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D E C I S I O N
of 28 June 2000

Case Number: T 0566/96 - 3.3.5

Application Number: 90201254.1

Publication Number: 0398446

IPC: B01J 35/02

Language of the proceedings: EN

Title of invention:

Catalyst for hydrogenation and/or dehydrogenation

Applicant:

Engelhard De Meern B.V.

Opponent:

-

Headword:

Catalyst/ENGELHARD

Relevant legal provisions:

EPC Art. 84, 54, 111(1)

Keyword:

"Novelty (yes after amendment) - support by the description
(yes)"

Decisions cited:

-

Catchword:



Case Number: T 0566/96 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 28 June 2000

Appellant: Engelhard De Meern B.V.
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 25 January 1996
refusing European patent application
No. 90 201 254.1 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: R. K. Spangenberg
Members: M. M. Eberhard
M. B. Günzel

Summary of Facts and Submissions

I. European patent application No. 90 201 254.1 was refused by a decision of the examining division posted on 25 January 1996. The decision was based on amended claim 1 filed on 8 November 1995, and claims 2 to 11 as originally filed, as the main request, and on claims 1 to 9 submitted on 8 December 1995 as the auxiliary request.

II. The grounds for the refusal were lack of novelty and lack of support by the description. The examining division held that the subject-matter of claim 1 according to the main request lacked novelty over the disclosure of either of D4 (US-A-4 124 537), D5 (EP-A-9 068), D6 (EP-A-0 199 509). The process of claim 1 according to the auxiliary request lacked novelty with respect to D4 which clearly described the use of the catalyst in a hydrotreating or hydrodesulphurisation process. These processes were hydrogenation processes according to the definition of "hydrogenation" given in the Hawley's Condensed Chemical Dictionary.

The examining division considered that the requirements of Article 84 EPC were not met since the claims were only partly supported by the description. As there was a general consensus in the catalyst field that the properties of a catalyst were unexpected, it could not be recognised on the basis of only 2 or 3 examples that an effect automatically existed for all the 133 catalyst compositions defined in claim 1.

III. The appellant lodged an appeal against this decision. With the statement of grounds of appeal, he submitted

additional experiments and requested that a patent be granted on the basis of the auxiliary request filed on 8 December 1995, which accordingly became the main request. In a communication pursuant to Article 11(2) RPBA, the appellant was informed of the provisional opinion of the board regarding allowability of the amendments, clarity, support by the description and novelty. In reply thereto, the appellant filed five sets of amended claims in replacement of all the previous requests. Additional test reports were submitted on 26 June 2000. Oral proceedings took place on 28 June 2000. At the oral proceedings the appellant filed two sets of amended claims as a main request and a first auxiliary request respectively. Claim 1 and claim 8 of the main request read as follows:

"1. A process for hydrogenation and/or dehydrogenation of an organic compound in the presence of one or more sulfur compounds, using a catalyst having improved resistance against deactivation by sulfur compounds, said catalyst comprising:

- a support material selected from the group of aluminium oxide, silicon oxide, silicon oxide-aluminium oxide, titanium dioxide, zirconium oxide, magnesium oxide, mixtures of two or more of these support materials and active carbon, said support material having a particle size up to 200 μm .
- at least one hydrogenation component selected from the group of nickel, cobalt, copper, platinum, palladium, rhodium, ruthenium and mixtures of two or more of these metals, said hydrogenation component being present in an amount of more than

0.1 wt.% calculated on the weight of the support material and the hydrogenation component in reduced form, and

- at least one metal oxide containing component, said metal oxide having been selected from the group consisting of oxides of silver, lanthanum, antimony, nickel, bismuth, cadmium, lead, tin, vanadium calcium, strontium, barium, cobalt, copper, tungsten, zinc, molybdenum, manganese, iron and mixtures of two or more of these oxides,

at least part of the hydrogenation component being present as separate particles on said support material and at least part of the metal oxide component being present as separate particles on the said support material, there being only very limited direct contact of the hydrogenation component with the metal oxide containing component, the particles of both the hydrogenation component and the metal oxide component being homogeneously distributed in the catalyst, the composition of the catalyst per 0.01 mm³ not differing more than 15% from the total composition of the catalyst and the hydrogenation component being different from the metal oxide containing component, said catalyst being obtainable by the separate preparation of a support material with the hydrogenation component applied to it and a support material with the metal-oxide material applied to it, followed by mixing the thus loaded support materials, or said catalyst being obtainable by applying the said hydrogenation component or precursor therefor to the support material, followed by drying, converting to a catalytically active material, and applying the said metal oxide containing component to said hydrogenation

component containing support material, followed by drying and conversion to a catalytically active material."

"8. A catalyst for hydrogenation and/or dehydrogenation having improved resistance against deactivation by sulfur compounds, suitable for use in the process of any of the claims 1-7, said catalyst comprising:

- a support material selected from the group of aluminium oxide, silicon oxide, silicon oxide-aluminium oxide, titanium dioxide, zirconium oxide, magnesium oxide, mixtures of two or more of these support materials and active carbon, said support material having a particle size up to 200 μm .
- at least one hydrogenation component selected from the group of nickel, cobalt, copper, platinum, palladium, rhodium, ruthenium and mixtures of two or more of these metals, said hydrogenation component being present in an amount of more than 0.1 wt.% calculated on the weight of the support material and the hydrogenation component in reduced form, and
- at least one metal oxide containing component, said metal oxide having been selected from the group consisting of oxides of silver, lanthanum, antimony, nickel, bismuth, cadmium, lead, tin, vanadium calcium, strontium, barium, cobalt, copper, tungsten, zinc, molybdenum, manganese, iron and mixtures of two or more of these oxides,

at least part of the hydrogenation component being

present as separate particles on said support material and at least part of the metal oxide component being present as separate particles on the said support material, the particles of both the hydrogenation component and the metal oxide component being homogeneously distributed in the catalyst, the composition of the catalyst per 0.01 mm³ not differing more than 15% from the total composition of the catalyst and the hydrogenation component being different from the metal oxide containing component, said catalyst being obtainable by the separate preparation of a support material with the hydrogenation component applied to it and a support material with the metal-oxide material applied to it, followed by mixing the thus loaded support materials."

IV. The appellant put forward inter alia the following arguments:

The requirement of support by the description was met since a number of different materials had already been shown to give the inventive effect. The general case law on catalysts did not seem to be applicable to the present case as the invention concerned the discovery and reduction to practice of a more general principle. Furthermore the additional experiments in the test report submitted on 26 June 2000 showed that the present invention was operative for various types of metal oxide and hydrogenation components and was applicable to the whole breadth of the claims.

The claimed process and catalysts were novel with respect to D4. Using the conditions described in the examples of D4 resulted in a product wherein the two components were intimately mixed as shown by the

appellant's test reports submitted with the statement of grounds of appeal and on 26 June 2000. The process of preparation used in these examples led to a Co-Mo-sulphide active phase. Cobalt, molybdenum and sulphide were homogeneously mixed on an atomic scale in each particle. In example 2 of D4 the cobalt and molybdenum compounds were dissolved in the aqueous nitric acid solution during the mixing step. This example was equivalent to example 1 insofar as both compounds were in solution. The separate addition of the components as defined in claim 1 with fixation of the hydrogenation component therebetween was totally different from the simultaneous impregnation disclosed in D4 and resulted in the hydrogenation component and the metal oxide component being present as separate particles on the support instead of forming a single Co-Mo-sulphide phase.

- V. The appellant requested that the decision under appeal be set aside and that the case be remitted to the examining division for further prosecution on the basis of the main request and first auxiliary request submitted at the oral proceedings before the board.

Reasons for the Decision

1. The appeal is admissible.

Main request

2. The amended claims 1 to 8 of the main request meet the requirements of Article 123(2)EPC. Claims 1 and 8 are based on a combination of the features stated in claims 1, 3, 4, 6, 9 and 11 as originally filed with

features disclosed in the original description. The upper limit of 200 μm for the particle size of the support material and the amount of hydrogenation component are indicated on page 8, lines 16 to 18, and page 7, lines 26 to 29, respectively. The homogeneous distribution of the components expressed in terms of the composition per 0.01 mm^3 is disclosed on page 2, lines 18 to 26. The feature concerning the limited direct contact of the different components is based on page 3, lines 17 to 26. The preparation method incorporated into both claims 1 and 8 is described in the passages on page 8, lines 22 to 25, and page 9, line 12 to page 10, line 3, of the original description. The second method of preparation introduced into claim 1 is directly and unambiguously derivable from page 9, lines 12 to 22, of the description. Dependent claims 2 to 7 are based on original claims 3, 4, 5, 7, 8, 10 and 11.

3. Concerning the lack of support by the description objected to by the examining division, additional evidence was submitted by the appellant in the test report of 26 June 2000. In the application as filed the combination of Ni (as the hydrogenation component) with iron oxide (as the metal oxide component) is exemplified. It is also shown that this combination leads to an improved resistance against deactivation by sulphur compounds in particular with respect to a catalyst containing the hydrogenation component (ie nickel) but no metal oxide component. In the test report filed with the appellant's letter of 22 June 1995, it was further proved that an improvement is also achieved with the combination of Pt with MoO_3 . In the test report of 26 June 2000, the following combinations of hydrogenation and metal oxide components were

tested: Pd/Pt/Ni + MoO₃/Fe₂O₃; Pd/Pt/Ni + MnO₂; Pd/Pt/Ni + V₂O₅; Pd/Pt/Ni + ZnO. These combinations were shown to exhibit a better resistance to deactivation by a sulphur compound (2,5-dimethyl-thiophene) than a catalyst containing the hydrogenation component (ie Ni or Pd/Pt/Ni) but no metal oxide component. Thus, the improved resistance against deactivation indicated in claim 1 and in the general part of the description has been proved to be achieved with six combinations which differ considerably from each other since they contain metals of five different groups (IIB, VB, VIB, VIIB and VIII) as the metal oxide component, and three different hydrogenation components. These combinations, although very different from each other, all exhibit an improved resistance against deactivation by sulphur compounds with respect to a catalyst not containing the metal oxide component. In these circumstances, the board considers that the objection of unreasonable generalisation raised by the examining division on the basis that the properties of a catalyst are unexpected, cannot be maintained. Therefore, the requirement of support by the description set out in Article 84 EPC is fulfilled. This, of course, does not mean that an improvement of resistance against deactivation was shown to be achieved with respect to the catalysts of the closest prior art; however, this question has to be examined in connection with the requirement of inventive step and not with the issue of support by the description.

4. D4 discloses an improved catalytic composite for use in hydrotreating or hydrodesulphurisation processes (see col. 4, lines 12 to 29). Hydrotreating is, by definition, a catalytic process for sulphur, nitrogen, and heavy metal removal and hydrogenation of unsaturated

hydrocarbons in petroleum feedstocks. Such a process falls within the scope of the process defined in claim 1, especially when considering the statement on page 10 of the description that "the process according to the invention comprises in its most general sense reactions in which hydrogenation and/or dehydrogenation occurs". Examples I and II of D4 disclose a supported catalyst comprising an alumina support with a particle size lying within the claimed range, cobalt as the hydrogenation component, and molybdenum oxide as the metal-oxide containing component. The calcined catalyst was contacted with an atmosphere containing a vacuum gas oil and hydrogen at a temperature of 385°C for several hours (see col. 5, lines 11 to 24). The amounts of cobalt stated in examples I and II of D4 also fall within the range indicated in claim 1.

- 4.1 The appellant argued that the preparation method used in example I of D4 resulted in a single Co-Mo-sulphide phase with cobalt and molybdenum atoms being homogeneously mixed on an atomic scale in each particle, contrary to the methods defined in claim 1. The appellant explained that the simultaneous impregnation of the metal oxide and hydrogenation components on the support material did not in fact lead to both these components being present as separate particles on the support, contrary to the statement on page 9 of the present application, which should be deleted.

In the appellant's test report of 26 June 2000, the catalyst of example I of D4 was reproduced using the operating conditions indicated in this example, followed by a reduction in hydrogen at 400°C for two hours. The calcined samples and the reduced samples

were analysed by transmission electron microscopy with EDAX as indicated in the report. From the analyses, it could be determined that whenever a metal was present, both Co and Mo were present. This applied to both the calcined and the reduced catalysts. It was concluded that cobalt and molybdenum were mixed on an atomic level and that the preparation method used in example 1 resulted in a homogeneous distribution of cobalt and molybdenum and not in the production of separate particles of molybdenum oxide and cobalt. In view of this test report, the board can accept the appellant's arguments that the preparation method followed in example I of D4, in particular the simultaneous impregnation of the cobalt and molybdenum compounds and the calcination conditions, do not lead to a catalyst in which at least part of the hydrogenation component and at least part of the metal oxide component are present as separate particles on the support, contrary to the methods of preparation defined in claim 1.

As regards the catalyst according to example II of D4, the appellant did not reproduce this example. He argued that the operating conditions used in this example would also lead to both the cobalt and the molybdenum oxide not being present in the form of separate particles on the support. According to the appellant, the cobalt and molybdenum compounds would be dissolved in the 5% aqueous nitric acid solution during the intensive mixing carried out in example II so that this example was comparable to the situation in example I where both compounds were in solution during the mixing step. In the absence of evidence to the contrary, the board sees no reason not to accept these arguments. In these circumstances and in view of the fact that the simultaneous application of the components and the

calcination conditions used in example I result in the molybdenum oxide and the cobalt not being present as separate particles on the support, the board considers, in the absence of evidence to the contrary, that a comparable result would be obtained in example II of D4.

4.2 It follows from the above that the catalysts as defined in claim 1 of the main request differ from the catalysts of examples I and II of D4 by the metal oxide containing component as well as the hydrogenation component being present as separate particles on the support, with very limited direct contact of the hydrogenation component with the metal oxide component. Therefore the process according to claim 1 of the main request is new over the disclosure of D4.

4.3 The preceding considerations and conclusions apply likewise to the catalysts as defined in claim 8 of the main request. Although it is not explicitly stated in claim 8 that there is only limited direct contact of the hydrogenation component with the metal oxide containing component, this feature derives implicitly from the method of preparation stated in this claim. Therefore, claim 8 meets the requirement of novelty with respect to D4.

4.4 D5 concerns catalysts for the production of natural gas by the methanisation of CO. Claim 1 differs from the subject-matter of D5 at least by the different catalytic reaction. The catalyst according to claim 8 is also new with respect to the catalyst prepared by the method disclosed in example 3 of D5. The separate preparation of i) a support material with the hydrogenation component applied to it and ii) a support

material with the metal oxide applied to it, followed by mixing the thus loaded supports obviously leads to a catalyst which differs structurally from the catalyst prepared as indicated in example 3 of D4. Therefore, claims 1 and 8 also fulfil the requirement of novelty with respect to D5.

4.5 D6 discloses high temperature catalyst compositions for internal combustion engine, ie for oxidation of carbon monoxide and unburned hydrocarbons in an exhaust stream (see page 17, claim 1). The process according to claim 1 of the main request is novel over this disclosure at least in that it concerns a different catalytic reaction. Regarding the catalyst of claim 8, there is no doubt that the method of preparation as defined in this claim leads to a catalyst which is structurally different from the catalysts containing Pd supported on Ba, La-Ba, or La-RE-Ba stabilised alumina prepared by the methods disclosed in examples I, II, III and IV of D6. Therefore, the catalysts according to claim 8 are new with respect to those of D6.

4.6 The board has also checked that the disclosure of the remaining documents cited in the search report and during the examining procedure does not destroy the novelty of the process and catalysts according to claims 1 and 8 of the main request. Therefore, these claims meet the requirement of novelty set out in Articles 52(1) and 54 EPC.

5. The issue of inventive step was not considered at all in the communications from the examining division, nor was it discussed at the oral proceedings before it. Furthermore, the appellant has indicated at the oral proceedings before the board that the catalysts of D4

were less resistant to deactivation than catalysts containing a hydrogenation component but no metal oxide component and, thus, less resistant to deactivation than the claimed catalysts. The appellant has proposed to file evidence showing this improvement, if necessary. In these circumstances, the board, in the exercise of its discretionary power pursuant to Article 111(1) EPC, finds it appropriate to remit the case to the examining division for further prosecution.

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.

The Registrar:

The Chairman:

S. Hue

R. Spangenberg