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**D E C I S I O N**  
**of 4 April 1995**

**Case Number:** T 0550/91 - 3.3.2  
**Application Number:** 85200971.1  
**Publication Number:** 0168091  
**IPC:** B01J 23/74  
**Language of the proceedings:** EN

**Title of invention:**  
Nickel/alumina catalyst, its preparation and use

**Patentee:**  
UNILEVER N.V., et al

**Opponent:**  
Hoechst Aktiengesellschaft  
Engelhard De Meern B.V.  
Süd-Chemie AG

**Headword:**  
Ni/Alumina catalyst/UNILEVER

**Relevant legal provisions:**  
EPC Art. 83, 84, 54

**Keyword:**  
"Lack of clarity not arising from amendment"  
"Article 84 no legal basis for revocation"  
"Parameter without definition of its method of measurement in the description"

**Decisions cited:**  
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**Catchword:**  
-



Case Number: T 0550/91 - 3.3.2

**D E C I S I O N**  
**of the Technical Board of Appeal 3.3.2**  
**of 4 April 1995**

**Appellant:**  
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**Decision under appeal:**

**Decision of the Opposition Division of the  
European Patent Office dated 21 February 1991  
posted on 4 July 1991 revoking European patent  
No. 0 168 091 pursuant to Article 102(1) EPC.**

**Composition of the Board:**

**Chairman:** P. A. M. Lançon  
**Members:** M. M. Eberhard  
S. C. Perryman

### Summary of Facts and Submissions

I. European Patent No. 0 168 091 based on application No. 85 200 971.1 was granted on the basis of 14 claims.

II. The Respondents (Opponents 1, 2 and 3) filed Notices of Opposition requesting the revocation of the patent on the grounds of lack of novelty, lack of inventive step and insufficiency of disclosure. Of the documents cited by the parties during the opposition procedure only the following ones were relied upon in the present appeal:

- E1: M. B. Borisova and al., Kinetika i Kataliz, Volume 15, No. 2, pages 488 to 496 (1972), English translation pages 425 to 439
- E3: J. W. E. Coenen and al., Proc. 3rd Int. Congr. Catalysis, Volume II, Amsterdam 1965, pages 1387 to 1399
- E4: E. G. M. Kuijpers and al., J. Catalysis, 112 (1988), pages 107 to 115
- E5: N. E. Buyanova and al., Kinetika i Kataliz, Volume 8, No. 4, pages 868 to 877 (1967), English translation pages 737 to 746
- C3: J. W. E. Coenen and B. G. Linsen, Physical and Chemical Aspects of Adsorbents and Catalysts, Acad. Press (1970) pages 472 to 527
- D1: Declaration of Prof. J. W. E. Coenen
- D2: Declaration of Dr J. C. Oudejans
- D3: Declaration of Prof. J. W. Geus

III. In the course of the opposition procedure, the Appellants (Patentees) submitted an amended set of claims. Claim 1 thereof reads as follows:

"1. A nickel/alumina catalyst satisfying the following combination of features:

- 1) a nickel/aluminium atomic ratio between 4 and 10;
- 2) an average pore size between 4 and 20 nanometres;
- 3) an active nickel surface between 90 and 150m<sup>2</sup>/g nickel;
- 4) nickel crystallites with an average diameter between 1 and 5 nanometres."

IV. The Opposition Division revoked the patent on the ground that Claim 1 as amended did not meet the requirement of clarity of Article 84 EPC. In its decision it held that the active nickel surface area defined in Claim 1 was already known from E1 since, on the one hand, the O<sub>2</sub>-chemisorption described in E5 and used in E1 for measuring the nickel surface area was a reliable method and, on the other hand, the experimental data submitted by the Patentees in D2 did not prove that the values of nickel surface area stated in E1 were incorrect. The Opposition Division took the view that the catalyst of Claim 1 comprised one feature which was formally new over the catalyst of E1, namely the average pore size of 4 to 20 nm. However this feature could neither be used for limiting the scope of Claim 1 nor for distinguishing the claimed catalyst from the known catalyst because it was unclear. In the absence of any information in the patent about the shape of the pores and about the formulae applied to calculate the average pore size, the range of 4 to 20 nm was vague and might extend over both of its limits up to the factor 2 and 0,5 respectively.

V. The Appellants lodged an appeal against this decision and filed four sets of claims as main request, first, second and third auxiliary requests. Claim 1 of the main request is identical to Claim 1 upon which the decision was based. They also submitted two additional declarations in the course of the appeal procedure, namely the declaration of Dr C. M. Lok (D4) and the declaration of Dr W. T. Koetsier (D5), as well as further documents, in particular:

K4 E. P. Barret, L. G. Joyner and P. P. Halinda, J.  
Amer. Chem. Soc., 73 (1951), pages 373 to 380

K2 Reporting Physisorption Data for Gas/Solid Systems  
IUPAC recommendations 1984; Pure & Appl. Chem.,  
Vol. 57, No. 4, pages 611 to 613, April 1985.

Respondent II provided the complete IUPAC recommendations mentioned above, i.e. pages 603 to 619 (hereinafter also designated K2).

Oral proceedings were held on 4 April 1995.

VI. The Appellants argued as regards the average pore size that there was no apparent reason to suppose that the catalysts of the invention, which were obtainable by a precipitation process including the in-situ formation of the support, would have pores with a form other than the slit shape obtained with the co-precipitation process of C3. In case of any doubt as to the pore model to be used in determining the average pore size, it would have been easy to repeat the examples of the patent in suit and to determine the total pore volume of the catalysts thus obtained and their surface area. As the BET surface area and the average pore size were disclosed in the patent in suit for each example, it would have been clear at once which pore model would have to be used to establish the scope of protection of Claim 1. It was further

pointed out that in the sixties the size of the pores was usually characterised assuming cylindrical pores (cf. K4), and that the slit-shaped pore model was developed later on. At the filing date the skilled person who wanted to determine the size of the pores had to make an assumption as to the appropriate model to be used, i.e. the cylindrical pore model or the slit-shaped pore model. For the present catalysts electron microscopy provided evidence that the slit-shaped pore model was more appropriate. In the case of a cylindrical pore model the formula  $2.V_p/S$  would have given the mean radius  $r$  of the assumed cylindrical pores whereas with the slit-shaped pore model the value of  $2.V_p/S$  would have represented the mean width  $d$  of the pores (wall to wall distance).

As to the novelty issue, it was contended that E1 neither disclosed the average pore size of the catalyst nor its pore volume. The range of 0.5 to 1 cm<sup>3</sup>/g for the pore volume of supported catalysts assumed by Respondent II could not be regarded as typical since pore volumes of 0.2 to 2 cm<sup>3</sup>/g had already been obtained by changing the conditions of preparation. Furthermore a broad pore volume range would normally have been associated with a certain range of BET surface area and not with a single value. Therefore, the Respondent's calculation did not prove that the claimed catalyst lacked novelty with respect to E1.

In connection with feature (3) of Claim 1 the Appellants argued that the experiments in D2 and D4 showed that the active nickel surface area of the E1 catalyst measured by H<sub>2</sub>-chemisorption was smaller than the surface area measured by O<sub>2</sub>-chemisorption. The experiments in D4, which were performed under the operating conditions reported in E1, demonstrated the unreliability of the data quoted in E1. Furthermore, E4 disclosed that the

chemisorption of oxygen always exceeded the monolayer coverage and no evidence has been submitted that the authors of E1 took the dangers of excess coverage into account in their measurements. As pointed out in D5 the mean number of  $1.3 \times 10^{19}$  molecules of oxygen /m<sup>2</sup> stated in E5 corresponded to a surface of the nickel atom of 7.692 Å<sup>2</sup> which value was substantially higher than that used in D1 for calculating the nickel surface area. Calculations based on the same mean surface area of the Ni atom for both the claimed catalyst and the catalyst of E1 would have led to a nickel surface area of the E1 catalyst lying outside the claimed range.

The Appellants further contended that Article 84 could not be used as a legal basis for revoking the patent since the objection based upon Article 84 did not arise out of amendments to the feature deemed to be vague and indefinite. Reference was made to the decision T 301/87 in this respect.

VII. The Respondents' arguments can be summarised as follows:

It was argued that the claims were silent about the method of determination of the nickel surface area. It derived from D2 and D3 that the method of determination of the nickel surface area by H<sub>2</sub>-chemisorption was not generally adopted before the priority date and that it could be carried out under different conditions of temperature and pressure. However the patent in suit did not give any data as to how the H<sub>2</sub>-chemisorption was performed; thus the claimed nickel surface areas were arbitrary. As the Ni crystallite size was calculated from the Ni surface area, this parameter was also undefined all the more so that an assumption had to be made for the crystallite shape (cf. D1) and the patent in suit gave no information in this respect.



The Respondents further contended that the values of nickel surface area disclosed in E1 were correct. It was derivable from E4 that the O<sub>2</sub>-chemisorption constituted a reliable method for determining the nickel surface area like the H<sub>2</sub>-chemisorption provided that some specific rules in both cases were observed. The statement in E1 that the crystallite size obtained by X-Ray determination was in agreement with the O<sub>2</sub>-adsorption data also proved that the O<sub>2</sub>-chemisorption led to correct values of the nickel surface. The experiments in D2 could not question the reliability of the result reported in E4 and E5 since the catalysts were not prepared according to the method of preparation described in E1. The result obtained in D4 showed the unreliability of these additional experiments since the Ni surface area for sample 1 differed by about 20% from that of sample 2. Contrary to the allegation in D5, the method of E5 did not involve the use of the value 7.692 Å<sup>2</sup> for the surface of the nickel atom since a calibration was first effected by physical adsorption of argon. Therefore, the nickel surface areas given in E1 did not need not to be corrected by the factor stated in D5.

As regards the average pore size it was argued that E3 and C3 did not disclose that the pore structure of Ni-SiO<sub>2</sub> catalysts was the same as that of Ni-Al<sub>2</sub>O<sub>3</sub> catalysts because of the same method of preparation, namely co-precipitation of the support and metal compound. According to C3 the shape of the pores depended upon the conditions of the co-precipitation. Furthermore, Claim 1 was not restricted to catalysts prepared by a specific process but encompassed all possible preparation methods. The Respondents also contested the two methods of determination of the average pore size presented in items 5 and 6 of D5. In this respect they stressed that the use of only one

formula for calculating the average pore size whatever the pore shape represented a gross and unacceptable simplification. This was also not in agreement with the IUPAC recommendations K2 which distinguished two different pore shape models (cylindrical or slit-shaped pores) and accordingly gave two different formulae for the calculation of the mean pore size or for the determination of the pore size distribution. In the latter case two different pore size distribution curves were obtained when considering the multilayer thickness  $t$  as indicated in K2 (cf. graph submitted at the oral proceedings).

As regards novelty of the feature (2) Respondent II argued that the average pore size was indeed not mentioned in E1 but could be calculated from the  $S_{\text{BET}}$  values given in E1 by using the known formula  $d_p = 4000 \times V_p / S_{\text{BET}}$  for the cylindrical pore model. Although the total pore volume was not given in E1, the typical range for the total pore volume of supported catalysts was generally 0.5 to 1.0 cm<sup>3</sup>/g. These lower and upper limits led to an average pore size of 8 nm and 16 nm respectively for a BET surface area of 241 m<sup>2</sup>/g, i.e. an average pore size lying within the claimed range. With the BET surface area of 135 m<sup>2</sup>/g average pore sizes of 14.8 nm and 29.6 nm were calculated. The broader range of 0.2 to 2 cm<sup>3</sup>/g for the pore volume gave a calculated average pore size of 4 nm with the lower value of the pore volume and a BET surface area of 200 m<sup>2</sup>/g, i.e. also a value which was novelty destroying for the claimed range.

In connection with the use of Article 84 as legal basis for the decision, it was argued either that the reasons given by the Opposition Division amounted to an

objection of insufficiency of the description in the sense of Article 100(b) or that the result of the Opposition Division's findings was in fact a lack of novelty.

- VIII. The Appellants requested that the decision under appeal be set aside and that a patent be maintained on the basis of the main request or one of the first or second auxiliary requests all filed with the Statement of Grounds of Appeal with the corrections thereto requested in the letter dated 1 June 1992, or of the third auxiliary request filed with the letter dated 1 June 1992. The Respondents requested that the appeal be dismissed.

#### **Reasons for the Decision**

1. The appeal is admissible.
2. *Amendments*

There are no objections under Article 123(2) and (3) EPC to the amended claims of the main request. The combinations of features recited in the claims are disclosed in the originally filed documents (cf. the original claims and the description page 2, lines 22 and 23 and page 5, lines 10 to 14). The amendments in Claims 1 and 3 also do not broaden the scope of protection of the granted claims. In connection with the terms "average pore size" in the dependent Claim 3, it should be noted that the replacement of the word "radius" of the granted Claim 4 by the word "size" does not contravene Article 123(3) in view of the fact that this amendment clearly represents a correction of an obvious mistake as correctly indicated in the decision of the Opposition Division and not contested by the Respondents.

3. *Legal basis for the decision*

3.1 According to the jurisprudence of the Boards an objection under Article 84 cannot in principle be a proper basis for the revocation of a patent if this objection does not arise out of the amendments made to the claims (cf. decisions T 301/87, OJ EPO, 1990, 335; T 472/88 EPOR, 1991, 487). However, objections to lack of clarity under Article 84 are relevant to the opposition proceedings insofar as they can influence the decision on issues under Article 100 EPC, for example novelty or inventive step. In such cases these objections need not be investigated further than is necessary to enable assessment of the issues already at hand (cf. decisions T 127/85, OJ OEB, 1989, 271, and T 525/90 unpublished).

3.2 In the present case Claim 1 of the main request differs from the granted Claim 1 only in that the lower value of 2 for the Ni/Al atomic ratio has been replaced by 4. Feature (2) (average pore size), which is considered in the decision under appeal as vague and unclear, was not amended during the opposition procedure and the question whether or not this feature fulfils the requirement of clarity of Article 84 does not arise out of the amendments made to the lower value of the Ni/Al ratio. Therefore, Article 84 could not in principle form a proper basis for the revocation of the patent in suit. However, it appears from the reasons given in the decision under appeal that the clarity objection to the average pore size range was investigated in connection with the novelty issue. The catalyst of Claim 1 was firstly compared with the catalysts of the prior art document E1 and as the average pore size range was found to constitute the sole possible distinguishing feature over the catalyst of E1, or in other words the sole possible new feature, the clarity and the meaning of

said feature were examined. In the last paragraph of page 13 of the decision it was stated that feature (2) was meaningless and could not be used for distinguishing the claimed catalysts from the prior art catalysts. From this paragraph read in the context of decision it implicitly follows that the catalyst of Claim 1 was considered as lacking novelty over the catalyst of document E1. In these circumstances the Board concludes that the novelty issue has implicitly been decided upon and thus that the decision of revocation is not only based upon Article 84 but also implicitly upon Article 54 EPC.

4. *Sufficiency of disclosure*

4.1 The objection that the patent in suit does not give information as to how the parameters stated in Claim 1 were determined raises the question whether the person skilled in the art would have been able, without undue burden, to carry out the invention as defined in Claim 1 on the basis of the instruction given in the description and of the common general knowledge.

4.2 As regards the **nickel surface area**, it derives from the description of the patent in suit, page 4 lines 1 to 2, that said surface area was determined by hydrogen chemisorption in the examples. The patent in suit indeed contains no instruction as to the operating conditions used for the H<sub>2</sub>-chemisorption, however the determination of the nickel surface area of supported catalysts by H<sub>2</sub>-chemisorption was a well-known method before and at the filing date of the patent application. This method was already described in standard textbooks published in 1970 (cf. C3 pages 494 to 495) and the content of such textbooks has to be considered as forming part of the common general knowledge of the skilled person. C3 discloses at page 494 the operating conditions, i.e. the

adsorption temperature, the hydrogen pressure and the equilibration time used at the Unilever Research Laboratory for the determination of the Ni surface area of supported catalysts by H<sub>2</sub>-chemisorption. Furthermore, according to page 494, 3rd paragraph, considerable differences occur between the details of the methods applied by different investigators with respect to adsorption temperature, hydrogen pressure and equilibration time, however the ultimate results generally are not very different. The average area taken up by one nickel atom in the surface is also indicated at page 495 as well as the resulting formula applied for calculating the active nickel surface area. Therefore the skilled person would have been able to repeat the examples of the patent in suit and to measure the active nickel surface area of the resulting catalysts by hydrogen chemisorption using the known operating conditions and formulae disclosed in textbooks available at the filing date of the patent, such as C3.

- 4.3 As to the **average diameter of the nickel crystallites**, it also derives from page 4, lines 1 to 2, of the patent in suit that it was calculated from the measured nickel surface area. Although the corresponding formula is not given in the patent in suit, it is disclosed at page 498 of the standard textbook C3 and must therefore be considered as forming part of the common general knowledge of the skilled person in this technical field. Furthermore it was well-known before the filing date that the crystallite size could also be determined from X-Ray line broadening (cf. C3, pages 490, 491 and 498). Therefore the skilled person would have been able to determine the nickel crystallite size even without indication of the formula in the patent in suit.

4.4 As regards **the average pore size**, there is no indication in the patent in suit of how this parameter was determined. However the physical adsorption of gases, in particular the nitrogen adsorption isotherms, were a well-known and commonly applied method for studying the pore structure at the filing date. At the oral proceedings it was not contested by the parties that the average pore size may be calculated from the total pore volume ( $V_p$ ) and the surface area ( $S$ ) by the formula  $d_p = 2V_p/S$  in the case of a slit-shaped pore model,  $d_p$  representing the average pore width, whereas, in the case of a cylindrical pore model, the average pore size is given by the formula  $d_p = 4V_p/S$  where  $d_p$  is the average pore diameter of the assumed cylindrical pores ( $\times 10^4$  in  $\text{Å}$ ). It was not disputed that these formulae formed part of the common general knowledge of the person skilled in the art of catalysis at the filing date of the patent application. The formula  $d_p = 2V_p/S_{\text{BET}}$  for the slit-shaped pore model is mentioned in E3 (cf. page 1390) which was published about twenty years before the filing date and which can also be considered as illustrating the common general knowledge. However, as argued by the Respondents, the patent in suit is silent about the pore shape model assumed for the determination of the average pore size. Therefore, the question arises whether the skilled person would have been in a position to determine the missing information without undue burden. The patent in suit contains six examples describing the detailed preparation of six different catalysts as defined in Claim 1 and indicating the BET surface area as well as the average pore size of each of the resulting catalysts. Thus, as argued by the Appellants, the skilled person would have been able to prepare an exemplified catalyst, to measure its total pore volume and its surface area by the known usual methods and to calculate its average pore size using the well-known formulae given above for the cylindrical and

slit-shaped pore models. It was not contested by the Respondents that a comparison of the resulting values with the value of the average pore size given in the considered example would have permitted one to deduce which of the two pore models had been used for the calculation of the average pore size. Electron microscopic observation, which is mentioned at pages 2 and 4 of the patent in suit in connection with the pore structure, would also have provided information as to which of these two pore models was the most appropriate. Therefore, in the absence of evidence to the contrary, it is considered that the skilled person would have been able to establish, without any undue burden, on the basis of the information in the patent in suit and of the common general knowledge which of the two pore shape models usually considered for the determination of the average pore size before the filing date had been assumed in the examples of the patent in suit.

4.5 For the preceding reasons, the requirement of sufficiency of disclosure is fulfilled as regards the catalysts defined by the four parameters recited in Claim 1 of the main request.

## 5. *Novelty*

5.1 The catalysts defined in Claim 1 have an average pore size between 4 and 20 nm. As there is no indication in the patent in suit of the method of determination of this parameter or of the pore shape model assumed for the determination of said range, this claim is construed as relating to catalysts having an average pore size between 4 to 20 nm whatever the assumed pore model and the method of determination, in combination with the other features (1), (3) and (4).



5.2 Document E1 discloses two nickel/alumina catalysts having a Ni/Al atomic ratio, an active nickel surface area and an average diameter of the nickel crystallites which all fall within the ranges indicated in Claim 1: see Table 1, the catalysts having a Ni/Al atomic ratio of 7.85, an active nickel surface area of 84 m<sup>2</sup>/g of catalyst and a mean size of nickel crystallites of either 3.7 nm or 4.4 nm, which were reduced in hydrogen at either 360°C or 400°C respectively. The average pore size of these catalysts is not indicated in E1. As the operating conditions used to prepare these catalysts are neither identical nor similar to those indicated in the patent in suit, it cannot be derived from E1 that the average pore size would necessarily lie within the claimed range.

5.3 The calculations of Respondent II based on the formula  $d_p = 4000V_p/S_{BET}$  for cylindrical pores and on an assumed pore volume of 0.5 to 1 cm<sup>3</sup>/g or 0.2 to 2 cm<sup>3</sup>/g for supported catalysts (cf. point VII above) do not convince the Board that the two catalysts of E1 considered above exhibit an average pore size lying within the claimed range. The values of the total surface area reported in Table 1 of E1 for the catalysts having a Ni/Al atomic ratio of 7.85, namely 241 m<sup>2</sup>/g and 135 m<sup>2</sup>/g, are those of the catalysts calcined in nitrogen at 330°C and 500°C and not of the reduced catalysts. Therefore, these values cannot be used for calculating the average pore size of the reduced catalyst. Furthermore, the BET surface area of 200 m<sup>2</sup>/g associated with the assumed pore volume of 0.2 cm<sup>3</sup>/g or 2 cm<sup>3</sup>/g is not disclosed in Table 1 or elsewhere in E1 for a reduced catalyst having a Ni/Al atomic ratio of 7.85. In addition, the assumed range for the total pore volume is usually associated with a range of BET surface area and not with a single value as pointed out by the Appellants. In these circumstances and in the

absence of evidence to the contrary the Board comes to the conclusion that the catalysts defined in Claim 1 of the main request differ from those of E1 by their average pore size and thus are novel with respect to this prior art.

5.4 In view of these findings there is no need to investigate whether or not the active nickel surface area indicated in Table 1 of E1 for the catalysts with a Ni/Al ratio of 7.85 is correct.

5.5 The catalysts of Claim 1 are also new with respect to the other documents cited in the opposition procedure. As this was not disputed by the Respondents at the appeal stage, it is not necessary to consider this matter in details.

6. The question whether or not the catalyst as defined in Claim 1 of the main request involves an inventive step has not been examined by the Opposition Division. In particular it is still not clear which technical problem has been solved by the catalyst of Claim 1 with respect to the closest prior art. In these circumstances, the Board finds it appropriate, in accordance with Article 111(1) EPC, to remit the case to the Opposition Division for further prosecution, in order to avoid loss of one instance.

**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 for further prosecution.

The Registrar:

The Chairman:

P. Martorana

P. A. M. Lançon