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Title of invention:

A catalyst for converting hydrocarbons

Applicants:

- 1) China Petroleum & Chemical Corporation
- 2) Research Institute of Petroleum Processing, Sinopec

Headword:

Catalyst for converting hydrocarbons / CHINA PETROLEUM and SINOPEC

Relevant legal provisions:

EPC Art. 84, 111(1)

Keyword:

Clarity (yes) Remittal (yes)

Decisions cited:

T 0728/98, T 2152/10

Catchword:



Beschwerdekammern Boards of Appeal Chambres de recours

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Case Number: T 0740/16 - 3.3.06

D E C I S I O N
of Technical Board of Appeal 3.3.06
of 15 March 2017

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Decision under appeal: Decision of the Examining Division of the

European Patent Office posted on 29 October 2015

refusing European patent application No. 06791146.1 pursuant to Article 97(2) EPC.

Composition of the Board:

S. Fernández de Córdoba

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Summary of Facts and Submissions

- I. This appeal lies from the decision of the Examining Division to refuse the European Patent Application No. 06 791 146.1.
- II. In the decision under appeal, the Examining Division arrived at the conclusion that each claim 1 according to the then pending main request and auxiliary requests 1 to 4 lacked clarity (Article 84 EPC), for the reasons as follows:
 - (a) Claim 1 defined the chemical composition of the zeolite beta component <u>following incorporation in</u> <u>the catalyst</u> (point 1.2 of the reasons) (underlined by the Board).
 - (b) Even though, at the filing date of the application as filed, X-ray fluorescence (XRF), as indicated in the application, was an appropriate analytical technique for determining the amounts of the various elements constituting the modified zeolite beta (a^1) of the claimed catalyst composition, and X-ray diffraction (XRD) was a suitable analytical technique for distinguishing between various zeolite structures, as shown in D9 (A.W. Chester and E.G. Derouane (eds.), Zeolite Characterization and Catalysis, 2009, Chapter 1, "Powder Diffraction in Zeolite Science. An Introductory Guide", by Allen W. Burton, pages 1 and 2), the skilled person, even applying these techniques, would not have been able to quantify the elements belonging to the modified zeolite beta (a¹) present in a mixture comprising the other zeolites (a^2) and (a^3) and components (b) and (c), all of which comprised similar elements; moreover, the skilled person

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would not have been able either to quantify the amounts of each component containing such elements in the mixture (points 1.3 to 1.6 and 1.10 to 1.12 of the reasons).

- (c) The catalyst of claim 1 had not to be necessarily made by first preparing a modified zeolite beta (a¹) and then mixing it with the other components, as shown in the examples of the application, but it could be prepared by mixing all components, including an unmodified zeolite beta, and then modifying the zeolite beta in situ, as shown for example in D2 (WO 95/02653 A1) (example 1), D4 (EP 1 076 598 A1) (examples 1-4) and D1 (US 2006/014630 A1) (example 5). Therefore, an analytical method for the determination of the chemical composition of the modified zeolite beta (a¹) in the finished catalyst was necessary.
 - Moreover, the wording of claim 1 could not be read as being limited to a catalyst wherein the modified zeolite beta (a^1) had a specific structure due to the particular method of preparation illustrated in the application (points 1.7 to 1.9 of the reasons).
- (d) In the absence of any experimental evidence it was not credible that the modified zeolite beta (a¹) could be isolated by a reverse engineering process from the final catalyst in order to enable its analysis (points 1.13 to 1.15 of the reasons).
- (e) The chemical expression, i.e. the anhydrous chemical formula, of the modified zeolite beta (a¹) contained in the claimed catalyst composition was unclear since, taking into consideration the evidence available on file, the skilled person, at the filing date of the application, would not have

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been in the position to determine the quantities of the elements constituting component (a^1) in the final composition (points 1.16 and 1.17 of the reasons).

- (f) Decision T 2152/10 of 13 March 2013, concerning a case wherein in a complex multicomponent system the skilled person was not in a position, either in the light of the application or in view of common general knowledge, to determine the respective quantities of the same element independently, was thus relevant to the present case (points 1.18 to 1.20 of the reasons).
- III. In their notice of appeal, the Appellants requested the reversal of the decision of the Examining Division and the grant of a patent. Oral proceedings were auxiliarly requested in the event that the former request could not be granted.
- IV. With the statement of grounds of appeal of 7 March 2016 the Appellants (re)filed the sets of claims according to the main request and auxiliary requests 1 to 4 upon which the Examining Division had decided and submitted an example of a SEM-EDX (Scanning Electron Microscopy Energy Dispersive X-Ray) analysis and an XRD analysis of a catalyst comprising beta zeolite, Y-zeolite and a matrix consisting of silica and alumina.

They inter alia argued that claim 1 according to the main request was clear and complied with Article 84 EPC.

Contrary to the Examining Division's view, the skilled person was in a position, at the filing date of the application, to determine the various quantities of the -4- T 0740/16

catalyst ingredients and of the various elements of the modified zeolite beta in the final catalyst composition. For example, the skilled person would have been able to perform the identification of the various ingredients in the final catalyst by means of known methods such as a combination of an SEM-EDX analysis with an XRD analysis.

V. In the communication of 22 December 2016 (point 4), the Board expressed its provisional opinion that claim 1 according to the main request appeared to be clear, apart from the inconsistent definition of the metal M in the wording of the claim. The Board referred also to the following document:

D10: A.W. Chester and E.G. Derouane (eds.), Zeolite Characterization and Catalysis, 2009, Cover pages and Preface: pages v, vi and xvii.

The Board also indicated (point 5) that it was inclined not to examine the other patentability requirements and to remit instead the case to the Examining Division for further prosecution.

Moreover, it requested the Appellants to indicate within a time limit of two months if they still maintained their auxiliary request to be heard in oral proceedings if their main request, i.e. reversal of the decision and grant of a patent, was not accorded by the Board, and hence to restate their requests.

VI. By letter of 22 February 2017 the Appellants filed an amended set of claims as their main request, allegedly overcoming the issue raised by the Board concerning the inconsistent definition of the metal M.

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- VII. The set of 9 claims according to the main request, filed with letter of 22 February 2017, contains one independent claim, claim 1, reading as follows:
 - "1. A hydrocarbon conversion catalyst, which comprises, based on the total weight of the catalyst,

10-50 wt% of a zeolite mixture,

10-70 wt% of a thermotolerant inorganic oxide selected from alumina, silica and amorphous silica-alumina, and 0-60 wt% of clay,

wherein said zeolite mixture comprises, based on the total weight of said zeolite mixture,

1-75 wt% of a zeolite beta modified with phosphorus and a metal M_{\star}

25-99 wt% of a zeolite having a MFI structure and 0-74 wt% of a large pore zeolite selected from faujasite, zeolite L, zeolite Ω , mordenite and ZSM-18 zeolite,

wherein the anhydrous chemical formula of the zeolite beat modified with phosphorus and the metal M is represented in the mass percent of the oxides as $(0-0.3)\,\mathrm{Na}_2\mathrm{O}\cdot(0.5-10)\,\mathrm{Al}_2\mathrm{O}_3\cdot(1.3-10)\,\mathrm{P}_2\mathrm{O}_5\cdot(0.7-15)\,\mathrm{M}_{\mathrm{x}}\mathrm{O}_{\mathrm{y}}\cdot(64-97)\,\mathrm{SiO}_2,$

in which the metal M is one or more selected from the group consisting of Fe, Co, Ni, Cu, Mn, Zn and Sn; x represents the atom number of the metal M, and y represents a number needed for satisfying the oxidation state of the metal M."

Dependent claims 2 to 9 relate to particular embodiments of the claimed hydrocarbon conversion catalyst.

VIII. The Appellants requested that the decision of the Examining Division be set aside and that the case be

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remitted to the Examining Division for further prosecution on the basis of the main request filed with letter dated 22 February 2017. Auxiliarly, if this request could not be granted, oral proceedings were requested.

Reasons for the Decision

Main Request

- 1. Construction of Claim 1
- 1.1 Claim 1 (see VII, supra) concerns a hydrocarbon conversion catalyst (a physical entity, a product), which comprises, based on the total weight of the catalyst,
 - a) 1-60 wt% of a zeolite mixture,
 - b) 5-99 wt% of a thermotolerant inorganic oxide and
 - c) 0-70 wt% of clay,

wherein a) comprises, based on the total weight of said zeolite mixture,

 a^{1}) 1-75 wt% of a zeolite beta modified with phosphorus and a transition metal M, wherein the anhydrous chemical formula is represented in the mass percent of the oxides as

 $(0-0.3) \text{Na}_2\text{O} \cdot (0.5-10) \text{Al}_2\text{O}_3 \cdot (1.3-10) \text{P}_2\text{O}_5 \cdot (0.7-15) \text{M}_x\text{O}_y \cdot (64-97) \text{SiO}_2$

in which the metal M is one or more selected from the group consisting of Fe, Co, Ni, Cu, Mn, Zn and Sn; x represents the atom number of the metal M, and y

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represents a number needed for satisfying the oxidation state of the metal $\mathbf{M}_{\boldsymbol{r}}$

- a^2) 25-99 wt% of a zeolite having a MFI structure and
- a^3) 0-74 wt% of a large pore zeolite selected from faujasite, zeolite L, zeolite Ω , mordenite and ZSM-18 zeolite.
- 1.2 For the Board, no ambiguity arises from the individual features of claim 1, as all the components (a^1) , (a^2) , (a^3) , (b) and (c) of the claimed catalyst are classes of compounds known to the skilled person at the filing date of the application; moreover, the skilled person would not have had any difficulty in understanding the chemical formula representing component (a^1) .
- 1.2.1 In fact, the wording "catalyst ... which comprises... said zeolite mixture comprises ... 1-75 wt% of a zeolite beta modified with phosphorus and a metal M, wherein the anhydrous chemical formula is represented in the mass percent of the oxides as $(0-0.3) \, \text{Na}_2\text{O} \cdot (0.5-10) \, \text{Al}_2\text{O}_3 \cdot (1.3-10) \, P_2\text{O}_5 \cdot (0.7-15) \, M_x\text{O}_V \cdot$ (64-97)SiO₂ in which the metal M is one or more selected from the group consisting of Fe, Co, Ni, Cu, Mn, Zn and Sn", unambiguously requires that a chemically modified zeolite beta of the given structure (a¹) be present in the zeolite mixture, independently from any possible chemical modification of the other zeolites. Hence, claim 1 requires the modified beta zeolite to be included in the zeolite mixture as such, i.e. as modified, as represented by the formula given. Claim 1 does not require that the other two zeolites of the mixture be modified with phosphorus and metal M, let alone together with zeolite beta. This construction

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- of Claim 1 is not relativised or contradicted by the other claims and the description.
- 1.2.2 The dependent claims merely define preferred embodiments of the modified zeolite beta, of the other two zeolites and of clay. None of them relates to any chemical modification of the other two zeolites or of the clay.
- 1.2.3 The application as filed contains ten examples of preparation and characterization (by XRF) of the modified zeolite beta <u>before inclusion in the mixture</u>, because this zeolite is critical to the invention.
- 1.2.4 Instead, the application as filed does not disclose any process of treating a mixture of the three different zeolites in order to chemically modify at least one of them (as disclosed in D1, D2 and D4) (see II(c), supra). Indeed, the application as filed suggests also to use already modified zeolites having a MFI structure or being large pore (see page 11, lines 24-25, "ZRP zeolites containing phosphorus and a transition metal"; page 11, lines 30-32). Consequently, the present application, which teaches the use of chemically modified zeolites, cannot, and does not, disclose any method for characterizing the entire mixture of zeolites in order to find out whether the specific modified zeolite beta within the mixture fulfils the given formula of Claim 1.
- 1.2.5 It follows from the foregoing that no interpretation whatsoever of claim 1, encompassing the treatment of the entire mixture of zeolites to chemically modify the beta zeolite, can be based on the application as filed.

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- 1.2.6 Therefore, Claim 1 defines a specific modified zeolite beta which is prepared and characterized as such before inclusion in the catalyst mixture, rather than "following incorporation into the catalyst", as alleged by the Examining Division.
- 1.2.7 This means, for the Board, that the further, usual treatments used in the catalyst preparation, such as drying and calcining, should not substantially alter the oxide composition of the incorporated zeolites.
- 2. Compliance with the requirements of Article 84 EPC (Clarity) - Claim 1
- 2.1 As regards the Examining Division's view (see II(e), supra) that the chemical expression, i.e. the anhydrous chemical formula, of the modified zeolite beta (a¹) contained in the zeolite mixture of the claimed catalyst was unclear since, taking into consideration the evidence available on file, the skilled person, at the filing date of the application, would not have been in the position to determine the quantities of the elements constituting component (a¹) in the final composition, the Board finds that this conclusion implies that the skilled person, having prepared a catalyst composition by following the teaching of the application, would not be able to know whether it is working within the ambit of the claimed invention or not.

Hence, this objection might appear to concern sufficiency of disclosure rather than clarity, since it already assumes that the skilled person would be able to understand the elements constituting the chemical formula of component (a^1) and its relative quantities but would not know how to determine them.

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2.1.1 However, in the Board's view, the Examining Division's objection reported above can be understood as concerning a lack of clarity arising from a not compliance of claim 1 with the principle of legal certainty, which has been expressed in some decisions of the Boards with respect to Article 84 EPC.

For example, in T 728/98 of 12 May 2000 (OJ 2001, 319, point 3.1 of the reasons) the Board entrusted with that case decided that "Article 84 in combination with Rule 29(1) EPC stipulates the requirements that the claims shall be clear and define the matter for which protection is sought in terms of the technical features of the invention. Those requirements serve the purpose $\circ f$ ensuring that the public is not left in any doubt as to which subject-matter is covered by a particular claim and which is not. From this principle of legal certainty... it follows that a claim cannot be considered clear in the sense of Article 84 EPC if it does not unambiguously allow this distinction to be made (see decisions G 2/88, OJ EPO 1990, 93, point 2.5 of the reasons; T 337/95, OJ EPO 1996, 628, points 2.2 to 2.5 of the reasons)... This applies all the more if the unclear feature is essential with respect to the invention in the sense that it is designed for delimiting the subject-matter claimed from the prior art, thereby giving rise to uncertainty as to whether or not the subject-matter claimed is anticipated."

Thus, for the Board, the Examining Division decided in this respect that claim 1 lacked clarity because the wording of the claim, particularly the definition of component (a¹), was allegedly found to give rise to uncertainty as to whether or not the subject-matter claimed is anticipated, if the composition of the

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zeolite beta defined could not be established with certainty in the finished catalyst.

- 2.1.2 In fact, also all the other specific arguments concerning lack of clarity on which the decision under appeal is based (see points II (a) to (d), supra) confirm that, in the Examining Division's view, the skilled person was not in a position at the filing date of the application to determine the various quantities of the elements belonging to the modified zeolite beta (a¹) in a mixture comprising the other zeolites (a²) and (a³) and components (b) and (c), which all might comprise similar elements and also to determine the amounts of each component containing such elements in the mixture used as catalyst for the claimed process.
- 2.2 However, the Board remarks also in this respect that the position adopted by the Examining Division is not only unsubstantiated but also does not appear to be in accordance with common general knowledge.
- 2.2.1 Document D9, which undisputedly discloses the suitability of XRD analysis for the identification of structurally different zeolites and their compositional characteristics (see chapter 1.1 on pages 1 and 2 of D9), is part of a more comprehensive publication (book) illustrating analytical methods for the identification of different zeolite types. The contents of the entire publication is reported on page xvii of D10. Moreover, as indicated in the introductory part of this publication (see, in particular D10, page vi, lines 3 to 11), "the first five chapters of the book provide tutorials in the major areas of zeolite characterization: X-ray powder diffraction, NMR, temperature programmed desorption and adsorption calorimetry, electron microscopy, and infrared

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spectroscopy. All these techniques provided major contributions to the development of zeolite science, particularly XRD, IR, and measurement of acid-base properties in the early days (1955-1980). In the 1980s, electron microscopy and NMR started to become more prominent and now are equally important."

The introductory guide of D9 (page 2, figure 1.1, and lines 1-10), filed by the Applicants at the oral proceedings before the Examining Division, stresses that XRD enables the skilled person to phase and structure identification of zeolites.

Therefore, even though the book, of which D9 and D10 are excerpts, was published in 2009, after the filing date of the present application, it is credible that the analytical techniques indicated above, including also the XRF analysis cited in the application as filed (page 22, lines 19 to 22), were available at the filing date of the application and part of the common general knowledge of the skilled person. These techniques enabled thus the skilled person to identify unmistakably structure and composition of zeolites in isolation and within a mixture and also to identify the position and quantity of an element within a specific zeolite structure.

- 2.2.2 The suitability of the combination of XRD and SEM-EDX for the analysis of the catalyst has convincingly been presented with evidence in the statement setting out the grounds of appeal.
- 2.2.3 The decision under appeal itself (Point 1.13 of the reasons) acknowledges the evidence brought forward by the Applicants in order to prove that the skilled

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person could characterize the zeolitic material present in the finished catalyst.

- 2.2.4 In these respects, apart from casting unsubstantiated doubts on to whether chemical modifications took place or not during the preparation of the catalyst after calcination, the Examining Division has not provided any item of common general knowledge in support thereof.
- 2.2.5 Documents D1, D2 and D4 specifically deal with preparation of catalysts, comprising a chemical treatment of all of the zeolites together, and as such are not comparable to the present application, which does not foresee any such treatment.
- 2.2.6 For the Board, the skilled person, reading claim 1 at issue and having to determine the composition of the final catalysts, would in any case not be at a loss as all of the zeolites defined in Claim 1, and more particularly disclosed in the application as filed, concern known (the source is either given in the application as filed or can be retrieved from the standard references such as the atlas of zeolites, now available also online) and distinct materials (see Figure 1.1 of D9), for which thus e.g. diffractograms and other data are available. It has not been shown either that calibration standards with defined amount of chemical modifiers such as phosphorus and metal M cannot be produced in order to accelerate the identification of e.g. a modified zeolite beta present in the catalyst.
- 2.2.7 Hence, for the Board, contrary to the Examining
 Division's opinion (II(b), supra), and in accordance
 with the position of the Applicants, a skilled person

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would even have been able, at the filing date of the application, to choose an adequate combination of analytical techniques in order to identify and quantify the different components of a catalyst including a zeolite mixture as that of claim 1 at issue.

- 2.3 Moreover, even accepting, for the sake of argument, the Examining Division's view (II(c), supra) that the wording of claim 1 cannot be read as being limited to a catalyst wherein the modified zeolite beta (a¹) has a specific structure due to the particular method of preparation illustrated in the application, e.g. because the wording of claim 1 is in fact silent about the method of preparation to be used for modifying the zeolite beta and for preparing the catalyst and about the position of the added elements within the modified zeolite beta structure, the Board is convinced that the skilled person would still have been able, at the filing date of the application, to identify and quantify the modified zeolite beta (a1) present in the final catalyst, even after having been submitted to the preparation process of the catalyst.
- The Board is in particular not convinced that the skilled person, as alleged by the Examining Division (II(d), supra), could not characterise the final catalyst, e.g. a spray-dried catalyst as exemplified in the application (e.g. example 11), and determine its constituents, for example, by reverse engineering, as argued by the Applicants. The Board thus shares the opinion of the Appellants (IV, supra) that the final catalyst is a mixture which can be analysed.
- 2.5 Decision T 2152/10 of 13 March 2013, considered by the Examining division to support its decision (II(f), supra), concerns the clarity of a claim directed to a

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catalyst comprising a modified ZSM-5 zeolite, wherein the quantities of the modifiers P (phosphorus) and specific metal M are given by means of the anhydrous chemical expression of the zeolite, and comprising also specific quantities of P additives and M additives (see point 1.1 of the reasons). In this invention both the modifiers and the additives are possibly introduced into the catalytic structure in the same step (see points 1.3, 1.3.1 and 1.3.2 of the reasons). Therefore, the difference between the P and M modifiers and the P and M additives in a given catalyst could only depend on the position in which these elements were located in the final product, i.e. within the pore system of the ZSM-5 zeolite or in a different location of the catalyst (points 1.5 and 1.6 of the reasons). It was thus decided (point 1.8 of the reasons) that "With no method of measurement in the application as filed and in the absence of evidence that well-known methods of measurement were part of the common general knowledge of the skilled person, the Board can only conclude that the skilled person at the relevant date of filing of the application under analysis was not able to measure the separate quantities of the modifiers and of the additives in a catalyst as the claimed one, so that the features relating to these quantities are not clear."

2.5.1 However, this case is, for the Board, not directly comparable with the present one since the point at stake concerned in that case the possibility of identifying and quantifying the possibly identical elements introduced into the catalytic structure in the same step and belonging either to the modified zeolite or being present as additives outside of the zeolite, which possibility was considered not to have been convincingly proved by evidence.

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The present case instead concerns the identification and quantification of elements within a zeolite structure and in a mixture including zeolites, which elements, according to the application, are not introduced into the catalytic structure in the same step (see points 1.2 to 1.2.7, supra), which identification and quantification, as explained above (2.2.7 to 2.4), was convincingly part of common general knowledge.

Therefore, T 2152/10 is not relevant to the present case.

- 2.6 Summarizing, for the Board, common general knowledge enabled the skilled person, at the filing date of the application, if necessary, to identify and quantify each zeolite and other component present even in the final catalyst composition.
- 2.6.1 Therefore, the public cannot be left in any doubt as to which subject-matter falls under claim 1.
- 2.7 Claim 1 according to the main request complies thus with the requirements of Article 84 EPC.
- 3. Compliance with the requirements of Article 84 EPC (Clarity) Claims 2 to 9
- 3.1 The dependent claims 2 to 9 concern specific more limited embodiments of the catalyst of claim 1, in particular with respect to components (a^1) (claims 2 to 5), (a^2) (claim 6), (a^3) (claim 7) and (c) (claims 8 and 9).

For the Board, the wordings of these claims do not introduce any additional clarity issue.

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- 3.2 Therefore, also claims 2 to 9 comply *mutatis mutandis* with the requirements of Article 84 EPC.
- 4. Remittal
- 4.1 The decision under appeal only dealt with the issue of lack of clarity.
- 4.2 Since the primary purpose of the appeal proceedings is to review the decision under appeal, the Board considers it thus appropriate, in the present case, not to examine the other patentability requirements and to remit instead the case (Article 111(1) EPC) to the Examining Division for examination of the outstanding patentability issues.

Order

For these reasons it is decided that:

- 1. The decision under appeal is set aside.
- 2. The case is remitted to the Examining Division for further prosecution on the basis of the main request filed with letter of 22 February 2017.

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The Registrar:

The Chairman:



D. Magliano G. Santavicca

Decision electronically authenticated