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Datasheet for the decision
of 29 September 2015

Case Number:	T 1159/12 - 3.3.06
Application Number:	08786527.5
Publication Number:	2175991
IPC:	B01J29/06, B01J29/18, B01J29/40, C07C4/06, C07C4/10, C10G11/05
Language of the proceedings:	EN

Title of invention:
CRACKING OF OLEFINS ON PHOSPHORUS MODIFIED MOLECULAR SIEVES

Applicant:
TOTAL RESEARCH & TECHNOLOGY FELUY

Headword:
Cracking of olefins on P-modified molecular sieves/ Total Research & Technology Feluy

Relevant legal provisions:
RPBA Art. 13(3), 12(1)
EPC Art. 84, 54(1), 123(2), 56

Keyword:
Late-filed auxiliary requests - admitted (yes) - occasioned by the Board's communication and the debate during oral proceedings
Claims - clarity (no) - Main Request
Amendments - allowable (no) - 1st Auxiliary Request
Novelty - (no) - 2nd Auxiliary Request
Inventive step - (no) - obvious solution - 3rd Auxiliary Request

Decisions cited:

Catchword:

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Case Number: T 1159/12 - 3.3.06

DECISION
of Technical Board of Appeal 3.3.06
of 29 September 2015

Appellant:
(Applicant)
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted on 23 December 2011 refusing European patent application No. 08786527.5 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman E. Bendl
Members: G. Santavicca
S. Fernández de Córdoba

Summary of Facts and Submissions

I. The appeal lies from the decision of the Examining Division to refuse European patent application No. 08786527.5 (European publication number 2 175 991).

II. In the decision under appeal, the Examining Division came to the conclusion that the Olefin Cracking Process (OCP) defined in Claim 1 of the Main Request filed during the oral proceedings held on 20 October 2011 lacked an inventive step over D1 (Zhao Guoliang et al, "*Effect of phosphorous on HZSM-5 catalyst for C_4-olefin cracking reactions to produce propylene*", Journal of Catalysis 248 (2007) 29-37, available online at www.sciencedirect.com from 5 April 2007), taken as the closest prior art, in combination with D2 (EP 1 797 951 A1).

III. With its statement setting out the grounds of appeal, the Appellant (re)submitted the claims dealt with in the decision under appeal and filed experimental results. Claim 1 according to the Main Request reads as follows (amendments to Claim 1 as originally filed made apparent by the Board):

"*1. Process for the catalytic cracking of an olefin-rich feedstock* ***comprising 10 to 100% olefins*** *which is selective towards light olefins in the effluent, the process comprising contacting a hydrocarbon feedstock containing one or more olefins, with a catalyst made of a phosphorus-modified zeolite (A), to produce an effluent with an olefin content of lower molecular weight than that of the feedstock, wherein said phosphorous modified zeolite (A) is made by a process comprising in that order:*

- selecting a zeolite with low Si/Al ratio among H^+ or NH_4^+-form of MFI, MEL, FER, MOR, clinoptilolite;
*- steaming at a temperature ranging from ~~400~~**420** to 870°C for ~~0.01~~**0.05**-200h;*
*- leaching with an aqueous acid solution containing the source of P at conditions effective to remove ~~a substantial part~~ **at least 10.0%** of Al from the **parent** zeolite and to introduce at least 0.3 wt% of P;*
- separation of the solid from the liquid;
- an optional washing step or an optional drying step or an optional drying step followed by a washing step;
- a calcination step,
Wherein the leaching period is 0.5 to 24 hours and the aqueous acid solution containing the source of P in the leaching step has a pH of 3 or lower.".

IV. In a communication in preparation for oral proceedings, dated 21 July 2015, the Board conveyed its provisional opinion on some of the issues to be dealt with and *inter alia* raised objections under Article 84 EPC against the feature "*low Si/Al ratio*".

V. With letter dated 26 August 2015, the Appellant replied to the provisional opinion and the objections raised by the Board and submitted a set of further amended claims as its Auxiliary Request. Claim 1 of the Auxiliary Request contains amendments as follows (amendments in comparison to Claim 1 as originally filed made apparent by the Board):

"*1. Process for the catalytic cracking of an olefin-rich feedstock* ***comprising 10 to 100% olefins*** *which is selective towards ~~light olefins~~* ***ethylene and propylene*** *in the effluent, the process comprising contacting a hydrocarbon feedstock containing one or more olefins* ***at a LHSV ranging from 0.5 to 30 hr^{-1}, at a total absolute***

pressure in the reactor ranging from 0.5 to 10 bars and at an inlet temperature of the feedstock of from 400 to 650°C, *with a catalyst made of a phosphorus-modified zeolite (A), to produce an effluent with an olefin content of lower molecular weight than that of the feedstock, wherein said phosphorous modified zeolite (A) is made by a process comprising in that order:*

- *selecting a zeolite with ~~low~~* ***an initial*** *Si/Al* ***atomic*** *ratio* ***ranging from 4 to 30*** *among* H^+ *or* NH_4^+*-form of MFI, MEL, FER, MOR, clinoptilolite;*
- *steaming at a temperature ranging from ~~400~~**420** to 870°C for ~~0.01~~**0.05**-200h;*
- *leaching with an aqueous acid solution containing the source of P at conditions effective to remove ~~a substantial part~~* ***at least 10.0%*** *of Al from the* ***parent*** *zeolite and to introduce at least 0.3 wt% of P;*
- *separation of the solid from the liquid;*
- *an optional washing step or an optional drying step or an optional drying step followed by a washing step;*
- *a calcination step,*

Wherein the leaching period is 0.5 to 24 hours and the aqueous acid solution containing the source of P in the leaching step has a pH of 3 or lower.".

VI. Oral proceedings were held on 29 September 2015. The debate focused on clarity of Claim 1 according to the Main Request and the allowability of the amendments under Article 123(2) EPC made in Claim 1 according to the Auxiliary Request. The Appellant then filed two sets of further amended claims, respectively labelled 2nd and 3rd Auxiliary Requests. The subsequent debate focused on the issues of novelty over D1 and inventive step over D1, taken as the closest prior art, in combination with D2.

VII. Claim 1 according to each of 2nd and 3rd Auxiliary Requests respectively contains the following further amendments, compared to Claim 1 according to the Auxiliary Request (now 1st Auxiliary Request):

2nd Auxiliary Request

"*1. Process for the catalytic cracking of an olefin-rich feedstock comprising 10 to 100% olefins which is selective towards* ***~~ethylene and~~ propylene*** *in the effluent, ...*".

3rd Auxiliary Request

"*1. Process for the catalytic cracking of an olefin-rich feedstock comprising 10 to 100% olefins which is selective towards* ***~~ethylene and~~ propylene*** *in the effluent, ...;*
- separation of the solid from the liquid ***by filtration;***
...".

VIII. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the claims according to the Main Request submitted with the statement setting out the grounds of appeal or, in the alternative, on the basis of the claims according to the Auxiliary Request (now 1st Auxiliary Request) submitted with letter of 26 August 2015, or on the basis of the claims according to the 2nd or 3rd Auxiliary Requests, both filed during oral proceedings.

IX. The Appellant's arguments of relevance for the present decision can be summarised as follows:

Main Request

Clarity - Article 84 EPC

The feature "*low Si/Al ratio*" in Claim 1 was clear, as it was known to the skilled person that this ratio was advantageously below 30, as disclosed in the application as originally filed.

1st Auxiliary Request

Article 123(2) EPC

The amendment "*selective towards ethylene and propylene*" in Claim 1 was based on the application as originally filed (reference was made to Pages 13, lines 2-3, and 15, lines 6-7). Although the objective defined on Page 3, lines 7 and 19, focused on propylene, it was impossible to exclude ethylene from the low olefins contained in the effluent of an olefin cracking process (OCP) reactor. Thus, the claims were in compliance with Article 123(2) EPC.

2nd Auxiliary Request

Novelty

The catalyst prepared by using the preparation method defined in Claim 1 differed from the catalyst according to D1, for the following reasons:

1. A steaming was performed before treatment with phosphoric acid, whereas in D1 there was no steaming. In this respect, the experimental results submitted with the statement setting out the grounds of appeal not only showed that the duration of the steaming step

was relevant, but also that a steaming before treatment with phosphor compounds removed aluminium from the framework, and produced extra framework alumina, which was then removed by leaching. Thus, these steps of the preparation according to Claim 1 at issue necessarily led to a different catalyst, compared to D1.

2. In the preparation method of Claim 1 at issue, the introduction of phosphor followed a different pathway, compared to that of D1. According to the claimed invention the step of leaching of the zeolite was followed by a step of separation of the solid from the liquid. Instead, in D1 the solid was merely impregnated with the solution containing the desired amount of phosphor, then water was removed by evaporation. Thus, Al nevertheless remained on the zeolite, which was only subsequently steamed. Hence, at least because of the large quantity of phosphorous compound used in the claimed preparation method, the method of D1 could not lead to a catalyst as claimed, i.e. could not remove at least 10% of Al species from the parent zeolite.

3. The catalyst of D1 was extruded with 50 wt% of silica (as a binder) prior to the introduction of phosphor. Hence, part of the phosphorous compound used would stay on the silica, rather than coming in contact with the zeolite. Thus, the catalyst as disclosed by D1 was definitely not the same as the claimed catalyst.

3rd Auxiliary Request

Inventive step

As regards inventive step, D1 described the closest prior art but concerned the cracking of olefins over modified ZSM-5 zeolites in an OCP reactor.

The problem solved by the present invention was an improvement in the yield of propylene and in the purity of the C_3-cut. This had been shown by the experiments submitted.

D2, which described the cracking of saturated hydrocarbons on modified zeolites in Fluid Catalytic Cracking (FCC) processes, had nothing to do with olefin cracking in an OCP process. It disclosed the use of an additive catalyst, which thus should be used together with the main FCC catalyst. Although D2 disclosed that ZSM-5 zeolite might crack olefins, it then taught to modify these zeolites. However, the modification with phosphor aimed to improve the hydrothermal stability. Instead, in the claimed process, the catalyst was used alone, and the introduction of phosphor aimed to improve the selectivity towards propylene. Since the skilled person found no hint in D2 to use the additive catalyst for the FCC process in an OCP process, he had no motivation to combine D1 with D2.

Consequently, the claimed process was not obvious.

Reasons for the Decision

Main Request

Admissibility

1. The claims according to the Main Request are identical to those of the Main Request dealt with in the decision under appeal. The Main Request is thus admissible (Article 12(1)(a) RPBA).

Allowability of the amendments

Clarity - Article 84 EPC

2. As highlighted in the Board's preliminary opinion, the feature "*low Si/Al ratio*" in Claim 1 at issue does not have a generally recognised univocal meaning, let alone the one invoked by the Appellant. In this respect, the application as originally filed (Page 6, lines 17-19; paragraph bridging pages 6 and 7) merely states a preference for the particular ratio disclosed, without acknowledging any generally recognised meaning. Hence, the term "*low*" as such is undefined.

2.1 Further, it is not apparent what Si/Al ratio (initial or final? molar or atomic?) is meant. In this respect, the application as originally filed (Page 6, lines 18 and 31, merely mentions an initial atomic ratio.

2.2 This lack of clarity does not enable a skilled person to decide whether or not a catalytic cracking process using a given catalyst falls within the process according to Claim 1. For instance the comparison with D1, which discloses a P-modified ZSM-5 Zeolite with an initial SiO_2/Al_2O_3 molar ratio of 40 is not reasonably possible.

2.3 Thus, Claim 1 according to the Main Request lacks clarity (Article 84 EPC).

2.4 Consequently, the Main Request is not allowable.

1st Auxiliary Request

Admissibility

The 1st Auxiliary Request was filed in reaction to the communication of the Board, raising new objections, e.g. under Article 84 EPC. Thus, the Board admits this request into the proceedings (Article 12(1)(c) RPBA).

Allowability of the amendments

Clarity - Article 84 EPC

3. The Board is satisfied that the Appellant succeeded to overcome the clarity objections discussed *supra* by amending the objected wording of Claim 1 to "*initial Si/Al atomic ratio*". Given the grounds of refusal relating to the requirements of Article 123(2) EPC *infra*, a more detailed reasoning is not deemed to be necessary in this respect.

Article 123(2) EPC

4. Claim 1 according to the 1st Auxiliary Request *inter alia* comprises the following amendment, compared to the respective claim of the application as filed: "*selective towards ethylene and propylene*".

4.1 The argument by the Appellant that this amendment was based on the application as filed (reference was made to Pages 13, lines 2-3, and 15, lines 6-7, and 13), is not convincing, for the following reasons:

4.1.1 The passage on Page 13, lines 2-3, explains that in a specific embodiment of the invention the effluent of an XTO reactor (i.e. an organics-to-olefins reactor) may be used in part or completely as feedstock for the process of the invention. This effluent *inter alia* comprises light olefins, whereby "*light olefins*" means ethylene and propylene. The passage does not address selectivity at all, let alone in OCP reactors.

4.1.2 The passage on Page 15, lines 6-7, has to do with the separation/recovery of compounds contained in the effluent from the OCP reactor, *inter alia* comprising light olefins, i.e. ethylene and propylene. Also this passage does not address selectivity towards ethylene.

4.1.3 Instead, the passage on Page 15, lines 12-14, reading "*in order to adjust the propylene to ethylene ratio, ethylene in whole or in part can be recycled over the OCP reactor and advantageously converted into more propylene*" unambiguously teaches that propylene rather than ethylene is the desired objective.

4.1.4 The argument of the Appellant that it was impossible to exclude ethylene from the low olefins in the effluent of an olefin cracking process (OCP) reactor is out of point, as selectivity in the context at issue does not mean the exclusive production of propylene.

4.2 Moreover, if the objectives defined in the application as filed are considered, as disclosed e.g. on Page 2, line 7, Page 3, lines 7 and 19, Page 9, lines 29-30 it is apparent that they only focus on propylene selectivity.

4.3 This is in particular apparent from the examples of the application as originally filed, all of which illustrate the high propylene/ethylene ratio obtainable by the claimed process, and the low amount of ethylene produced.

4.4 Therefore, the claims of the 1st Auxiliary Request are not in compliance with Article 123(2) EPC.

4.5 Consequently, the 1st Auxiliary Request is not allowable.

2nd and 3rd Auxiliary Requests

Admissibility

5. The 2nd and 3rd Auxiliary Requests were filed in reaction to the debate during the oral proceedings. They did not raise issues which the Board could not immediately deal with, nor required adjournment of the oral proceedings. Thus, despite their late filing, the 2nd and 3rd Auxiliary Requests were admitted into the proceedings (Article 13(3) RPBA).

2nd Auxiliary Request

Allowability of the amendments

Articles 123(2) and 84 EPC

6. The 2nd Auxiliary Request is identical to the 1st Auxiliary Request apart from the further amendment consisting of the deletion of the expression "*ethylene and*". This further amendment is considered to be clear and overcomes the objection raised by the Board under Article 123(2) EPC, namely that the application as filed merely discloses a selectivity towards propylene (*supra*, Point 4). Taking into account the reasoning concerning novelty *infra*, a more detailed argumentation as to these requirements is not deemed to be necessary.

Novelty

7. In the decision under appeal novelty was only conditionally acknowledged over the disclosure of D1.

7.1 It was acknowledged that the process of preparation of the catalyst defined in Claim 1 then at issue differed from the known process described in Dl in that it comprised a steaming step at a temperature from 420°C to 870°C and for 0.05 to 200 h, <u>before leaching with an aqueous acid solution of phosphoric acid</u> (emphasis added by the Board). However, although the processes of preparation according to Claim 1 then at issue and that disclosed in Dl were different, there was no evidence that the final catalyst therefrom would be different. This was because also the catalyst of Dl was dealuminated, albeit by different process steps, which could potentially provide different silica-alumina frameworks. Said catalysts could thus be different in the (final) Si/Al ratio or in the type of structure/ framework obtained after dealumination. Neither of them were however defined in Claim 1 then at issue.

7.2 The Board shares these objections, as well as the fact that in such situations the applicant has the burden of proof that the catalysts of the application and those of Dl are effectively different (i.a. Case Law of the Board of Appeal (2013), page 607, item Vl.H.5.1.2).

7.3 In the present case, the applicant has not proven that the catalyst obtained by the process of preparation defined in Claim 1 at issue is novel over the catalyst disclosed by Dl, for the following reasons:

7.3.1 As already pointed out by the Board in its written preliminary opinion on page 7, the experimental evidence submitted with the statement setting out the grounds of appeal is not suitable for proving that the treatment defined in Claim 1 leads to catalysts which are (consistently) different from those of D1.

7.3.2 Even if the Board accepts that this evidence appears to show that pre-steaming, for the particular example made, has an effect, no comparative example has been made over the catalyst disclosed in D1, on the basis of which the conditional objection of lack of novelty by the Examining Division was raised. Moreover, in the experiment, the post-steamed catalyst disclosed by D1 (Page 34, left-hand column, last paragraph, second sentence) has not been considered at all. Thus, it is not proven that pre-steaming as claimed consistently provides a clear, distinctive dealumination (with effect on the phases of Al) whatever parent zeolite is chosen, let alone over a zeolite which is post-steamed, thus leading to a catalyst which is consistently different from that of D1.

7.3.3 As regards the other features of the (preparation) process defined in Claim 1, none of them has been invoked, let alone shown, to be able to provide a clear, distinctive feature to the zeolite material as such (e.g. a different final Si/Al ratio, a different framework and porosity, or different Al phases), which is suitable to distinguish the catalyst prepared as defined in Claim 1 from the catalyst of D1.

7.3.4 This particularly applies to the initial atomic ratio ranging from 4 to 30 as defined in Claim 1 at issue, which is a different from the initial Si/Al molar ratio atomic ratio of 40 disclosed by D1. Nevertheless, this difference is not suitable to limit in any way the final Si/Al range, which evidently depends on the extent of dealumination. In fact, this difference in the starting molecular sieve has not been invoked as a feature imparting novelty to the claimed process by the Appellant.

7.3.5 The alleged difference between the catalyst of D1 in comparison to the one of the invention caused by the processing step of evaporation/drying of the Al-containing solution, rather than by its complete removal can also not be considered by the Board as a distinguishing feature, as Claim 1 at issue refers to the "*separation of the solid from the liquid*", which includes also the evaporation mentioned in D1 (as shown by the experimental evidence submitted by the Appellant).

7.3.6 Finally, the extrusion of the catalyst with SiO_2, as described on Page 30, left-hand column, last paragraph of D1, is also not excluded by the wording of Claim 1 of the 2nd Auxiliary Request and can therefore not be a distinguishing feature either.

7.4 Hence, the Appellant has not convincingly demonstrated that the cracking process as defined in Claim 1 at issue is novel vis-à-vis D1 (Articles 54(1) and (2) EPC).

7.5 Therefore, the 2nd Auxiliary Request is not allowable either.

3rd Auxiliary Request

Allowability of the amendments

Articles 123(2), 84 and 54 EPC

8. Claim 1 according to 3rd Auxiliary Request, compared to Claim 1 according to the 2nd Auxiliary Request, comprises the further feature "*separation of the solid from the liquid by filtration*", which is disclosed on Page 7, line 12, of the application as filed, in the context of the removal of the extra-framework aluminium oxide, formed by the steaming step and dissolved in the acid solution containing the source of phosphor. In the light of this amendment the Board is satisfied that Claim 1 according to 3rd Auxiliary Request is clear, novel and fulfils the requirements of Article 123(2) EPC. However, since the 3rd Auxiliary Request is not allowable for lack of an inventive step (*infra*), the Board does not see any need to give further details in these respects.

Inventive step

The invention

9. The present invention concerns a process for the cracking of olefins on phosphorous modified molecular sieves (P-modified molecular sieves), in particular a process for cracking an olefin-rich hydrocarbon feedstock which selectively redistributes the olefin content of the feedstock in the resultant effluent towards propylene (page 1, lines 9-15; page 2, line 7; page 3, lines 17-20).

The closest prior art

10. D1, in particular the disclosure of the C_4-olefin cracking process using the molecular sieve catalysts prepared according to Pages 30 (Point 2.1 Catalyst preparation) and 34 (left-hand column, last paragraph, second sentence), is the closest prior art document for assessing inventive step. This was not disputed by the Appellant.

The relevant disclosure of D1

11. D1 (Point 2.1 "Catalyst preparation"; Point 2.3 "Catalytic testing"; Page 34, left-hand-column, last paragraph, second sentence; Figure 7) illustrates a process for the catalytic cracking of a C_4-olefin feedstock which is selective towards propylene (D1, Title; page 30, left-hand column, first full paragraph, second and third sentences; page 35, right-hand column, last full paragraph, second sentence); the process comprising contacting the said feedstock, at a weight hourly space velocity (WSHV) of 3.2 h^{-1} and atmospheric pressure (Page 31, left-hand column, first paragraph, second sentence), as well as at a temperature of 500°C (see Figure 11), with a catalyst made of a P-modified molecular sieve (HZSM-5) to selectively produce propylene. Hence an effluent with an olefin content of lower molecular weight than that of the feedstock was produced, wherein said P-modified molecular sieve is made by a preparation process comprising in that order:
- selecting an (MFI-structure) NH_4ZSM-5 zeolite (first two lines of Point 3.1 "Effect of P on the structure of the catalyst") with a silica/alumina molar ratio of 40 (Point 2.1, "catalyst preparation", first sentence).

- impregnating (leaching) with an aqueous acid solution of phosphoric acid (see page 30, left-hand column, lines 15 to 24 of Point 2.1 "catalyst preparation");
- drying at 50°C for 24h under vacuum and 120°C for 4h, hence separating the solid from the liquid;

and,

- calcining at 600°C for 3 h.

Since D1 discloses the preparation of aqueous solutions having the desired amounts of phosphoric acid, and since phosphoric acid is a strong acid, it is to be concluded that its pH was of 3 or lower.

11.1 According to D1 (conclusion provided by the discussion concerning the results given in Table 1 on page 32, right-hand column, lines 3 to 6 under the said Table 1), the impregnation (leaching) conditions were effective to remove Al from the parent zeolite and to introduce at least 0,6 wt% of P (see also items 0.6PZ0, 0.9PZ, 1.5PZ and 2.1 PZ of Table 1), i.e. modified MFI zeolite having 0.6 wt% of P, 0.9 wt% of P, 1.5 wt% of P and 2.1 wt% of P, respectively. Hence, the leaching step was carried out for the time necessary to obtain P-modified HZSM-5 samples containing up to 2.1 wt.% phosphor.

11.2 Still according to D1 (page 34, left-hand column, last paragraph, second sentence), in order to investigate the hydrothermal stability of parent and P-modified HZSM-5 zeolites, a steaming (100% steam, 4h, 600°C) was carried out on these (already P-treated zeolites), i.e after leaching or impregnation with P. The steaming was found to improve hydrothermal stability significantly.

11.3 The process of Claim 1 according to the 3rd Auxiliary Request thus appears to be distinguished therefrom in particular in that it requires, before the step of impregnating (leaching), a step of steaming at a temperature ranging from 420 to 870°C for 0.05 to 200h.

The technical problem according to the Appellant

12. According to the statement setting out the grounds of appeal, the problem to be solved is the providing of an improved process, in particular resulting in a much better selectivity in C_4 olefins conversion at iso-sum of ethylene and propylene, particularly less by-products such as aromatics, paraffins and methane, as well as higher ratio to propylene and a significantly higher purity of C_3's cut (Page 4/14 of the statement setting out the grounds of appeal). During the oral proceedings before the Board the Appellant particularly stressed on the higher purity of C_3's cut.

The solution

13. The application at issue proposes to solve this problem by a process for the catalytic cracking of an olefin rich feedstock as defined in Claim 1 according to the 3rd Auxiliary Request, which is in particular characterized in that it comprises a step of steaming the selected zeolite at a temperature ranging from 420 to 870°C for 0.5 to 200 h **before** the step of leaching with an aqueous acid solution containing the source of phosphor.

The alleged success of the solution

14. As regards the improvement in propylene yield and purity of the C_3's cut of the claimed process, in comparison to the process of D1, the Appellant essentially relied on the catalyst tests included in

the experiments submitted with the statement setting out the grounds of appeal (see Pages 4 and 5 of the statement setting out the grounds of appeal).

14.1 In its catalyst tests, the Appellant had checked the performance of two catalysts, respectively prepared from steamed and phosphor modified precursors and non-steamed but phosphor modified precursors, whereby the precursor material was a sample of commercially available Zeolyst CBV 2314 NH_4-ZSM-5 zeolite, having a Si/Al ratio of 12. In particular, the steamed and phosphor treated catalyst provided a purity C_3's of 92.2, compared to the 84.6 C_3's purity provided by the non-steamed catalyst.

14.2 It is immediately apparent that the tested zeolites are different from those used in D1, at least in the initial Si/Al ratio. Also, the comparison made did not consider a catalyst according to D1, which had been post-steamed.

14.3 Moreover, the meaning of the "purity C_3's" mentioned in the catalyst tests results is not given, let alone the way it was determined. In this respect the Board notes that D1 was acknowledged in the application as filed (see page 2), and that only the technical problem of providing an improved process in respect of propylene purity is mentioned on page 3 (lines 6-7 and 10-11) of the application as filed (see also page 7, last paragraph, last sentence). Hence, the application as filed addresses the problem of providing propylene purity (i.e. at least of chemical grade), not however any improvement in the purity of the C_3's cut, whatever is meant thereby.

14.4 It follows from the foregoing that, since the catalysts tested were not according to D1, it is not clear that the showed improvements only relate to the pre-steaming step.

14.5 Furthermore, since also D1 (e.g. Page 35, last full paragraph, second sentence) addresses and solves a problem of providing a process for catalytic cracking of olefins with a hydrothermally stable catalyst having high selectivity towards propylene, in the absence of any truly comparative evidence on file, showing a whatsoever better performance across the breadth of Claim 1 at issue, the disclosure of the application as filed is insufficient at rendering plausible the improvement over D1 invoked by the Appellant.

14.6 Therefore, the Board cannot accept as plausible that the invoked, ambitious problem of providing an improved process over D1 is effectively solved by the process of Claim 1 at issue.

Reformulation of the technical problem

15. Since the problem effectively solved cannot be formulated in terms of an improvement over the closest prior art D1, it has to be reformulated in a less ambitious way.

15.1 In line with the objects described on Page 3 (lines 6 to 20) of the application as filed, which do not mention an improvement over D1, it can be seen in the providing of an alternative process for the catalytic cracking of an olefin-rich feedstock which is stably selective towards propylene.

15.2 Considering the results of Examples 1 to 5 of the application as filed, as well as the results of the catalyst tests annexed to the statement setting out the grounds of appeal, all showing high yield and C_3/C_2 ratio, the Board accepts it as plausible that the less ambitious problem is effectively solved by the process of Claim 1 at issue.

Obviousness

16. It thus remains to be decided whether the skilled person starting from the process described by D1, and aiming to solve the less ambitious problem solved, would have found any motivation to consider D2, and if yes, whether he would have found any suggestion towards the process as defined in Claim 1 at issue.

16.1 The Appellant essentially argued that, since D2 has to do with fluidised catalytic cracking (FCC), it does not concern cracking of an olefin rich feedstock, but of saturated hydrocarbons on modified zeolites, i.e. a heavier feed. It seems that the Board has no reason to dispute this, as it considers that this is not the point, in so far the disclosure of D2 not only concerns the base zeolite catalysts for the FCC process (zeolite Y) but particularly the catalyst "additive" which comprises ZSM-5 type zeolites (see paragraph [0003], more particularly its preparation).

16.2 In respect of this catalyst "additive", the disclosure of D2 appears to be very motivating and relevant for the skilled person, for the following reasons:

16.2.1 D2 relates to a catalytic cracking catalyst additive (title) and to a catalytic cracking process (paragraph

[0001]). According to D2 it was known that, in the FCC process:
- zeolite Y was used as the base catalyst to crack the heavy feedstocks (paragraph [0002])
- for producing light-olefins, an additive catalyst was added, which had to comprise ZSM-5 (Paragraph [0003]);
- this additive catalyst was suitable for boosting the propylene yield, as it *inter alia* <u>selectively catalysed the formation of propylene out of heavy olefins</u> (paragraph [0004], emphasis added by the Board).

16.2.2 Still according to D2 (Paragraphs [0021] to [0025]), the additive catalyst should however also achieve high propylene purity in any C_3 fraction in the effluent.

16.2.3 Therefore, D2, which concerned the same objectives as the application at issue, and which was thus motivating for the skilled person, would have been considered, even if it relates to FCC processes, as the FCC process per se plays no role.

16.3 According to D2 (paragraph [0026]), the problem of providing more propylene of high purity is solved by a process for producing the catalyst additive which comprises the steps of providing a MFI or MEL aluminosilicate with a Si/Al atomic ratio of 10 to 250; and, dealuminating the MFI or MEL aluminosilicate by extracting from 20 to 40 wt% of the alumina therefrom.

16.4 As disclosed in paragraph [0033] of D2, the dealumination is carried out by both, heating in steam and then contacting the MFI or MEL with a phosphorous containing mineral acid to remove aluminium from throughout the crystalline silicate framework and extract aluminium from the catalyst, and to deposit phosphorous in the dealuminated aluminosilicate. In these respects, the preferred acid is phosphoric acid and the treated dealuminated aluminosilicate contains from 0.1 to 0.3 wt% of phosphorous (paragraphs [0034] and [0035]) (see also paragraph [0094] on the effect achieved by incorporating phosphor).

16.5 As apparent from e.g. Paragraph [0079] of D2, the use of this additive catalyst *inter alia* increases the purity of the C_3-cut, as well as the ratio of propylene to total C_3s.

16.6 Summing up, the disclosure of D2 concerns the features (pre-steaming) distinguishing the claimed subject-matter from D1 and, as such, is a readily available option to the skilled person seeking to solve the less ambitious technical problem posed.

16.7 Consequently, the claimed process was obvious over D1, taken as the closest prior art, in combination with D2.

16.8 Therefore, the Board has no reason to deviate from the decision under appeal.

Conclusion

17. The issues of lack of clarity (Main Request), added subject-matter (1st Auxiliary Request), lack of novelty (at least 2nd Auxiliary Request) and, more particularly, lack of an inventive step (at least 3rd Auxiliary Request) are prejudicial to the grant of a patent on the application at issue.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

Beschwerdekammern des Europäischen Patentamts
Boards of Appeal of the European Patent Office
Chambres de recours de l'Office européen des brevets

D. Magliano E. Bendl

Decision electronically authenticated