 to 11 (auxiliary request No. 2) or on the basis of Claims 1 to 11 (auxiliary request No. 3), likewise submitted in the course of oral proceedings.

Claim 1 as granted reads as follows:

"A process for converting alkyl aromatic hydrocarbons by hydrodealkylation which comprises contacting an alkyl aromatic hydrocarbon feed under conversion conditions with a catalyst, characterised in that the catalyst comprises an intermediate pore crystalline aluminosilicate zeolite-based catalyst composition having an internal pore diameter of less than 6.0 Angstroms but greater than 4.5 Angstroms incorporated within a matrix material and on which has been placed a molybdenum compound or a tungsten compound."

Claim 1 according to the first auxiliary request reads as follows:

"A process for converting alkyl aromatic hydrocarbons at a temperature of from 95°C to 800°F (425°C) by hydrodealkylation which comprises contacting an alkyl aromatic hydrocarbon feed containing xylenes and ethylbenzene under conversion conditions with a catalyst, whereby to isomerise xylenes and convert

ethylbenzene substantially by hydrodeethylation, characterised in that the catalyst comprises an intermediate pore crystalline aluminosilicate zeolite-based catalyst composition having an internal pore diameter of less than 6.0 Angstroms but greater than 4.5 Angstroms incorporated within a matrix material and on which has been placed a molybdenum compound or a tungsten compound."

Claim 1 of the second auxiliary request differs from that of the first auxiliary request in that the reaction temperature is from 95°C to 700°F (371°C).

In Claim 1 according to the third auxiliary request the temperature range is from 95°C to 319°C.

The Respondent requested that the appeal be dismissed.

VII. At the conclusion of the oral proceedings, the Board's decision to maintain the patent on the basis of the second auxiliary request was announced.

### Reasons for the Decision

1. The appeal is admissible.
2. The Board has no objections to any of the versions of the claims. Claim 1 of the first, second or third auxiliary request represents a combination of granted Claims 1 and 8 and page 2, lines 4 to 7 (cf. Claims 1, 6 and 7 and page 1, lines 7 to 12 of the application as filed). The lower temperature limit is disclosed in Claim 12 as filed and granted. The upper temperature limits of the first and second auxiliary requests are disclosed in document (3) and are intended to overcome a novelty objection vis-à-vis the disclosure of this

document. The upper temperature limit of the third auxiliary request corresponds to the reaction temperature of Example 1 of the disputed patent.

The dependent claims of the auxiliary request correspond to Claims 2 to 7 and 9 to 12 as granted (cf. also Claims 2 to 6 and 8 to 12 as filed).

- 2.1 The passage on page 3, lines 27 to 38 relied on by the Respondent to support his allegation that Claim 1 of each of the three auxiliary requests is unallowable, discloses operating conditions for the isomerisation of xylene mixtures with concurrent conversion of ethylbenzene by hydrodeethylation. By way of guidance, three temperature ranges with the corresponding ranges for hydrogen to hydrocarbon ratios, weight hourly space velocities and pressure are disclosed as appropriate conversion conditions are broadly disclosed.

Clearly, these four parameters are to some extent interrelated and the course of the reaction is influenced to some extent by the combination of these parameters used during the reaction. However, in the Board's judgment, the requirement of Claim 1 that the process is carried out under conversion conditions to isomerise xylenes and convert ethylbenzene substantially by hydrodeethylation takes into account the interrelationship between these four parameters in a manner which would be understood by the skilled reader in the light of the cited passage.

3. The first question to be decided is whether the subject-matter claimed according to the various requests is novel having regard to the disclosure of document (3).

- 3.1 In accordance with the established jurisprudence of the Boards of Appeal (cf. Decision T 124/87

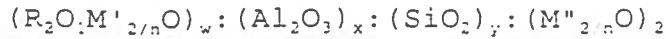
"Copolymer/DUPONT", OJ EPO 1989, 491, paragraph 3.2; T 12/81 "Diastereomers, OJ EPO 1982, 262, paragraph 5; T 198/84 "Thiochloroformates", OJ EPO 1985, 209, paragraph 4; T 666/89 "Washing Composition/UNILEVER, Headnote published OJ EPO 1992/06 and T 12/90 of 23 August 1990, reported in 1991, EPOR 312), in order to answer this question it is necessary to consider whether the disclosure of document (3) is such as to make the process claimed in accordance with the various requests available to the skilled person in the form of a technical teaching. Therefore, it is necessary to determine the nature and the extent of the information actually imparted to the skilled person by this document as distinct from its literal disclosure.

3.2 Document (3) discloses a process for isomerising a monocyclic methyl-substituted aromatic hydrocarbons, for example, xylenes, using a catalyst comprising from about 0.1 to about 5% by weight of a crystalline aluminosilicate zeolite, such as ZSM-5, which may be combined with a support or binder material which acts as a diluent, at a temperature in the range of about 800°F to about 1 000°F (cf. Claims 1, 2 and 17 and column 9, lines 17 to 26). According to this document, at these temperatures and using the catalyst disclosed therein ethylbenzene present in the feedstock reacts primarily via hydrodeethylation to yield benzene and ethane (cf. column 10, lines 51 to 54). Since ZSM-5 aluminosilicate zeolite is suitable for use in the process of the disputed patent (cf. Claim 10 as granted), document (3) discloses the use of an intermediate pore crystalline aluminosilicate zeolite based catalyst composition having an internal pore diameter of less than 6.0 Angstroms but greater than 4.5 Angstroms.

According to the paragraph bridging columns 4 and 5 of document (3), a crystalline aluminosilicate zeolite, the



X-ray diffraction pattern of which shows substantially the significant lines for zeolite ZSM-5, is identified, in terms of mole ratios of oxides and in the anhydrous state, as follows



wherein  $w/x$  is from greater than 0.5 to less than 3;  $y/x$  is greater than 20;  $z/x$  is from greater than zero to less than about 100, R is a nitrogen-containing cation, n is the valence of M' or M'', M' is a metal from Group 1A of the Periodic Table, ammonium, hydrogen or mixtures thereof, and M'' is a selected from rare earth metals, chromium, vanadium, molybdenum, indium, boron, mercury, tellurium, silver and one of the platinum group metals.

In the course of oral proceedings, the Appellant admitted that the expression "on which has been placed a molybdenum compound" could not serve to distinguish the catalyst used in the process of the disputed patent from the above catalyst in which the symbol M'' represents molybdenum.

Having regard to the above-mentioned jurisprudence, the Board finds that document (3) makes available to the public in the form of a technical teaching a process falling within the terms of Claim 1 as granted. Therefore, the Appellant's main request must be refused.

- 3.3 The process claimed in Claim 1 of the first auxiliary request differs from that of the granted Claim 1 in that it is specified that the hydrocarbon feed contains xylenes and ethylbenzene, the reaction is carried out at a temperature of from 95°C to 800°F under conversion conditions such that the xylenes are isomerised and the ethylbenzene is substantially converted by hydrodeethylation.

However, according to column 10, lines 45 to 60 at temperatures of from greater than about 800°F to about 1 000°F with the above-defined dilute catalyst composition not only are the xylenes in the feedstock isomerised but ethylbenzene reacts primarily via dealkylation to benzene and ethane rather than via disproportionation to benzene and diethylbenzene.

The decision with respect to the novelty of the subject-matter claimed according to this request depends upon the true meaning of the phrase "greater than about 800°F", in its context. In the Board's view, this expression is clearly intended to include reaction temperatures of 800°F and just below, i.e. certainly down to 799°F. Therefore, in the Board's judgment, the subject-matter of Claim 1 lacks novelty having regard to the disclosure of document (3).

3.4 With respect to the subject-matter of the second and third auxiliary requests, the Board finds that it is novel in the light of the cited prior art. Since the Respondent admitted that the subject-matter of the claims of these two requests was novel, it is not necessary to give detailed reasons for this finding.

3.5 In his facsimile of 9 June 1993, the Respondent alleged that at least granted Claims 1 to 4, 7 to 10 and 12 lacked novelty with respect to document (9) when considered in the light of document (10). Document (9) discloses the use of an AMS-IB crystalline borosilicate-based catalyst incorporated within a matrix material and on which is impregnated a molybdenum compound for converting alkyl-substituted aromatic hydrocarbons such as xylenes and ethylbenzene (cf. Claims 1 and 2). Document (10), which is incorporated by reference in document (9) (cf. the Examples), discloses the preparation of such AMS-IB crystalline borosilicates.

Since aluminium may be present in the starting materials for preparing these catalysts, the Respondent alleged that they could be considered to be crystalline aluminosilicates falling within the definition of the catalysts used in the present process. This allegation was strenuously denied by the Appellant who maintained the claims in accordance with any of the requests could not be construed as including the use of the catalyst referred to in these documents.

In the Board's judgment, borosilicates containing traces of aluminium would not be understood by the skilled person to be aluminosilicates. Therefore, the Board agrees with the Appellant and has decided not to take these late-filed documents into consideration.

Since the decision under appeal was based on the fact that the claimed subject-matter lacked novelty in the light of the combined disclosure of documents (3) and (6), the Board finds it would be useful to indicate that, when assessing novelty, it is not permissible to combine separate items of prior art. However, in a case where there is a specific reference in one prior art document to a second prior art document, when construing the former document the presence of such a specific reference may necessitate that part or all of the disclosure of the first document to be considered as part of the disclosure of the first document (cf. T 153/85 "Alternative claims/AMOCO CORPORATION, OJ EPO 1988, 1, paragraph 4.2).

In the present case the reference to US application No. 878 588 filed 17 January 1978 in document (3) (cf. column 4, lines 46 and 47 and column 8, lines 53 and 54) cannot be regarded as a specific reference within the sense of the above decision which would allow the disclosure of documents (3) and (6) to be combined,

since it is impossible for the reader of document (3) to correlate this unpublished US patent application with the published British patent application without comparing the latter with the information given in document (3) concerning the contents of the unpublished document. Due to the fact that it was intended to refer to United States application serial No. 878 558 of 17 February 1978 (cf. Respondent's letter filed on 30 October 1992) it was impossible for the reader to have traced any published document containing the information disclosed in document (3) in the paragraph bridging columns 5 and 6 and in column 8, line 53 to column 9, line 16.

4. It still remains to be decided whether the subject-matter claimed according to the second and third auxiliary requests involves an inventive step.

4.1 According to the second auxiliary request the disputed patent relates to a process for the isomerisation of mixed xylenes with concurrent conversion of ethylbenzene substantially by hydrodeethylation using an intermediate pore crystalline aluminosilicate zeolite-based catalyst composition. Document (3), which is considered to represent the closest prior art, also discloses such a process.

According to this document using the catalyst composition disclosed therein it is necessary to operate the process at reaction temperatures from greater than about 800°F (427°C) to about 1 000°F (538°C) to ensure that ethylbenzene reacts primarily via hydrodeethylation rather than via disproportionation (cf. column 10, lines 45 to 56).

In the interest of energy conservation it would be advantageous to achieve the same result with respect to

the conversion of ethylbenzene to benzene and ethane at lower temperatures. Therefore, in the light of this closest state of the art, the technical problem underlying the patent in suit is to provide a more economical process for the isomeration of mixed xylenes and the conversion of ethylbenzene substantially by hydrodeethylation.

According to the disputed patent the technical problem is essentially solved by carrying out the reaction at a temperature of from 95°C to 700°F (371°C) using an intermediate pore crystalline aluminosilicate zeolite-based catalyst as defined in Claim 1 incorporated within a matrix and on which has been placed a molybdenum compound or a tungsten compound.

In the light of the results obtained in Example 1 of the disputed patent, the Board is satisfied that this technical problem has been solved.

- 4.2 According to document (3) using catalysts of the type referred to in Claim 1 of the second auxiliary request at temperatures in the range of 600° to 700°F result in the conversion of ethylbenzene primarily by disproportionation (cf. column 10, lines 23 to 34). In order to achieve conversion of ethylbenzene via hydrodeethylation this document teaches the use of a catalyst composition containing less than about 5% by weight active zeolite at reaction temperatures of from greater than about 800°F to about 1 000°F (cf. column 10, lines 45 to 56).

Therefore, from the teaching of this document the skilled person would not contemplate operating the process at temperatures below about 800°F. Hence, this document would not lead the skilled person to the

proposed solution to the above-defined technical problem.

- 4.3 Document (6) discloses a crystalline aluminosilicate zeolite composition having in the anhydrous state, a formula, in terms of mol ratios of oxides given in paragraph 3.2 above in connection with document (3) (cf. Claim 1). In an example a xylene mixture is isomerised at 680°F (360°C) using the hydrogen form of the zeolite prepared using platinum (cf. page 21, lines 25 to 39, including Table 14). Although 23% by weight of the ethylbenzene present in the feedstock is converted, it is impossible to decide whether the conversion has proceeded mainly by disproportionation or primarily by hydrodeethylation. However, in view of the teaching of document (3), the skilled person would deduce that the predominant reaction would be disproportionation.

Therefore, the teaching of this document alone or the combined teaching of documents (3) and (6) would not provide the skilled person with any incentive to consider trying a catalyst comprising an intermediate pore crystalline aluminosilicate zeolite composition incorporated within a matrix material and on which has been placed a molybdenum compound or a tungsten compound, would solve the technical problem underlying the disputed patent.

- 4.4 Document (7) discloses a process for isomerising a xylene mixture with concurrent conversion of ethylbenzene to xylene or to compounds readily separated by distillation from eight carbon atom aromatics by contacting the feedstock in the vapour phase with a catalyst comprising a zeolite of the ZSM-5 type, zeolite ZSM-12 or zeolite ZSM-21 at a temperature of 500 to 1 000°F (260° to 538°C), optionally in the presence of hydrogen (cf. Claims 1 and 2).

According to lines 17 to 28 of page 9 of this document, if the process is carried out in the presence of hydrogen, the catalyst also includes a metal having hydrogenation capabilities, such as the metals of Group VIII of the Periodic Table, particularly nickel. However, this document is silent with respect to molybdenum. Furthermore, if the reaction is to be carried out at temperatures much below 800°F it is preferred to use catalysts containing nickel rather than ones containing a metal of the platinum group (cf. page 10, lines 12 to 21). Finally, it is stated in this document that it is important for the purposes of the invention that the side chain of ethylbenzene is cracked to yield benzene and ethylene (cf. page 10, lines 60 to 62). Naturally, in the presence of hydrogen, any ethylene produced is hydrogenated to yield ethane.

In the Board's judgment, the skilled person reading document (7) in the light of the teaching of the later published document (3) as outlined above would conclude that the conversion of ethylbenzene substantially by hydrodeethylation at temperatures much below 800°F is only possible in the presence of aluminosilicate zeolite catalysts containing nickel; i.e. nickel containing catalysts are exceptions to the teaching of document (3) that the reaction temperature must be above about 800°F to achieve hydrodeethylation of ethylbenzene.

4.5 By referring to the paragraph at lines 11 to 15 of page 11 of document (1), the Respondent alleged that nickel, molybdenum and tungsten were equivalent hydrogenation/dehydrogenation components of zeolite catalyst since all three appeared in a list of elements which may be used to prepare hydrocarbon conversion catalyst from zeolite FU9 by ion exchange or impregnation with cations or oxides.

However, the Board cannot accept this allegation since the previous paragraph at lines 5 to 10 reads:

"If zeolite FU9 in any cationic form or as a catalytic composite is exchanged or impregnated with hydrogenation/dehydrogenation components, such as Ni, Co, Pt, Pd, Re, Rh, hydrocracking and reforming catalysts can be made, especially if the Na<sub>2</sub>O content is less than 0.1% w/w".

Hence, in the absence of any reference to molybdenum or tungsten in this paragraph, it is clear that in this document only nickel was regarded as being equivalent to cobalt, platinum, palladium, rhenium and rhodium with respect to hydrogenation/dehydrogenation activity in zeolite catalysts.

- 4.6 Therefore, the proposed solution to the above-defined technical problem is not obvious in the light of the cited prior art. Hence, the subject-matter of Claim 1 according to the second auxiliary request involves an inventive step. Claim 1 of this request and dependent Claims 2 to 11 which relate to preferred embodiments of the process of Claim 1, are allowable.
5. In the light of the above it is not necessary to consider the third auxiliary request.




Order

For these reasons, it is decided that:

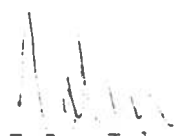
1. The Opposition Division's decision is set aside.
2. The case is remitted to the Opposition Division with the order to maintain the patent on the basis of auxiliary request No. 2, with corresponding amendments to the description.

The Registrar:



E. Görgmaier

The Chairman:



K.J.A. Jahn