

Veröffentlichung im Amtsblatt	J/Nein
Publication in the Official Journal	Yes/No
Publication au Journal Officiel	Oui/Non

Aktenzeichen / Case Number / N^o du recours : T 27/89 - 3.3.1

Anmeldenummer / Filing No / N^o de la demande : 81 301 529.4

Veröffentlichungs-Nr. / Publication No / N^o de la publication : 0 062 123

Bezeichnung der Erfindung: De-aluminization of aluminosilicates

Title of invention:

Titre de l'invention :

Klassifikation / Classification / Classement : C01B 33/28

ENTSCHEIDUNG / DECISION

vom / of / du 5 October 1989

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /

Titulaire du brevet :

Mobil Oil Corporation

Einsprechender / Opponent / Opposant :

OI Degussa AG

OII Akzo GmbH

OIII Grace GmbH

Stichwort / Headword / Référence : De-aluminization/Mobil

EPÜ / EPC / CBE Articles 54, 56 and 123 EPC

Schlagwort / Keyword / Mot clé :

"Novelty (affirmed after amendment)

Inventive step (denied) - obvious

alternative,

Deletion of inessential feature allowed"

Leitsatz / Headnote / Sommaire



Case Number : T 27 /89 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 5 October 1989

Appellant : Mobil Oil Corporation
(Proprietor of the patent) 150 East 42nd Street
New York
New York 10017 (US)

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

Respondent : Degussa AG
(Opponent OI) Zweigniederlassung Wolfgang
Rodenbacher Chaussee 4
Postfach 13 45
D-6450 Hanau 1 (DE)

Respondent : Akzo GmbH
(Opponent OII) Postfach 10 01 49
D-5600 Wuppertal 1 (DE)

Other party to
the proceedings: Grace GmbH
(Opponent OIII) Erlengang 31
D-2000 Norderstadt 1 (DE)

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Decision under appeal: **Decision of Opposition Division of the European Patent Office dated 4 November 1988 revoking European patent No. 0 062 123 pursuant to Article 102(1) EPC.**

Composition of the Board:

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

Summary of Facts and Submissions

- I. The mention of the grant of European patent No. 0 062 123, in respect of European patent application No. 81 301 529.4 filed on 8 April 1981, was announced on 2 July 1986 (cf. Bulletin 86/27).
- II. Notices of opposition were filed on 3 February 1987, 19 March 1987 and 20 March 1987 requesting the revocation of the patent on the ground that its subject-matter lacked novelty and did not involve an inventive step. Opponent OIII also requested revocation on the ground that the patent did not disclose the invention in a manner sufficiently clear and complete for it to be carried out by the skilled person. The opposition was supported, inter alia, by the following documents:
- (1) US-A-3 644 220, and
 - (2) Catalysis by Zeolites, H.K. Beyer and I. Belenykaja, pages 203 to 210, 1980.
- II. By a decision dated 4 November 1988, the Opposition Division revoked the European patent. The Opposition Division considered that the reference in Claim 1 to aluminosilicate zeolites having a major pore dimension greater than 5nm was an obvious error, the correction of which rendered the objection under Article 100(b) EPC untenable. However, the Opposition Division concluded that the claimed subject-matter lacked novelty in the light of the disclosure in document (1). The question of whether the opposition filed by Opponent OI was admissible was left in abeyance.
- IV. An appeal was lodged against this decision by a duly confirmed telefax on 10 January 1989 and the appeal fee

duly paid. With the Statement of Grounds of Appeal, filed on 13 March 1989 by a duly confirmed telefax, the Appellant submitted a schedule of amendments. The Appellant contended that the subject-matter of the amended Claim 1 was novel and involved an inventive step in the light of the cited prior art.

- V. Both Respondents have argued that the proposed amendments to Claim 1 are formally unallowable. Furthermore, the Respondents contended that the subject-matter of the amended claim lacks novelty. Respondent OII has also alleged that the subject-matter of a claim restricted to the de-aluminisation of ZSM-5 having a constraint index of 8.3 would not involve an inventive step.
- VI. In his letter filed on 21 August 1989, the Appellant informed the Board that he would be taking no further part in the prosecution of the appeal. In the Statement of Grounds of Appeal the Appellant requested that the decision under appeal be set aside and a patent maintained on the basis of the schedule of amendments filed on 13 March 1989. In accordance with this schedule, Claims 1 and 10 read as follows:

"1. A process for removing aluminum from crystalline aluminosilicate zeolites having a constraint index between 1 and 12 which comprises contacting the hydrogen form of the aluminosilicate at elevated temperature with an inorganic halide or oxyhalide comprising a halogen and non-halogen component which latter is capable of substitution for aluminium in the aluminosilicate structure, forming an aluminum halide, introducing said non-halogen component into the aluminosilicate structure and volatilizing and removing the formed aluminum halide.

10. The process of any preceding claim wherein the zeolite is ZSM-5, ZSM-11 or ZSM-12."

The Respondents requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible. The grounds of appeal were filed on 13 March 1989 by a duly confirmed telefax. Therefore, the argument of Respondent OII that the grounds of appeal were not filed within four months after notification of the decision is without foundation.
2. The present Claim 1 differs from the granted Claim 1 in that the lower limit for the constraint index of the crystalline aluminosilicate used a starting material in the claimed process is specified and the requirement that it should have a major pore dimension greater than 5nm has been deleted.
 - 2.1 A basis for limiting the disputed patent to the treatment of zeolites having constraint indices between 1 and 12 is to be found in Examples 1 and 3 (cf. page 9, lines 1 to 5 and page 11, lines 7 to 11 of the published patent application and page 4, lines 3 to 5 and 49 to 51 of the patent specification).

Although these Examples disclose the preparation of ZSM-5, which has a constraint index of about 8.3, the cited passages clearly disclose that this value of about 8.3 fulfils the general requirement that zeolites suitable for treatment in accordance with the claimed process should

preferably have constraint indices between 1 and 12. Therefore, the request to replace the word "below" in line 3 of the Claim 1 as granted by the expression "between 1 and" is formally allowable.

- 2.2 Furthermore, the deletion of the expression "and a major pore dimension greater than 5nm" is allowable under Article 123(3) EPC. Since it is well-known that crystalline zeolites which have a constraint index of between 1 and 12 cannot have a major pore dimension greater than 5nm (50Å), the deletion of the phrase in question cannot be considered to extend the scope of protection conferred. Moreover, since crystalline aluminosilicates which have constraint indices between 1 and 12 necessarily have a major pore dimension greater than 0.5nm (5Å), the recital of this dimension in Claim 1 is redundant and its deletion could not be regarded as offending against Article 123(3) EPC.

The amendments to Claim 10 and page 2, line 41 and lines 49 to 52 and the deletion of Examples 5 and 6 are a consequence of restricting the process to the treatment of aluminosilicates having constraint indices between 1 and 12. These amendments are, therefore, also allowable.

3. The patent relates to a process for removing aluminium from crystalline aluminosilicate zeolites. Document (2), which may be regarded as representing the closest state of the art, discloses the dealuminisation of aluminosilicate zeolites such as Na-Y zeolite and (NH₄, Na)-Y zeolite (cf. first paragraph on page 204 and the third paragraph on page 205). This prior art process comprises dehydrating the zeolite at 377°C in a stream of nitrogen and reacting the dehydrated zeolite with gaseous silicon tetrachloride diluted with nitrogen at temperatures between 457° to 557°C. The dealuminated product is washed with water to

remove chloride ions and dried (cf. page 204, lines 23 to 34). If the treatment with gaseous silicon tetrachloride is carried out at temperatures of 507° to 557°C, zeolites with silica to alumina ratios of 40 to 100 are obtained (cf. page 205, lines 2 to 4).

- 3.1 In the light of this closest prior art, the technical problem underlying the patent in suit may be seen in providing a further process for dealuminating crystalline aluminosilicate zeolites.

According to the disputed patent this technical problem is essentially solved by contacting the hydrogen form of an aluminosilicate having a constraint index between 1 and 12 at elevated temperature with an inorganic halide or oxyhalide and volatilising and removing the thus formed aluminium halide.

In the light of results obtained in Examples 2 and 4, the Board is satisfied that the above-defined technical problem is plausibly solved.

4. After examination of the cited documents, the Board has concluded that the subject-matter of the amended Claims 1 and 10 is novel.
- 4.1 Example 8 of document (1) describes the treatment of previously calcined H-faujasite with silicon tetrachloride at a temperature of 204°C for a period of 1.5 hours. The treated H-faujasite was finally washed with dilute ammonium hydroxide. However, since it is accepted that faujasite has a constraint index of less than 1, this Example does not anticipate the subject-matter of the amended Claims 1 and 10. Moreover, the constraint indices of the zeolites listed in column 2, lines 19 to 25 of this document do not have constraint indices falling within the

defined range. Although, according to GB-A-1 446 522, tetraethylammonium mordenite has a constraint index of 1.7 (cf. Table 1a on page 7), the Board has the view that the reference to mordenite in document (1) (cf. column 2, lines 25 and 41) cannot be construed as including TEA-mordenite within its ambit.

4.2 Document (2) discloses the dealumination of faujasite-type zeolites, which do not have constraint indices between 1 and 12. Therefore, this document does not anticipate the presently claimed subject-matter.

5. It still remains to be examined whether the requirement of inventive step is met by the claimed subject-matter.

5.1 As previously mentioned document (2) describes the dealuminisation of faujasite-type zeolites by treating them with gaseous silicon tetrachloride at elevated temperatures. Although, according to the equation on page 204 of this document the cation present in the zeolite lattice may be any suitable cation, the experiments described in this document were carried out using Na-Y zeolite and (NH₄,Na)-Y zeolite, prepared from Na-Y zeolite by ion exchange with ammonium chloride (degree of exchange 65%) (cf. first paragraph on page 204).

5.2 The first sentence of the fourth paragraph on page 205 specifically states that (NH₄,Na)-Y zeolite can be dealuminated under the same conditions as those employed to dealuminate Na-Y zeolite. These conditions include heating the zeolite in a stream of nitrogen at 377°C for 2 hours (cf. first three lines of the fourth paragraph on page 204). During this heating process the ammonium ions will decompose and the resulting zeolite will be in the H⁺/Na⁺ form. Thus, this document teaches that a Na-Y zeolite

in which 65% of the sodium ions have been replaced by hydrogen ions can be successfully dealuminated using gaseous silicon tetrachloride.

Therefore, the use of aluminosilicate in the hydrogen form as a starting material in the prior art process known from document (2) would be an obvious alternative to the skilled person.

5.3 Moreover, the skilled person would be positively encouraged to treat zeolites in the hydrogen form since he is aware from the equation on page 204 and the discussion in the first two paragraphs of page 205 of document (1) that the use of this particular form of the zeolite as the starting material in this prior art process would eliminate the necessity of washing the resulting dealuminated zeolites.

5.4 The Appellant's argument that there is little incentive to dealuminise zeolites having constraint indices of between 1 and 12 in view of their relatively high silica to alumina ratios in the as-synthesised state cannot be accepted, since the present Claim 1 makes no reference to the silica to alumina ratio of the zeolite starting material and zeolites with constraint indices falling within the specified range are known whose silica to alumina ratios are much lower than that of synthetic zeolites, such as ZSM-5. Thus, for example, trimethylammonium offretite has a constraint index of 3.7 (cf. the table in column 4 of US-A-4 137 148) and a silica to alumina ratio of 3.4 to 4.25 (cf. Table 4.26 on page 350 of Zeolite Molecular Sieves by Donald W. Breck).

Furthermore, it is well-known in the art that the tolerance of crystalline aluminosilicate to acidic and thermal environments are influenced by the structural

silica to alumina ratio. Thus, the increase in stability of zeolites towards acids and heat resulting from an increase in the silica to alumina ratio would provide the skilled person with the incentive to contemplate the dealuminisation of zeolites with relatively high silica to alumina ratios.

6. In the Board's judgment the proposed solution to the problem of providing an alternative process for the dealuminisation of crystalline aluminosilicate zeolites is obvious. Therefore, the subject-matter of Claim 1 does not involve an inventive step.

7. Dependent Claims 2 to 10, which relate to preferred embodiments of the process in accordance with Claim 1, do not contain any independent inventive features and are, therefore, unpatentable in the absence of an allowable main claim.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar:



The Chairman:

