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

Case Number: T 0503/93 - 3.3.5

Application Number: 86302857.7

Publication Number: 0198720

IPC: C10G 35/095

Language of the proceedings: EN

Title of invention:

Zeolitic reforming catalyst and method of producing the same

Patentee:

CHEVRON RESEARCH AND TECHNOLOGY COMPANY

Opponent:

Exxon Chemical Patents Inc.

Headword:

Zeolitic reforming catalyst/CHEVRON

Relevant legal provisions:

EPC Art. 83, 54, 56

Keyword:

"Sufficiency of disclosure (yes)"

"Novelty and inventive step (yes)"

Decisions cited:

T 0511/92, T 0450/89

Catchword:



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Boards of Appeal

Chambres de recours

Case Number: T 0503/93 - 3.3.5

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

Appellant: CHEVRON RESEARCH AND TECHNOLOGY COMPANY
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 22 March 1993
revoking European patent No. 0 198 720 pursuant
to Article 102(1) EPC.

Composition of the Board:

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

Summary of Facts and Submissions

- I. European patent No. 0 198 720 based on application No. 86 302 857.7 was granted on the basis of seven claims.
- II. The Respondent (Opponent) filed a notice of opposition requesting revocation of the patent on the grounds of lack of novelty, lack of inventive step and insufficiency of disclosure (Article 100(a) and (b) EPC). The following documents were cited by the parties during the opposition proceedings:
- D1: EP-A-0 096 479,
 - D2: GB-A-2 116 450,
 - D3: US-A-3 766 056,
 - D4: Product data sheet of "Catalyst Base 30-200" from Linde Molecular Sieves (1978),
 - D5: Letter from Du Pont de Nemours GmbH dated 3 June 1992, relating to the "Iron Content of Ludox HS-30 and Ludox HS-40 as a Function of Time",
 - D7: EP-A-0 107 389 and
 - D8: Product data sheet of "Zeolite L - KL1" from Laporte Industries Ltd.
- III. The Opposition Division revoked the patent on the grounds of lack of novelty and inventive step. The decision was based on an amended set of 5 claims referring to a reforming catalyst and a method for

producing this catalyst as a main request, and as an auxiliary request on one single claim referring to the use of a promoter.

Concerning the main request, the Opposition Division found that the catalyst according to Claim 1 of the main request was not novel over Example 16 of D1 in view of a calculation presented by the Respondent. This calculation was based on D5 which showed a constant iron content in the silica source "Ludox HS-40" for the years 1987 to 1991. It was deduced therefrom that the iron content must have been similar in the Ludox product used in 1982, the priority year of D1, for the preparation of the zeolite of Example 16. Reference was made to D4 and D8 and to Example 1 of the patent in suit which all showed that commercial type L zeolites contain considerable amounts of iron and titanium impurities.

The subject-matter of the use claim of the auxiliary request was held to be novel, but not to be based on an inventive step having regard to the teaching of D2 in combination with D3.

- IV. The Appellant (Proprietor) lodged an appeal against this decision. With his statement of grounds of appeal he filed new claims and made reference to seven further citations in support of his arguments in the statement of grounds of appeal. From the said citations, only the following is of importance for the present decision:

D15: James H. Gary, Glenn E. Handwerk, Petroleum Refining, Marcel Dekker Inc., New York, 1975, pp. 65-85, 114-141.

V. In response to a communication from the Board, in which some possible objections under Rule 57a EPC and Article 123(2) EPC were raised, the Appellant filed a new set of five claims comprising two independent claims, Claim 1 reading as follows:

"1. A dehydrocyclisation catalyst comprising a type L zeolite containing platinum metal and at least one promoter metal selected from iron, cobalt, titanium and europium, wherein said catalyst has a platinum to promoter metal mole ratio of less than 10:1."

Independent Claim 2 relates to a method of producing the dehydrocyclisation catalyst of Claim 1.

VI. The Appellant submitted that the Respondent's calculation of the iron content was based on the analytical data of D5 which dated from much later than D1 itself. The iron impurities in the starting materials and in particular in the silica source were, however, dependent on factors like source of raw material, source of water, corrosion condition of equipment used and kind of procedure for making silica sol. These factors created considerable uncertainty as to the actual amount of iron in Du Pont's Ludox product in 1983 (date of D1). Even if the iron content had remained constant between 1987 and 1991, it could have substantially changed between 1983 and 1987. Further, it appeared from D5 that Du Pont did not analyse the iron content in the product before 1987.

D4 and D8 were completely irrelevant because they related to other commercial zeolites and there was no evidence of any similarity with the zeolite of D1. Likewise, the reference to example 1 of the patent in suit was irrelevant because the iron content of Ludox at the effective date of D1 could not be derived

therefrom. Hence, the subject-matter defined in the claims was not anticipated by D1.

Concerning inventive step, the Appellant held that, contrary to the decision under appeal, the teaching of D3 was incompatible with that of D2. The latter concerned catalytic reforming, in particular dehydrocyclisation, whereas D3 concerned primarily hydrocracking and hydrotreating. Reference was made inter alia to D15 with respect to the differences. Therefore, a person skilled in the art would not have been led into the field of hydrocracking and hydrotreating to solve problems associated with reforming.

Finally, the Appellant submitted that the claims directed to the method of making the catalyst were also inventive having regard to the prior art because they represented a method of making a catalyst which was itself inventive.

VII. With letter dated 16 December 1996, the Respondent withdrew the opposition.

Before withdrawing the opposition, he maintained his objection concerning sufficiency of the disclosure, raised during the opposition proceedings. In his opinion there was no teaching in the patent as to how the claimed catalyst containing promoter metal could be obtained, because the conditions given in the contested patent for preparing and testing the catalyst were not suitable for reducing the promoter compounds to the corresponding metal.

In respect of novelty, he argued that normally the quality of products manufactured over an extended period of time was improved so that any amounts of

impurities were decreased. It must, therefore, be expected that Ludox contained still more impurities at the priority date of D1 than indicated in D5. In addition, D4 and D8 demonstrated that, depending on the impurities of the starting materials, zeolites generally contained iron. In zeolite synthesis, these starting materials were always at least very similar.

The claimed subject-matter was furthermore not novel over D3, which disclosed a catalyst containing L zeolite impregnated with promoters such as Fe, Co, Ni, Mg, Ca and rare earth cations in an amount of at least 0.5%wt and containing a hydrogenation component (Mo, W or Group VIII metal). Since platinum in an amount of preferably 0.1 to 2%wt was mentioned as a suitable hydrogenation component, present Claim 1 directly read on the catalyst of D3.

Concerning inventive step, the Respondent argued that improved stability, whether obtained by using alkaline earth metals as in D2 or iron as in D3 as stabilizers, led to the desired maintenance of the high initial activity. D3 did therefore not lead away from the claimed invention.

VIII. The Appellant requested that the decision be set aside and the patent be maintained on the basis of the new set of claims.

Reasons for the Decision

1. The appeal is admissible.
2. *Allowability of the amendments made to the claims*

There are no objections under Article 123(2) EPC with respect to the amended Claims 1-5. A basis for these claims can be found in the application as originally filed (see claims, page 1, lines 7 to 20 and 26 to 31, and page 2, lines 18 to 25).

There are also no objections under Article 123(3) EPC: The amendments made to Claims 1-5 consist a) in a restriction to europium as far as the promoter is a rare earth metal and b) in the new definition of the catalyst as a dehydrocyclisation catalyst instead of a reforming catalyst. Dehydrocyclisation is, however, one of several chemical reactions which come under the definition of reforming (see e.g. D15, page 66). The amendments therefore constitute a limitation of the extent of protection conferred by the patent.

3. *Sufficiency of disclosure*

The Board agrees with the Opposition Division's position that there exists a problem of uncertainty in Claim 1 insofar as it is not unequivocally clear whether the platinum and the promoter contained in the catalyst are present in metallic form, elemental form or in oxidic form. In view of the description of the patent specification, in particular the description of the catalyst preparation given in the examples, the above uncertainty in Claim 1 is in the Board's judgment not prejudicial to the maintenance of the patent in view of Article 100(b) EPC. Contrary to the Respondent's position, it is held that the terms

"platinum metal" and "promoter metal" as used in present Claim 1 merely define a content of these metals in the same sense as used in the examples of the patent specification, namely in the sense of the content of the chemical elements in the catalyst, but do not necessarily imply that the metals must be present in **metallic or elemental form.**

4. *Novelty*

4.1 Novelty was contested in respect of D1, in particular in view of Example 16, which discloses a type L zeolite derived from a synthesis gel comprising "Ludox HS40" as a silica source in an amount such that the gel contains 40 moles of SiO₂ and containing a conventional platinum loading of 0.6 wt% (see in D1, Example 16 in combination with page 37, lines 19 and 20, page 46, first paragraph and Fig. 5). As stated in D1 (see page 27, lines 7 to 9) the catalysts of D1 are suitable for dehydrocyclisation reactions. Therefore, the above objection is not overcome by the amendment of Claim 1 during the appeal proceedings.

From D5, where the amounts of iron impurities in the Ludox HS40 products between the years 1987 and 1991 are indicated, as well as from D4, D8 and Example 1 of the patent specification, the Respondent had inferred that the zeolite disclosed in D1 contained enough iron impurities to provide a platinum to iron mole ratio of less than 10:1.

However, from the fact that the iron content of Ludox HS-40 was constant from 1987 to 1991, it cannot, without further proof, be deduced that the same content was also present in 1982, the priority date of D1. Neither the mere expectation that the quality of a product was improved over the years, nor the fact that

other commercial zeolites, like those of D4 and D8, contained substantial amounts of iron, can be taken as such a further proof. Likewise it is of no importance for the disclosure of D1 that in Example 1 of the patent in suit it is shown that a significant amount of Fe and/or Ti can be present without their deliberate addition. The Respondent's line of argument therefore fails to show that the claimed subject-matter was directly and unambiguously derivable from D1 (see also T 511/92 of 22 May 1993, reasons No. 2.2).

In addition, the manufacturers of Ludox HS-40 have indicated in D5 that the iron content is not a specification item and not determined on a routine basis. In the Board's judgment, it is therefore clear that the iron content was not held to be critical in the Ludox products. Hence, the Board is unable to infer from this document any reason to make an effort to decrease or even control the amount of iron impurities in the Ludox product. Rather, it is reasonable to assume that the iron content largely depends on the quality of the starting materials and equipment, as set out by the Appellant. Therefore, in the absence of any evidence concerning the actual content of iron in the Ludox product delivered by Du Pont in 1982, D1 does not, in the Board's judgment, clearly and unmistakably disclose an iron content within the ambit of Claims 1 and 4 (see also T 450/89 of 15 October 1991, reasons No. 3.11), or, in other words, such an amount of iron impurity in the type L zeolite of the catalyst according to Example 16 of D1 that would inevitably have been enough to give a platinum to promoter mole ratio of less than 10:1.

- 4.2 The claimed subject-matter is also novel over D3 for the following reasons:

D3 discloses the preparation of a catalyst containing a zeolite, a polyvalent metal cation and a hydrogenation component (see column 1, lines 16-20). The polyvalent metal is selected from a variety of bi- and trivalent metal cations. Preferred is the iron group, but cobalt or nickel are particularly preferred. The preferred zeolites are type X or Y, but type L is also mentioned (see column 2, lines 32-45 and examples). A variety of possible hydrogenation components are suggested, inter alia platinum, but with molybdenum being preferred (see column 1, lines 44-63, column 2, lines 44-63 and column 3, lines 49-68). The only example with zeolite L (see example 7) uses cobalt and nickel as polyvalent metal and molybdenum as hydrogenation component. No other information or guidance is given as to which specific combination of materials out of these three groups are contemplated. Hence, D3 does not teach to combine the specific features characterizing the catalyst according to the patent in suit.

5. *Inventive step*

The patent in suit is concerned with the provision of a highly active and highly selective type L zeolite-supported platinum reforming catalyst for use in dehydrocyclisation reactions (see specification page 2, lines 6 to 20).

From the cited prior art only D1, D2 and D7 refer to L type zeolite-supported platinum reforming catalysts, in particular for use in dehydrocyclisation reactions (see in D1, page 7, first paragraph; in D2, page 1, lines 3-5; in D7, page 1, lines 2-21). D2 and D7 both disclose the necessity of a stabilizer to be present in

the catalyst to maintain its catalytic activity over an extended period of time and to improve activity and selectivity (see in D7, page 2, lines 10 to 25; in D2, page 6, lines 53/54 and examples, in particular Tables I to III and VI). Hence, the Board shares the view expressed in the decision under appeal that the terms "stabilizer" and "promoter" are equivalent within the present context of improving catalyst activity and/or selectivity. It follows therefrom that D2 and D7 represent the closest state of the art.

Since the Appellant did not advance any improvements over the catalysts of D2 and D7 and since no such improvements are apparent from the patent specification, the problem to be solved in respect of this state of the art consists in the provision of further stable catalysts for dehydrocyclisation reactions.

Present Claim 1 suggests solving this problem by the addition of a promoter selected from iron, cobalt, titanium and europium to an L-type zeolite containing platinum in an amount such that the platinum to promoter mole ratio is less than 10:1. Having regard to the examples of the contested patent, the Board is satisfied that the stated problem has thereby been solved.

D2 uses barium, strontium or calcium, which is introduced by impregnation or ion exchange (see page 2, lines 44-53 and page 6, lines 50-58), D7 uses sodium, lithium, potassium, rubidium, barium or cesium (see page 2, lines 32-37). D1 says nothing about such additives. These documents cannot, therefore, suggest solving the above-mentioned problem by the addition of the stabilizers (promoters) indicated in present Claim 1.

Such stabilisers are, however, mentioned in D3, which proposes the incorporation of a polyvalent metal cation into a zeolite catalyst containing a hydrogenation component in order to overcome the destabilizing effect of said hydrogenation component. The stabilizer is preferably selected from the iron group, in particular from cobalt and nickel (see point 4.2 above).

D3 mainly focuses on catalysts for hydrogenation, in particular in hydrocracking and hydrotreating processes, but also mentions reforming (see column 6, line 68 to column 7, line 3). According to D15 reforming includes both dehydrogenation and hydrocracking (see page 66, lines 17-20), but dehydrogenation reactions like dehydrocyclisation differ from hydrocracking in many aspects. For example, in the latter case hydrogen is consumed, while dehydrogenation produces hydrogen. Still more important is the fact that hydrocracking consumes the starting material for dehydrocyclisation, i.e. paraffins with a minimum necessary number of carbon atoms (see D15, pages 66 to 70, see also patent specification page 2, lines 6-10). It is therefore perfectly convincing that, as set out in the patent specification (see page 2, lines 11-13) it is important for the catalyst to be selective either for hydrocracking or for a dehydrogenation reaction. As can be seen from D15, page 74, lines 3/4, platinum supported on a silica or silica-aluminium base is thought to serve as a catalytic site for both hydrogenation and dehydrogenation reactions. A person skilled in the art would, therefore, normally not consider platinum to be suitable to provide the respective selectivity. Concerning the addition of a stabiliser, D3 rather suggests that the iron group metals generally impart selectivity for hydrocracking, irrespective of whether the zeolite is type X, Y or L (see Examples 1-8).

Since, moreover, nothing else in D3 suggests that any specific combination of a dehydrogenation component and iron or cobalt as a stabilizer could render a type L zeolite catalyst or any other zeolite catalyst selective for dehydrogenation reactions, D3 does not contain any information concerning the solution of the technical problem solved by the patent in suit.

6. None of the other citations anticipates the claimed invention or renders it obvious, neither alone nor in any combination with the other documents. Since the Respondent did not rely on these documents in his argumentation against the independent claims, it is not necessary to give reasons for this finding.

7. The above considerations also apply to the method of Claim 2 as well as to dependent Claims 3 to 5 which relate to particular modifications of the method according to Claim 2.

It follows therefrom that the grounds of opposition raised by the Respondent do not prejudice the maintenance of the patent as amended.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the Opposition Division with the order to maintain the patent on the basis of Claims 1 to 5 and the adapted description submitted with letter of 30 April 1998.

The Registrar:

S. Hue

The Chairman:

R. Spangenberg

