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D E C I S I O N
of 3 June 1997

Case Number: T 0923/94 - 3.3.2

Application Number: 85309442.3

Publication Number: 0187522

IPC: C01B 33/34

Language of the proceedings: EN..

Title of invention:
Zeolite beta preparation

Patentee:
Exxon Research and Engineering Company

Opponent:
Grace GmbH

Headword:
Zeolite beta/EXXON

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step - yes"

Decisions cited:
-

Catchword:
-



Case Number: T 0923/94 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 3 June 1997

Appellant:
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 6 October 1994
revoking European patent No. 0 187 522 pursuant
to Article 102(1) EPC.

Composition of the Board:

Chairman: P. A. M. Lançon
Members: G. J. Wassenaar
R. E. Teschemacher

Summary of Facts and Submissions

I. The appeal lies from a decision of the Opposition Division to revoke European patent No. 0 187 522, which was granted with 8 claims in response to European patent application No. 85 309 442.3.

II. A notice of opposition was filed against the European patent by the Respondent. Revocation of the patent was requested in accordance with Article 100(a) EPC. The only substantiated ground was lack of inventive step within the meaning of Article 56 EPC.

The opposition was supported, inter alia, by the following documents:

(D1): EP-A-55 046

(D2): US-A-3 308 069

III. The decision under appeal was based on the set of claims as granted.

The Opposition Division held that claim 1 was new but lacked an inventive step over D2 in combination with the teaching of D1. The only difference with respect to the process disclosed in D2 was regarded to be the ratio between sodium oxide and the organic template (tetraethylammonium (TEA)) in the reaction mixture. They considered that the technical problem underlying the invention was to reduce the amount of organic template and that there was a clear indication pointing the skilled person to the claimed solution.

IV. In the statement of the grounds of appeal, the Appellant maintained that the subject matter of claim 1 was not only new but also involved an inventive step.

The Appellant argued essentially as follows:

D2 was the closest prior art. The problem underlying the invention was as set forth in the opposed patent and not the technical problem as defined in the impugned decision, which was defined with hindsight. The statement in D1 that "zeolite Nu-2 can be prepared from reaction mixtures far richer in sodium oxide than zeolite beta", would not encourage the skilled person to reduce the amount of template if he wanted to prepare zeolite beta.

After the summons for oral proceedings, which were held on 3 June 1997, the Appellant submitted a new set of claims 1 to 7 as its main and only request. New main claim 1 corresponded to claim 2 as granted.

The only independent claim 1 reads as follows:

A process for the preparation of zeolite beta, in which a reaction mixture comprising sources of sodium, aluminium, tetraethylammonium (TEA), silicon and water, utilising the molar ratio (expressed as oxide) $\text{SiO}_2/\text{Al}_2\text{O}_3$ from 10 to 200 and optionally with a seeding quantity of zeolite beta, is heated at a temperature above 75°C until zeolite beta is formed:

characterised in that the reaction mixture comprises the components in the following molar ratios (expressed as oxide):

$\text{Na}_2\text{O}/\text{TEA}_2\text{O}$ 'from 0.3 to 1,0

$\text{TEA}_2\text{O}/\text{SiO}_2$ from 0.01 to 0.1

$\text{H}_2\text{O}/\text{TEA}_2\text{O}$ from 25 to 150

and is formed by first mixing powdered sources of sodium oxide, silica and alumina (optionally with zeolite beta seeds), and without the addition of water in such a quantity that the initial mixture becomes a solution or sol, and thereafter combining the mixed powder with an aqueous solution containing TEA cations.

During oral proceedings the Appellant argued with respect to this new claim 1, that the premixing of the solid components as now claimed made it possible to reduce the water content and thereby reduce the production costs since more solid matter could be processed in the generally pressurised reactors and less caustic effluent water had to be treated. Thus more effective use could be made of the expensive equipment, and environmental investment for water treatment could be reduced. For the skilled person it was surprising that under the claimed conditions zeolite beta could be obtained at all. It was completely unexpected that it could be obtained in high purity and high yields as found by the patentee.

- V. The Respondent maintained that the subject matter of present claim 1 lacked an inventive step. The problem how to reduce the cost of zeolite beta was solved in the most obvious way by reducing the relative amount of the most expensive reaction component, i.e. the tetraethylammonium component. Moreover, since according to D2 tetraethylammonium is used in substantial excess over the equivalent amount of cation required per aluminium atom, it was obvious to the person skilled in the art that it should be possible to reduce the amount of tetraethylammonium if necessary and still obtain zeolite beta. It was further argued that the only critical requirement stated in D2 was that at least some tetraethylammonium ions must be present in the reaction mixture, which suggested that reaction compositions outside the preferred range might be used

as long as tetraethylammonium ions were present. With respect to the premixing of the solids it was argued during oral proceedings that this was only one of the two alternatives for making zeolite beta as disclosed in D2, column 4, lines 5 to 11. It might be true that the volume of water to be treated was reduced, but this water was a more concentrated caustic solution so that there was no overall saving in disposing of the effluent. The improved yield and the saving in water treatment brought forward by the Appellant were just allegations without factual basis.

- VI. The Appellant requested that the decision under appeal be set aside and the patent be maintained in amended form on the basis of the set of claims 1 to 7, submitted on 5 May 1997.

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.
2. Amended claim 1 corresponds to claim 2 as granted and amended claims 2 to 7 correspond to claims 3 to 8 respectively as granted. The amendments fulfil the requirements of Article 123(2) and (3) EPC.
3. The subject matter of the claims is new. Novelty was, in fact, no longer disputed.
4. *Inventive step*
 - 4.1 The Board agrees with the parties that D2 represents the closest prior art. It discloses a process for the preparation of zeolite beta by reacting in aqueous

media, at a temperature from about 75°C to about 200°C, amorphous silica solids or sols and a soluble aluminate along with aqueous solutions of tetraethylammonium hydroxide (TEAOH). The composition of the reaction mixture, expressed in terms of mol ratios, preferably falls within the following ranges (column 3, line 56 to column 4, line 20):

$\text{SiO}_2/\text{Al}_2\text{O}_3$ from about 10 to about 200,

$\text{Na}_2\text{O}/\text{TEAOH}$ from about 0.0 to about 0.1,

$\text{TEAOH}/\text{SiO}_2$ from about 0.1 to about 1.0,

$\text{H}_2\text{O}/\text{TEAOH}$ from about 20 to about 75.

These ranges are also mentioned in the broadest method claim 5.

The mol ratio of $\text{Na}_2\text{O}/\text{TEAOH}$ of 0.0 to 0.1 corresponds with a mol ratio of $\text{Na}_2\text{O}/\text{TEA}_2\text{O}$ of 0.0 to 0.2. On the basis of the broadest ranges disclosed in D2, the highest ratio of $\text{Na}_2\text{O}/\text{TEA}_2\text{O}$ is 0.2. The ratios actually used in the examples were much lower. From Example 2 it can be calculated that the ratio of $\text{Na}_2\text{O}/\text{TEA}_2\text{O}$ was only about 0.03 (0.3/9.5). The molar ratio of TEA_2O to the total composition, exclusive of water, in this example is 0.095 (9.5(TEA_2O) / [0.3(Na_2O) + 0.9(Al_2O_3) + 9.5(TEA_2O) + 89.2(SiO_2)]).

According to Example 1, a solution of sodium aluminate was added to a solution of TEAOH plus silica gel, which mixture was then processed whereby zeolite beta was obtained. A similar preparation of the reaction mixture is also disclosed in Example 6.

According to Example 2, aluminium turnings were dissolved in a solution of TEAOH. This solution was mixed with a silica sol also containing Na_2O .

According to Example 7 sodium aluminate was added to a solution of TEAOH. To this solution was added a silica sol. All the exemplified preparations of the reaction mixture involve the mixing of two solutions or the mixing of a solution with a sol. None of the examples discloses the yield or the purity of the obtained zeolite beta.

- 4.2 Starting from D2, the technical problem underlying the claimed invention is, in agreement with the statements in the patent in suit, considered to be a reduction of the production costs of zeolite beta. This problem is solved by the reaction composition and the mixing procedure as claimed in claim 1.

Important cost factors in the production of zeolite beta are the high price of TEAOH and the investment in equipment, especially the autoclaves for the high pressure synthesis; see the patent specification, page 2, line 51 to page 3, line 8.

The increased ratio of $\text{Na}_2\text{O}/\text{TEA}_2\text{O}$ implies a reduction in the total amount of TEAOH needed for the preparation of zeolite beta. In fact, according to Example 1 of the patent in suit the molar ratio of TEA_2O to the total composition, exclusive of water, is $0.078 (2.8(\text{TEA}_2\text{O}) / [1.2(\text{Na}_2\text{O}) + 2.8(\text{TEA}_2\text{O}) + 30.7(\text{SiO}_2) + 1(\text{Al}_2\text{O}_3)])$. This ratio is clearly lower than the calculated ratio for Example 2 of D2.

By first mixing the solid components to a powder mixture and then combining the mixed powder with an aqueous solution containing TEA cations, the amount of water introduced into the reaction system is

substantially reduced. The lowest amount of water in the examples of D2 was used in Example 7. On the basis of the weight of the components and a silica content of 30% in the silica sol (see Example 2), the weight ratio between the water and the rest of the components could be calculated. It turned out to be substantially higher than the weight ratio between water and the rest of the components calculated for Example 1 of the patent in suit. In fact, the Respondent has not contested that TEA₂O and water content could be reduced by the process of present claim 1. By reducing the amount of water, the filling of the reactor with the components forming zeolite beta can be improved. If the conversion and reaction time are not reduced by reducing the amount of TEA and water, more efficient use can be made of the reactors. With respect to the amount of SiO₂ introduced in the autoclave, the yield of the dried product in the examples of the patent in suit lies between 75% (Example 7, pure beta with a crystallinity of 83% after 6 days at 150°C) and 120% (Example 12, pure beta with a crystallinity of 82% after 7 days at 150°C). These figures show that SiO₂ was converted to a high degree into zeolite beta. Due to a lack of comparable data for the Comparative Example 1, performed according to the process of D2, it was not possible to confirm the Appellant's allegation that purity and overall yields of the products obtained with the process of the invention were substantially improved over those of the products made with the process of D2. On the other hand the Respondent has not provided evidence or convincing arguments to show that, with the process of present claim 1 the conversion of the reaction mixture into zeolite beta is inferior to that of the process of D2. Under these circumstances, the Board accepts that the yield of pure zeolite beta obtained with a process according to present claim 1 is at least as good as that of the zeolite beta obtained according to D2. This means that the reduction in TEA and water in the

reaction mixture results in a reduction of the costs for the preparation of zeolite beta. The Board is, therefore, satisfied that the said technical problem is actually solved by the claimed process.

- 4.3 It remains therefore to be decided if, for solving the above stated problem, the claimed solution would have been obvious to a person skilled in the art.

D2 does not present specific limitations as to how the components of the reaction mixture should be mixed. In column 4, lines 5 to 8 it is indicated that the method comprises reacting in aqueous media, amorphous silica solids or sols and a soluble aluminate along with aqueous solutions of TEAOH. The Board agrees with the Respondent that this statement does not exclude the mixing procedure as now claimed, but sees therein no positive indication for first forming a powder mixture of the solid components and combining the mixture with an aqueous solution of TEAOH. In all the examples of D2 an aqueous liquid containing TEAOH is mixed with another aqueous liquid containing at least one of the other components. The examples of D2 thus point away from the claimed solution. The Respondent's argument that the claimed mixing method is the only alternative for the liquid/liquid mixing disclosed in D2 cannot be accepted. Other mixing procedures could for instance be envisaged such as adding all the solid components separately to an aqueous solution of TEAOH and forming a solution or slurry thereof, or forming a solution of sodium aluminate and TEAOH in water and mixing the solution with solid silica. More important, however, is the fact that there was no indication that the claimed way of mixing could have a beneficial effect, let alone that it could solve the problem underlying the invention.

- 4.4 D1 does not provide a hint at the claimed solution

either. D1 discloses the preparation of zeolite material, designated as zeolite Nu-2, which was believed to be a member of the zeolite beta family of zeolites (page 5, lines 14 and 15). Despite the similarities in X-ray pattern, zeolite Nu-2 was considered to be distinct from zeolite beta disclosed in D2. Contrary to zeolite beta, zeolite Nu-2 showed only slight absorption for cyclohexane (page 5, line 14 to page 6, line 24). With respect to its preparation it is said that zeolite Nu-2 can be prepared from aqueous mixtures containing the oxide components and at least an alkylated quaternary ammonium compound (TEAOH); see page 10, lines 20 to 28. No specific conditions for the mixing procedure are disclosed. The examples disclose only liquid/liquid mixing. If solid components are used, they are added separately to an aqueous solution. In Example 9 of D1, solid sodium hydroxide is dissolved in an aqueous solution of TEAOH followed by the addition of alumina powder. To this aqueous mixture is then added colloidal silica (a sol). Thus D1 provides no hint for first mixing the solid components to a powder mixture and then combining the mixed powder with an aqueous solution containing TEA cations.

- 4.5 The other citations mentioned in the opposition proceedings do not contain any hint for the claimed mixing procedure either. Since with respect to the set of claims under consideration the Respondent only relied on the above-mentioned citations, it was not necessary to discuss the other documents in this decision. The claimed mixing procedure, being an essential feature of the invention does not, therefore follow in an obvious manner from the state of the art. If one essential feature of a claim is non-obvious in the context of the claim, the whole claim should be regarded as non-obvious. In the present circumstances it can, therefore, be left aside whether the other essential feature of increased $\text{Na}_2\text{O}/\text{TEA}_2\text{O}$ ratio was

obvious or not. The Board wants, however, to remark, that the Respondent's main argument, that it is obvious to reduce the most expensive component, i.e. the organic template (TEAOH), if you want to reduce the costs, is not convincing. Much research on zeolite beta has been done after the publication of D2 in 1967. Directly after its publication it should have been evident to persons skilled in the art that reduction of the organic template would reduce the costs. Nevertheless it took more than 17 years, until the priority date of the patent in suit, before the proposal came to reduce the amount of organic template below the limits indicated in the broadest method claim of D2. This indicates that the person skilled in the art thought that it was impossible to obtain zeolite beta from a reaction mixture beyond the limits of claim 5 of D2.

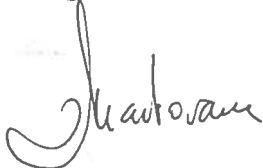
- 4.6 In view of the preceding considerations, the subject-matter of claim 1 involves an inventive step within the meaning of Article 56 EPC. Since the dependent claims 2 to 7 relate to particular embodiments of claim 1, the same applies to said claims.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the first instance with the order to maintain the patent in amended form with claims 1 to 7 as submitted on 5 May 1997, and a description yet to be adapted.

The Registrar:



P. Martorana

The Chairman:



P. A. M. Lançon

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