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D E C I S I O N
of 10 February 1999

Case Number: T 0605/96 - 3.3.5

Application Number: 88307951.9

Publication Number: 0306238

IPC: C01B 33/34

Language of the proceedings: EN

Title of invention:
ZSM-5 and its synthesis

Patentee:
Mobil Oil Corporation

Opponent:
Grace GmbH

Headword:
ZSM-5/MOBIL

Relevant legal provisions:
EPC Art. 54, 56

Keyword:
"Novelty - yes"
"Inventive step - no, arbitrary selection"

Decisions cited:
-

Catchword:
-



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Boards of Appeal

Chambres de recours

Case Number: T 0605/96 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 10 February 1999

Appellant: Grace GmbH
(Opponent) Erlengang 31
22844 Norderstedt (DE)

Representative: von Kameke, Allard, Dr.
Uexküll & Stolberg
Patentanwälte
Beselerstrasse 4
22607 Hamburg (DE)

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

Representative: Kador & Partner
Corneliusstrasse 15
80469 München (DE)

Decision under appeal: Interlocutory decision of the Opposition Division of
the European Patent Office posted 20 May 1996
concerning maintenance of European patent No. 0 306 238
in amended form.

Composition of the Board:

Chairman: R. K. Spangenberg
Members: G. J. Wassenaar
W. Moser

Summary of Facts and Submissions

I. The appeal is from the decision of the Opposition Division to maintain European patent No. 0 306 238 in amended form on the basis of claims 1 to 8, submitted on 1 February 1996 during oral proceedings. Claim 1 thereof is identical to claim 1 as granted and reads as follows:

"ZSM-5 in the form of a crystal having two dimensions of at least about 0.05 micrometer and a third dimension of less than about 0.02 micrometer, wherein the pore structure of the ZSM-5 comprises tortuous channels running substantially in the direction of said third dimensions and wherein the mesitylene sorption capacity of the ZSM-5 is at least 3 weight %."

Claim 2, which was amended with respect to claim 2 as granted, reads as follows:

"A process for preparing ZSM-5 as claimed in claim 1 comprising the steps of:

- a) providing an aqueous crystallization reaction mixture which is free of organic directing agents and which includes sources of silica, alumina and hydroxyl ions such that the solids content of the reaction mixture is at least 35 weight percent and the OH^-/SiO_2 molar ratio is at least 0.11;
- b) effecting crystallization of said reaction mixture at a temperature of 88° to 104°C (190° to 230°F) while continuously agitating the mixture; and
- c) recovering ZSM-5 crystals from the mixture."

II. In the decision, inter alia, the following prior art documents were cited:

D1: EP-A-0 202 797

D2: US-A-3 926 782

D3: EP-A-0 110 650

D5: Hydrocarbon Adsorption Characterization of Some High Silica Zeolites, Wu ea, AD-6-2, pages 547-554.

The Opposition Division held that D1 did not destroy the novelty of claim 1 because run D of Table 4 of D1 did not give any information regarding the dimensions of the particles, the type of particles and the mesitylene sorption of the particles obtained. They were not convinced that the preparation conditions in said run D would have inherently led to a product according to claim 1. They further held that the subject-matter of claim 1 involved an inventive step because the specific morphological characteristics of the product according to claim 1 were not deducible from the prior art and no document gave any indication how the claimed particles could be obtained. Claim 2 was held to be patentable because it related to a process for preparing a patentable product.

III. In the statement of the grounds of appeal, the appellant (opponent) disputed the novelty of claim 1 on the basis of D1, D2 and D3. Lack of inventive step was argued on the basis of the same documents. During oral proceedings, which were held on 10 February 1999, the

objections were maintained. The arguments put forward during the written and oral proceedings can be summarised as follows:

With respect to lack novelty it was argued that D1 disclosed a preparation of ZSM-5 under conditions fulfilling all the requirements of granted claim 2 (run D of Table 4). Since according to the patent in suit such conditions led to the product of claim 1, the same must have been the case for the product obtained by said run D. It was further argued that both D2 and D3 disclosed ZSM-5 in the form of small platelets. Included were platelets with a diameter corresponding to the requirement of claim 1 of having two dimensions of at least about 0.05 micrometer (0.1 to 0.25 micrometer in D2, ex. 9, and 0.02 to 0.1 micrometer in D3, ex. 5). At least some of said known small platelets must have had a thickness of less than about 0.02 micrometer. Since the mesitylene sorption only depended on the surface area of the platelets, the known platelets also had the mesitylene sorption capacity required by claim 1.

With respect to lack of inventive step it was argued that the product of claim 1 was merely a product, obtained by selecting preparation conditions given in D1, having no surprising properties. It was obvious to choose a high solid content and low temperatures because it was known that these conditions provided small crystals.

IV. The respondent (patentee) refuted the appellant's arguments. An affidavit was submitted to show that the technical information in D1 was contradictory so that

the reasoning put forward in the grounds of the appeal were made on an incorrect assumption as to the disclosure of D1. During oral proceedings auxiliary requests with amended claims 1 and 2 were filed. In auxiliary request 1, claim 1 was amended by the further requirement that the product was obtainable by a process comprising the steps of claim 2 of the main request as indicated above. A further auxiliary request was filed whereby the product claim (claim 1) was deleted.

With respect to granted claim 1, the respondent's arguments can be summarized as follows:

None of the cited documents disclosed ZSM-5 crystals having the morphology and properties required by claim 1. The appellant, on whom the burden of proof lay, did not demonstrate that the process of run D of D1 yielded a product within the scope of claim 1. With respect to inventive step it was argued that the claimed invention was not a selection from D1. The claimed product had a unique new structure resulting in improved catalytic properties due to enhanced diffusivity. There was no pointer in the prior art to the claimed structure. The process of claim 2 had the further advantage of producing the product of claim 1 in a reliable way without the use of an organic template. There was no indication in the prior art that the claimed combination of process features would lead to a product with the unique structure of claim 1.

With respect to the process claims it was further argued that they were not properly examined by the Opposition Division. The patentability of claim 2, was

acknowledged on the basis of the patentability of the product according to claim 1. The process of claim 2, however, involved an inventive step even if the product would not have been inventive. Thus if the patentability of the product were rejected, the case should be remitted to the first instance for proper consideration of inventive step of the process claims before two instances. A question for referral to the Enlarged Board of Appeal was submitted to challenge the competence of this Board to decide on the patentability of the process claims without a proper decision of the Opposition Division on this point.

- V. The appellant requested that the decision under appeal be set aside and that the patent be revoked.

The respondent requested that the appeal be dismissed or that the decision under appeal be set aside and that:

- (a) the patent be maintained on the basis of claims 1 and 2 submitted during oral proceedings as first auxiliary request and claims 3 to 8 received on 1 February 1996; or
- (b) that the following question be referred to the Enlarged Board of Appeal as second auxiliary request:

"Where an Opposition Division has decided that a claim of a patent is patentable on a purely formal ground (in this case, that the claim contains a reference to an earlier claim already found by the Division to be patentable), and not on the basis

of any determination in relation to the prior art of the novelty or inventiveness of that part of the subject matter of the claim which is additional to that reference: is the Board of Appeal entitled to finally decide on the ground that that part of the subject-matter of the claim is not patentable over the prior art, or must the case first be remanded to the Opposition Division for a decision on such patentability?", or

- (c) the patent be maintained with claim 2 of the first auxiliary request submitted during oral proceedings and claims 3 to 8 received on 1 February 1996 as claims 1 to 7 according to the third auxiliary request.

Reasons for the Decision

- 1. The appeal is admissible.
- 2. *Main request*
 - 2.1 Novelty
 - 2.1.1 Novelty has been disputed on the basis of documents D1, D2 and D3.

D1 relates to a method for the synthesis of zeolites and discloses that in the absence of an organic template (directing agent) ZSM-5 can be crystallized from a reaction mixture which preferably has the following molar composition:

SiO ₂ /Al ₂ O ₃	28.5 to 100
H ₂ O/SiO ₂	1 to 200
OH ⁻ /SiO ₂	0.02 to 0.4 and
M _{2/n} O/SiO ₂	0.02 to 0.5.

The crystallization of ZSM-5 can be carried out under either static or stirred conditions in polypropylene jars or in stainless steel autoclaves at 50 to 250°C for 2 to 3 hours to 150 days, preferably 5 to 100 hours. When no organic template is present, the preferred temperature is 140 to 200°C (column 6, line 1 to column 8, line 6). Under these conditions crystals of very different sizes may be obtained. Disclosed are rather coarse crystals of 1 to 5 μm (run A), medium sized crystals of 0.1 to 0.5 μm (run C) and fine crystals of 0.01 to 0.05 μm (run B); see Table 3. Specifically disclosed is also a process whereby a synthesis mixture having the molar composition

SiO ₂ /Al ₂ O ₃	29.4
H ₂ O/SiO ₂	5.05
OH ⁻ /SiO ₂	0.108
OH ⁻ /H ₂ O	0.0215

and 36.4 wt% solids is reacted at 120°C under stirring at 250 rpm for 145 hours to form ZSM-5 (run D, Table 4). No properties such as crystal size, morphology and sorption capacity have been disclosed

for run D. Considering that the value of 0.108 is equivalent to 0.11, the composition and reaction conditions of run D fulfil all the requirements which according to claim 2 as granted are necessary for obtaining the product of claim 1. The reaction temperature is also in agreement with the temperature range of 38 to 121°C mentioned in the patent in suit (page 3, lines 23 to 24). The appellant deduced therefrom that the product of said run D also must have had the same morphology and properties as the product of claim 1. The Board cannot accept this conclusion for the following reasons: Although it is not totally to be excluded that the product of said run D fulfils all the requirements of claim 1, it need not **necessarily** be so. The process parameters of granted claim 2 are obligatory conditions for obtaining the product of claim 1. They cannot be regarded as sufficient conditions which result automatically into the product of claim 1. In the case of multiple parameters it cannot be expected that any combination of parameter values leads to the desired result. This is especially the case if a combination of extreme values of the parameter ranges is chosen. In run D of D1, the values for the molar ratio of OH^-/SiO_2 of 0.108 and the temperature of 120°C are both extreme values in the corresponding ranges mentioned in the patent in suit. Moreover, the patent in suit requires a reaction time from 60 to 120 hours (page 3, lines 26 to 27), which condition is not met by run D of D1. In the absence of any proof that the product of run D of D1 had indeed the morphology and properties required by claim 1, the Board considers the subject-matter of claim 1 to be novel over D1. This issue is not affected by the respondent's submissions that temperatures above 104°C

turned out to be unsuitable for obtaining the product of claim 1, and that the OH^-/SiO_2 molar ratios given in the examples of D1 were erroneous.

2.1.2 D2 discloses that ZSM-5 crystals with a crystallite diameter of 0.005 to 0.1 μm are especially suitable as catalysts for hydrocarbon conversions (column 6, lines 12 to 34). In Example 9, the example which comes closest to the subject matter of the patent in suit, crystallites in the form of platelets having diameters of 0.1 to 0.25 μm are specifically disclosed (column 12, lines 52 to 55). The thickness of the platelets is not disclosed. There is no proof that some of the platelets have a thickness of less than 0.02 μm . D2 is also silent about the mesitylene sorption of the products. It is known in the art and accepted by the respondent that mesitylene cannot enter the pores of ZSM-5 so that only surface adsorption is possible. According to D5, even small ZSM-5 particles have a mesitylene sorption capacity of only 1.4 wt% (Table 3). The high sorption capacity of the products of the examples in the patent in suit is therefore probably due to the non-crystalline part of the products, which is at least 40%. Since the product of Example 9 of D2 consists of 100% ZSM-5, it is unlikely that it will adsorb at least 3 wt% of mesitylene.

2.1.3 D3 concerns the preparation of zeolites. Disclosed is ZSM-5 in the form of thin platelets (page 3, lines 1 to 4 and page 4, lines 14 to 21). Example 1 specifically discloses a product consisting for 100% of ZSM-5 in the form of platelet crystals of 0.2 to 1.0 μm as the maximum dimension. Here again, there is no proof that the product comprises platelets with a thickness of

less than 0.02 μm . D3 is also silent about the mesitylene sorption capacity. In Example 5 of D3 smaller ZSM-5 crystals of 0.02 to 0.1 μm are obtained. The morphology is, however, not disclosed. There is no evidence that the product of Example 5 comprises crystals in the form of platelets having a diameter of at least 0.05 μm and a thickness of less than 0.02 μm and has a mesitylene sorption capacity of at least 3 wt%. The Board, therefore, concludes that there is no evidence that any of the products according to the cited prior art fulfils all the requirements of claim 1, so that the subject-matter of claim 1 must be considered novel.

2.2 Inventive step

2.2.1 Both parties indicated that they considered D1 to represent the closest prior art. The Board agrees that D1 is a suitable starting point for the evaluation of inventive step. According to the patent in suit, the ZSM-5 produced by the present process can advantageously be employed in a variety of organic conversion reactions and in particular in the conversion of alcohols and ethers to gasoline boiling range hydrocarbons (page 4, lines 28 to 33). The patent contains no information from which it can be deduced that the product of claim 1 has a better performance in such conversions than the products of D1 or that it has any other technically relevant advantages over known ZSM-5 products. The respondent has provided no evidence showing any improvement, but has merely alleged that the product of claim 1 has a unique structure and high diffusivity. The alleged unique structure of the ZSM-5 crystals of claim 1 consisted in the tortuous channels

running substantially in the direction of the smallest dimension of the platelets. The Board cannot share this view. On the basis of common general knowledge in the field of chemistry and crystallography, and the fact that the claimed ZSM-5 is prepared by a process very close to the preparation methods of the above mentioned prior art ZSM-5 crystals, it is unlikely that the direction of the channels in the claimed crystals is different from that in the prior art products. There is no evidence for a unique structure. There is also no evidence for improved diffusivity. The high mesitylene sorption cannot be regarded as evidence for high diffusivity because, as indicated above, the mesitylene sorption must be at least partly attributed to the impurities in the claimed product.

The problem underlying the invention can therefore only be seen in the provision of a new ZSM-5 comprising product. It is evident that this problem has been solved by the product according to present claim 1.

2.2.2 It remains to be decided whether it was obvious to a person skilled in the art to solve the said problem by providing a product having the properties required by claim 1. As illustrated in the above mentioned prior art documents, the crystal morphology and crystallinity of ZSM-5 are changed by minor deviations in starting composition and process conditions. Thus, starting from D1 it was obvious for a skilled person to produce a new ZSM-5 product by operating within the general conditions mentioned in D1 for obtaining ZSM-5 thereof, but not following exactly any of the examples disclosed therein. When there is no organic template present, D1 requires a reaction mixture having preferably the

following composition:

a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 28.5 to 100,

a $\text{H}_2\text{O}/\text{SiO}_2$ molar ratio of 1 to 200,

a OH^-/SiO_2 molar ratio of 0.02 to 0.4 (column 6, lines 17 to 31). As relevant reaction conditions for obtaining ZSM-5 there are mentioned: a solids loading of preferably at least 20% up to 40% by weight and stirring at 50 to 250°C for preferably 5 to 100 hours (column 7, line 9 to column 8, line 1).

The preferred composition and reaction conditions according to the patent in suit are:

an aqueous reaction mixture free of organic directing agents, comprising a source of silica, a source of alumina and a source of hydroxyl ions having a solid content of at least 35%,

a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 25 to 50,

a $\text{H}_2\text{O}/\text{SiO}_2$ molar ratio of less than 10,

a OH^-/SiO_2 molar ratio of at least 0.11,

a crystallisation temperature of 88 to 104°C and,

a crystallisation time of 80 to 100 hours (page 2, lines 37 to 39 and page 3, lines 13 to 30).

These conditions fall within the ranges mentioned in D1 to obtain ZSM-5. Thus all the relevant composition and process conditions necessary for obtaining the product of claim 1 are indeed selected from the conditions mentioned in D1. The respondent's argument, that the present product is not a selection from D1, was based on its alleged unique structure. Since a unique structure has not been made credible, this argument must fail. Since the product obtained by the selected process conditions of the patent in suit has no surprising properties and is merely an alternative to the known products, the product of claim 1 must be considered as being the result of an arbitrary selection of known process conditions which does not involve an inventive step within the meaning of Article 56 EPC.

3. *First auxiliary request*

Claim 1 of the first auxiliary request differs from claim 1 of the main request by further indicating that the product is obtainable by a process according to claim 2 as amended during oral proceedings before the Opposition Division. The Board is unable to see any limitation of the product claim by this extension. In the absence of any further explanation in this respect from the side of the respondent, the Board holds that the scope of claim 1 of the first auxiliary request is identical to the scope of claim 1 of the main request, so that the reasons against inventive step given above equally apply to the first auxiliary request.

4. *Second auxiliary request*

Under Article 111(1) EPC a Board of Appeal has a discretion during appeal proceedings before it, either to "exercise any power within the competence of the department which was responsible for the decision appealed (here: the Opposition Division) or (to) remit the case to that department for further prosecution." In accordance with the jurisprudence of the Boards of Appeal (cf. T 79/89 [OJ EPO 1992, 283], reasons 2.2), this provision confers the power upon a Board of Appeal to act *inter alia* as the first and only instance in deciding upon a new request, without the possibility of further appellate review. Hence it follows that the question of law according to the respondent's second auxiliary request does not need to be referred to the Enlarged Board of Appeal because the Board hearing the present case considers itself able to answer it beyond any doubt by reference to Article 111(1) EPC and the above jurisprudence of the Boards of Appeal relating to that provision, ensuring thereby a uniform application of the law. For these reasons, the present Board deems a decision of the Enlarged Board of Appeal as not necessary and the respondent's second auxiliary request is refused.

5. *Third auxiliary request*

The main claim of the third auxiliary request is a process for preparing the product of claim 1 of the main request with the process steps as defined in amended claim 2 as maintained by the Opposition Division.

As already explained above (point 2.2.2), the claimed process conditions, providing a product lacking

surprising properties, are an arbitrary selection from the conditions disclosed in D1 and therefore do not involve an inventive step. The respondent's argument that the claimed process had the advantage of producing ZSM-5 crystals without an organic template in a reliable way is not relevant for inventive step since there is no evidence that the present process is in this respect more reliable than the prior art processes. On the contrary, the crystallinity of at most 60% for the examples according to the patent in suit is rather poor compared with the crystallinity of 90% indicated for prior art ZSM-5 products also obtained without organic template; see D3, Example 5.

Since there is no set of claims on file which fulfils the requirements of the EPC, the patent cannot be maintained.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The patent is revoked.

The Registrar:

The Chairman:

S. Hue

R. Spangenberg