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**D E C I S I O N**  
of 29 May 2002

**Case Number:** T 0685/97 - 3.3.7

**Application Number:** 92870056.6

**Publication Number:** 0507761

**IPC:** B01J 29/70

**Language of the proceedings:** EN

**Title of invention:**

Modified zeolite alkylation catalyst and process employing same

**Patentee:**

FINA TECHNOLOGY, INC.

**Opponent:**

Mobil Oil Corporation

**Headword:**

-

**Relevant legal provisions:**

EPC Art. 54, 56, 84, 123

EPC R. 57(a)

**Keyword:**

"Amendments - broadening of a claim (no) - added subject-matter (no) "

"Novelty (yes) "

"Inventive step (yes) - closest prior art, problem and solution"

**Decisions cited:**

T 0153/85, T 0291/85

**Catchword:**

-



Case Number: T 0685/97 - 3.3.7

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.7  
of 29 May 2002

**Appellant:** Mobil Oil Corporation  
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**Decision under appeal:** Interlocutory decision of the Opposition Division  
of the European Patent Office posted  
24 April 1997 concerning maintenance of European  
patent No. 0 507 761 in amended form.

**Composition of the Board:**

**Chairman:** R. E. Teschemacher  
**Members:** G. Santavicca  
B. L. ter Laan

## Summary of Facts and Submissions

- I. The mention of the grant of European patent 0 507 761, in respect of European patent application 92 870 056.6 filed on 3 April 1992 and claiming a right of priority in the United States of America of 5 April 1991 (US 681 035), was published on 22 March 1995. The patent as granted comprised 18 claims; independent claims 1, 6 and 11 read as follows:

"1. A molecular sieve catalyst comprising lanthanum/beta zeolite wherein the lanthanum content is between 0.05 and 0.5 weight percent, wherein the surface area of said molecular sieve, based upon the crystalline structure of said lanthanum/beta zeolite, is at least 500 m<sup>2</sup>/g and wherein said lanthanum/beta zeolite has a sodium content of less than 0.04 weight percent Na<sub>2</sub>O."

"6. A method for the preparation of a lanthanum/beta zeolite derived by modification of an alkali metal containing zeolite beta synthesized by the hydrothermal digestion of a reaction mixture comprising silica, alumina, an alkali metal oxide, and an organic templating agent, comprising:

- (a) treating said synthesized zeolite beta with an ion-exchange medium to protonate a portion of the active sites in said zeolite by exchanging said alkali metal;
- (b) calcining under anhydrous conditions said ion exchanged zeolite at a temperature within the range of 400°-700°C for a period within the range of 2-10 hours;
- (c) treating said calcined zeolite with an ion exchange medium to protonate an additional portion of the active sites in said zeolite by exchanging said alkali metal;

- (d) calcining under anhydrous conditions said ion-exchanged zeolite from step (c) at a temperature within the range of 400°-700°C for a period within the range of 2-10 hours;
- (e) incorporating lanthanum into said zeolite system by treating the ion-exchanged zeolite from step (d) with an ion-exchange medium comprising a lanthanum salt solution to obtain a lanthanum/beta zeolite;
- (f) mixing said lanthanum/beta zeolite with a binder to produce a mulled lanthanum/beta zeolite binder mixture; and
- (g) pelletizing said lanthanum/beta zeolite binder mixture and drying the resulting pellets."

"11. A process for the liquid phase alkylation of aromatic compounds, comprising:

- (a) supplying a feedstock containing an aromatic substrate into a reaction zone and into contact with a lanthanum/beta zeolite alkylation catalyst wherein the lanthanum content is between 0.05 and 0.5 weight percent, wherein the surface area of said molecular sieve, based upon the crystalline structure of said lanthanum/beta zeolite, is at least 500 m<sup>2</sup>/g and wherein said lanthanum/beta zeolite has a sodium content of less than 0.04 weight percent Na<sub>2</sub>O;
- (b) supplying a C<sub>2</sub>-C<sub>4</sub> alkylation agent to said reaction zone;
- (c) operating said reaction zone at temperature and pressure conditions to maintain said aromatic substrate in the liquid phase causing alkylation of said aromatic substrate by said alkylating agent in the presence of said catalyst; and
- (d) recovering alkylated aromatic substrate from said reaction zone."

The dependent claims were directed to preferred embodiments of the molecular sieve catalyst defined in claim 1 (claims 2 to 5), of the method for the preparation of a lanthanum/ beta zeolite defined in claim 6 (claims 7 to 10) and of the process for the liquid phase alkylation of aromatic compounds defined in claim 11 (claims 12 to 18), respectively.

II. A notice of opposition was filed on 20 December 1995. Revocation of the entire patent was requested on the grounds of Article 100(a) EPC that the claimed subject-matter lacked novelty and an inventive step having regard to the following documents:

D1: EP-A-0 485 683

D2: US-A-3 308 069

D3: US-A-3 251 897

D4: EP-A-0 055 046

D5: US-A-4 086 186

D6: US-A-4 891 458

D7: WO-A-91/00 777

III. In an interlocutory decision posted on 24 April 1997, the Opposition Division found that the subject-matter of the claims according to the main request filed during the oral proceedings held on 4 March 1997 fulfilled the requirements of the EPC. In that request, claims 1 and 10 as granted had been amended by the limitation of the surface area of the zeolite material to at least 650 m<sup>2</sup>/g, and a reference to the zeolite defined in claim 1 had been included in claim 5.

IV. According to that decision:

- (a) The amendments to the patent fulfilled the requirements of Articles 84 and 123(2) and (3) EPC;

(b) As to novelty, D1 did not disclose any lanthanum/beta zeolite, nor did it disclose any method for preparing such a catalyst. D2 did not disclose the surface area of the zeolite beta, nor did it disclose the combination of the specific amount of lanthanum together with the specific amount of sodium oxide. A combination of D2 with D1 against novelty was not acceptable, since D1 did not refer to D2 in the context of modifying a zeolite beta with lanthanum. Hence, the subject-matter of claim 1 was novel;

(c) As regards inventive step, D6 represented the closest prior art. The technical problem defined in the opposed patent was to produce ethyl benzene with little or no formation of xylene; table II in the patent showed that that problem had been solved. The general expressions "high purity" and "high yields" in D6 did not mean that this specific technical problem had been identified. Moreover, the zeolite beta of D6 had too high a sodium content. The combination with D4, which disclosed a low sodium content, was based on hindsight and would not have been made by the skilled person. Consequently, the claimed subject-matter involved an inventive step.

V. On 23 June 1997, the opponent lodged an appeal against that decision and paid the prescribed appeal fee. With the statement of grounds of appeal, received on 21 August 1997, the appellant filed a further document, namely D8 (Breck, Zeolite Molecular Sieves, John Wiley & Sons, New York, 1974, pages 507 to 508).

VI. In a communication in preparation for oral proceedings, the Board detailed the points to be discussed.

VII. By a letter received on 8 April 2002, the appellant announced that they would not be represented at the scheduled oral proceedings, which were, accordingly, held on 29 Mai 2002 in the absence of the appellant's representative, in compliance with Rule 71(2) EPC.

During the oral proceedings, the proprietor (respondent) filed an amended set of claims and a description brought in line with these claims. Independent claims 1, 5 and 10 read as follows:

"1. A molecular sieve catalyst comprising lanthanum/beta zeolite wherein the lanthanum content is between 0.05 and 0.5 weight percent, wherein the surface area of said molecular sieve, based upon the crystalline structure of said lanthanum/beta zeolite, is at least 650 m<sup>2</sup>/g and wherein said lanthanum/beta zeolite has a sodium content of less than 0.04 weight percent Na<sub>2</sub>O."

"5. A method for the preparation of a lanthanum/beta zeolite as defined in any one of claims 1 to 4 derived by modification of an alkali metal containing zeolite beta synthesized by the hydrothermal digestion of a reaction mixture comprising silica, alumina, an alkali metal oxide, and an organic templating agent, comprising:

- (a) treating said synthesized zeolite beta with an ion-exchange medium to protonate a portion of the active sites in said zeolite by exchanging said alkali metal;
- (b) calcining under anhydrous conditions said ion-exchanged zeolite at a temperature within the range of 400°-700°C for a period within the range of 2-10 hours;
- (c) treating said calcined zeolite with an ion-exchange medium to protonate an additional portion

- of the active sites in said zeolite by exchanging said alkali metal;
- (d) calcining under anhydrous conditions said ion-exchanged zeolite from step (c) at a temperature within the range of 400°-700°C for a period within the range of 2-10 hours;
  - (e) incorporating lanthanum into said zeolite system by treating the ion-exchanged zeolite from step (d) with an ion-exchange medium comprising a lanthanum salt solution to obtain a lanthanum/beta zeolite;
  - (f) mixing said lanthanum/beta zeolite with a binder to produce a mulled lanthanum/beta zeolite binder mixture; and
  - (g) pelletizing said lanthanum/beta zeolite binder mixture and drying the resulting pellets."

"10. A process for the liquid phase alkylation of aromatic compounds, comprising:

- (a) supplying a feedstock containing an aromatic substrate into a reaction zone and into contact with a lanthanum/beta zeolite alkylation catalyst wherein the lanthanum content is between 0.05 and 0.5 weight percent, wherein the surface area of said molecular sieve, based upon the crystalline structure of said lanthanum/beta zeolite, is at least 650 m<sup>2</sup>/g and wherein said lanthanum/beta zeolite has a sodium content of less than 0.04 weight percent Na<sub>2</sub>O;
- (b) supplying a C<sub>2</sub>-C<sub>4</sub> alkylation agent to said reaction zone;
- (c) operating said reaction zone at temperature and pressure conditions to maintain said aromatic substrate in the liquid phase causing alkylation of said aromatic substrate by said alkylating agent in the presence of said catalyst; and
- (d) recovering alkylated aromatic substrate from said reaction zone."



VIII. The appellant's arguments, which however did not take into account the amendments to the claims made during the oral proceedings, can be summarised as follows:

- (a) The subject-matter of all claims of the patent lacked novelty having regard to D1 augmented by so much of D2 as was incorporated by reference into D1. Since D1 contained no description of the preparation of a zeolite beta other than that incorporated by reference to D2, all of the examples of D2 which concerned the preparation of zeolite beta from raw materials should be interpreted as being incorporated into D1. In this respect, attention was drawn to decision T 0153/85 (OJ EPO 1988, 1);
- (b) The subject-matter of claims 1 to 4 of the patent lacked an inventive step having regard to D2, D4 or D6. In particular, in the impugned decision, the technical problem had not been identified correctly and had not been solved within the entire scope of the claims;
- (c) The subject-matter of claims 5 to 9 lacked an inventive step in view of D2, D4 or D6 in the light of D8, which showed that the multiple exchange technique, which was the sole distinction over D2, was well known;
- (d) The subject-matter of claims 10 to 16 of the patent in suit lacked an inventive step over D5, D6 or D7.

IX. The arguments of the respondent during the appeal proceedings can be summarised as follows:

- (a) Regarding novelty, D2 could be incorporated into D1, for the purpose of assessing novelty, only to

the extent that there was a specific reference to D2 in D1, as established for instance in decisions T 0153/85 (*supra*) and T 0291/85 (OJ EPO 1988, 302).

The parts of D1 that mentioned document D2 aimed at indicating the basic procedures for the preparation of the crystalline zeolite beta as synthesized. Some passages also indicated that the zeolite beta according to D1 was characterised by a sodium content substantially lower than the corresponding sodium contents disclosed by D2.

Although D2 mentioned lanthanum in a long list of elements that could be used to replace the sodium ion in the original sodium form of zeolite beta, all the exemplified treatments were however carried out with rare earths in general, without any specific reference to lanthanum. Therefore, D1 did not contain any specific reference to a procedure for making any beta/lanthanum zeolite.

Therefore, even if taking the reference into account, D2 could not supplement the teaching of D1 to arrive at the claimed subject-matter, which, as a consequence, was novel;

- (b) As regards inventive step, D6 represented the closest state of the art. It dealt with C<sub>2</sub>-C<sub>4</sub> alkylation of aromatics by using zeolite beta that could be treated by ion exchange and calcination and which could have a binder.

Since the xylene isomers were difficult to separate, and since the produced ethyl benzene was to be dehydrogenated to styrene and polymerised to polystyrene, it was undesirable to have xylene isomers present in the polystyrene end product.

This technical problem was specifically stated in the patent in suit, namely a lanthanum/beta zeolite catalyst exhibiting better selectivity to xylene formation than did its hydrogen form (page 3, lines 38-39).

The results shown in the figure and in table II demonstrated that a lanthanum/beta zeolite formed much lower xylene than its hydrogen form that had not been treated with lanthanum. Hence, the problem had been solved.

None of the documents cited by the opponent specifically addressed this problem. Although rare earths were cited in some documents, the disclosure either concerned mixtures of rare earth elements, as in D3, or was generic, as in D4 and D7. D4 concerned zeolite materials which did not contain the specifically claimed amount of lanthanum, as did D7, which in fact concerned boron beta zeolite. D5 concerned ZSM-34 zeolite and D8 described zeolite Y, ie different zeolite materials.

The expression "rare earth" as such was not an incentive to specifically use lanthanum, since the rare earth family comprised elements with different properties. Nor had the low content of lanthanum as claimed been disclosed. In this respect, the low lanthanum content that had been chosen to reduce xylene formation, also introduced extra acidity into the zeolite material.

Moreover, D6 did not address the same technical problem as the patent in suit. The examples in D6 did not mention xylene formation. Hence, D6 did

not hint at the low sodium content and the low lanthanum content as claimed in the patent in suit in order to form less xylene.

Therefore, the claimed subject-matter was not obvious.

X. The appellant (opponent) requested that the decision under appeal be set aside and that the patent be revoked.

XI. The respondent (patentee) requested that the appeal be dismissed and that the patent be maintained in the version appearing in the order.

### Reasons for the Decision

1. The appeal is admissible

2. *Amendments*

2.1 Compared with the claims as granted, the claims according to the main request contain the following modifications:

(a) In claims 1 and 10 (feature (a)), the surface area of the molecular sieve, based upon the crystalline structure of said lanthanum/beta zeolite, is at least 650 m<sup>2</sup>/g;

(b) In claim 5, a reference to the lanthanum/beta zeolite indicated in any of claims 1 to 4 has been inserted in the first line, before "derived by".

(c) In claim 11, the number in the reference to the preceding claim has been corrected.

(d) The description, page 3, has been brought in line with the new claims.

2.2 All the amendments have their basis in the original application:

(a) The surface area of the molecular sieve, based upon the crystalline structure of said lanthanum/beta zeolite, of at least 650 m<sup>2</sup>/g, as defined in claims 1 and 10 (feature (a)), has a basis in the additional features of original claims 4 and 14, respectively.

(b) The new reference to the zeolite of claim 1 in claim 5 not only aims at consistency between these claims, but also at overcoming the ground of opposition under Article 100(a) EPC as to lack of novelty and inventive step by inclusion in claim 5 of the specific features of the zeolite as defined in claim 1 (Rule 57a EPC). Basis for that amendment in the original application is given in claim 8 as well as on page 7, first line of the last paragraph, to page 12, first paragraph.

2.3 The amendments therefore fulfil the requirements of Articles 84 and 123(2) EPC as well as those of Rule 57a EPC. Since they restrict the scope of the granted patent, Article 123(3) EPC is also complied with.

### 3. Novelty

3.1 D1 is a European patent application filed on 13 November 1990, ie before the priority date of the patent in suit, but published on 20 May 1992, ie after the filing date of the patent in suit. Therefore, this earlier European patent application belongs to the prior art pursuant to Article 54(3) EPC.

D1 concerns an alkylation-transalkylation process, comprising the steps of:

- (a) supplying a feedstock containing an aromatic substrate into a reaction zone containing a molecular sieve aromatic alkylation catalyst;
- (b) supplying a C<sub>2</sub>-C<sub>4</sub> alkylating agent to said reaction zone;
- (c) operating said reaction zone at temperature and pressure conditions to maintain said aromatic substrate in the liquid phase and causing alkylation of said aromatic substrate by said alkylating agent in the presence of said catalyst to produce an alkylated product comprising a mixture of monoalkylated and polyalkylated aromatic products;
- (d) recovering said alkylated product from said reaction zone and supplying said product from said reaction zone to a separation zone for the separation of said aromatic substrate;
- (e) operating said separation zone to produce a lower boiling fraction comprising said aromatic substrate and a higher boiling fraction comprising a mixture of monoalkylated aromatic-polyalkylated aromatic mixture;
- (f) supplying said higher boiling fraction from said separation zone to a second separation zone;
- (g) operating said second separation zone to produce a second lower boiling fraction comprising monoalkylated aromatic product and a higher boiling fraction comprising heavier polyalkylated aromatic product;

- (h) supplying at least a portion of said polyalkylated aromatic product including substantially all of the dialkylated and trialkylated aromatics in said polyalkylated product to a transalkylation reaction zone containing a molecular sieve transalkylation catalyst;
- (i) supplying said aromatic substrate to said transalkylation zone;
- (j) operating said transalkylation reaction zone under temperature and pressure conditions to maintain said aromatic substrate in the liquid phase and effective to cause disproportionation of said polyalkylated aromatic fraction to arrive at a disproportionation product having a reduced polyalkylated aromatic content and an enhanced monoalkylated aromatic content;
- (k) supplying at least a portion of said disproportionation product to said first recited separation zone (claim 1).

The aromatic substrate comprises benzene and said alkylating agent is an ethylating or propylating agent, such as an olefin, eg ethylene (claims 2 to 5).

The alkylation catalyst comprises a molecular sieve selected from the group consisting of zeolite beta, zeolite omega and zeolite Y and the transalkylation catalyst comprises a molecular sieve selected from the group consisting of zeolite Y, zeolite beta and zeolite omega (claim 6). Preferably, the alkylation catalyst comprises zeolite beta (claim 7) and the alkylating agent is ethylene (claim 8).

The molecular sieve comprising the hydrogen form of zeolite beta has a surface area based upon the

crystalline structure of the zeolite beta of at least 650 m<sup>2</sup>/g as well as a sodium content of less than 0.04 wt.% Na<sub>2</sub>O (claims 18 and 20).

The molecular sieve catalyst can comprise the hydrogen form of zeolite beta in combination with a binder, wherein the surface area of the zeolite beta, based upon the composite of the molecular sieve including said binder, is at least 450 m<sup>2</sup>/g (claim 19).

The subject-matter of any of independent claims 1, 5 and 10 of the patent in suit is distinguished over the disclosure of D1 by the presence of between 0.05 and 0.5 weight percent of lanthanum in the composition of the beta zeolite forming the molecular sieve catalyst.

Therefore, the claimed subject-matter of the patent in suit is novel over D1 (Article 54(3) EPC).

3.2 The distinction of the claimed invention from D1 stated above was not contested by the appellant, who instead argued that:

- D1 referred to D2 in the context of the preparation of the zeolite beta and, consequently, all of the examples of D2, including those that mentioned the incorporation of lanthanum, were included by reference in the disclosure of D1.
- That combined disclosure took away the novelty of the claimed subject-matter.



3.2.1 D2 discloses a crystalline synthetic zeolitic material having the calculated composition:



wherein X is less than 1, Y is greater than 5 but less than 100, W is up to about 4, and TEA represents tetraethylammonium ion, said material being characterized by an X-ray powder diffraction pattern essentially the same as that shown in Table 4 of D2 (claim 1).

The method for synthesizing such a crystalline material comprises preparing a reaction mixture whose composition expressed in terms of mol ratios falls within the following ranges:

SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub>	from about 10 to about 200
Na <sub>2</sub> O/TEA hydroxide	from about 0.0 to about 0.1
TEA hydroxide/SiO <sub>2</sub>	from about 0.1 to about 1.0
H <sub>2</sub> O/TEA hydroxide	from about 20 to about 75

and maintaining said mixture at a temperature within the range of 75°C to 200°C until said crystalline material is formed (claim 5).

Then, the crystals may be separated from the mother liquor and subjected to ion exchange with a metal selected from the group consisting of the metals of Groups 1a, 2a, 3a, 3b, 4b, 7b and 8 of the periodic classification of the elements (claim 8).

According to D2, a catalytic material can be prepared by replacing the major portion of the sodium in the zeolite with other metallic and/or ammoniacal ions (column 5, lines 17 to 20). The crystals can be ion exchanged with the rare earth metals such as cerium, lanthanum, praseodymium, neodymium, samarium and

mixtures thereof (column 5, lines 51 to 53). No proportion or extent of the cation exchange is defined in these passages, which is the only place in D2 where lanthanum is mentioned as such, as part of a list of several rare earth metals.

Cation exchange with rare earth metals is illustrated in examples 5 and 9. These examples, however, do not specify which rare earth metal is used. Also, the proportion of the rare earth metal used in example 9 (3.74% RE<sub>2</sub>O<sub>3</sub>), the only place in D2 where a proportion of said elements can be found at all, is outside the specific range of proportions as defined in the claims of the patent in suit.

3.2.2 It follows that D2 does not disclose the information that the zeolite beta of the molecular sieve catalyst should contain lanthanum in an amount of between 0.05 and 0.5 weight percent, as defined in the claims of the patent in suit.

3.3 Therefore, even if D2 were to be combined with D1, the combined disclosure of D1 and D2 could not anticipate the subject-matter of the independent claims of the patent in suit.

3.4 The further cited documents have not been used to attack the novelty of the claimed subject-matter (statement of grounds for appeal, page 14). The Board has no reason to take a different position.

3.5 The subject-matter of the amended claims of the patent in suit is thus novel under Article 54 EPC.

#### 4. *Inventive step*

4.1 The patent in suit relates to a modified zeolite alkylation catalyst and a process employing the same.

4.2 Such catalysts and processes have been described in D6, which was considered to represent the closest prior art by the Opposition Division and the respondent. The appellant, however, also mentioned D4 and D7 as appropriate starting points.

4.2.1 D6 discloses a process for the alkylation of an aromatic hydrocarbon which comprises contacting a stoichiometric excess of the aromatic hydrocarbon with a C<sub>2</sub> to C<sub>4</sub> olefin under at least partial liquid phase conditions and in the presence of a catalyst comprising zeolite beta (claim 1).

The aromatic hydrocarbon may be benzene, toluene and xylene, or mixtures thereof; preferably the aromatic hydrocarbon is benzene (claims 3 and 4).

The olefin may be ethylene, propylene, butene-1, trans-butene-2 and cis-butene-2, or mixtures thereof; preferably the olefin is propylene or ethylene (claims 5 to 7). In one embodiment, the aromatic hydrocarbon is benzene and the olefin is ethylene (claim 16).

The alkylation reaction is carried out at a temperature in the range of about 250°F to 450°F and a pressure in the range of about 50 psig to 1000 psig, and at a weight hourly space velocity of about 0.5 to 50 (claims 8 to 9).

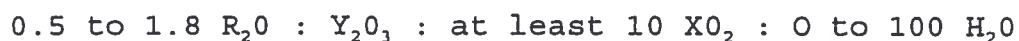
A major portion of the cation sites in the zeolite beta may be occupied by hydrogen ions and/or rare earth ions (claim 10). In this regard, it is especially preferred that at least 80% of the cation sites be occupied by hydrogen ions and/or rare earth ions (column 4, lines 29 to 31). Lanthanum is mentioned among the rare earth ions which are suitable for ion exchange (column 4, lines 18 to 19).

The zeolite beta used in the production of ethyl benzene by the alkylation of benzene with ethylene (example 6) was made according to a procedure resulting in a surface area of 640 m<sup>2</sup>/g and a sodium content of 0.3 wt.% (example 1).

According to example 7, a catalyst comprising zeolite beta in the form of an extrudate with 80% zeolite beta and 20% alumina binder and a sodium content of 200 ppm is much more active in transalkylation than eg rare earth Y zeolite.

In summary, as far as zeolite beta materials suitable for alkylation of aromatics such as benzene, toluene or xylene with lower olefines such as ethylene or propylene are concerned, the general teaching of D6 is that at least 80% of the cation sites should be occupied by hydrogen ions and/or rare earth ions, ie the zeolite beta material may be metal modified. That modified zeolite beta material is capable of providing monoalkylated products in high yield and high product purity for much longer periods of time than other zeolite catalysts (column 2, lines 41 to 50).

4.2.2 D4 discloses a synthetic zeolite material having a molar composition expressed by the formula:



wherein R is a monovalent cation or 1/n of a cation of valency n, X is silicon and/or germanium, Y is one or more of aluminium, iron, chromium, vanadium, molybdenum, arsenic, manganese, gallium or boron, and H<sub>2</sub>O is water of hydration additional to water notionally present when R is H and having, as made, an X-ray diffraction pattern substantially as shown in Table 1 (claim 1).

A catalyst comprising a synthetic zeolite material as defined above and a catalytic process employing that catalyst are also part of the disclosure of D4 (claims 10 and 11).

The zeolite described by D4 is a member of the zeolite-beta family (page 5, lines 14 to 15), as confirmed by D6 (column 3, lines 42 to 48).

To produce the hydrogen form of the zeolite, D4 teaches a sequential or multiple ion exchange/calcination protocol (page 12 line 23 to page 13 line 5), whereby rare earths are among the possible replacing cations (page 12 line 35 to page 13 line 1).

Example 9 shows that this sequential procedure leads to a hydrogen beta zeolite with 0.03% w/w of sodium, obtained via repeated ion exchange with hydrochloric acid. In example 10, the use of a catalyst comprising that zeolite beta is illustrated, however, in the process of toluene disproportionation.

Nevertheless, alkylation of aromatics is mentioned on page 10, line 9, whereby the useful catalysts therefor may have been treated with any of the metals listed on page 10, lines 2 to 3, inter alia a rare earth metal such as Re.

In summary, as far as alkylation of aromatics and metal exchange with rare earth metals are concerned, the general teaching of D4 is that the catalyst is preferably in its acid form (page 10, lines 4 to 9), although it may have been treated with Re.

- 4.2.3 D7 discloses a zeolite having a mole ratio of an oxide selected from silicon oxide, germanium oxide, and mixtures thereof to an oxide selected from boron oxide, or mixtures of boron oxide with aluminum oxide, gallium

oxide or iron oxide, greater than 10:1 and wherein the amount of aluminum is less than 0.10% by weight and having the X-ray diffraction lines of Table 1(b) (claim 1).

That zeolite is referred to as "Low-Aluminum Boron Beta Zeolite" or "(B) Beta zeolite" (page 3, lines 7 to 12).

The zeolite can undergo ion exchange with hydrogen, ammonium, rare earth metal, group IIA metal, or group VIII metal ions (claim 8), whereby said metals can be occluded in the zeolite (claim 9).

D7 also concerns a zeolite composition comprising that zeolite and an inorganic matrix (claim 10).

D7 describes, *inter alia*, a process for alkylating an aromatic hydrocarbon that comprises contacting under alkylating conditions at least a molar excess of an aromatic hydrocarbon with a C<sub>2</sub> to C<sub>20</sub> olefin under at least partial liquid phase conditions and in the presence of a zeolite as defined in claim 1 (claim 29).

In that process, the aromatic hydrocarbon may be benzene, toluene and xylene, or mixtures thereof (claim 31). Suitable olefines include ethylene, the preferred olefin being propylene (page 27, last paragraph). Ethyl benzene is one of the reaction products mentioned on page 28, lines 19 to 20.

For high catalytic activity in the alkylation or transalkylation of an aromatic hydrocarbon, the (B) Beta zeolite of D7 should predominantly be in its hydrogen form (page 27, lines 1 to 2). Also, it is preferred that at least 80% of the cation sites are occupied by hydrogen and/or rare earth ions (page 27, lines 7 to 9).

An amount of sodium of less than one percent is mentioned in connection with hydrocracking catalysts on page 15, lines 27 to 29.

In summary, the general teaching of D7 is that a particular zeolite beta catalyst with a reduced amount of aluminum may be prepared by use of a particular template, ie a diquaternary ammonium used instead of a tetraethyl ammonium as a template. That new material has interesting hydrocarbon conversion properties, particularly reforming properties with high sulfur tolerance (page 5, first full paragraph), and is suitable for alkylation of benzene. In the latter case, for high catalytic activity, the boron zeolite beta should be predominantly in its hydrogen ion form (page 27, lines 1 to 2).

- 4.3 In selecting the closest prior art, the first consideration is that it should be directed to the same purpose or effect as the invention (Case Law of the Boards of Appeal of the European Patent Office, fourth edition, 2001, I.D.3.1).

The patent in suit aims at providing a metal-modified alkylation catalyst and a process for employing such a catalyst in the alkylation of aromatic substrates with relatively low molecular weight alkylating agents (page 2, lines 53 to 55), whereby that metal-modified catalyst shows better selectivity than its hydrogen counterpart in the formation of xylene (page 3, lines 38 to 39).

It appears from the above discussion of D6, D4 and D7, that D6 not only relates to the same technical field as the patent in suit, it actually concerns a similar catalyst used in the same kind of alkylation of the same aromatics with the same lower olefines, whereas D4 and D7 are less specific regarding the alkylation

process and the catalyst used therefor. Therefore, D6 represents the closest prior art, as also acknowledged in the patent in suit (page 2, lines 47 to 49).

- 4.4 Although D6 describes that zeolite beta in its hydrogen form is a very good catalyst for liquid phase alkylation, the selectivity could still be improved.

The patent in suit thus addresses the problem of improving such a zeolite beta in order to provide an alkylation catalyst having not only high activity and good stability (page 6, lines 31 to 33), but also exhibiting better selectivity to xylene formation (page 3, lines 38 to 39).

- 4.5 According to the patent in suit, that problem is solved by a molecular sieve catalyst comprising a lanthanum/beta zeolite as defined in claim 1.

The examples in the patent in suit (page 4 line 55 to page 6 line 33) and the results thereof in table II and in Figure 1 show a comparison between a lanthanum/beta zeolite catalyst according to the invention and a hydrogen beta zeolite containing no lanthanum, under identical alkylating conditions for producing ethyl benzene. According to these results, the lanthanum/beta zeolite according to the invention produces less xylene (ppm) relative to ethyl benzene.

In view of this, the Board is satisfied that the problem thus defined is effectively solved by the claimed solution.

- 4.6 It remains to be decided whether the claimed subject-matter is obvious having regard to the documents on file.



4.6.1 In D6, the suggestion of incorporating rare earth elements into the zeolite has to do with the purpose of obtaining a high catalytic activity of the resulting catalyst, not to obtain higher selectivity from that catalyst, let alone less xylene formation (column 4, lines 21 to 31).

Although D6 addresses the problem of high purity of the product in general, it does not address the more specific problem of reducing the xylene formation. There is no mention of xylene in D6. Example 6, which concerns the production of ethyl benzene by alkylation of benzene with ethylene, teaches the use of a hydrogen/beta zeolite without any metal incorporation, and mentions that the product of the reaction was not pure, since it comprised polyethylenebenzenes and other non-specified products.

Despite the suggested preference that at least 80% of the cation sites be occupied by hydrogen ions and/or rare earth ions, ie that the zeolite beta material may be metal modified, D6 neither discloses the use of lanthanum to modify the zeolite beta, nor the actual extent of any such modification with a rare earth element, let alone in combination with a corresponding very low amount of sodium oxide, as defined in claim 1 of the patent in suit.

Example 7 of D6 shows that the hydrogen form of the zeolite beta is superior to the comparative rare earth Y zeolite (Linde SK-500) in respect of activity and selectivity (table 4), in the transalkylation process for producing cumene.

Therefore, the claimed subject-matter of the patent in suit is not obvious over D6 taken alone.

4.6.2 The same conclusion is valid for the other documents as cited, in particular D2, D4 and D7:

D2 mentions the possibility of metal modifying the beta zeolite but does not disclose the preparation of catalysts for alkylation of aromatics. Only n-hexane cracking is mentioned in examples 4 to 5 and 8 to 9.

D4 and D7, as discussed above, neither address the same technical problem as the patent in suit, nor give any hint at the claimed solution.

Hence, none of these documents refer to the desirability of reducing the formation of xylene. In particular, neither of D2, D4 and D7 suggest ion exchange of the zeolite beta with lanthanum, let alone the low amount thereof in combination with a very low amount of sodium oxide, as claimed in the patent in suit.

D3 and D5 do not concern molecular sieve catalysts comprising zeolite beta material. In fact they were cited as evidence of the composition of the rare earth mixtures, and of typical zeolite surface areas respectively. D3 merely suggests the use of mixtures of rare earth metals, in higher amounts than in the patent in suit, for the purpose of increasing the activity of catalysts made of zeolite X, Y, mordenite, etc. (column 1, line 47). D5 was only cited to show the surface area of the zeolite material. Moreover, D5 exemplifies catalysts for cracking of n-hexane and propylene polymerization.

D8 has been referred to in the statement of grounds to show that the feature of multiple exchange with intervening calcination to achieve thorough elimination of sodium and enhance stability had been known since at least 1968. The cited parts however mention zeolite Y.

Thus, neither of D2, D3, D4, D5 and D7 can provide the features missing in D6 to arrive at the combination of features now being claimed.

Therefore, any combination of D6 with any of said cited documents would not result in another conclusion, since none of them would lead to the claimed subject-matter.

- 4.6.3 The same arguments apply when D4 or D7 would be chosen as the closest prior art, ie lack of suggestion of ion exchange of zeolite beta material to obtain a catalyst with low content of lanthanum and low content of sodium, and the skilled person would not arrive at the claimed subject-matter either.
- 4.7 It follows that the subject-matter defined in claim 1 of the patent in suit involves an inventive step. Consequently, the subject-matter of the other claims, which relate to the product of claim 1, or depend on the product of claim 1, similarly involve an inventive step.
- 4.8 Therefore, the sole request fulfils the requirements of the EPC.

**Order**

**For these reasons it is decided that:**

1. The decision under appeal is set aside.
2. The case is remitted to the department of the first instance with the order to maintain the patent in the following version:

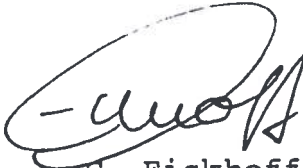
**claims:** 1 to 16 as submitted during the oral proceedings before the Board;

**description:** page 2 as submitted during the oral proceedings before the Opposition Division;  
 page 3 as submitted during the oral proceedings before the Board;  
 pages 4 to 6 of the patent as granted;


**drawings:** Figure 1 of the patent as granted.



The Registrar:

The Chairman:

  
 C. Eickhoff



  
 R. Teschemacher

 25.10.2002  
 28.10.02.