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Datasheet for the decision of 23 June 2020

Case Number: T 0911/16 - 3.3.02

Application Number: 03785754.7

Publication Number: 1567617

IPC: C10G49/04, B01J35/10, B01J23/85

Language of the proceedings: EN

Title of invention:
HYDROPROCESSING OF HYDROCARBON USING A MIXTURE OF CATALYSTS

Applicant:
Albemarle Netherlands B.V.
Nippon Ketjen Co., Ltd

Headword:

Relevant legal provisions:
EPC Art. 56, 84

Keyword:
Claims - clarity (yes)
Inventive step - (yes)

Decisions cited:

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Catchword:

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Case Number: T 0911/16 - 3.3.02

D E C I S I O N
of Technical Board of Appeal 3.3.02
of 23 June 2020

Appellant: (Applicant 1)	Albemarle Netherlands B.V. Barchman Wuytierslaan 10 3818 LH Amersfoort (NL)
Appellant: (Applicant 2)	Nippon Ketjen Co., Ltd Seavans North, 2-1, 1-chome Shibaura Minato-ku, Tokyo 105-6791 (JP)
Representative:	Hoyng Rokh Monegier LLP Rembrandt Tower, 31st Floor Amstelplein 1 1096 HA Amsterdam (NL)
Decision under appeal:	**Decision of the Examining Division of the European Patent Office posted on 10 November 2015 refusing European patent application No. 03785754.7 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman	M. O. Müller
Members:	M. Maremonti
	P. de Heij

Summary of Facts and Submissions

I. The appeal by the applicants (hereinafter "appellants") lies from the decision of the examining division to refuse European patent application No. 03 785 754.7.

II. The main request refused by the examining division contained ten claims. Independent claims 1 and 7 were substantially identical to claims 1 and 7 as filed and read as follows (amendments to claims 1 and 7 as filed highlighted by the board):

"*1. A process for hydroprocessing a heavy hydrocarbon oil, comprising contacting a heavy hydrocarbon oil in the presence of hydrogen with a mixture of hydroprocessing catalyst I and hydroprocessing catalyst II wherein catalyst I comprises a Group VIB metal component and optionally a Group VIII metal component on a porous inorganic carrier, said catalyst having a specific surface area of at least 100 m^2/g, a total pore volume of at least 0.55 ml/g, at least 50% of the total pore volume in pores with a diameter of at least 20 nm (200 Å) and at least 65% of the total pore volume in pores with a diameter of 10-120 nm (100-1200 Å), and catalyst II comprises a Group VIB metal component and optionally a Group VIII metal component on a porous inorganic carrier, said catalyst having a specific surface area of at least 100 m^2/g, a total pore volume of at least 0.55 ml/g, 30-80% of the pore volume in pores with a diameter of 10-20 nm (100-200 Å), and at least 5% of the pore volume in pores with a diameter of at least 100 nm (1000 Å), ~~with~~ **wherein the** catalyst I ~~having~~ **has** a larger percentage of its pore volume in pores with a diameter of at least 20 nm (200 Å) than catalyst II.*"

"*7. A mixture of catalysts comprising a catalyst I which comprises a Group VIB metal component and optionally a Group VIII metal component on a porous inorganic carrier, and 0.5 to 6 wt.% of a Group VIII metal component, calculated as oxide on the weight of the catalyst, on a porous inorganic carrier, said catalyst having a specific surface area of at least 100 m^2/g, a total pore volume of at least 0.55 ml/g, at least 50% of the total pore volume in pores with a diameter of at least 20 nm (200 Å) and at least 65% of the total pore volume in pores with a diameter of 10-120 nm (100-1200 Å), and a catalyst II which comprises a Group VIB metal component and optionally a Group VIII metal component on a porous inorganic carrier, said catalyst having a specific surface area of at least 100 m^2/g, a total pore volume of at least 0.55 ml/g, 30-80% of the pore volume in pores with a diameter of 10-20 nm (100-200 Å), and at least 5% of the pore volume in pores with a diameter above 100 nm (1000 Å), ~~with~~* ***wherein the*** *catalyst I ~~having~~* ***has*** *a larger percentage of its pore volume in pores with a diameter of at least 20 nm (200 Å) than catalyst II.*"

The examining division also refused an auxiliary request. In claims 1 and 7 of this auxiliary request, the only amendment to above claims 1 and 7 was the restriction of the surface area of catalyst I to range from 100 to 180 m^2/g.

III. Document US 6,086,749 (D1) was the only document cited in the appealed decision.

The examining division came to the following conclusion, *inter alia*:

- The subject-matter of claims 1 and 7 of the main request and the auxiliary request did not involve

an inventive step in view of D1 taken as the closest prior art.

IV. In these appeal proceedings, the appellants contested the reasoning of the examining division and submitted that the subject-matter of claims 1 and 7 as filed before the examining division involved an inventive step in view of document D1 taken as the closest prior art.

V. The appellants were summoned to oral proceedings. In preparation for these proceedings, the board issued a communication in which it expressed, *inter alia*, the preliminary opinion that the subject-matter of claims 1 and 7 was not clear under Article 84 EPC.

VI. In a communication dated 4 June 2020, the appellants were informed that oral proceedings would be held by videoconference according to their request dated 28 May 2020.

VII. Oral proceedings were held on 23 June 2020 by videoconference. During the oral proceedings, the appellants submitted a new set of claims named main request, annex A. In response to clarity objections raised by the board, the appellants filed a new set of claims named main request, annex B.

VIII. Final request

The appellants request that the decision under appeal be set aside and that a European patent be granted on the basis of the claims of the main request, annex B, as filed during the oral proceedings on 23 June 2020.

IX. The appellants' arguments, where relevant to the present decision, are summarised as follows:

Clarity of the claims:

- Claims 1 and 7 had been amended by inserting the features expressing the methods used to measure the total pore volume, the pore size distribution and the specific surface area of catalysts I and II. In this way, the clarity objection raised by the board in its preliminary opinion had been overcome.

- Additionally, the term "*total*" had been inserted into claims 1 and 7 with reference to the pore volume of catalyst II. In claim 7, the weight percent of the Group VIII metal had been removed. In claims 2 and 8, the term "*essentially*" had been deleted. In this way, the clarity objections raised by the board during oral proceedings had been overcome.

- It had to be concluded that the subject-matter of the main request, annex B, met the requirements of Article 84 EPC.

Inventive step:

- Although not directed to the same purpose of the claimed invention, document D1 might be regarded as the closest prior art.

- Starting from D1, the objective technical problem had to be considered to be the provision of an improved process for hydroprocessing a heavy hydrocarbon oil with a reduced sediment formation whereby the hydrodesulfurisation activity was not sacrificed.

- In fact, the results reported in table 3 on page 20 of the application as filed showed that the claimed combination of catalysts I and II was not arbitrary

but instead was purposely selected to achieve asphaltene removal and sediment reduction while maintaining high hydrodesulfurisation (HDS) activity at the same time.

- The obtained performance was higher than expected, i.e. higher than the average of the performance obtained when catalysts A and B were used alone.

- D1 was the only document cited in the impugned decision. It did not contain any reference to the reduction of sediment formation. Moreover, although D1 disclosed the possibility of using a mixture of two different catalysts, it did not disclose any specific examples of a catalyst mixture, let alone a process carried out with such a catalyst mixture. There was no example in D1 that could have been reworked to show the advantages of the claimed process.

- D1 did mention in the passage from column 32, line 66 to column 33, line 11, preferred pore size distribution, pore volume and specific surface area of the catalyst. However, this was done in very general terms, without giving any indication as to how these parameters might be chosen when using a mixture of two catalysts.

- Therefore, D1 did not contain any teaching pointing to the claimed solution for the posed technical problem.

- It had to be concluded that the claimed subject-matter involved an inventive step.

Reasons for the Decision

1. Independent claims 1 and 7 of the main request, annex B, filed during the oral proceedings on 23 June 2020, read as follows, with the amendments to claims 1 and 7 of the main request refused by the examining division (which were practically identical to claims 1 and 7 as filed, see point II *supra*) having been highlighted by the board:

"*1. A process for hydroprocessing a heavy hydrocarbon oil, comprising* [...] *and catalyst II comprises a Group VIB metal component* [...] *30-80% of the* ***total*** *pore volume in pores with a diameter of 10-20 nm (100-200 Å), and at least 5% of the* ***total*** *pore volume in pores with a diameter of at least 100 nm (1000 Å), wherein* [...] *than catalyst II,*

wherein the determination of the total pore volume and the pore size distribution is effected via mercury penetration at a contact angle of 140° with a surface tension of 480 dynes/cm and the surface area is determined in accordance with the BET method based on N_2 adsorption."

"*7. A mixture of catalysts comprising a catalyst I which comprises a Group VIB metal component and optionally a Group VIII metal component on a porous inorganic carrier~~, and 0.5 to 6 wt.% of a Group VIII metal component, calculated as oxide on the weight of the catalyst, on a porous inorganic carrier~~, said catalyst* [...] *and a catalyst II which comprises a Group VIB metal component* [...] *30-80% of the* ***total*** *pore volume in pores with a diameter of 10-20 nm (100-200 Å), and at least 5% of the* ***total*** *pore volume in pores with a diameter above 100 nm (1000 Å), wherein* [...] *than catalyst II,*

wherein the determination of the total pore volume and the pore size distribution is effected via mercury penetration at a contact angle of 140° with a surface tension of 480 dynes/cm and the surface area is determined in accordance with the BET method based on N_2 adsorption."

Additionally, in dependent claims 2 and 8, the term "*consisting essentially of*" was amended to "*consisting of*".

Main request, annex B - compliance with Article 123(2) EPC

2. Claims 1 and 7 (point 1 *supra*) are based on claims 1 and 7 as filed. The insertion of the term "*total*" with reference to the pore size distribution of catalyst II is based on page 7, lines 1 to 6 of the application as filed. In claim 7, the deletion of the weight percent of the Group VIII metal component is allowable in view of the fact that this component is defined in the claim as being optional and that, as is disclosed on page 5, lines 11 to 14 of the application as filed, the weight percent is merely preferred. In claims 1 and 7, the features specifying the methods used to measure the total pore volume, the pore size distribution and the surface area are disclosed on page 5, lines 20 to 30 of the application as filed. Finally, the amendment of "*consisting essentially of*" to "*consisting of*" in claims 2 and 8 with reference to the carrier of catalysts I and II is based on page 6, lines 21 to 24 of the application as filed. In fact, here, the presence of additional components besides alumina in the carrier is disclosed as being purely optional: "*minor amounts of other components* ***may be present***" (emphasis added by the board).

The board thus concludes that the main request, annex B, complies with the requirements of Article 123(2) EPC.

Main request, annex B - compliance with Article 84 EPC

3. The board is satisfied that the claims of the main request, annex B, are clear within the meaning of Article 84 EPC for the following reasons.

3.1 The insertion of the term "*total*" in claims 1 and 7 clarifies that the pore size distribution of catalyst II is related to the total pore volume as previously defined in both claims as being at least 0.55 ml/g.

3.2 In claim 7, the deletion of the feature expressing the weight percent of the Group VIII metal component removes the contradiction arising from the fact that this component is previously defined in the claim as only optionally being contained in the claimed mixture.

3.3 In claims 1 and 7, the features specifying the methods used to measure the total pore volume, the pore size distribution and the surface area have been included. This insertion avoids ambiguities resulting from the fact that there are different measurement methods for the mentioned parameters, which may lead to different results.

3.4 The amendment of "*consisting essentially of*" to "*consisting of*" in claims 2 and 8 clarifies that according to this specific embodiment, the carrier of catalysts I and II is only alumina. This avoids uncertainties as to which further components may be present and in which amount.

3.5 The board thus concludes that the claims of the main request, annex B, meet the requirements of Article 84 EPC.

Main request, annex B - novelty under Article 54 EPC

4. In the impugned decision, the examining division did not raise any objection to the novelty of the claimed subject-matter. The board sees no reason to take another stance. The subject-matter of the main request, annex B, is thus novel (Article 54 EPC).

Main request, annex B - inventive step under Article 56 EPC

5. The closest prior art

5.1 In the impugned decision (point 3), the examining division indicated that document D1 was the closest prior art. Although the appellants disputed that D1 might represent the most relevant state of the art, they did not indicate any other prior art document. Moreover, they also started from D1 in their argumentation concerning inventive step.

5.2 Document D1 discloses (column 1, lines 11 to 61; column 24, line 64 to column 25, line 9; column 28, lines 20 to 27; column 29, lines 17 to 25; column 30, lines 16 to 67; column 31, line 39 to column 32, line 24; column 32, line 66 to column 33, line 11; column 41, lines 27 to 67) a process for hydroprocessing a heavy hydrocarbon oil, comprising contacting a heavy hydrocarbon oil in the presence of hydrogen with a mixture of two catalysts, both comprising a Group VIB metal component and a Group VIII metal component on a porous inorganic carrier, both catalysts having a specific surface area of at least 200 m^2/g to 600 m^2/g and a total pore volume ranging from 0.8 to 3 ml/g.

According to D1 (column 32, line 66 to column 33, line 11), at least 50% of the total pore volume is in pores with a diameter from 100 to 300 Å, while less than 20% of the total pore volume is in pores with a diameter of 0 to 100 Å.

D1 specifies (*loc. cit.*) that the two catalysts forming the disclosed mixture consist of catalyst particles with different catalytic characteristics, such as different physical and/or catalytic properties. Each of the catalysts would typically have a particular conversion reaction capability that differs from the particular conversion reaction capability of the other catalyst. Thus, by way of example, one of the catalysts could possess hydrodemetallation capabilities, whereas the other catalyst could possess hydrodenitrification and/or hydrodesulfurisation capabilities. These aims are also mentioned in the present application (page 3, lines 9 to 21).

The board concludes that D1 does represent a suitable starting point for the assessment of inventive step.

6. The technical problem

6.1 D1 does not disclose any **specific** pair of catalysts and does not disclose any pore size or surface area characteristics of **each** of the two catalysts forming such a pair. As set out above, D1 only discloses that both catalysts have a specific surface area of at least 200 m^2/g to 600 m^2/g. Without specifying that this applies to a pair of catalysts, D1 further discloses a total pore volume ranging from 0.8 to 3 ml/g, and discloses that at least 50% of the total pore volume is in pores with a diameter from 100 to 300 Å, while less than 20% of the total pore volume is in pores with a diameter of 0 to 100 Å.

6.2 The disclosure of D1 can thus at best be interpreted as referring to a pair of two catalysts each having the disclosed surface area and pore size characteristics. The subject-matter of claims 1 and 7 differs from this teaching of D1 in that the second catalyst is specified in the claims to have

(a) 30-80% of the pore volume in pores with a diameter of 10-20 nm (100-200 Å) and

(b) at least 5% of the pore volume in pores with a diameter of at least 100 nm (1000 Å),

and in that

(c) the first catalyst has a larger percentage of its pore volume in pores with a diameter of at least 20 nm (200 Å) than the second catalyst.

6.3 Table 3 of the application as filed (page 20) shows the results obtained by using a catalyst mixture according to claim 7 (50%/50% of catalysts A and B corresponding to catalysts I and II according to claim 7) in a process according to claim 1, especially in terms of hydrodesulfurisation (HDS), asphaltene removal and sediment formation. The results are compared with the performance of the two catalysts when used alone.

The results show that catalyst A (catalyst I in claim 7), when used alone, produces very low sediment formation (0.02) in comparison with catalyst B (catalyst II in claim 1) when used alone (0.29). The asphaltene removal is also higher (100 vs. 81). Instead, the HDS activity is higher for catalyst B (126 vs. 100). When a catalyst mixture according to claim 7 is used, a good balance is obtained between asphaltene and sediment reduction and hydrodesulfurisation activity. In fact, performance that is higher than the

average between the performance of the single catalysts is achieved in terms of HDS and asphaltene and sediment reduction.

6.4 On the basis of these results, the board concludes that the objective technical problem lies in the provision of a catalyst mixture for the hydroprocessing of heavy hydrocarbon oils resulting in a good balance between asphaltene and sediment reduction and hydrodesulfurisation.

6.5 In the appealed decision (points 5 to 7), the examining division stated that the effect of the distinguishing features over D1 could not be ascertained from the application, especially since a bi-catalytic system was compared with mono-catalytic systems. In the absence of a technical effect, the technical problem was regarded as the mere provision of an alternative catalyst mixture for hydroprocessing, with arbitrary properties.

6.6 The board would agree with the examining division had D1 disclosed a specific pair of catalysts which differed in surface area and pore size characteristics. In that case, a comparison of the performance achieved with this pair of catalysts with that achieved by the claimed pair of catalysts might indeed have been necessary to establish any technical effect. However, as set out above, D1 does not disclose any specific embodiment of a catalyst pair to be used in the hydroprocessing of a heavy hydrocarbon oil that would allow a comparison with the claimed subject-matter. D1 (column 32, line 66 to column 33, line 11) only provides very general teaching of, *inter alia*, the pore size distribution of the catalyst to be used.

6.7 The results in table 3 show that by selecting two catalysts with specific surface area and pore size

characteristics as claimed from the very general and broad disclosure of D1, a good balance between asphaltene and sediment reduction and hydrodesulfurisation is achieved. Hence, it is on this basis that the objective technical problem needs to be formulated.

7. Obviousness of the claimed solution

7.1 As set out above, although D1 discloses that a mixture of two catalysts can be used in the hydroprocessing of heavy hydrocarbon oils, it does not contain any specific examples of such a mixture. In fact, examples I to III (columns 42 to 45) are directed to demonstrating the functionalities of an ebullating bed. In example IV (columns 45 to 47), a hydrocarbon feed is directed to a catalyst bed that only contains a single catalyst. The obtained performance is not reported.

7.2 D1 does mention e.g. in column 33, line 66 to column 34, line 18, that each catalyst has the appropriate catalytic activity for the specific application, e.g. demetallation, denitrification, hydrodesulfurisation etc., and that the catalytic activity also depends, among various parameters, on pore volume and pore size distribution. However, no specific teaching is provided in D1 that would have prompted the skilled person to select different pore size distributions for the two catalysts to be used, let alone distributions appropriate for solving the posed technical problem. In fact, while asphaltenes are briefly mentioned as contaminants (column 26, lines 30 to 35), D1 does not indicate how, i.e. by means of which specific catalyst characteristics, these can be removed. Additionally, the problem of sediment formation is not addressed in

D1, which does not mention anywhere how sediment formation can be reduced.

7.3 The very general teaching (point 6.4 *supra*) of the pore size distribution of the catalyst to be used, as reported in column 32, line 66 to column 33, line 11 of D1, is to be regarded as nothing more than an invitation for the skilled person to perform a research programme to find out, *inter alia*, the pore size distributions of the two catalysts making it possible to solve the posed technical problem. Such a research programme would have required the skilled person to exercise inventive skill.

7.4 D1 is the sole document cited in the appealed decision. For the reasons mentioned above, the board is convinced that the skilled person would not have been prompted by D1 and/or common general knowledge to use a catalyst mixture as defined in claim 7 in a process as defined in claim 1 to solve the posed technical problem.

The board thus concludes that the subject-matter of claims 1 and 7, and of claims 2 to 6 and 8 to 10, which are dependent on these claims, involves an inventive step within the meaning of Article 56 EPC.

Conclusion

8. The appellants' main request, annex B, is allowable under the EPC.

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 with the order to grant a patent on the basis of the following claims and a description to be adapted thereto:

   Claims:

   Claims 1 to 10 of the main request, filed during the oral proceedings of 23 June 2020 as annex B.

The Registrar: The Chairman:

N. Maslin M. O. Müller

Decision electronically authenticated