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Datasheet for the decision
of 10 April 2024

Case Number: T 0190/22 - 3.3.06

Application Number: 10290480.2

Publication Number: 2428547

IPC: C10G3/00, C10L1/08

Language of the proceedings: EN

Title of invention:
Process for the continuous hydrogenation of triglyceride containing raw materials using a nickel and molybdenum based catalyst

Patent Proprietor:
IFP Energies Nouvelles

Opponent:
Neste Oyj

Headword:
Catalyst with low nickel/molybdenum atomic ratio/IFP

Relevant legal provisions:
EPC Art. 100(a), 54, 56

Keyword:

Novelty - (yes) - catalyst atomic ratio not implicitly disclosed
Inventive step - (yes) - non-obvious solution of the underlying technical problem

Decisions cited:

Catchword:

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Case Number: T 0190/22 - 3.3.06

DECISION
of Technical Board of Appeal 3.3.06
of 10 April 2024

Appellant: (Patent Proprietor)	IFP Energies nouvelles 1 & 4 avenue de Bois-Préau 92500 Rueil-Malmaison (FR)
Representative:	IFP Energies nouvelles Département Propriété Industrielle Rond Point de l'échangeur de Solaize BP3 69360 Solaize (FR)
Respondent: (Opponent)	Neste Oyj Keilaranta 21 02150 Espoo (FI)
Representative:	Zacco Denmark A/S Arne Jacobsens Allé 15 2300 Copenhagen S (DK)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted on 7 December 2021 revoking European patent No. 2428547 pursuant to Article 101(3)(b) EPC.**

Composition of the Board:

Chairman	J.-M. Schwaller
Members:	L. Li Voti
	C. Heath

Summary of Facts and Submissions

I. The proprietor's appeal is against the decision of the opposition division to revoke European patent no. 2 428 547, claim 1 thereof (main request) reading:

"*1. Process for the continuous hydrogenation of triglyceride containing raw materials like vegetable oils to produce diesel and/or kerosene fraction in a fixed bed reactor system having several catalyst beds comprising hydrogenation catalyst, which beds are arranged in series, wherein the raw material feed, hydrogen containing gas and diluting agent are passed together through the catalyst beds of the reactor system at hydrogenation conditions, wherein*
a) the raw material feed stream is divided into a number of different partial stream F1 to Fn identical to the number of catalyst beds n in the reactor system at a temperature of less than 80°C, the stream of hydrogen containing gas is also divided into the same number of different partial stream H1 to Hn at a temperature of less than 110°C, then partial raw material feed stream and partial stream of hydrogen containing gas H1 are passed to the first catalyst bed, and before the first partial mixed stream F1+H1 enters the first catalyst bed, it is mixed with added diluting agent which is a specific fraction of the hydrogenation product obtained by the present process leaving the last bed, partial raw material feed stream F2 and partial stream of hydrogen containing gas H2 are passed to the second catalyst bed and so on, if n is greater than 2, each partial stream of raw material feed following in downstream direction being that much larger than the preceding one, that the weight ratio of diluting agent to raw material feed is

essentially the same at the entrance of all catalyst beds and does not exceed 4:1, said diluting agent comprising the hydrogenation product formed in every catalyst bed as well as the specific fraction recycled to the reactor inlet at the first catalyst bed and is referred to as added diluting agent,
b) the temperature at the reactor inlet at the first catalyst bed after start up phase is adjusted by adding diluting agent called added diluting agent, which is recovered from the reactor exit product mixture by separation in one or two steps without controlled pressure reduction,
c) added diluting agent is only added to the streams of raw material feed F1 and hydrogen containing gas H1 entering at the reactor inlet and passing through the first catalyst bed, and
d) hydrogen is used in excess of the theoretical hydrogen consumption,
wherein the hydrogenation takes place at hydrogen partial pressures in the range of 1 to 8 MPa, at temperatures of 145 to 280°C, and with the LHSV of the raw material feed in each bed from 0.1 to 5 m^3/m^3 *catalyst/h*
characterized in that a hydrogenation bulk or supported catalyst comprising an active phase constituted by at least one element from group VIB and at least one element from group VIII, said elements being in the sulfide form, and the atomic ratio of the metal or metals from group VIII to the metal or metals from group VIB being strictly more than 0 and less than 0.095, is used in at least the first catalyst bed of the fixed bed reactor system."

Dependent claims 2 to 11 relate to particular embodiments of the above process.

II. With its grounds of appeal the appellant defended the patent as granted (main request) and filed inter alia five sets of claims as auxiliary requests 1 to 5 as well as Supplemental Examples 2 and 3 (in the following D6) and Supplemental Example 4 (in the following D9).

III. In its reply the opponent/respondent argued that claim 1 as granted lacked novelty over D8 (EP 2 226 375 A1) and that all the requests on file lacked an inventive step starting from document D1 (US 2009/0318737 A1) or D5 (US 2010/0163458 A1) as representing the closest prior art. It also referred to the "APPENDIX" contained under point 6 of its reply (pages 33-34) and to documents D3 (US 2009/0326285 A1) and D4 ("*Deoxygenation of vegetable oils over sulfided Ni, Mo and NiMo catalysts*" by D. Kubička et al., Applied Catalysis A: General 372 (2010) pages 199-208).

IV. At the oral proceedings held on 10 April 2024, the appellant confirmed that it requests that the decision under appeal be set aside and the patent be maintained as granted or, as an auxiliary measure, that the patent be maintained in amended form on the basis of one of auxiliary requests 1 to 5 filed with the grounds of appeal.

The respondent also confirmed that it requests the appeal to be dismissed.

Reasons for the Decision

1. *Main request - Novelty*

1.1 D8 was published on 8 September 2010, i.e. on the filing date of the patent in suit, and may thus be cited only against the novelty of the claimed subject-matter in virtue of Article 54(3) EPC. In its example 3, D8 discloses a process for the continuous hydrogenation of triglyceride-containing raw materials in a fixed bed reactor system having five catalyst beds arranged in series comprising a commercial NiMo hydrogenation catalyst, with the applied process conditions being similar to those in claim 1 at issue. Taking into account the teachings in paragraphs [0026] and [0033] and claim 1 that the catalysts are preferably sulfided and that the weight ratio of recycled diluting agent to raw material feed is essentially the same at the entrance of all catalyst beds and does not exceed 4:1, the process of example 3 thus contains all features of claim 1 at issue with the exception of the atomic ratio of the metal from group VIII (Ni) to the metal from group VIB (Mo) of strictly more than 0 and less than 0.095.

1.2 According to the respondent, since:
- the process of example 3 yields (table 2 in paragraph [0056]) non detectable or only trace amounts of CO and CO_2, and so almost completely suppressing decarboxylation/decarbonylation (DCO) reactions,
- such CO and CO_2 amounts were even inferior to those obtained in example 1 of the patent in suit (paragraph [0075], Table 2: 0.15 and 0.35 wt.% on feed) wherein a catalyst having an atomic ratio Ni/Mo of 0.02 was used, and
- according to the patent in suit (paragraph [0009]), the chosen atomic ratio enhanced the selectivity for hydrodeoxygenation (HDO) reactions and limited DCO,

it followed that the NiMo catalyst in example 3 of D8 was necessarily one similar to that used in the contested patent, i.e. one having a Ni/Mo atomic ratio of more than 0 and less than 0.095. Thus, a skilled person trying to rework example 3 of D8 would find it reproducible only by using a NiMo catalyst having an atomic ratio according to claim 1 at issue.

This conclusion was supported by the experimental data summarised in the APPENDIX filed with its reply to the grounds of appeal and showing that a constant reduction of the Ni/Mo atomic ratio constantly increased HDO reactions against DCO. In particular, a low atomic ratio of 0.02 like in A6 (example 1 of the patent) or in A7 (example in the not prepublished US 2010/0240942 A1 (D7)), or the absence of Ni like in A8 (example of D5) were able to suppress the DCO reactions to a great extent, whilst higher atomic ratios of 0.4, 0.395 or 0.25 like in A1 (comparative example of D7), A2 (example 2 of D6), A3 (comparative example of D5) or A4 (example of D9) were by large less efficient. Finally A5, another example of D7, relating to a catalyst with an atomic ratio of 0.05, provided DCO results in-between those mentioned above.

It followed that an atomic ratio as required by claim 1 at issue was implicitly disclosed in example 3 of D8.

1.3 The board disagrees therewith and remarks that D8 (paragraphs [0057]-[0058]) explicitly teaches that the achieved suppression of DCO reactions is due to the selected sequence of process steps and does not contain any disclosure suggesting that a particular atomic ratio of the used NiMo catalyst might have an influence in this respect. Furthermore, in example 2 of D8 wherein **the same catalyst as in example 3** was used under different process conditions (inter alia higher reactor temperature and weight ratio of recycled diluting agent to raw feed), increased amounts of CO and CO_2 were obtained (0.29 and 7.73 wt% on feed according to table 2) and which were higher than those obtained in example 1 of the patent in suit (see above). Thus, even by using the same catalyst as in example 3, considerable DCO reactions still occurred. Therefore, in the board's view, a skilled reader in the art would not unequivocally gather from the overall context of D8 that a specific NiMo catalyst is responsible for the suppression of DCO in example 3.

1.3.1 The board notes that there is also no evidence for the respondent's allegation that D8/example 3 would only be reproducible with a catalyst as required by claim 1 at issue. Moreover, since the opposition division acknowledged the novelty of claim 1 at issue in the contested decision, it was up to the opponent/appellant to demonstrate that these findings were wrong, e.g. by reworking the cited example of D8.

1.3.2 As regards the experimental evidence considered by the respondent and summarised in the APPENDIX, this also does not include any reworking of the process of example 3 of D8, which requires five catalyst beds and palm oil as starting product, whilst the experiments in APPENDIX (including example 1 of the patent in suit) all use **rapeseed oil** as a starting material and either **three** catalyst beds (A2, A4 and A6), or only **one** catalyst bed without any recycling of the reactor product mixture and control of the ratio of diluting agent to raw material feed (A1, A3, A5, A7, A8). Therefore, none of these processes is, in the board's view, directly comparable with that of D8/example 3, so that it cannot be concluded from this data that - contrary to the explicit teaching of D8 - the excellent DCO suppression in its example 3 is due to the atomic ratio of the NiMo catalyst used.

1.3.3 The board further observes that the NiMo catalyst used in D8 is a commercial one, and no evidence was provided that the skilled person at the publication date of D8 would have considered said catalyst to be the one required by the patent in suit.

The respondent cited in this respect document D3, published about one year before D8, but D3 merely discloses (paragraphs [0026]-[0027]; claims 1-3 and 10-12) that hydrotreating catalysts containing a Group IV-VIII transition metal, for example Mo, may contain a promoter metal selected from Ni, Co or mixtures thereof, in an amount ranging from 0.5 wt% to 15 wt% of the weight of the metal in the sulfided metal catalyst (corresponding to a Ni/Mo atomic ratio of 0.01 to 0.29 in a hypothetical NiMo catalyst), without however suggesting that any specific catalyst encompassed by this broad range was commercially available. In fact, the only example in this document (see paragraph [0038]) relating to a catalyst with a broad range (1:100 to about 1:2) of molar ratios of Ni or Co to Mo is a **hypothetical** one (see first line of this paragraph) which cannot support the assumption that all catalysts encompassed by the disclosed broad range of Ni/Mo atomic ratios were commercially available.

1.3.4 It follows from the above considerations that D8 does not directly and unambiguously disclose a catalyst as claimed, so that the process disclosed in D8 cannot be considered to inevitably comprise all the features of claim 1 as granted.

1.4 Therefore, claim 1 as granted and, consequently also claims 2 to 11 dependent thereon, are novel over D8 and comply with the requirement of Article 54 EPC.

2. *Main request - Inventive step*

2.1 The object of the invention (paragraphs [0007]- [0008] of the patent) is the provision of a hydrotreating process using less recycle and/or avoiding deoxygenation of triglycerides through decarb-reactions, maximising the base gas oil and/or kerosene yield and promoting selectivity towards HDO against DCO while limiting hydrogen consumption.

2.2 While D1 was chosen as the starting point for the assessment of inventive step by both parties during opposition and in the decision under appeal, the respondent argued that also D5 could be considered to represent a suitable starting point.

D1 (paragraph [0002]) relates to a process for producing paraffinic hydrocarbons useful as fuel from renewable feedstock, such as the triglycerides found inter alia in plant oils, which process involves hydrogenation and DCO and/or HDO in two or more reaction zones, each reaction zone having an associated feedstock. In particular in its paragraphs [0005] and [0018], the process of D1 allows for the volume of recycle and hydrogen consumption to be significantly reduced so that it deals explicitly with at least part of the goals of the patent in suit and is thus a suitable starting point.

2.3 During oral proceedings the respondent agreed that the technical problem to be solved over this prior art could be formulated as the provision of a further process for the continuous hydrogenation of triglyceride-containing raw materials like vegetable oils in a fixed catalytic bed reactor system having several catalyst beds comprising hydrogenation catalysts which maximises the yield in targeted hydrocarbons and promotes selectivity towards HDO against DCO.

The board, for the sake of argument in its favour, agrees that by taking into account the entire description of D1 and in particular the process features disclosed in paragraphs [0002], [0015], [0016], [0018], [0022], claims 1 and 16, the process disclosed more in detail in figures 1 and 2 and their corresponding parts of the description (paragraphs [0023]-[0036]) can be considered to mainly differ from the process of claim 1 at issue in that it does **not** disclose the use of a catalyst with an atomic ratio of the metal or metals from group VIII to the metal or metals from group VIB strictly more than 0 and less than 0.095.

2.4 As to the question whether the above problem has been effectively solved, a comparison of the process according to example 1 of the patent in suit with similar processes in D6 and D9, in which at least the catalyst of the first catalyst bed has a higher Ni/Mo atomic ratio than claimed, confirms that the claimed process more effectively enhances HDO and suppresses DCO reactions and thus maximises the yield of targeted hydrocarbons.

Even though these results were not in dispute, the respondent argued that there was no evidence that the same effect would be obtained in a process wherein **only** the first catalyst bed contained a catalyst with the required Ni/Mo atomic ratio. In its view, the other catalytic beds could contain a catalyst having higher atomic ratios outside the range of claim 1 at issue, and thus promote more DCO and so jeopardise the solution of the above technical problem.

2.4.1 The board however notes that the decision under appeal had already stated in paragraph 1.2.4 that "*a potentially surprising technical effect has only been shown for the type of catalyst used, in particular for the Ni:Mo ratio, in the experimental data supplied during the examination of the underlying patent application, present as D6 in the opposition proceedings...For the choice of catalyst, the effect has been shown to lead to a high HDO selectivity at high conversion, as also recognised by the* ***opponent*** *and division.*"

Moreover, the respondent accepted in its reply to the grounds of appeal (points 3.50-3.52) that the technical problem underlying the claimed invention had been solved by reducing the Ni content in the NiMo catalyst.

Under these circumstances, it is incumbent on the respondent/opponent to show that the above otherwise recognised technical advantage could not be achieved throughout the entire scope of claim 1.

In this respect, no evidence however was provided that in a process as claimed, wherein the product exiting the reactor is recycled as diluting agent to the inlet of the first catalyst bed, the possibly promoted DCO selectivity in the further catalyst beds would consistently offset the strongly enhanced HDO selectivity achieved in the first catalyst bed. Moreover, the claimed process certainly provides more HDO selectivity than the process of D1, which (see below) does not contain any preference for the catalyst to be used and suggests inter alia the use of catalysts based on Ni, Pd or Pt, which provide mainly DCO.

Therefore, in the absence of any evidence, the board cannot accept the respondent's argument and can only conclude that the above technical problem has been convincingly solved throughout its entire scope by means of the process of claim 1 at issue.

2.5 It remains to be evaluated whether it was obvious for the skilled person starting from D1 to select the catalyst of claim 1 at issue in order to solve the above formulated technical problem.

2.5.1 It is not in dispute that D1 (paragraph [0015]) does not have any preference for the hydrogenation catalyst to be used, which thus can be any of those well known in the art, such as Ni or NiMo dispersed on a high surface area support, or any other noble metal catalyst, such as those based on Pt and/or Pd dispersed on gamma-alumina. However, as D1 (paragraph [0018]) suggests that a reduction of the operating pressure advantageously increases DCO and reduces HDO, the skilled person wishing to increase the selectivity towards HDO would thus rather increase the operating pressure within the limits indicated in this document, which are also those within the limits of the patent in suit, but it would not necessarily select a particular hydrogenation catalyst. Therefore, D1 does not contain any teaching that could lead the skilled person to select the combination of features of claim 1 at issue and more particularly the claimed catalyst with the specific Ni/Mo atomic ratio with the expectation of solving the technical problem formulated above.

2.5.2 The respondent argued that the selection of the catalyst as claimed was obvious from the disclosure of D4 or D5.

2.5.3 D4 (abstract; page 200, paragraphs 2.1 and 2.2; page 208, paragraph 4. Conclusions) is a study about deoxygenation of vegetable oils in a fixed bed reactor containing one catalyst bed under pressure, temperature and LHSV conditions similar to those used in the patent in suit. The tested catalysts were Ni or Mo on alumina, or NiMo on alumina with an atomic Ni/(Ni+/Mo) ratio in the range 0.2-0.4, i.e. an atomic Ni/Mo ratio between 0.25 and 0.667, which is above the upper limit of claim 1 at issue. D4 discloses that the NiMo catalysts tested were be more active than the monometallic catalysts and provided a higher yield of hydrocarbons at a given conversion. Moreover, they yielded a mixture of DCO and HDO products whilst the Mo catalyst yielded almost exclusively HDO hydrocarbon products and the Ni catalyst only DCO products. D4 also explicitly discloses that the effect of the atomic Ni/(Ni+/Mo) ratio in the range 0.2-0.4 on activity and selectivity was not significant.

The board notes that D4 is silent about the behaviour of NiMo catalysts with lower Ni/Mo atomic ratios and contains no suggestion that a reduction of this ratio would suppress more consistently DCO and thus provide a significant advantage over the catalysts specifically tested in D4, which are similar to those tested in the comparative examples of D6 and D9 and promote more effectively DCO than a catalyst according to claim 1 at issue.

Also figures 8 and 9 of D4 do not suggest that an atomic ratio as claimed could provide the convincingly proven technical advantage of maximising the yield in targeted hydrocarbons and further promoting selectivity towards HDO. Figure 8 merely shows that the Mo catalyst does not promote almost any DCO, that the Ni catalyst promotes almost only DCO and that the NiMo catalyst with an atomic ratio Ni/(Ni+/Mo) ratio of 0.3 promotes both HDO and DCO, which results are in-between those of the other two catalysts and slightly closer to those of the monometallic Mo catalyst. However, there is no reason to assume, in view of this isolated experiment and in the absence of further corroborating experimental evidence, that a further lowering of the Ni/Mo atomic ratio would provide results significantly closer to those of the monometallic Mo catalyst. In fact, figure 9 shows that the catalyst with the lowest Ni/(Ni+/Mo) atomic ratio, namely 0.2, shows only a slightly increased HDO selectivity and reduced DCO selectivity with respect to the other two catalysts with a ratio of 0.3 and 0.4, which different selectivity is explicitly considered not to be significant in D4 (see abstract). Therefore, in the absence of corroborating evidence that a lowering of the Ni/Mo atomic ratio would continuously increase HDO and reduce DCO selectivity, the skilled person would not derive from D4 any suggestion that would have prompted him to try a catalyst as claimed instead of i) either the best HDO catalyst tested in this document, i.e. Mo on alumina, or ii) those catalysts shown in D4 to provide the best hydrocarbon yield together with an acceptable DCO reduction such as one of the NiMo catalysts with a Ni/(Ni+/Mo) atomic ratio between 0.2 and 0.4 outside the range of claim 1 at issue.

2.5.4 D5 (paragraphs [0010]-[0012], [0021], [0023]-[0027]) discloses a hydrogenation process able to promote HDO and reduce DCO by using a catalyst having as active phase only one sulfided group VIB element, like Mo. In particular, the example of D5 with a process carried out in a fixed bed reactor containing one catalyst bed under pressure, temperature and LHSV conditions similar to those of the patent in suit, shows that a Mo catalyst (HDT1) provides much better selectivity towards HDO in comparison to a NiMo catalyst (HDT2) having a Ni/Mo atomic ratio of 0.395, i.e. well above the limits of claim 1 at issue.

D5 is however completely silent about the activity of NiMo catalysts having possibly lower Ni/Mo atomic ratios and its teaching is thus similar to that of D4. Moreover, in view of its paragraph [0023] which states that the selectivity towards HDO reactions can be **surprisingly** better controlled by means of a monometallic catalyst, this teaching would have rather prompted the skilled person faced with the technical problem formulated above to try the monometallic Mo catalyst and not to try further bimetallic catalysts, let alone those having a lower Ni/Mo atomic ratio than that of example HDT2.

2.5.5 The board remarks also that the only experimental data reported in the APPENDIX and representing the state of the art, are the examples of D5 already discussed above (A3 and A8 in the APPENDIX). Therefore, this document does not need further discussion.

2.5.6 The board thus finds that the overall teaching of the cited prior art would have led the skilled person, faced with the above technical problem to try a monometallic Mo catalyst or, as a less favourable alternative, the NiMo catalysts disclosed in D4 which provide very good hydrocarbon yield combined with an acceptable DCO suppression and were manifestly better than other known hydrogenation catalysts disclosed in D1, such as Ni, Pd or Pt.

Consequently, the skilled person would not have been led by the prior art to try a catalyst with a Ni/Mo atomic ratio as required by claim 1 at issue and would have arrived at the claimed subject-matter only by using hindsight.

2.5.7 Finally, taking **D5** as starting point for the evaluation of inventive step, the closest prior art is represented by the process of its example, which is carried out in a fixed bed reactor containing one catalyst bed under pressure, temperature and LHSV conditions similar to those of the patent in suit, and wherein the catalyst is Mo on alumina.

2.5.8 Even though the skilled person faced with the technical problem formulated above in view of the description of D5 (paragraphs [0057]-[0058]) could have decided to modify this process by using more than one catalyst bed and control the reaction temperature, and in the case it would have found a teaching in D1 of how to adapt the process conditions, the skilled person for the reason already exposed above would not have found any suggestion in D1 or D4 to try the catalyst of claim 1 at issue, instead of the monometallic Mo catalyst of D5 which has the best HDO selectivity or, as a less favourable alternative, the known NiMo catalysts disclosed in D4 (and in the comparative HDT2 example of D5), which provide a good hydrocarbon yield and acceptable DCO suppression.

Therefore, the board cannot agree with the respondent that the skilled person seeking to find other suitable catalysts would have routinely added Ni to the Mo catalyst of D5 and gradually increased its content (as suggested under point 3.77 of the respondent's reply to the grounds of appeal), so as to arrive at a catalyst having a Ni/Mo atomic ratio as required by claim 1 at issue and disclosed in D3 (which disclosure concerns, as explained above, only hypothetical catalysts and not commercially available ones).

2.5.9 The board thus concludes that also starting from D5 the skilled person would have arrived at the claimed subject-matter only by using hindsight.

2.6 It follows form the above considerations that the subject-matter of claim 1 at issue and, consequently, that of dependent claims 2 to 11, involve an inventive step under Article 56 EPC.

3. The board therefore concludes that the grounds of opposition under Article 100(a) EPC in combination with Articles 54 and 56 EPC do not prejudice the maintenance of the patent as granted.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the department of first instance with the order to maintain the patent as granted.

The Registrar: The Chairman:

Beschwerdekammern des Europäischen Patentamts
Boards of Appeal of the European Patent Office
Chambres de recours de l'Office européen des brevets

A. Pinna J.-M. Schwaller

Decision electronically authenticated