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Datasheet for the decision of 13 March 2013

Case Number: T 2152/10 - 3.3.07

Application Number: 05824020.1

Publication Number: 1867388

IPC: B01J 29/46, B01J 27/18, C10G 11/05

Language of the proceedings: EN

Title of invention:
A catalyst and a hydrocarbon oil cracking method

Applicants:
China Petroleum & Chemical Corporation
RESEARCH INSTITUTE OF PETROLEUM PROCESSING, SINOPEC

Headword:
-

Relevant legal provisions:
EPC Art. 84
RPBA Art. 13

Keyword:
"Clarity - all requests (no)"
"Admissibility of sixth to fifteenth auxiliary requests (yes)"

Decisions cited:
-

Catchword:
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Case Number: T 2152/10 - 3.3.07

D E C I S I O N
of the Technical Board of Appeal 3.3.07
of 13 March 2013

Appellants: (Applicants)
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 26 April 2010 refusing European patent application No. 05824020.1 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman: J. Riolo
Members: D. Semino
W. Ungler

Summary of Facts and Submissions

I. The appeal lies from the decision of the examining division announced at the oral proceedings on 25 March 2010 refusing European patent application No. 05 824 020.1.

II. The decision was based on a single set of amended claims filed as main request with letter of 22 March 2010. Independent claim 1 according to that request read as follows:

"1. A catalyst, which comprises, calculated by dry basis:
10-65 wt% ZSM-5 zeolite,
0-60 wt% clay,
15-60 wt% inorganic oxide binder selected from one or a mixture of more than one of pseudoboehmite, alumina sol, silica-alumina sol, water glass and phosphorus-alumina sol,
0.5-15 wt% one or more metal additives selected from the metals of Group VIIIB, and 2-25 wt% P additive, in which the metal additive and the P additive are calculated by oxide;
in which the ZSM-5 zeolite is modified by P and one of the metals M selected from Fe, Co or Ni, the anhydrous chemical expression, calculated by oxide, is $(0\text{-}0.3)Na_2O \cdot (0.5\text{-}5)Al_2O_3 \cdot (1.3\text{-}10)P_2O_5 \cdot (0.7\text{-}15)M_xO_y \cdot (70\text{-}97)SiO_2$, in which x is the atom number of M and y is a number needed to satisfy the oxidation state of M, and in which the contents of metal additive of Group VIIIB and P additive do not include the contents of transition metal and P in the modified ZSM-5 zeolite."

III. In the decision under appeal the following documents were cited in addition to some examples filed with letter of 25 February 2010:

D3: WO-A-2005/097950
D8: CN-A-1 465 527
D10: J.M. Thomas and W.J. Thomas, Principles and Practice of Heterogeneous Catalysis, VCH Verlagsgesellschaft mbH, 1997, pages 213 and 214

IV. According to the decision the claimed subject-matter did not meet the requirements of Article 84 EPC, because the amounts of phosphorous and transition metals introduced in the claimed catalyst as "modifiers" and the amounts of phosphorous and Group VIIIB metals introduced in the claimed catalyst as "additives" could neither be determined by indication in the description nor by objective procedures which were usual in the art. In document D10 it was indeed reported that X-rays emitted during electron microscopy analysis made available the elemental composition of the analysed material, but it was still not clear how it would be possible in the complex heterogeneous system under study to distinguish between transition metals and P deriving from the zeolite modification from transition metals and P introduced as additives by making use of the technique of D10. In an *obiter dictum* lack of inventive step was objected in view of D3 and D8, each taken alone or in combination.

V. The appellants (applicants) filed a notice of appeal against the above decision. With the statement setting out the grounds of appeal, the appellants submitted six

set of claims as main and first to fifth auxiliary requests.

Claim 1 according to the main request was identical to claim 1 of the main request on which the decision was based with the addition that ZSM-5 zeolite listed as first ingredient was indicated to be a "modified" one. Claim 1 of the first auxiliary request corresponds to claim 1 of the main request with the addition that the metal additive and the P additive "are present in the matrix of the catalyst".

Claim 1 according to the second auxiliary request read as follows:

"1. A method of manufacturing a catalyst, which comprises:
adding one or more transition metal additives selected from the metals of Group VIIIB and P additive, together with a modified ZSM-5 zeolite, which is modified by P and one of the metals selected from Fe, Co or Ni, and has an anhydrous chemical expression, calculated by oxide, of $(0-0.3)Na_2O\cdot(0.5-5)Al_2O_3\cdot(1.3-10)P_2O_5\cdot(0.7-15)M_xO_y\cdot(70-97)SiO_2$, in which M is one of the metals selected from Fe, Co or Ni, x is the atom number of M, and y is a number needed to satisfy the oxidation state of M, a clay and an inorganic oxide binder selected from one or a mixture of more than one of pseudoboehmite, alumina sol, silica-alumina sol, water glass and phosphorus-alumina sol, to form the catalyst;
wherein the catalyst comprises, calculated by dry basis,
10-65wt% of the modified ZSM-5 zeolite,
0-60wt% of the clay,
15-60wt% of the inorganic oxide binder;

0.5-15wt% of one or more transition metal additives selected from the metal of Group VIIIB and 2-25wt% P additive;
wherein the transition metal additive and the P additive are calculated by oxide."

Claim 1 according to the third auxiliary request read as follows:

"A method of manufacturing a catalyst, wherein the catalyst comprises, calculated by dry basis:
10-65 wt% modified ZSM-5 zeolite, which is modified by P and one of the metals M selected from Fe, Co or Ni, the anhydrous chemical expression, calculated by oxide, being $(0-0.3)Na_2O\cdot(0.5-5)Al_2O_3\cdot(1.3-10)P_2O_5\cdot(0.7-15)M_xO_y\cdot(70-97)SiO_2$, wherein x is the atom number of M and y is a number needed to satisfy the oxidation state of M;
0-60 wt% clay;
15-60 wt% inorganic oxide binder selected from one or a mixture of more than one of pseudoboehmite, alumina sol, silica-alumina sol, water glass and phosphorus-alumina sol;
0.5-15 wt% one or more metal additives selected from the metals of Group VIIIB, and 2-25 wt% P additive;
wherein the metal additive and the P additive in the catalyst are calculated by oxide, and
wherein the manufacturing method comprises combining the metal additive of Group VIIIB, the P additive, the clay, the inorganic oxide binder and the modified ZSM-5 zeolite to form the catalyst."

Claim 1 of the fourth auxiliary request corresponded to claim 1 of the second auxiliary request with the

specification that the catalyst is formed "via a process of spray-drying". Claim 1 of the fifth auxiliary request corresponded to claim 1 of the third auxiliary request with the replacement of the step of combining the ingredients to form the catalyst with the specification that "in the manufacturing method the Group VIIIB metals are introduced by adding transition metal compounds before spray-drying, or are introduced after spray-drying through immersion or chemical adsorption of transition metal compounds and calcination, and the P additives are introduced by at least one of the following methods:
(i) adding phosphorous compounds before spray-drying;
(ii) by the phosphorus-alumina sol inorganic oxide binder;
(iii) after spray-drying through immersion or chemical adsorption of phosphorous compounds, optional solid-liquid separation, and drying and calcination.".

VI. In a communication sent on 24 January 2013 in preparation of oral proceedings the Board, as far as the main request was concerned, addressed *inter alia* the issue of clarity related to the lack of a method of measuring the quantity of the modifiers and of the additives in the final catalyst and the issue of inventive step with respect to document D3 as the closest prior art. As to the auxiliary requests, it was noted *inter alia* that in the claims defining the method of manufacturing of a catalyst the quantities of the ingredients were still defined with respect to the final product and not with respect to the compounds to be mixed. The computer translation of D8 (D8') was annexed to that communication.

VII. With letter dated 19 February 2013 the appellants filed some additional test data and 10 sets of claims as sixth to fifteenth auxiliary requests.

Claim 1 of the sixth auxiliary request corresponded to claim 1 of the second auxiliary request, wherein the "adding" step was preceded by a "modifying" step for the ZSM-5 zeolite to give a modified zeolite with the anhydrous chemical expression present in claim 1 according to all request. Claim 1 of the seventh auxiliary request corresponded to claim 1 of the sixth auxiliary request wherein the wording "adding to the modified ZSM-5 catalyst" was modified into "combining the modified ZSM-5 catalyst".

Claim 1 according to the eighth to fifteenth auxiliary requests corresponded to claim 1 according to the main and to the first to seventh auxiliary requests respectively, wherein the expression "comprises" with reference to the composition of the catalyst had been amended into "consists of".

VIII. Oral proceedings were held on 13 March 2013.

IX. The arguments of the appellants, as far as relevant to the present decision, can be summarised as follows:

Clarity of the requests filed with the statement of grounds

(a) The view that the skilled person could not distinguish the modifiers from the additives in the final catalyst was not tenable because at the effective filing date one of average skill in

catalysis was in a position to discriminate between them. In particular Fe, Co, Ni and P within the pore system of the ZSM-5 zeolite would count as modifier in the final catalyst, whereas they were counted as additives when present in different locations of the catalysts. SEM-EDX was a standard experimental tool for making that distinction as illustrated in D10. In the SEM-EDX examination of a sample one would focus on a specific grain, see what it is (a ZSM-5 grain or part of the matrix) and determine the amount of transition metal and P; by repeating the measurement at different spots, the average content of the additives and of the modifiers could be determined. The skilled person could therefore distinguish between modifiers and additives and measure their quantities using standard techniques, which were part of the common general knowledge, so that lack of clarity did not arise for claim 1 of the main request. In addition, according to T 0578/06 of 29 June 2011 (not published, see in particular point 21 of the reasons) the examining division had the burden of proof for the objections it had raised and the objection of lack of clarity was not substantiated, as the doubts expressed concerning the use of the technique in D10 were simple speculation.

(b) The lack of clarity issue was rendered moot in the first auxiliary request by the specification that the additives were present in the matrix of the catalyst and in claim 1 according to the second to fifth auxiliary requests by means of the replacement of the product claims with claims

directed to the methods of manufacturing the catalyst. In those claims, modified ZSM-5 zeolite was used as a starting material and additives were added in the appropriate amounts so as to obtain the claimed contents thereof in the final catalyst. Even if the amounts were defined with reference to the final product, it was clear to the skilled person that they were the amounts to be added in the manufacturing method.

Admissibility and clarity of the late filed requests

(c) The auxiliary requests filed with letter of 19 February 2013 were a legitimate reaction to the communication of the Board, where for the first time doubts were raised on the lack of clarity of the method claims (no method claims were decided upon in the appealed decision) and on lack of inventive step with respect to document D3 (inventive step was only dealt with in an *obiter dictum* in the decision). The sixth and seventh auxiliary requests addressed the issue of clarity by further specifying the presence of a preliminary modifying step, and the eighth to fifteenth auxiliary requests addressed the issue of inventive step by re-establishing the validity of the priority claim and excluding therefore document D3 from the state of the art.

X. The appellants requested that the decision under appeal be set aside and a patent be granted on the basis of the main request or the first to fifth auxiliary requests filed with the grounds of appeal or on the

basis of the sixth to fifteenth auxiliary requests filed with letter dated 19 February 2013.

Reasons for the Decision

Clarity

1. *Main request*

1.1 Claim 1 of the main request concerns a catalyst which comprises among others a ZSM-5 zeolite modified by P and one of the metals M selected from Fe, Co or Ni, one or more metal additives selected from the metals of group VIIIB (including therefore Fe, Co and Ni) and P additive. The quantity of the modifiers is given by means of the anhydrous chemical expression of the zeolite (including (1.3-10)P_2O_5 and (0.7-15)M_xO_y) and the quantity of the additives by means of wt% (0.5-15 wt% of the metal additives and 2-25 wt% of P additive, calculated by the respective oxides) with the specification that the contents of the additives do not include the contents of the modifiers. In all the examples of the application Fe, Co or Ni are used as metal additives.

1.2 It is evident from the definition of the catalyst in claim 1 of the main request that the same elements are present both as modifiers and as additives in the claimed product and that it is necessary both to understand the difference between modifiers and additives and to be able to measure the quantities of the two, as specific and separate ranges for the

quantities of the modifiers and of the additives are given in the claim.

1.3 It appears from the evidence on file not only that the modifier and additive compounds are largely overlapping, but also that the methods of introducing them into the catalytic structure do not substantially differ.

1.3.1 In the examples of D8, which is cited in the application under analysis as reference for the production of the modified ZSM-5 zeolite (page 4, lines 25 to 31 of the original application), it is shown that Fe, Co, Ni and P compounds (typically the inorganic salts of the metals and phosphoric acid or its ammonium salts) are introduced into a slurry containing the unmodified ZSM-5 zeolite, which is dried and calcined after mixing (see embodiments 1 to 7 in D8').

1.3.2 Similarly, in the examples of the application under analysis Fe, Co, Ni and P compounds (again the inorganic salts of the metals and phosphoric acid or its ammonium salts) are added to a slurry containing ZSM-5 zeolite and other ingredients (typically clay and other inorganic binders), which is then spray-dried and calcined (see examples 1 to 19 in the application as filed).

1.4 If it is the case that the ZSM-5 zeolite is modified by Fe, Co, Ni and P in D8, then it is inevitable that at least part of the Fe, Co, Ni and P compounds which are meant to be introduced as additives into the catalyst of the application under analysis will also attach to the ZSM-5 zeolite as modifiers.

1.5 In order to draw a line between modifiers and additives the appellants supported the view that Fe, Co, Ni and P within the pore system of the ZSM-5 zeolite would count as modifiers in the final catalyst, whereas they are counted as additives when present in different locations of the catalysts.

1.6 While this explanation is qualitatively reasonable and in line with the wording of claim 1 of the main request, which mentions a modified zeolite and additives for the overall catalyst, it makes it clear in view of the method of production that the difference between modifiers and additives does not depend on the point in time in which they are introduced into the catalyst (i.e. during the preliminary modification of the zeolite or during the subsequent manufacture of the finished catalyst), but on the position in which these elements are located in the final product (the pore system of the zeolite or the parts of the catalyst external to the zeolite).

1.7 In any case, as specific and separate quantities for the modifiers and the additives are indicated in claim 1 of the main request (see point 1.1, above), a method of measurement is necessary to distinguish them quantitatively in the final product and determine their quantities.

1.7.1 The application as filed does not provide any method of measurement of these quantities. In the examples the composition of the starting zeolite and the quantities of the ingredients to be mixed to manufacture the catalysts are given (examples 1 to 19). Indications are also given of the quantities of the additives in the

final product, but no information is provided concerning how these quantities have been measured, nor as to what the composition of the zeolite (including in particular its modifiers) in the final product is.

1.7.2 The appellants supported the view that techniques for accomplishing those measurements were part of the general knowledge of the person skilled in the art at the relevant date of filing of the application under analysis. Reference was made in particular to scanning electron microscopy (SEM-EDX) as illustrated in D10. In the SEM-EDX examination of a sample one would focus on a specific grain, see what it is (a ZSM-5 grain or part of the matrix) and determine the amount of transition metal and P; by repeating the measurement at different spots, the average content of the additives and of the modifiers could be determined.

1.7.3 Document D10 is an extract of a general textbook on catalysis and relates to electron microscopy (section 3.7.5 on page 213) as a technique for characterising catalysts (see heading on top of page 213). Together with generically emphasising the powerfulness of the technique (introductory paragraph in section 3.7.5) and schematically showing the way it works (figure 3.44 on page 214), D10 provides the generic information in the list of accomplishments of the technology that "the X-rays emitted during electron-microscopy analysis tell us the composition of the material under study" (page 213, point 3).

1.7.4 That generic information of D10, which does not refer to any specific system, let alone to one in which the same elements are present in different parts of the

structure and their quantities should be identified independently, does not provide the needed information on a method of measurements for the separate quantities of the modifiers and the additives in a catalyst as the one claimed in the present application and cannot be considered as sufficient evidence that the method described qualitatively in the submissions of the appellants indeed belongs to the common general knowledge of the skilled person at the relevant date of filing of the application under analysis.

1.7.5 A further confirmation that the submissions of the appellants in this respect are only unsubstantiated allegations with the evidence available on file is given by the fact that the method they qualitatively described was never applied to any of the examples in the application under analysis, nor to any of the additional examples provided during the examination and appeal procedures (the examples filed with letter of 25 February 2010 contained no compositions and the test data filed with letter of 19 February 2013 indicated the compositions without specifying the method of measurement used).

1.8 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.

1.9 During the oral proceedings the appellants additionally argued that the examining division had the burden of proof for the objection of lack of clarity related to the lack of a method of measurement, in particular due to the evidential document (D10) filed by the appellants in relation to that objection. However, as already pointed out above (see points 1.7 to 1.7.5 and 1.8), the burden of proof is in the present case on the appellants to show that it was possible to distinguish between modifiers and additives and measure their quantities using standard techniques which were part of the common general knowledge, as they alleged. As the Board has come to the conclusion that the evidence on file is not sufficient to prove this allegation, the burden of proof has not been discharged by the appellants in this respect.

1.10 On that basis, claim 1 of the main request is not clear contrary to the requirements of Article 84 EPC.

2. *First auxiliary request*

2.1 The amendment in claim 1 according to the first auxiliary request that the additives "are present in the matrix of the catalyst", which is in line with the explanation of the difference between modifiers and additives given by the appellants (see point 1.5, above) does not solve the problem that no method of measurements for the quantities of modifiers and additives is present in the application under analysis, nor has been shown to be known from the common general knowledge.

2.2 Claim 1 of the first auxiliary request is thus not clear for the same reasons as detailed for claim 1 of the main request.

3. *Second auxiliary request*

3.1 Claim 1 of the second auxiliary request concerns a method of manufacturing a catalyst. However, the claim does not specify the quantities of the different ingredients which are mixed during the manufacturing process, but defines the composition of the final product by means of the wording "wherein the catalyst comprises" followed by the list of compounds and their quantities with reference to the end product of the method.

3.2 Also in this case the quantities of modifiers and additives in the final product are features of the claim in spite of the change of claim category. The lack of a method of measurements for those features is thus a clarity issue for claim 1 of the second auxiliary request as it is for the main request.

3.3 The argument of the appellants that, even if the amounts are defined with reference to the final product, it is clear to the skilled person that they are the amounts to be added in the manufacturing method, cannot be followed by the Board, as it does not correspond to a sensible reading of the claim, in which there is no doubt that the quantities specified are those in the product to be obtained by the method, and in view of the fact that the difference between modifiers and additives does not depend on the point in time in which they are introduced into the catalyst during

manufacture, but on the position in which these elements are located in the final product (see points 1.3 to 1.6, above).

3.4 In view of this claim 1 of the second auxiliary request is not clear for the same reasons as detailed for claim 1 of the main request.

4. *Third auxiliary request*

4.1 Claim 1 of the third auxiliary request still concerns a method of manufacturing a catalyst. In spite of the quite different wording with respect to claim 1 of the second auxiliary request, it also defines the composition of the final product by means of the wording "wherein the catalyst comprises" followed by the list of compounds and their quantities with reference to the end product of the method and not the quantities of the different ingredients which are mixed during the manufacturing process.

4.2 Claim 1 of the third auxiliary request is thus not clear for the same reasons as detailed for the second auxiliary request.

5. *Fourth and fifth auxiliary requests*

5.1 Claim 1 of the fourth and fifth auxiliary requests correspond to claim 1 of the second and third auxiliary requests respectively with amendments which concern the definition of some method steps (see point V, last paragraph, above), but not the composition of the final product.

5.2 Claim 1 according to the fourth and fifth auxiliary requests is thus not clear for the same reasons as detailed for claim 1 according to the second and third auxiliary requests respectively.

Admissibility of the sixth to fifteenth auxiliary requests

6. The sixth to fifteenth auxiliary requests were filed by the appellants a few weeks after the communication of the Board (see points VI and VII, above), in which for the first time it was made clear that the clarity issue for the product claim could be equally relevant for the method claims (no method claims were decided upon in the contested decision) and that the Board intended to address the issue of inventive step on the basis of document D3 as the closest state of the art (inventive step appeared only in an *obiter dictum* in the contested decision).

6.1 They can be seen as a legitimate reaction to the points raised in the communication of the Board as in claim 1 according to the sixth and seventh auxiliary request an attempt was made to reformulate the method claim in order to solve the clarity issue and in the eighth to fifteenth auxiliary requests amendments were introduced which aimed at re-establishing the validity of the priority claim, so that document D3 did not belong to the state of the art.

6.2 As these requests are a legitimate reaction of the appellants to a new situation, the Board considers it appropriate to exercise its discretion according to Article 13 of the Rules of Procedure of the Boards of

Appeal by admitting the late filed requests into the proceedings.

Clarity

7. *Sixth and seventh auxiliary requests*

7.1 The methods according to claim 1 of the sixth and seventh auxiliary requests both include with respect to the method of claim 1 of the second auxiliary request a preliminary step in which a ZSM-5 zeolite is modified by P and one metal among Fe, Co and Ni. In both cases, however, the claim still contains as a feature the composition of the final product, which still implies specific limitations on the quantities of modifiers and additives.

7.2 The clarity objection raised for claim 1 of the second auxiliary request therefore equally holds for claim 1 according to the sixth and seventh auxiliary requests.

8. *Eight to fifteenth auxiliary requests*

8.1 The amendments introduced in the eighth to fifteenth auxiliary requests with respect to the main and the first to seventh auxiliary requests respectively were not meant to solve the clarity issue. Indeed, the appellants did not add any new argument in relation to the clarity issue as to these requests and the change of the wording "comprises" into "consists of" does not have any bearing on the lack of a method of measurement for the quantities of modifiers and additives.

8.2 Claim 1 according to the eighth to fifteenth auxiliary requests is thus not clear for the same reasons as given for claim 1 of the main and the first to seventh auxiliary requests respectively.

Conclusion

9. As claim 1 according to all the requests on file is not clear in the sense of Article 84 EPC, there is no need to analyse any other issue and the appeal is to be dismissed.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar The Chairman

S. Fabiani J. Riolo