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**Datasheet for the decision  
of 6 November 2012**

**Case Number:** T 0046/11 - 3.3.07

**Application Number:** 03075070.7

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**Language of the proceedings:** EN

**Title of invention:**  
Shaped hydrogenation catalysts and processes for their  
preparation and use

**Applicants:**  
BASF Catalysts LLC

**Headword:**  
-

**Relevant legal provisions:**  
EPC Art. 56

**Keyword:**  
"Inventive step - obvious alternative (all requests)"

**Decisions cited:**  
-

**Catchword:**  
-



Case Number: T 0046/11 - 3.3.07

**DECISION**  
of the Technical Board of Appeal 3.3.07  
of 6 November 2012

**Appellants:** BASF Catalysts LLC  
(Applicants) 100 Campus Drive  
Florham Park, NJ 07932 (US)

**Representative:** Fisher, Adrian John  
Carpmaels & Ransford  
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**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office posted 6 August 2010  
refusing European patent application  
No. 03075070.7 pursuant to Article 97(2) EPC.

**Composition of the Board:**

**Chairman:** J. Riolo  
**Members:** F. Rousseau  
D. T. Keeling

## Summary of Facts and Submissions

- I. The appeal lies from the decision of the Examining Division posted on 6 August 2010 refusing European patent application No. 03 075 070.7, which is a divisional application from application No. 96 911 778.7.
- II. The contested decision was based on a Main Request and an Auxiliary Request, submitted on 18 June 2010 and during the oral proceedings on 22 July 2010, respectively. Claim 1 of the Main Request read as follows:
- "1. A shaped catalyst composition for hydrogenating aldehydes, ketones, carboxylic acids, carboxylic acid esters and nitroaromatic compounds, said composition comprising a uniform mixture of (i) from 30% by weight up to 75% by weight of nickel, said nickel being in the form of nickel oxide; (ii) from 10% by weight up to 40% by weight of calcium silicate; and (iii) from 1% by weight up to 30% by weight of at least one clay material, wherein said composition is free of added alumina other than that which may be present resulting from the clay material, and free of chromium and barium."
- III. According to the contested decision, novelty was acknowledged, in particular over the prior art documents D1 (WO 95/01949) and D3 (EP-A-0 131 460) disclosing the commercial Ni-based catalyst designated Ni 3266 E, which catalyst comprised in view of the information provided in D1 and D2 (Prabhu et al, Applied Catalysis A: General, Elsevier Science,

Amsterdam, Vol. 183, no. 2, 1999, pages 241-252) 50 wt.-% of nickel and a calcium silicate/bentonite clay/alumina support. As regards inventive step, the closest prior art was D1, in particular its example 5 relating to hydrogenation of an aldehyde using that catalyst. The claimed catalysts differed from catalyst Ni 3266E in that the amount of calcium silicate and the amount of clay had been specified, and alumina other than that, which may be present resulting from the clay material, was not employed, the commercial catalysts used in the examples of the application being far removed from the prior art illustrated in D1. The Applicants had failed to demonstrate the existence of any technical effect brought about by those distinguishing features. Furthermore, the alumina contained in Ni 3266E served as binder and the exclusion of "added alumina" according to Claim 1 did not mean that the binding function could not be fulfilled by replacing alumina by another inorganic binder, D1 already suggesting variation of the components of the support material. An inventive step was therefore denied for the Main Request. An inventive step was also denied for the subject-matter according the Auxiliary Request, which was directed to a process for preparing the shaped catalyst including *in situ* formation of the calcium silicate. The decision under appeal also contained *obiter dicta* concerning formal issues in relation to the requirements of Article 84 EPC and Article 123(2) EPC.

IV. With their statement setting out the grounds of appeal dated 15 December 2010, the Applicants (hereinafter the Appellants) submitted three sets of amended claims on the basis of which grant of a patent was requested. In

response to the Board's communication dated 20 September 2012 addressing issues to be discussed at the oral proceedings in respect of the requirements of Articles 76(1), 123(2), 84 and 56 EPC, the Appellants with their letter dated 5 October 2012 rearranged the order of the requests then on file and submitted three further sets of amended claims, a complete set of the Main and First to Fifth Auxiliary Requests being enclosed in their letter. The respective independent Claims 1 of those requests read as follows:

*Main, First and Fifth Auxiliary Requests*

"1. A shaped catalyst composition for hydrogenating aldehydes, ketones, carboxylic acids, carboxylic acid esters and nitroaromatic compounds, said composition comprising a uniform mixture of (i) nickel; (ii) calcium silicate; and (iii) at least one clay material, wherein said composition is free of added alumina other than that which may be present resulting from the clay material, and free of chromium and barium."

*Second Auxiliary Request (amendments compared to Claim 1 of the present Main Request are underlined).*

"1. A shaped catalyst composition for hydrogenating aldehydes, ketones, carboxylic acids, carboxylic acid esters and nitroaromatic compounds, said composition comprising a uniform mixture of (i) from 30% by weight up to 75% by weight of nickel, said nickel being in the form of nickel oxide; (ii) from 10% by weight up to 40% by weight of calcium silicate; and (iii) from 1% by weight up to 30% by

weight of at least one clay material, wherein said composition is free of added alumina other than that which may be present resulting from the clay material, and free of chromium and barium."

Claim 1 of the present Second Auxiliary Request is identical to Claim 1 of the Main Request on the basis of which the contested decision was based.

*Third Auxiliary Request (amendments compared to Claim 1 of the present Second Auxiliary Request are indicated in bold and strikethrough)*

- "1. A shaped catalyst composition for hydrogenating aldehydes, ketones, carboxylic acids, carboxylic acid esters and nitroaromatic compounds, said composition comprising a uniform mixture of (i) from 30% by weight up to 75% by weight of **only one metal selected from the group consisting of copper, nickel, manganese, zinc, cobalt and iron, wherein said only one metal is** nickel ~~being~~ in the form of nickel oxide; (ii) from 10% by weight up to 40% by weight of calcium silicate; and (iii) from 1% by weight up to 30% by weight of at least one clay material, wherein said composition is free of added alumina other than that which may be present resulting from the clay material, and free of chromium and barium."

*Fourth Auxiliary Request (deletions compared to Claim 1 of the Main Request are indicated in strikethrough)*

- "1. A shaped catalyst composition for hydrogenating aldehydes, ketones, carboxylic acids, carboxylic

acid esters and nitroaromatic compounds, said composition comprising ~~a uniform mixture~~ of (i) nickel; (ii) calcium silicate; and (iii) at least one clay material, wherein said composition is free of added alumina other than that which may be present resulting from the clay material, and free of chromium and barium."

V. Oral proceedings took place on 6 November 2012 in the course of which the present decision was announced.

VI. The arguments of the Appellants, as far as they are relevant for the present decision, can be summarised as follows:

(a) None of the cited prior art documents disclosed a catalyst composition as specified in Claim 1. D1 and D3 each described a catalyst Ni-3266E, which contained a silica/alumina-supported metal catalyst. Neither D4 nor D5 disclosed a catalyst composition comprising calcium silicate, and those documents also failed to teach a catalyst composition which was free of chromium, barium and added alumina. Novelty of the claimed subject-matter was therefore given.

(b) In respect of inventive step, the commercial catalyst designated Ni 3266E and its use for hydrogenation of an aldehyde in example 5 of D1 constituted the closest prior art. Catalyst Ni 3266E as demonstrated by D2 comprised nickel on a calcium silicate/bentonite clay/alumina support.

(c) It could not reasonably be doubted that the catalyst compositions of the present invention were in fact effective for hydrogenations reactions. Although the experiments shown in the present application related to copper catalysts, these results were not exclusive to copper catalysts. Rather, they were common to catalysts prepared without an alumina support, so they could be extrapolated to other catalysts, such as nickel-containing catalysts, prepared without an alumina support. The examples of US-A-6 342 464 (D6) demonstrated the efficacy of the catalyst compositions according to present Claim 1. Nickel catalysts produced without alumina support, for examples, using the preparation methods employed in Examples 2 and 3 of D6 were superior catalysts to nickel catalysts produced in a traditional manner (e.g. Ni-3266E). With the results appearing in Figures 1 and 3 of D6 nickel catalysts produced without alumina support had been shown to have a higher average catalytic activity for the hydrogenation of 3-hydroxypropanal, than a standard, off-the-shell nickel catalyst produced with alumina support. Additionally, the hydrogenation reaction was more stable with nickel catalysts produced without alumina support compared to a standard, off-the-shelf nickel catalyst produced with alumina support. It was held that the beneficial results shown for the copper catalysts obtained with Examples 2 and 3 of D6 could be extrapolated to the presently claimed nickel catalysts.

(d) Even if the claimed catalyst composition provided no unexpected advantages over catalyst Ni 3266E,



the catalyst composition of the present invention would involve an inventive step as being a non-obvious alternative to that prior art catalyst. It was well established in the Case Law that the person of ordinary skill in the art was unimaginative and did not really venture into unexplored territory. For the skilled person, the presumption was that the calcium silicate, the bentonite clay and the alumina support in Catalyst Ni 3266E were there for a purpose. A vendor of catalyst did not include components which are not required. *Prima facie*, therefore, it would not have been obvious to dispense with the alumina support, without some indication in the prior art that the alumina was unnecessary. There was no such disclosure in the prior art.

- (e) The catalysts claimed in the Main Request were therefore inventive.
  
- (f) As regards the catalysts defined in the Auxiliary Requests, an inventive step was also to be acknowledged for at least the reasons given in relation to the Main Request. It was in this context acknowledged that Claims 1 of Auxiliary Requests 3 and 4 despite of having a different wording had the same meaning.

VII. The Appellants requested that the decision under appeal be set aside and that a patent be granted on the basis of the claims of the Main Request or of one of the First to Fifth Auxiliary Requests, which were all attached to the Appellants' letter dated 5 October 2012.

VIII. The decision was announced at the oral proceedings.

### **Reasons for the Decision**

1. The appeal is admissible.

#### *Main Request*

2. Although the conformity of the claims of the Main Request in respect of the requirements set out in Articles 76(1), 132(2) and 84 EPC would have required a decision, in particular in view of the presence in Claim 1 of the terms "uniform mixture" and "free of added alumina other than that which may be present resulting from the clay material", it is more appropriate in the present appeal proceedings to deal with the issue on the basis of which the contested decision was taken, namely lack of inventive step.

#### *Inventive step*

#### *Closest prior art*

3. The present invention relates to catalysts which are useful as hydrogenation catalysts (page 1, first paragraph of the application). D1 discloses in Claim 1 hydrogenation catalyst compositions suitable for converting a 3-alkoxypropionaldehyde dialkyl acetal to the corresponding 1,3-dialkoxypropane, said catalyst composition comprising at least one catalytic metal selected from the group consisting of Pd, Ni, Co, Pt, Rh and Ru and a support material, said support material being one or more materials selected from the group

consisting of silica, alumina, silica-alumina, aluminosilicates and carbon. D1 also provides on page 5 examples of commercially available catalysts that are useful for the above reaction, in particular the commercial catalyst designated Ni 3266E which according to page 5, lines 10-13 of D1 contains a calcium promoted silica/alumina supported metal catalyst having a nickel content of about 50 wt.-% and a surface area of about 150m<sup>2</sup>/g. According to D2 this catalyst comprises nickel on a calcium silicate/bentonite clay/alumina support. It is not disputed that the shaped catalyst compositions defined in Claim 1 of the present Main Request differ from commercial catalyst Ni 3266E only in that they are free of added alumina other than that which may be present resulting from the clay material.

4. Thus, in agreement with the Examining Division and the Appellants, the Board is satisfied that the disclosure of D1, which in particular relates to the use of the shaped catalyst Ni-3266E for the catalytic hydrogenation of 3-alkoxypropionaldehyde dialkyl acetal represents a suitable starting point for assessing inventive step.

*Problem successfully solved and solution*

5. Having regard to this prior art document, the Appellants submitted that the problem solved by the presently claimed subject-matter over D1 should be formulated taking into account the technical effects addressed with the experiments presented in the present application in relation to copper catalysts, namely improved activity, selectivity and crush strength. They

took the stand that the technical effects shown with these experiments were not exclusive to copper catalysts, but were common to catalysts prepared without an alumina support, so they could be extrapolated to other catalysts, such as nickel-containing catalysts, prepared without an alumina support.

6. The results of the catalytic hydrogenation tests presented in the application, which concern the catalytic hydrogenation of ethylpropylacrolein, furfural, nitrobenzene and of a C<sub>12</sub> methylester, are respectively summarized in Tables 1 to 4. The comparison presented in Table 1 concerns the hydrogenation of ethylpropylacrolein. It is meant to demonstrate that copper catalysts using only clay or commercial synthetic calcium silicate as a binder do not impart an improvement in respect of activity, selectivity and crush strength, which improvement is obtained when using copper catalysts comprising a combination of those binders. As regards the comparative tests shown in Table 2, they are alleged to demonstrate an improved crush strength and similar activity and selectivity of the copper catalysts of the parent application for the hydrogenation of furfural, when compared to a commercial non-chromium copper catalyst described to be "Engelhard Cu-0320T: 61% Cu, 20% Na<sub>2</sub>SiO<sub>3</sub>". The comparison shown in Table 3 relates to the hydrogenation of nitrobenzene into aniline and is reported to show that the copper catalysts of the parent application give better activity than a commercial copper chromite catalyst (Engelhard Cu-1152T: 29% Cu, 26% Cr, 7% Ba, 15% CaSiO<sub>3</sub>), while having higher strength and lower bulk density. Finally, the copper

catalysts of the parent application, when compared in Table 4 with a commercially available reference copper chromite catalyst (Engelhard Cu-1987 T1/8: 36% CuO, 33% Cr, 3% Mn), are alleged to have better activity and selectivity for the conversion of a C<sub>12</sub> methylester into the corresponding C<sub>12</sub> alcohol. It is therefore conspicuous to the Board that the comparisons offered in Tables 1 to 4 of the present application have not been shown to have been made with a support corresponding to that used for catalyst Ni-3266E, i.e. a support which contrary to the reference catalysts used in Tables 1 to 4 contains a mixture of calcium silicate, bentonite clay and alumina.

7. According to the established case law, if comparative tests are relied on to demonstrate an inventive step on the basis of an improved effect, it is *inter alia* required that the nature of the experimental evidence offered must be such that the alleged advantage or effect is convincingly shown to have its origin in the feature(s) distinguishing the invention from the closest prior art. It follows therefore that the experimental results presented in the application, which are not based on a comparison with a catalyst having a binder system corresponding to that of Ni-3266E, let alone in the framework of nickel based catalysts, are not suitable to demonstrate any technical effect resulting from the replacement of the binder system of Ni-3266E by a binder system as presently defined in Claim 1. Under these circumstances, the experimental results contained in the parent and divisional applications do not support the Appellants' contention for the existence of technical effects resulting from the absence of "added alumina other than

that which may be present resulting from the clay material".

8. As to the experimental evidence constituted by Example 4 of D6, it concerns the hydrogenation of 3-hydroxypropanol using three catalysts A, B and D according to the invention of D6 and one reference catalyst C, described to be a standard off-the-shelf catalyst. The composition of those catalysts is only defined in Table 1 in terms of the content of their various metals, including nickel, silicon, aluminium, molybdenum and calcium. It is however not possible on the basis alone of the information presented in Table 1 to determine the nature of the binder system employed for those catalysts. Neither is it possible in respect of catalyst C on the basis of the Appellants' vague statement at the oral proceedings that catalyst C is believed to be a similar type of catalyst than Ni-3266E. Thus, the comparison offered with the results shown in Table 1 of D6 does not allow any conclusion to be drawn as regards the effect of the absence of "added alumina other than that which may be present resulting from the clay material" on crush strength, activity or selectivity of the catalyst.
  
9. Even if one assumed that two catalysts among catalysts A, B and D used in Example 4 of D6 corresponded to those prepared in Examples 2 and 3 of that document, and catalyst C were the catalyst obtained with the synthesis described in Example 1 which is defined to concern a conventional catalyst preparation, it would not be possible to attribute the difference in crush strength or activity pattern observed between catalyst C and catalysts A, B and D to the absence or presence

of "added" alumina, because the catalysts tested would also differ at least in the use of different amounts of molybdenum trioxide, in the use of a different clay and the absence or different amounts of calcium silicate, the influence of which on the crush strength and activity cannot be ignored. This is all the more the case, in view of the Appellants' argument that the presence or absence of the alumina binder in Ni-3266E has an influence on the catalytic activity of the catalyst and in view of the information provided in D6 according to which MoO<sub>3</sub> not only has a binder function, but also serves as an activity promoter.

10. Summing up, in view of the established case law that alleged but unsupported advantages cannot be taken into consideration in respect of the determination of the problem underlying the invention, the technical problem solved by the subject-matter of present Claim 1 over D1 must be seen merely in the provision of further catalysts which are effective for hydrogenation reactions. The Board has no doubt, in particular in view of the presence of nickel, that this problem is successfully solved over D1 by the subject-matter of present Claim 1.

*Obviousness*

11. It remains to be decided whether or not the skilled person starting from D1 and wishing to solve the above defined problem would have been guided by the available prior art to apply the solution defined in Claim 1 of the present Main Request, namely the use of a binder system which contains calcium silicate, at least one clay material and is free of "added alumina" other than

that which may be present resulting from the clay material and free of chromium and barium.

12. Claim 1 of D1 already indicates that the catalyst support is one or more materials selected from the group consisting of silica, alumina, silica-alumina, aluminosilicates and carbon, silica including calcium promoted silica as shown on page 5, lines 20-21 which provides a description of the binder system of Ni-3266E. D1 allows therefore binders which do not contain alumina in addition to that already contained in the clay material. Moreover, numerous inorganic materials are known to the skilled person as binder or support for catalysts. Hence, the skilled person starting from document D1 and wishing to provide further catalysts which are effective for hydrogenation reactions would have found it obvious in view of document D1 alone, or his general knowledge, to use instead of the binder system of Ni-3266E a binder system obtained with calcium silicate, at least one clay material and optionally further binder materials different from alumina. Replacing the binder system in the catalyst of D1 by a combination of materials known to perform the same function, for example those which as for Ni-3266E are also free of chromium and barium, would be obvious to the skilled person who is merely seeking to provide further catalysts which are effective for hydrogenation reactions and said catalytic activity is expected in view of the presence of nickel.
  
13. Consequently, the subject-matter of Claim 1 according to the Main Request does not involve an inventive step within the meaning of Article 56 EPC. The Main Request is therefore rejected.



*First Auxiliary Request*

14. The First Auxiliary Request must be also rejected for the same reasons, as its Claim 1 is identical to that of the Main Request.

*Second Auxiliary Request*

15. Compared to Claim 1 of the Main Request, Claim 1 of the Second Auxiliary Request additionally requires that the nickel be in the form of nickel oxide and that the amount of nickel, calcium silicate and the at least one clay material in the shaped catalyst be comprised in the range of from 30 wt.-% to 75 wt.-%, from 10% by weight up to 40% by weight and from 1% by weight up to 30% by weight, respectively. In respect of the definition of nickel being in the form of nickel oxide, which was already defined in Claim 1 of the Main Request underlying the contested decision, it was never argued that the definition of the oxide form for the nickel, which is a measure conventional in the art, provided any contribution to an inventive step, let alone any distinguishing feature over the commercial catalyst Ni-3266E. As to the ranges defining the amounts of calcium silicate and clay material now introduced into Claim 1, they have not been shown to be linked to any surprising technical effect and must, therefore, be considered as arbitrary limitations for which no inventive step can be acknowledged. As a result, the additional features inserted in Claim 1 of the Main Request do not change the conclusions of the Board as to the absence of an inventive step. Thus, the

catalysts claimed in accordance with the Second Auxiliary Request also lack an inventive step.

16. Moreover, Claim 1 of the Second Auxiliary Request defines that the shaped catalyst can comprise 75 wt.-% of nickel, the nickel being in the form of nickel oxide, which means that nickel oxide can be present according to that definition in an amount of 95 wt.-% for Nickel(II) oxide or even higher amounts for other forms of nickel oxide. Thus, the lower limits of 10 wt.-% for calcium silicate and of 1 wt.-% for the at least one clay material required by present Claim 1 are not compatible with an upper limit of 75 wt.-% of nickel, the nickel being in the form of nickel oxide, as it would amount to a percentage of the components in the catalyst in excess of 100 wt.-%. Under these conditions, the definition of the catalyst composition is ambiguous and Claim 1 of the Second Auxiliary Request is deemed to lack clarity within the meaning of Article 84 EPC. This also raises doubt whether or not the combination of features operated by the Appellants with Claim 1 of the Second Auxiliary Request would be considered to have a basis in the parent application in order to meet the requirements of Article 76(1) EPC. However, this question can be left unanswered in view of the fact that the Second Auxiliary Request is rejected on the ground that the subject-matter of its Claim 1 lacks, for the reasons given above, clarity and an inventive step within the meaning of Article 56 EPC.

*Third Auxiliary Request*

17. The Appellants acknowledged that Claim 1 of the Third Auxiliary Request and Claim 1 of the Second Auxiliary

Request had, despite a different wording, the same meaning. Under those conditions, the negative conclusions on clarity and inventive step apply for the same reasons to the Third Auxiliary Request, which therefore is also rejected.

*Fourth Auxiliary Request*

18. Claim 1 of the Fourth Auxiliary Request differs only from Claim 1 of the Main Request in that it is no longer required that the mixture of compounds (i) to (iii) be uniform. This amendment was only introduced in order to overcome an objection raised against the Main Request under the provisions of Articles 76(1) and 123(2) EPC. The suppression of this feature in Claim 1 of the Main Request, which provides no further distinction over the closest prior art, is therefore without any effect on the assessment of inventive step given above in respect of Claim 1 of the Main Request. Hence, the Fourth Auxiliary Request must be rejected as its Claim 1 does not involve an inventive step either.

*Fifth Auxiliary Request*

19. The Fifth Auxiliary Request must be also rejected for the same reasons as given in respect of the Main Request as their Claims 1 are identical.

**Order**

**For these reasons it is decided that:**

The appeal is dismissed.

The Registrar

The Chairman

S. Fabiani

J. Riolo