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D E C I S I O N
of 19 April 1994

Case Number: T 0585/90 - 3.3.2

Application Number: 84200028.3

Publication Number: 0114704

IPC: B01J 23/74

Language of the proceedings: EN

Title of invention:
Nickel-based catalyst, its preparation and its application

Patentee:
Unilever N.V., et al

Opponent:
Hoechst Aktiengesellschaft

Headword:
Ni-catalyst/UNILEVER

Relevant legal norms:
EPC Art. 56

Keyword:
"Non-obvious improvement over the closest prior art"

Decisions cited:
-

Catchword:
-

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Case Number: T 0585/90 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 19 April 1994

Appellant: (Proprietor of the patent)
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Representative:
-

Decision under appeal:
Decision of the Opposition Division of the European Patent Office dated 8 March 1990, posted on 25 May 1990 revoking European patent No. 0 114 704 pursuant to Article 102(1) EPC.

Composition of the Board:

Chairman: P.A.M. Lançon
Members: M.M. Eberhard
C.E.M. Holtz

Summary of Facts and Submissions

I. European patent No. 0 114 704 based on application No. 84 200 028.3 was granted on the basis of twenty claims.

II. The Respondent (Opponent) filed a notice of opposition requesting revocation of the patent on the grounds of lack of inventive step and insufficiency of disclosure. Of the documents cited during the opposition procedure only the following ones were relied upon in the present appeal:

(1) J.Am. Oil Chemists' Soc., Vol. 54, 1977
(4) GB-A-1 367 088.

III. The Opposition Division revoked the patent on the grounds that the catalyst and the method for its preparation as defined in the claims of the three requests filed on 8 March 1990 did not involve an inventive step. In connection with the process claim it was considered that the essential steps of the claimed process resided in a rapid precipitation step and a relatively long ageing step and that a similar process was disclosed in document (4). According to the decision, even if the precipitation step were to be carried out with a mechanical energy input considerably higher than that required for normal stirring, this feature would not be inventive since the result of rapid precipitation, e.g. smaller particles having a higher specific surface, was common knowledge, as was the effect of enhanced ageing.

IV. The Appellants (Patentees) lodged an appeal against this decision and filed an amended set of claims together with the appeal statement. Further sets of claims were

submitted in reply to two communications of the Board and at the oral proceedings which were held on 19 April 1994. Claim 1 of the single request maintained at the end of the oral proceedings reads as follows:

"A process for preparing a nickel-based hydrogenation catalyst containing 10 - 90 parts by weight of nickel/nickel compounds and 90 - 10 parts by weight of a water-insoluble carrier material, having an overall active nickel surface of 70 - 200 m^2 per gram of nickel and wherein the catalyst comprises nickel/nickel compound aggregates with an average particle size of 2 to 100 micrometers (µm), which process involves precipitation of nickel hydroxide/carbonate followed by ageing, separation, drying, and reduction of the precipitate in the presence of hydrogen, characterized in that
in a first step a solution of a nickel salt and a solution of an alkaline compound are fed into a precipitation reactor and nickel hydroxide/carbonate is rapidly precipitated in the presence of at least part of the insoluble carrier material under vigorous agitation with a mechanical energy input of 5 - 25 kW per 1000 kg of solution in the precipitation reactor with a mean residence time of 0.01 - 1 minute, during which the normality of the solution in the precipitation reactor containing excess alkali, is kept between 0.1 and 0.3 N and the temperature is from 20 to 55°C, and that in a following step the reaction mixture is transferred to a post-reactor for ageing, in which the mean residence time is 20 to 180 minutes and the temperature is at least 10°C higher than in the precipitation reactor and is kept between 60 and 100°C, after which the solids are separated and further processed."

The dependent Claims 2 to 12 relate to preferred embodiments of this process.

V. In connection with the claimed process the Appellants put forward *inter alia* the following arguments:

No prior art document is referred to in support of the statement in the appealed decision that rapid precipitation will result in particles having a higher specific surface. The mechanism of precipitation in the presence of a carrier is complex and may not be fully understood. The person skilled in the art could not predict the particle size of the precipitate merely from the residence time in the precipitation reactor.

Document (4) does not recite a separate ageing step in the sense of the present invention. No reason for using a second reactor is disclosed in this document. The fact that the temperature is the same in both reactors suggests that the second reactor must be considered as an additional precipitation reactor which was "sometimes" needed to increase the capacity of the first precipitation reactor in order to obtain the longer precipitation times of up to 60 minutes.

The claimed process provides catalysts having unexpected improved activity and selectivity in the hydrogenation of oils and/or fatty acids over the catalysts disclosed in document (4) as shown by table II of the patent in suit. The nickel content of the prior art catalyst (22% in fat suspension) is similar to that of the remaining examples and this catalyst was prepared according to the process of document (4). The Appellant does not know whether the preparation was performed in one or two reactors, however both alternatives were used at that time and the difference in quality of the resulting catalysts was minor. The combination of a first precipitation step at a lower temperature and a shorter residence time than in (4) with a longer ageing time leads to the improved catalyst. With the claimed process

the insoluble nickel compound is largely precipitated homogeneously owing to the vigorous agitation. As a result thereof the nickel aggregates are for the most part free of carrier particles and are accessible from all sides, whereas the process of document (4) provides a supported catalyst in which the nickel particles are present on the particles of the support material. The disclosure of (4) leads a person skilled in the art away from the claimed process by teaching that where two reactors are used the temperature should be identical in both reactors.

VI. The Respondent's arguments insofar as they concern the claimed process may be summarised as follows:

It is questionable whether the vague statement at page 2, lines 27 to 30, of the patent in suit may provide a support to the limitation in the amended Claim 1 that the temperature in the post-reactor is at least 10°C higher than in the precipitation reactor all the more so as this feature is not compatible with the upper values of 95°C and 100°C stated in the granted process claim.

The features recited in Claim 1 are either derivable from document (4) or trivial. In this document the precipitation is performed under usual stirring. There is no difference between stirring and vigorous agitation, and an energy input of 5 to 30 kW/1000 l for the agitation of liquids is usual. According to document (4) the residence time of the product in the precipitation reactor is less than 60 minutes. This range encompasses residence times of 0.01 to 1 minute. The limitation to 0.1 - 1 minute may distinguish the claimed process from that of document (4), however no surprising effect results from this feature (compare example 1 of the patent in suit with the remaining

examples). In the process of (4) the temperature in the second reactor lies within the claimed range and the residence time corresponds to the lower limit of the claimed range. If the same conditions are used, then the same effect must occur. Therefore the Appellant's allegation that the second reactor acts as a precipitation reactor in the process of (4) because of the same temperature of 90°C in both reactors is not correct. It is observed that the features set forth as essential features by the Appellant have no influence upon the quality of the catalyst, in particular upon its activity. It derives from the comparison of examples 1 and 6 of the patent in suit with the remaining examples that the residence time of 0.01 - 1 minute and the temperature of 20 - 55°C in the first reactor are not critical, i.e. they do not lead to a product with superior properties. In example 1 the operating conditions are very close to those of the prior art process, however the resulting catalyst has a better activity for the oil hydrogenation than the catalyst of example 7 which was prepared according to the claimed process. The Appellants have brought no evidence showing that the claimed process leads to catalysts having improved properties over the catalysts of (4). The prior art catalyst referred to in table II has a different nickel content. Furthermore the process itself also has no advantage over the process of document (4) since the lower temperature and the shorter residence time in the first precipitation step are balanced by a longer mean residence time and a higher temperature in the ageing step so that no improvement is obtained as regards the heating and the time consumption. The Respondent dropped its objection regarding sufficiency of disclosure at the oral proceedings.

VII. The Appellants requested that the decision under appeal be set aside and the patent be maintained on the basis of Claims 1 to 12 and a description adapted thereto, as submitted in the oral proceedings. The Respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

2. The amendments in the claims and in the description meet the requirements of Article 123(2) and (3) EPC. The subject-matter of Claim 1 is supported by Claims 1 and 7 as originally filed and the original description, page 2, lines 5 to 28, page 3, lines 18 to 23, page 5, lines 1 to 9, examples 2 to 5 and 7. The feature that the temperature in the post-reactor is at least 10°C higher than in the precipitation reactor is directly and unambiguously derivable from the original description, page 2, lines 23 to 25, which mentions a temperature difference of **10°C**, and from table I, examples 2, 3, 4, 5 and 7 in which the temperature in the ageing post-reactor is **at least** 10°C greater than that in the precipitation reactor, the temperature in the precipitation reactor lying within the range of 20 - 55°C and the mean residence time being of 0.5 or 1 minute. The dependent Claims 2 to 12 find a support in Claims 7, 8, 12, 10, 11, 13, 14, 9, 17, 18 and 19 as originally filed. Moreover, the protection conferred by the amended claims is clearly restricted in comparison to the granted claims.

3. The Respondent dropped its objection based on Article 100(b) at the oral proceedings. As the Board has also come to the conclusion that the patent in suit

meets the requirements of sufficiency of disclosure taking into account the general knowledge about stirrers and stirring technology, further details are not necessary.

4. It was not disputed that the process of Claim 1 is novel. The claimed process differs from that of document (4) at least by a lower temperature and a shorter mean residence time in the precipitation reactor and by the fact that the temperature in the post-reactor (ageing step) is kept at least 10°C higher than in the precipitation reactor. Furthermore the mechanical energy input of 5 to 25 kW per 1000 kg of solution for stirring is higher than the stirring intensity which is normally required to obtain an optimum stirring efficiency (cf. textbook "Grundoperationen chemischer Verfahrenstechnik" Rauck/Müller, 6th edition 1982, page 319). Thus, this feature is also a distinguishing feature over the process of document (4).

5. The Board considers, in agreement with the parties, that document (4) represents the closest prior art with respect to the claimed process.

Document (4) discloses a process for preparing a nickel-based catalyst which is particularly suitable as hydrogenation catalyst for example in the fatty acid hardening. This catalyst contains a nickel/nickel compound and a water-insoluble carrier such as silica, in particular kieselguhr, the nickel content of the catalyst and its specific nickel surface being for example of 45.5 wt% and 105 m^2 per gram of nickel respectively (cf. Claims 1, 6 and 29; page 2, lines 38 to 43, example 1 and table II).

The process of document (4) involves precipitation of nickel hydroxide/carbonate in a mixing vessel (precipitation reactor or first reactor) in the presence of the carrier by mixing a solution of a nickel salt, a solution of an alkali metal carbonate and the carrier such that throughout the precipitation a suspension is formed with a temperature in the range of 75°C to 95°C and a pH of 8 to 10. The mean residence time of the suspension in this precipitation reactor is of less that 60 minutes, preferably 5 to 20 minutes, during which the alkalinity (excess of alkali metal carbonate present during precipitation) is maintained between 0.01N and 0.2N, preferably 0.10N and 0.15N in a continuous process (cf. Claims 1, 15, 17 to 20, page 1, lines 60 to 68 and 80 to 90). This precipitation step may be followed by a post-reaction in a second reactor, which post-reaction is normally referred to as "**ageing**" as pointed out by the Appellant at page 2, lines 14 to 19, of the patent in suit. For example, the precipitation is carried out at 90°C in the first reactor and the suspension is transferred to the second reactor in which it is kept at 90°C for a mean residence time of 8 to 20 minutes. Then the solids are separated and dried and the catalyst is activated with hydrogen (cf. page 2, lines 29 to 34, page 4, lines 7 to 24, page 5, table III).

5.1 Starting from this closest prior art, the technical problem underlying the patent in suit can be seen in providing a process for the preparation of a nickel-based hydrogenation catalyst, which process leads to a catalyst having an improved activity and selectivity in the hydrogenation of oils and/or an improved activity in the hydrogenation of fatty acids.

It is proposed that this problem be solved by a process comprising the combination of features recited in the characterising part of Claim 1. This process differs in

particular from the process of document (4) in that the temperature in the precipitation reactor is from 20 to 55°C, the mean residence time of the mixture in this reactor is of 0.01 to 1 minute, the precipitation is carried out under a vigorous agitation with a mechanical energy input of 5 to 25 kW per 1000 kg of solution, and the temperature in the ageing post-reactor is at least 10°C higher than in the precipitation reactor.

5.2 In table II of the patent in suit, the hydrogenation characteristics of the catalysts prepared by the claimed process are compared to those of a "known catalyst". The Appellant indicated that this catalyst was prepared according to the process of document (4) and that the nickel content of 22% in a fat suspension corresponded to a nickel content similar to that of the catalyst of examples 1 to 7, i.e. about 52%. As regards the process of preparation of this known catalyst the Appellant was not able to give information as to whether the preparation was performed in only one precipitation reactor (first alternative disclosed in document (4)) or in two reactors (second and closest alternative of (4)). However, the Appellant pointed out at the oral proceedings that the difference of quality between a catalyst prepared according to the first alternative of (4) and a catalyst resulting from the second alternative thereof was minor. This was not contested by the Respondent. In these circumstances it can be considered that the hydrogenation characteristics (i.e. the hydrogenation time, the iodine value and the melting point) reported in the tables II and III of the patent in suit in connection with the "known catalyst" do represent the hydrogenation characteristics of a catalyst prepared according to the closest prior art. The comparison of these hydrogenation characteristics with those of the catalysts prepared according to the claimed process shows that the catalysts obtained by the

claimed process (examples 2, 3, 4, 5, 7) exhibit an improved activity and selectivity in the oil hydrogenation over the catalyst of document (4) as well as an improved activity in the fatty acid hydrogenation. Therefore, in the absence of evidence to the contrary it is plausible that the technical problem stated above has been solved by the combination of features defined in Claim 1.

The Respondent's arguments that the operating conditions used in example 7 of the patent in suit lead to a catalyst having a lower activity in the oil hydrogenation test than the catalyst of example 1 which was prepared under conditions very close to that of document (4) (cf. point V above) cannot change these findings for the following reasons. Although the temperature and the mean residence time used in the first step of example 1 are close to those disclosed in (4), this example neither belongs to the state of the art nor can it be regarded as corresponding to the closest prior art in view in particular of the operating conditions of the ageing step which considerably differ from those of document (4). Therefore, it cannot be concluded from the comparison of example 7 with example 1 that no improvement is achieved over the catalyst of the closest prior art. It should be noted that this conclusion would not be in agreement with the results reported in the two last columns of table II for example 7 and for the "comparative example" with the catalyst of document (4). As regards the examples 1 and 6, which now lie outside the scope of protection defined in Claim 1, they show that other combinations of features than the claimed one may result in an improvement of activity and selectivity over the catalyst of document (4), however nor can it be deduced

therefrom that the claimed combination of operating conditions does not solve the technical problem stated above.

5.3 Document (4) itself discloses that the temperature in the precipitation reactor is in the range of 75°C to 95°C. In the alternative, which involves the use of a second reactor, the temperature of the suspension is the same in both reactors, namely 90°C. In the examples 40 and 42 (cf. pages 10 and 11) the preparation of the catalysts was performed in one reactor with a mean residence time of 8 minutes at different precipitation temperatures, i.e. 90°C, 80°C and 70°C. From the results reported on table VIII it can be derived that at comparable Ni/SiO_2 ratios the activity of the catalyst prepared at 70°C is lower than that of the catalysts prepared at precipitation temperatures of 90°C and 80°C. Even if it is assumed that the activity of the catalysts was tested in the hydrogenation of benzene, the skilled person confronted with the problem stated above would not be encouraged in view of this teaching to decrease the temperature in the precipitating reactor to the claimed range of 20 - 55°C in order to improve the activity of the catalyst in the oil and/or fatty acid hydrogenation. Document (4) does not contain any information which could give the skilled person an incentive to reduce the temperature and the mean residence time in the precipitating reactor to values within the ranges of 20 - 55°C and 0.01 - 1 minute, to perform the first step under a more vigorous agitation and to carry out the ageing step at a temperature at least 10°C higher than in the precipitation reactor in order to obtain a catalyst with an improved activity and

selectivity in the hydrogenation of oils and/or an improved activity in the hydrogenation of fatty acids. In the Board's view it could not be expected that these modifications would lead to these improved properties.

5.4 Document (1) discloses a process for preparing silica supported nickel catalysts having a nickel content, a nickel surface and an average particle size which fall within the ranges stated in the preamble of Claim 1 (cf. page 1389 point 2.2 and page 1391, table 1). According to (1) nickel nitrate and sodium carbonate were used for the precipitation of nickel compounds in the presence of suspended silica, the precipitate was filtered, washed and dried at 120°C. Reduction was performed at 480°C in a stream of hydrogen (cf. page 1389, point 2.2). This document, which deals with the activity and the selectivity of these supported nickel catalysts in fatty oil hydrogenation, links the activity and selectivity results with the pore structure of the catalyst, the ratio of wide pores (d>25A°) to medium pores (d=20-25A°) being correlated with the selectivity and the nickel surface in the wide pores with the activity. However, as this document does not give any information about the operating conditions of the precipitating step and does not even mention an ageing step its teaching could not point towards the claimed combination of operating conditions even in combination with the disclosure of document (4).

5.5 During the appeal procedure the Appellant has not relied any more upon the remaining documents cited in the opposition procedure. After examination of these documents the Board has come to the conclusion that they also do not suggest the claimed combination of features in order to improve the activity and selectivity of the catalyst known from document (4).

5.6 It follows from the above that it was not obvious to the skilled person to perform the precipitation step and the ageing step under the operating conditions recited in Claim 1 of the request submitted at the oral proceedings.

6. The dependent Claims 2 to 12 which relate to preferred embodiments of Claim 1 derive their patentability from that of Claim 1.

7. The Board has noted that the adapted description submitted in the oral proceedings contains amended pages 2 to 7 and that figures 1 and 2 of the granted patent were not amended but are still part of the patent.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to maintain the patent on the basis of Claims 1 to 12 and a description adapted thereto, as submitted in the oral proceedings.

The Registrar: The Chairman:

P. Martorana P.A.M. Lançon