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D E C I S I O N
of 1 October 1998

Case Number: T 0267/95 - 3.3.5

Application Number: 85301045.2

Publication Number: 0156490

IPC: C10G 11/05

Language of the proceedings: EN

Title of invention:
ZSM-5 catalytic cracking process using large size ZSM-5 crystals

Patentee:
Mobil Oil Corporation

Opponents:
VAW Vereinigte Aluminium-Werke AG, Berlin und Bonn
Grace GmbH

Headword:
ZSM-5 cracking process/MOBIL

Relevant legal provisions:
EPC Art. 54, 56

Keyword:
"Main request: Novelty (no); disclosure of a document containing a reference to another document"
"Auxiliary request: Inventive step (no); arbitrary selection"

Decisions cited:
T 0153/85, T 0548/94

Catchword:

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0267/95 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 1 October 1998

Appellant: (Opponent)
VAW
Vereinigte Aluminium-Werke AG, Berlin und Bonn
Georg-von-Boeselager-Str. 25
53117 Bonn (DE)

Representative:
Müller-Wolff, Thomas, Dipl.-Ing.
Harwardt Neumann
Patent- und Rechtsanwälte
Postfach 14 55
53704 Siegburg (DE)

Other party: (Opponent)
Grace GmbH
Erlengang 31
22844 Norderstedt (DE)

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

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

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

Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted 14 March 1995 rejecting the opposition filed against European patent No. 0 156 490 pursuant to Article 102(2) EPC.**

Composition of the Board:

Chairman: R. K. Spangenberg
Members: G. Dischinger-Höppler
J. H. van Moer

Summary of Facts and Submissions

I. The appeal is from the Opposition Division's decision rejecting two oppositions against European patent No. 0 156 490, relating to a cracking process using large-size ZSM-5 crystals, which was granted with eight claims.

II. The Opposition Division considered 25 documents and held that in the cracking catalysts containing ZSM-5 disclosed in the prior art, the crystal sizes of the ZSM-5 generally ranged from 0.02 to 100 μm. In comparison, the crystal size of 0.2-5 μm in granted Claim 1 was considered as a selection which conferred novelty on the subject-matter of the patent in suit. Further, the examples of the patent in suit demonstrated that in comparison with a process using ZSM-5 of much smaller crystal size an increased octane number was obtained, in particular after steaming. None of the cited prior art documents gave any hint that such an improvement was possible by using large ZSM-5 crystals. Therefore, inventive step was acknowledged.

III. Both Appellants (Opponents) lodged an appeal against this decision. Oral proceedings were held on 1 October 1998, during which the Respondent (Proprietor) submitted an amended Claim 1 as a main request and a further amended Claim 1 as an auxiliary request. Claim 1 of the main request differs from granted Claim 1 in that it has been restricted to the use of an additive catalyst "consisting of" instead of "comprising" a zeolite dispersed in a matrix and in

that the crystal size range has been restricted to 0.2 to 0.5 μm. It reads as follows:

"1. A process for the catalytic cracking of a petroleum fraction in the presence of a cracking catalyst under cracking conditions, during which process coked catalyst is formed and is passed through a regenerator wherein the coke deposits are burned with formation of steam, carbon oxides and hot regenerated catalyst, characterised by adding to the cracking catalyst from 0.1 to 50 weight percent based on cracking catalyst of an additive catalyst consisting of a zeolite dispersed in a matrix, said zeolite having a constraint index of 1 to 12, a silica to alumina mole ratio greater than 12, and a crystal size of 0.2 to 0.5 μm (microns)."

Claim 1 of the auxiliary request differs therefrom in that the cracking catalyst has been defined to be one "which is selected from zeolite X, zeolite Y and naturally occurring faujasite or which is amorphous".

Of the documents cited in the opposition proceedings, only the following remain relevant to this decision:

D1: US-A-4 309 279
D3: US-A-3 702 886
D7: US-A-3 926 782
D8: EP-A-0 021 674
D9: EP-A-0 026 962 and
D10: EP-A-0 026 963.

IV. The Appellants contended that the crystal size of 0.2-0.5 μm was insufficiently defined and not suitable for distinction because crystal sizes generally depended on the method of determination, and because it was unclear whether the size was valid for any direction of the crystals.

They further submitted that the subject-matter of Claim 1 of the main request lacked novelty over the disclosure of D1, wherein reference was made to D3 with respect to the ZSM-5 class zeolite to be used in the additive catalyst, since the ZSM-5 synthesized in D3 had a crystal size within the claimed range. This was confirmed in D8 to D10. In addition, Claim 1 of the main request did not specify the cracking catalyst used, which could consequently contain any catalyst suitable for cracking and hence covered the presence of further ZSM-5 of any crystal size dispersed in a matrix. Such a zeolite could not, however, be distinguished from a zeolite added with the additive catalyst. The presence of ZSM-5 crystals outside the claimed range was, therefore, not excluded.

Concerning Claim 1 of the auxiliary request, lack of inventive step in view of D1 as the closest prior art was asserted. This claim proposed for the purpose of providing an improved additive catalyst for a cracking process, a ZSM-5 having a narrower crystal size range. However, no improvement was shown over the process taught in D1. Moreover, it was generally obvious to test different types of ZSM-5 in order to find the most suitable one, because it was known that too large crystals were less resistant to attrition and because

it was important for a catalyst in a cracking process to be stable in the presence of steam. Moreover, if in accordance with the Proprietor's assertion, there existed only two ranges of crystal size in commercial ZSM-5 products at the time of D1, the skilled person had only the choice between these two types of ZSM-5 for reworking the process of D1.

V. According to the Respondent, Claim 1 on its proper construction required that all dimensions of the crystal are within the claimed size range of 0.2 to 0.5 μm. Since the range was defined as an absolute physical value, the method of measuring was not important for definition, but for a determination of the size the most accurate method, ie scanning electron microscopy (SEM), not sorption, must be used.

Concerning novelty, it was pointed out that D1 did not indicate any specific crystal size. Further, the reference to D3 was merely for the purpose of identifying the class of zeolites in the sense of X ray diffraction characteristics. It was not allowable to construe such references as forming part of the disclosure of D1. The claimed subject-matter was in the Respondent's opinion in fact novel over the teaching of D1 because the skilled person had the possibility to select a particular ZSM-5 from numerous different documents. Even if he had only considered D3, he had the possibility to select from 27 examples. Only three of these mentioned a crystal size and none was definitely within the claimed range. Moreover, the disclosure of D8 to D10 was irrelevant for the purpose of assessing novelty. Further, the additive catalyst

was composed of a particular zeolite glued in a matrix and, therefore, distinguishable from any other zeolite present in catalyst mixture.

As to inventive step, the Respondent argued that the problem in view of D1 was to obtain in a catalytic cracking unit an octane gain over a prolonged period of time on stream. The solution consisted in increasing the hydrothermal stability by using in the additive catalyst solely a zeolite having a crystal size of 0.2-0.5 μm. The superiority of the claimed process using such a large size ZSM-5 over a process using small crystals was evident from the examples given in the patent specification. D1 did not mention a particular crystal size. The skilled person had, however, several reasons to select a small crystal size ZSM-5 for the purpose of D1, because of the inherent advantages concerning increased diffusion rate and reaction time on the one hand and reduced ageing and attrition on the other. The prior art did not provide any guidance which led to the significance of the crystal size for hydrothermal stability of a zeolite catalyst under the conditions in an FCC unit.

VI. The Appellants requested that the decision under appeal be set aside and that the patent be revoked.

The Respondent requested that the appeal be dismissed and the patent be maintained on the basis of the main or auxiliary request submitted during the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

2. In the Board's judgment there are no objections under Article 123(2) and (3) EPC with respect to the amended independent claims of the main and auxiliary requests. Since this was not contested by the Appellants, there is no need to give reasons for this finding.

3. *Main Request*

Novelty was attacked on the basis of D1. This document discloses a process for catalytic cracking of petroleum fractions in the presence of a conventional cracking catalyst comprising a zeolite as the catalytically active component, wherein a separate additive catalyst comprising a zeolite having a Constraint Index of 1-12 and a silica to alumina mole ratio greater than 12 is added to said conventional cracking catalyst (see Claim 1). This process as well as that according to the contested patent are performed in FCC-units, wherein a fluidized bed of catalyst circulates between the FCC reactor and the FCC regenerator. The purpose of the regenerator is to burn away coke deposited on the catalyst (see the patent in suit, page 3, lines 4 to 14 and examples; in D1, Claim 8, column 4, lines 28/29, column 6, lines 10 to 14 and examples). The features concerning regeneration of coked catalyst by burning with formation of steam, carbon oxides and hot regenerated catalyst are thus inherent in the process

known from D1. This interpretation of the disclosure of D1 was not disputed.

According to the examples of D1 the additive catalyst is composed of 25 wt% of commercially manufactured low sodium type ZSM-5 in a matrix (see Example 2) and added to the conventional cracking catalyst in an amount of 0.1 wt% based on the commercial cracking catalyst (see Example 4). As a consequence, no distinction between the process of D1 and that according to the patent in suit can be derived from the amount of additive catalyst.

Therefore, D1 discloses a process as claimed in Claim 1 of the main request, with the exception that no crystal size is mentioned. However in respect of ZSM-5, D1 mentions D3, the content of which is incorporated therein by reference (see column 10, lines 39 to 44).

The Respondent argued that in D1 the purpose of this cross-reference to D3 was merely for identifying a suitable class of zeolites in the sense of X-ray diffraction characteristics and not for identifying a specific zeolite and a process for obtaining this zeolite. In addition, he submitted that D1 contained a large number of references to different documents, so that it was absurd to assume that the disclosure of D1 included the content of all these documents. In his opinion, for the assessment of novelty the disclosure of D1 had to be considered in isolation, as was made clear in T 153/85 (OJ 1988, 1, reasons No. 4.2).

The Board agrees that it is only the actual content of

a document which may destroy novelty and that it is not permissible to combine for this purpose separate items of prior art. However, in a case where there is a specific reference in a first prior art document to a second prior art document, the presence of such specific reference may make it permissible to consider a part or the whole of the disclosure of the second document as being part of the disclosure of the first document. In T 153/85 (see reference above) it was decided that the teaching of a first document had to be construed to imply a particular method according to a second document, because it was specified in the first document that copolymers described therein "may be conveniently prepared" by said method. In the present case, D1 prefers ZSM-5 as the zeolite in the additive catalyst (see Claim 4 and column 11, lines 1 to 4). Only this type of zeolite is used in the examples where it is identified as a commercially manufactured low sodium type. D1 itself does not disclose how this ZSM-5 can be obtained, so that it is generally at the disposal of the reader of D1 to use any suitable prior art ZSM-5 product. However, the skilled person trying to rework the examples of D1 is in this respect expressly referred to D3. In addition, the ZSM-5 products of D3 are exactly those which are suitable for the examples of D1. As a consequence, it is, in the Board's judgment, certainly not appropriate to regard the purpose of said reference to D3 merely as a means for identifying a suitable class of zeolites and not as a disclosure of specific zeolites suitable for performing the process of D1. The Board holds, therefore, that it is legitimate in the present case to replace the reference to D3 in D1 by the relevant

content of that document. Having regard to the above circumstances, the fact that D1 incorporates by reference a number of further documents concerning inter alia other types of suitable zeolites (see column 10, lines 39 to 62) is not relevant to the proper assessment of the disclosure of D1 for the purpose of answering the question whether or not Claim 1 of the patent in suit covers subject-matter made available to the public by D1.

The crystal size of the ZSM-5 product is mentioned on three occasions in D3: in Example 2, where "microscopic examination showed the presence of very small crystals (of the order of 1 micron)"; in Example 26, where "microscopic examination showed mainly crystalline material < 1μ" and in Example 27, according to which "microscopic examination showed mainly large rod-shaped crystals to 8 x 20μ, some large cubes to 25μ". In agreement with the Respondent's view, D3, and therefore also D1, does not mention a crystal size range of exactly 0.2 to 0.5μm as claimed in the contested patent.

However, as correctly observed by the Appellants, the type of cracking catalyst to be used in the claimed process is not defined. Consequently, Claim 1 of the patent in suit covers the possibility to use any cracking catalyst. The Respondent did not contest that ZSM-5 of any crystal size was suitable for this purpose, but expressed the opinion that the additive catalyst was distinguishable insofar as it contained ZSM-5 of the above crystal size range within a matrix,

the latter having the effect of a glue surrounding the zeolite crystals. The Board is unable to accept this argument, since Claim 1 does not define whether or when such an effect of the matrix comes into action, but merely defines the additive catalyst to consist of a zeolite dispersed in a matrix. According to Claim 1, the additive catalyst can be added together with the cracking catalyst and need not be present in the form of particles separate from those of the cracking catalyst. Therefore, the particles of both catalysts may be dispersed in the same matrix, so that the Board agrees with the Appellant's submission that the catalyst mixture used in Claim 1 of the contested patent cannot be distinguished from any catalyst system comprising ZSM-5 of a crystal size which covers the claimed range and a matrix.

Concerning the contentious definition of the crystal size, the patent in suit fails to indicate any method for its determination and makes no distinction between the various dimensions of the crystals. Hence, no information is given concerning the accuracy of the claimed range of crystal size. The Board holds, therefore, that the accuracy of any prior art method for determining crystal sizes may be applied accordingly. As pointed out by the Appellants, D3 is a basic document concerning ZSM-5, to which reference is made in several later-published documents such as D8 to D10. In all these documents, which were published in the Respondent's name, D3 is referred to as disclosing an "average particle size diameter of less than 1/2 micron" (see in D8, page 1, lines 20 to 23; in D9, page 1, lines 21 to 23; in D10, page 1, lines 23 to

26). According to common linguistic usage, this expression clearly indicates that at least particles up to 0.5 μm must be present, since otherwise an average particle size below this value would be impossible. Contrary to the Respondent's opinion and although the terms "particle" and "crystal" usually have a different meaning, as was for example evident from D7 (see Example 9), the Board is satisfied that these terms are used synonymously in D8 to D10 (see in D8, page 2, lines 27 to 29 and page 5, lines 28 to 30; in D9, page 1, lines 23 to 26 and Claim 1; in D10, page 1, lines 26 to 31). The terms actually used in Examples 2 and 26 of D3 which indicate that crystals smaller than 1 μm in diameter must be present, and the statement in D3 that crystal size depends on the nature of the reaction mixture (column 6, lines 13 to 16) are not in contradiction thereto. Nevertheless, the Board agrees with the Respondent's argument that the microscopic methods at the time of D3 have in all probability been less accurate than the methods used later, such as scanning electron microscopy (SEM), so that the values given in D3 might be less precise. What matters in respect of the relevant question of novelty is, however, **not what is said** in D3 about the crystal size, but only which crystal size **was in fact obtained** as a result of performing the process disclosed in D3. The exact range of crystal sizes so obtained can be proved at any time after the publication of D3 by any kind of suitable evidence, including the references to D3 contained in D8, D9 and D10. The Board holds that the evidential weight of these documents is very high, since at the time of their publication the more

accurate method of SEM for the determination of the crystal size was available, and since these documents as well as D1 and D3 were published in the name of the Respondent, who should have the best knowledge of the relevant facts. This being taken into account, the Board cannot accept the Respondent's argument that by being published 10 years later D8 to D10 might be wrong with respect to the crystal sizes and could not, therefore, be used to establish the true crystal size obtained according to D3.

On this basis, the Board is convinced that the average crystal size of the products obtained according to D3 is correctly indicated in D8 to D10 which, by the way, were already available at the priority date of the patent in suit. The Board holds, therefore, that at least the ZSM-5 products prepared according to Examples 2 and 26 of D3 contain crystals within the claimed range. Further, as pointed out above, the disclosure of these products forms part of the relevant disclosure of D1. Consequently, the process according to Claim 1 of the main request of the patent in suit is not novel over the disclosure of D1, so that this request must fail.

4. *Auxiliary Request*

Unlike in Claim 1 of the main request, the cracking catalyst in Claim 1 of the auxiliary request does not contain crystalline ZSM-5, so that the presence of ZSM-5 having a crystal size outside the range of 0.2 to 0.5 μm is excluded. This claim was only attacked on the basis of inventive step in view of D1. It is undisputed

that this document represents the closest prior art.

As indicated in point V above, the Respondent submitted that in respect of this prior art the problem existed of obtaining by a catalytic cracking process a product having an increased octane number for a prolonged period of time on stream. However, this problem can only be taken into account for the assessment of inventive step if it is credible that it has in fact been solved by the claimed process. This being disputed by the Appellant, the Board has to examine that matter.

In the Respondent's view it was evident from the results shown in Tables 2 to 5 of the contested patent that the technical surplus provided by the use of a catalyst system containing ZSM-5 of a crystal size of exclusively 0.2-0.5 μm in accordance with the claimed process over the process according to D1 consisted in an increase in the gasoline octane number over a prolonged period of time on stream. Nevertheless, it was admitted by the Respondent that no comparisons have been made with embodiments exactly according to D1, taking into account the crystals sizes of D3. He argued that this was due to the fact that D1 (including D3) did not attach any importance to the crystal size and left various possibilities open. It was, therefore, in any case necessary to make a choice. A skilled person would, in the Respondent's view, in this case select a small crystal size because maximum conversion can be obtained very quickly as a result of the large target provided by the large surface area and of the minimum residence time within the pores of small crystals. In addition, small crystals were more resistant to ageing

and attrition. Therefore, comparisons have been made with those modifications which were outside the claimed subject-matter and would have been used by a person skilled in the art.

In fact, all the examples reflected in the above tables 2 to 5 show comparisons between a catalyst system with ZSM-5 having a crystal size within the claimed range of 0.2 to 0.5 μm in the additive catalyst and another system containing ZSM-5 of the much smaller crystal size of 0.02 to 0.05 μm. The preparation of these two kinds of additive catalysts is described in Examples 1 and 2. However, as correctly noticed by the Appellants, these methods of preparation differ with respect to the manufacturing of the matrices, the extent of the final ion exchange after addition of the respective ZSM-5 and the ion exchange solutions used therefor. It is, therefore, prima facie not possible to ascribe the different results presented in said tables solely to the different crystal sizes as a result of their superior hydrothermal stability as alleged by the Respondent. As a consequence, it is of no importance whether or not hydrothermal stability of a zeolite had been already linked in the art with the crystal size parameter. Concerning Tables 2 and 3, these circumstances are further aggravated by the fact that, in addition, different cracking catalysts (base catalysts) were used. The Respondent contested any effect of such differences on the performance of the catalyst. However, such an assertion is, in the absence of any support by evidence, in the present situation not sufficient for rebutting the Appellant's

objections, since as a matter of principle comparisons must be such that it is convincingly shown that any effect has its origin solely in the distinguishing feature (see Case Law of the Boards of Appeal of the European Patent Office, 1986, Chapter I, D-7.7.2).

Consequently, there is in the present case no reliable evidence that the problem addressed by the Respondent has in fact been solved, so that it cannot be taken into account for the assessment of inventive step. The Board considers, however, that the relevant problem in view of the disclosure of D1 can be seen in providing a further process for the catalytic cracking of petroleum. As indicated in point 3 above, D1 teaches a process according to present Claim 1 with the exception that the zeolites suitable in the additive catalyst are not restricted to a particular crystal size. Accordingly, Claim 1 of the auxiliary request proposes to select from these zeolites those which have a crystal size ranging from 0.2 to 0.5 μm. As is evident from the examples, the above problem has actually been solved by this feature.

It remains, therefore, to be decided whether or not such a selection was obvious for someone skilled in the art in order to provide such a further process.

As set out above, the selected range does not provide any effect which was hitherto unknown and unexpected. Further, in view of D8 to D10 a skilled person would have recognised that the crystal size of the ZSM-5 used in the examples of D1 was already of the same order of magnitude (see point 3 above). Picking out, within this order of magnitude, the particular range claimed, represents in the Board's judgment merely an arbitrary selection, which cannot involve an inventive step (see T 548/94 of 18 May 1998, reasons Nos. 2.5 and 2.6). Therefore, the Board concludes that the claimed process represents a hitherto undisclosed but obvious embodiment of the process disclosed in D1.

For this reason, the auxiliary request must also fail.

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