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**D E C I S I O N**  
of 13 June 1995

**Case Number:** T 0935/91 - 3.3.2

**Application Number:** 83104985.3

**Publication Number:** 0094693

**IPC:** C01B 33/28

**Language of the proceedings:** EN

**Title of invention:**

Crystalline aluminosilicate zeolite and process for its  
production

**Patentee:**

Teijin Petrochemical Industries Ltd.

**Opponent:**

Mobil Oil Corporation

**Headword:**

Zeolite/TEIJIN

**Relevant legal provisions:**

EPC Art. 54, 111(1), 102(3)

**Keyword:**

"Novelty"  
"Main request (no)"  
"Auxiliary request (yes)"  
"Product by process"

**Decisions cited:**

G 0009/91, G 0010/91

**Catchword:**

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Boards of Appeal

Chambres de recours

Case Number: T 0935/91 - 3.3.2

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.2  
of 13 June 1995

**Appellant:**  
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**Decision under appeal:** Decision of the Opposition Division of the  
European Patent Office dated 22 October 1991  
revoking European patent No. 0 094 693 pursuant to  
Article 102(1) EPC.

**Composition of the Board:**

**Chairman:** I. A. Holliday  
**Members:** G. J. Wassenaar  
J. A. Stephens-Ofner

### Summary of Facts and Submissions

- I. European patent No. 0 094 493 was granted in response to European patent application No. 83 104 985.3.
- II. A Notice of Opposition was filed against the European patent by the Respondent. Revocation of the patent was requested on the grounds of lack of novelty, lack of inventive step and insufficient disclosure (Articles 100(a) and (b) EPC).

Of the documents cited the following are relevant for the present decision:

- (1) GB-A-1 567 948
- (5) Chemistry Letters 1981, pages 169-172
- (6) An Experimental Report dated 8 May 1990 filed by the Appellant.

- III. The Opposition Division revoked the patent on the grounds of lack of novelty over (1). They considered that the process of Claim 7 as granted was completely anticipated by (1) and since it was expected that the same process would lead to the same result, the product obtained by the process of Claim 7, i.e. the product of granted Claim 1, must also be the same as that of (1).
- IV. The Appellant lodged an appeal against this decision; oral proceedings took place on 13 June 1995.

With the Statement of Grounds the Appellant filed an amended Claim 7 and Exhibits I and II showing the differences of the zeolite according to the patent in suit with respect to several prior art zeolites. On 31 October 1994 a new set of Claims 1 to 12 was filed as main request and a new set of Claims 1 to 11 as

auxiliary request I together with a corrected Exhibit I, a new Exhibit III and an expert's opinion. Exhibit III contained an experimental report wherein zeolites made according to the patent in suit were compared with zeolites ZSM-5 synthesized according to (5). It was argued that the zeolites of the invention had a unique combination of physical properties resulting in surprising catalytic properties.

During oral proceedings the Appellant filed two additional sets of auxiliary requests II and III.

- V. The Respondent disagreed with Appellant's submissions and maintained the novelty objection both with respect to the main and auxiliary request.

At the oral proceedings, the Respondent presented new evidence in the form of Table 1 of FR-A-2 289 444 and a paper containing Table A of the patent in suit together with X-ray diffraction patterns of zeolite zeta 1 and ZSM-5 taken from Table 1 of FR-A-2 289 444.

- VI. Claim 1 of the main request is identical to Claim 1 as granted and reads as follows:

\*1. A novel crystalline aluminosilicate zeolite which is characterized in that the zeolite has

- (a) a silica/alumina mole ratio of from 10 to 100,
- (a) an X-ray diffraction pattern as shown in the following Table A,

Table A

<u>X-ray lattice distance, d(Å)</u>	<u>Diffraction angle, 2θ</u>	<u>Relative intensity (I/I<sub>0</sub>)</u>
11,26	7,85	37
10,11	8,75	24
9,83	9,00	6
9,12	9,70	1
7,51	11,80	1
6,78	13,05	4
6,05	14,65	9
5,74	15,45	7
5,61	15,80	8
5,41	16,40	2
5,00	17,75	6
4,65	19,10	4
4,39	20,25	8
4,28	20,75	14
4,11	21,65	4
4,04	22,05	7
3,86	23,05	100
3,83	23,25	75
3,75	23,70	45
3,74	23,80	53
3,66	24,30	33
3,61	24,65	5
3,50	25,45	7
3,46	25,75	10
3,36	26,50	19
3,33	26,80	10
3,28	27,20	4
3,26	27,35	1
3,06	29,15	16
3,00	29,75	18
2,98	29,95	18
2,96	30,20	8

- (c) a specific n-hexane adsorption of at least 0.07 g/g, and
- (d) a (2-methylpentane/cyclohexane) adsorption ration of from 1.1 to 1.6,

and that when the intensity ( $I_0$ ) of the X-ray diffraction peak at  $d(\text{\AA})=3.86$  is taken as 100, the relative intensity ( $I/I_0$ ) of the X-ray diffraction peak at  $d(\text{\AA})=3.83$  is at least 70.

Claim 1 of auxiliary request I differs therefrom by the following additional requirement:

obtainable by a process comprising maintaining a silica source, an alumina source and a zeolite selected from zeolites ZSM-5 and zeolites having the characteristics specified above, in an aqueous solution containing 1 to 200 millimoles, per gram of said zeolite, of an alkali metal hydroxide, in such proportions that the silica source, the alumina source and the alkali metal hydroxide satisfy the following mole ratio in terms of  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$  and  $\text{OH}^-$ :

$$\begin{aligned}\text{SiO}_2/\text{Al}_2\text{O}_3 &= 5-100 \\ \text{OH}^-/(\text{SiO}_2+\text{Al}_2\text{O}_3) &= 0.5-1 \\ \text{OH}^-/\text{H}_2\text{O} &= 0.005-0.05\end{aligned}$$

and in such proportions of the silica source and the alumina source that 0.1 to 200 millimoles of  $\text{SiO}_2$  and 0.01 to 20 millimoles of  $\text{Al}_2\text{O}_3$  are used per gram of the zeolite, one hour to two days at a temperature of 120 to 200°C under autogenous pressure, the starting mixture being heated to the desired temperature and if required with stirring, being maintained at this temperature until a zeolite is formed, and after forming the zeolite crystals the reaction mixture being cooled to room temperature, filtered and washed with water until the ion conductivity of the washing reached 50  $\mu\text{u/cm}$  or below, then the crystals being separated and maintained at 50°C or higher for 5 to 24 hours under atmospheric or reduced pressure.

Claim 1 of auxiliary request II differs from Claim 1 of auxiliary request I only in that "obtainable" is replaced with "obtained".

Claim 1 of auxiliary request III differs from Claim 1 of auxiliary request II in that "obtained by a process comprising maintaining a silica source, an alumina source and a zeolite selected from zeolites ZSM-5 and zeolites having the characteristics specified above" is replaced with "obtained by a process comprising maintaining a silica source, an alumina source and a ZSM-5 zeolite".

VII. The Appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of the claims filed on 31 October 1994 by way of main and auxiliary request I, or on the basis of auxiliary requests II or III submitted in the course of the oral proceedings.

The Respondent requested that the appeal be dismissed.

#### **Reasons for the Decision**

1. The appeal is admissible.

2. *Main request*

2.1 Novelty

2.1.1 The zeolite of Claim 1 is characterised by features (a), (b), (c) and (d) and the additional feature that when the intensity ( $I_0$ ) of the X-ray diffraction peak at  $d=3.86$  is taken as 100, the relative intensity

( $I/I_0$ ) of the X-ray diffraction peak at  $d=3.83$  is at least 70. Said additional feature is hereinafter referred to as feature (e).

Feature (b) comprises a detailed X-ray diffraction pattern without indication of any tolerances. According to the Appellant, the claim is strictly limited to zeolites having a X-ray pattern as indicated, i.e. with all the lines even the weakest and the corresponding intensities and without any additional lines.

The Board cannot follow this interpretation for the following reasons. If Appellant's view were correct, feature (e) would be redundant because it is broader than feature (b) requiring a relative intensity of 75 for  $d=3.83$ . Thus feature (e) puts doubt upon the limitation imparted by feature (b). If a claim is not fully clear in itself, its meaning should be interpreted in the light of the description and drawings (Article 69(1) EPC).

According to the description of the granted patent, the zeolite provided by the invention has the X-ray lattice distances shown in Table A (page 5, line 23). It should be noticed that in this statement reference is only made to distances and not to the relative intensities of the diffraction lines. The splitting of the strongest prior art peak at  $d=3.85$  into two strong peaks at  $d=3.86$  and  $d=3.83$ , whereby the relative intensity of  $d=3.83$  is at least 70 (page 5, lines 25 to 28 and page 6, lines 59 to 61) is considered to be a significant difference with prior art ZSM-5 as disclosed in (4). Furthermore, according to the description, another great difference is that one peak observed at  $d=3.00$  in prior art ZSM-5 is observed as a bifurcated peak in most of the zeolites of the patent in suit, if not in all. This means that the patent does not exclude zeolites not having said



bifurcated peak, which puts in doubt not only the strictness of the relative intensities but also that of the lattice distances in Table A.

Thus the Board cannot regard the scope of Claim 1 to be strictly limited to zeolites having the exact X-ray diffraction pattern as indicated in Table A.

2.1.2 The description, however, leaves no room for any other interpretation of feature (e) of Claim 1 than the strict wording. Thus documents which do not disclose feature (e) do not destroy the novelty of Claim 1. Of the cited prior art documents only (5) shows feature (e). According to Appellant's own comparison of the most significant diffraction peaks shown in Figure 4 of (5) for Na-OEL-ZSM-5, the relative intensities of  $d=3.86$  and  $d=3.83$  are 100 and 74 respectively (Appellant's letter of 8 May 1990, page 9). Said Figure 4 further discloses all the diffraction angles ( $\theta$ ) corresponding to the significant lattice distances with generally the same intensity pattern. Since in the Board's view (see point 2.1.1 above) feature (b) does not exclude zeolites having a slightly different intensity pattern, features (b) and (e) are anticipated by (5).

2.1.3 Document (5) does not explicitly disclose feature (a). Disclosed is, however, a silica/alumina mol ratio of 42.5 in the reaction system from which the zeolite was crystallised. According to Appellant's own submission, Exhibit III, page 2, the molar ratio in the product was 38.0 so that feature (a) is also met by (5).

2.1.4 Document (5) does not disclose features (c) and (d), but according to said Exhibit III, Table 1, the zeolite C-1, made according to (5), satisfies the specific adsorption conditions mentioned in (c) and (d).

According to the Appellant's contention, the cyclohexane adsorption rate for zeolite C-1 was low and it took a long time to reach adsorption equilibrium, while  $V_{2-MP}/V_{CH}$  at the time of adsorption equilibrium was as low as 1.1 so that zeolite C-1 is different from zeolite B series (Exhibit III, page 3).

The Board does not doubt that zeolite C-1 is different from zeolite B, but for novelty, only the scope of the broadest claim is relevant. The values found for  $V_{2-MP}/V_{CH}$  after 2, 8 and 30 hours 1.5, 1.2 and 1.1 respectively, fall within the range of feature (d). The value after 2 hours should, in conformity with the measurements conditions given in the patent specification (page 7, lines 41 to 54), be considered as representative.

2.1.5 From the above it follows that (5) discloses a product which satisfies all the requirements of Claim 1, so that Claim 1 lacks novelty and the main request must be rejected.

3. *Auxiliary requests I and II*

Claim 1 of these auxiliary requests contain the new expression "a zeolite selected from zeolites ZSM-5 and zeolites having the characteristics specified above". The meaning of this expression in the context of the patent in suit is not entirely clear since it could mean the foregoing part of the claim, or the whole specification. Although clarity (Article 84 EPC) is not a ground for opposition, it should be taken into consideration for amended claims; Articles 102(3) and 111(1) EPC. Since the amendment introduced an ambiguity, the amended Claims 1 of auxiliary requests I and II lack clarity within the meaning of Article 84 EPC. These requests must accordingly be rejected.

4. *Auxiliary request III*

4.1 Clarity

By deleting the expression "zeolites selected from zeolites having the characteristics specified above" the clarity objection referred to under point 3 above has been removed.

4.2 Allowability under Article 123 EPC

Claim 1 has the form of a product by process claim and consists of a combination of granted product Claim 1 and process Claims 7 and 11 together with additional process features mentioned on page 4, lines 33 to 53 of the specification and Example 2. The amended lower limit of 0.5 for  $\text{OH}^-/(\text{SiO}_2 + \text{Al}_2\text{O}_3)$  is based on Example 2 disclosing values of 0.50 and 0.49 for said ratio. The scope of the claim is clearly limited with respect to granted Claim 1.

The amendments are also based on the patent application as filed; see the original application, page 7, line 28 to page 9, line 17 and page 10, line 35 to page 14, line 7, page 24, line 32 and page 25, line 34. Claim 1, therefore, satisfies Article 123(2) and (3) EPC.

4.3 Novelty

4.3.1 In the decision under appeal the Opposition Division argued that the product of Claim 1 lacked novelty since it could be obtained by a process which was not new. Such a reasoning is, however, only valid if the product is defined only by the result of a process. If, as in the present case, the product is defined by process features, structural features and physical properties all limitations resulting therefrom should be taken into consideration when judging novelty.

The process for obtaining such a product should be regarded as being limited to such conditions that a product is obtained having the required structural features and physical properties. Thus since only (5) discloses structural feature (e), only this prior art document is relevant for the novelty of present Claim 1.

- 4.3.2 The product of (5) is obtained by a process in which an organic cation (oxyethyl lactamide (OEL)) is used and which also contains said organic cation to a substantial amount in the dried state from which the X-ray diffraction pattern is obtained.

In the present case, however, no organic cation is added. Although it is not excluded that the ZSM-5 seeds used in the process still contain some organic cation (page 4, lines 1 to 5 of the specification) the product obtained will contain only a very limited amount, if any, of organic cation because of the high amount of alkali metal ions in the reaction mixture associated with the high alkalinity (high OH<sup>-</sup> concentration) of the reaction mixture.

Thus the dried product directly obtained by the process indicated in Claim 1 is clearly different from the product disclosed in (5).

- 4.3.3 The calcined hydrogen exchanged products suitable for the preparation of catalysts should also be considered to be different.

Although the product obtained by calcining the zeolite Na-OEL-ZSM-5 of (5) no longer contains substantial amounts of organic cation and thus, having a composition similar to that of the product of present Claim 1, its structure should be considered different because of differences in catalytic activity.

According to the comparative examples given in Exhibit III, the comparable zeolites obtained by the process of the patent in suit (zeolite B-2) have a higher cyclohexane decomposition index and a higher methylnaphtalene conversion than zeolite C-1 obtained according to (5); see Examples 3 and 4, Tables 2 and 3.

- 4.3.4 According to the respondent, the differences in catalytic activity could be explained by differences in the preparation of the final catalyst. For preparing the catalyst the hydrogen exchanged form of the zeolite was used. To obtain the H-form of zeolite B-2, the dried as prepared zeolite was treated with an aqueous ammonium chloride solution followed by washing and drying, whereafter the dried product was calcined at 450°C for 16 hours (Example 4 of Experimental Report dated 8 May 1990). To obtain the H-form of zeolite C-1, the dried as prepared zeolite was first calcined in air at 500°C for 16 hours before the zeolite was ion-exchanged as indicated above for zeolite B-2. The additional calcination would slightly degrade the crystal structure, which could explain the reduced shape selection.

The Board does not doubt that the reduced total calcination time for zeolite B-2 contributes to its performance, but sees in this an argument in favour of novelty. Since the ZSM-5 synthesis disclosed in (5), results in a product containing substantial amounts of organic cations, these cations must first be removed by a first calcination step before the H-form can be prepared by ion exchange.

Thus if the additional calcination step, necessary to obtain the H-form of prior art ZSM-5, implies a degradation of its crystal structure, the H-form of the zeolite obtained

according to present Claim 1 differs from the prior art in that its structure is not degraded. Novelty can accordingly be acknowledged.

#### 4.4 Inventive step

The decision under appeal does not contain any reasons with respect to inventive step. In order to guarantee the parties' right to appeal against a decision containing fresh reasons and following the reasoning set out in G 9 and G 10/91, (OJ EPO 1993, 408 and 420), the issue of inventive step has been left open and the Board exercises its power under Article 111(1) EPC to remit the case to the first instance for further prosecution.

#### **Order**

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

1. The decisions under appeal is set aside.
2. The case is remitted to the first instance for further prosecution on the basis of auxiliary request III.

The Registrar:

The Chairman:

E. Görgmaier

I. A. Holliday