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D E C I S I O N
of 24 June 2005

Case Number: T 0499/02 - 3.3.7

Application Number: 97917586.6

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Language of the proceedings: EN

Title of invention:

Preparation and use of non-chrome catalysts for Cu/Cr catalyst applications

Applicant:

ENGELHARD CORPORATION

Opponent:

-

Headword:

-

Relevant legal provisions:

EPC Art. 54, 84, 111(1), 123(2)

Keyword:

"Clarity (yes) - functional feature"

"Remittal (yes) - fresh case"

Decisions cited:

T 0068/85

Catchword:

-



Case Number: T 0499/02 - 3.3.7

D E C I S I O N
of the Technical Board of Appeal 3.3.7
of 24 June 2005

Appellant: ENGELHARD CORPORATION
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 5 December 2001
refusing European patent application
No. 97917586.6 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: R. E. Teschemacher
Members: B. J. M. Struif
B. L. ter Laan

Summary of Facts and Submissions

I. European patent application No. 97 917 586.6 originating from international application PCT/US97/04678 published as WO-A-97/34 694 has a filing date of 21 March 1997 and claims priority from US 60/013 824 dated 21 March 1996. The independent claims 1, 2, 9, 25, 29, 31 and 40 read as follows:

"1. A catalyst having the formula $n\text{CuO} \cdot \text{Al}_2\text{O}_3$ wherein n is between 0.14 and 5.13."

"2. A generally homogeneous catalyst consisting primarily of oxides of copper and aluminum, the catalyst having aluminum content calculated as alumina greater than about 20% by weight."

"9. A method of making a catalyst containing primarily oxides of copper and aluminum, comprising the steps of:
co-precipitating a Cu-Al-O precipitate from a solution containing a soluble copper salt and a soluble aluminum compound in the presence of a precipitating agent;
calcining the precipitate at an appropriate temperature for an appropriate length of time."

"25. A solid catalyst comprising an extrudate of a Cu-Al-O powder with LOD of thirty to fifty percent, the extrudate being formed with or without binder or lubricant, the extrudate being substantially free of chromium."

"29. A solid Cu-Al-O catalyst comprising a tablet being formed with 0 ~ 8% graphite powder substantially free of chromium, the catalyst comprising a tablet having a

pore volume from 0.2 ml/g to 0.6 ml/g and a bulk density of approximately 0.7 g/ml to approximately 1.5 g/ml."

"31. In a catalytic reaction catalyzed by a copper-chrome catalyst, the improvement comprising substituting for the copper-chrome catalyst a Cu-Al-O catalyst substantially free of chrome."

"40. A method of preparing a Cu-Al-O catalyst powder comprising the steps of:
co-precipitating copper nitrate and sodium aluminate in the presence of sodium carbonate to form a Cu-Al-O slurry;
filtering the slurry to form a filter cake
washing the filter cake;
drying the filter cake to a dried powder; and
calcining the dried powder."

II. The appeal lies from the decision of the examining division holding that the above patent application did not meet the requirements of Article 84 EPC. Furthermore, objections to novelty and inventive step were raised. The decision was based on claims 1 to 44 filed with letter dated 11 October 2000 as the sole request, of which independent claims 1 and 23 referred to a catalyst and its method of preparation, reading as follows:

"1. A Cu-Al-O catalyst having a homogeneous bulk composition, the catalyst consisting essentially of oxides of copper and aluminum, having been heated to a calcination temperature ranging from 400° to 900 °C to

produce the catalyst having less than 10% leachable copper ions."

"23. A method of making a catalyst containing oxides of copper and aluminum, comprising the steps of:

co-precipitating a Cu-Al-O precipitate from solutions containing a soluble copper salt and a soluble aluminum compound in the presence of a precipitating agent at a pH of about 7.4 ± 0.5 ;

washing the filter cake to lower the Na content in the catalyst below 1% by weight optionally drying the precipitate;

and calcining the precipitate at a temperature ranging from 400 to 900°C."

The decision was based *inter alia* on the following documents:

D2: EP-A-0 042 471

D3: EP-A-0 434 061

III. The decision can be summarized as follows:

- (a) The term "the catalyst having less than 10% leachable copper ions" in claim 1 was not clear, since the conditions of leaching were not specified. That term was a functional feature without any supporting features to state how it was achieved. Furthermore, at a calcination temperature of 400°C, the level of leaching was considerably higher than the claimed one. Thus, the requirements of Article 84 EPC were not met.

The following further objections were mentioned, without however forming the basis of the refusal:

(b) The subject-matter of claim 1 was not novel in view of D2, which disclosed a catalyst composition comprising Al and Cu oxides in which a mixture of nitrates of Al and Cu was brought together and the pH adjusted. Then, the product was washed, dried and calcined at 750 to 850°C to form a spinel structure. Furthermore, D3 was novelty destroying for claims 1 and 18, since the calcination of the Al and Cu compounds was carried out at 400°C and since reactions from the list of claim 18 were disclosed. Thus, the requirements of Article 54 EPC were not met.

(c) Even if process claim 23 were novel, it did not involve an inventive step. The process conditions of the examples of D2 were close to the pH range specified in claim 23 and normal washing would lead to a reduction in Na content of less than 1%. The only difference of process claim 23 from D2 was the higher pH range of 6.9 versus 6.5 for carrying out the reaction in D2. That difference was small and it was not apparent that it contributed to an inventive step. Thus, the claimed subject-matter did not comply with Article 56 EPC.

IV. On 5 February 2002 the applicant (appellant) filed a notice of appeal against the above decision, the prescribed fee being paid on the same day. With the statement setting out the grounds of appeal filed on 5 April 2002, the appellant submitted an amended set of

claims 1 to 41 as the sole request, as well as a test report dated 12 February 2002.

V. By letter dated 25 April 2005, in reply to a communication of the board, the appellant submitted an amended set of claims 1 to 35 and adapted description pages 1 to 4, 4A and 4B, replacing the previous requests on file.

VI. Oral proceedings were held on 24 June 2005. During the oral proceedings the appellant submitted an amended claim 1 (sole request) reading as follows:

"1. Use of a nonchrome homogeneous catalyst consisting primarily of oxides of copper and aluminum, the catalyst having an aluminum content calculated as alumina greater than 20 % by weight, and a CuO content of $61 \pm 10\%$ by weight, and wherein the catalyst includes crystal phase CuO and CuAl_2O_4 and has no more than 5% copper ions leachable as determined by reacting 100 ml 10% acetic acid with 10g of powder catalyst for one hour with continuous stirring, in a reaction chosen from the group consisting of hydrogenolysis of coconut fatty acid, oxoalcohol finishing, hydrogenation of nitriles to unsaturated secondary amines, hydration of acrylonitrile to acrylamide, hydrogenolysis of methyl laurate, alkylation of a phenol with alcohol, amination of an alcohol, dehydrogenation of an alcohol, hydration of a nitrile, hydrogenation of an aldehyde, hydrogenation of an amide, hydrogenation of a fatty acid via esterification and hydrogenolysis, selective hydrogenation of a fat, selective hydrogenation of an oil, hydrogenation of a nitrile, hydrogenation of a nitroaromatic hydrocarbon, hydrogenation of a ketone,

hydrogenation of furfural, hydrogenation of an ester, hydrogenation of carbon monoxide to methanol, oxidation of a vapor organic compound, oxidation of sulfur dioxide, oxidation of an alcohol, decomposition of nitric oxide, selective catalytic reduction of nitric oxide, and purification of a gas stream by the removal of oxygen."

VII. The appellant argued in substance as follows:

- (a) The amendments to claim 1 were based on original claims and the description and met the requirements of Article 123(2) EPC. Since the leaching conditions had been incorporated into claim 1, the clarity objection was overcome.

- (b) D2 disclosed a catalyst wherein 60 to 100% of the copper present was bound to aluminium oxide in the form of its spinel. The catalyst of D2 was used only for the conversion of carbon monoxide with water to provide hydrogen and concerned a technical use different from the claimed one. Thus, D2 did not disclose the use of a catalyst having a low amount of leachable copper ions and a specific high CuO content for the reactions now specified in claim 1.

According to D3, the amount of leachable copper prepared according to page 5, lines 25 to 33, was in excess of 35%, i.e. outside the claimed range, as shown by the experimental report of 12 February 2002. Furthermore, under the reaction conditions of D3, no crystalline copper oxide-aluminium oxide spinel was formed.

Thus, the claimed subject-matter was novel over D2 and D3.

- (c) The appellant agreed to a remittal of the case to the first instance if the board came to the conclusion that the objections raised in the decision under appeal had been overcome.

VIII. The appellant requests that the decision under appeal be set aside and that a patent be granted on the basis of claim 1 in the version as submitted during the oral proceedings and the other claims as well as the description yet to be adapted to amended claim 1.

Reasons for the Decision

1. The appeal is admissible.

Amendments

Article 123(2) EPC

2. The amendments to claim 1 are based on original claims 2, 6, 18, 21, 22 and 34 in connection with page 11, lines 16 to 20 (definition for the leachable cation measurement), page 12, lines 7 to 8 (leachable copper content), page 5, lines 1, 4, 7, 10, 12, 15, 18 and page 6, line 1 (non-chrome). Except for the term "no more than", which should read "less than", (see original claim 22 and page 12, lines 7 and 8), all the amendments can be directly and unambiguously derived from the application as filed. With the one exception

still to be corrected, the amended features meet the requirements of Article 123(2) EPC.

Clarity

3. The examining division had objected to the functional feature "the catalyst having less than ... leachable copper ions" as not being clear, since the conditions for measuring the leaching were not specified and since the feature referred to a result to be achieved without stating the means as to how to achieve the desired result.

3.1 According to T 68/85, OJ EPO 1987, 228, functional features defining a technical result are permissible in a claim if, from an objective viewpoint, such features cannot otherwise be defined more precisely without restricting the scope of the invention, and if these features provide instructions which are sufficiently clear for the expert to reduce them to practice without undue burden, if necessary with reasonable experiments.

3.2 The leachable content of copper ions is a feature of the catalyst with functional and structural aspects, which can be determined on a catalyst sample by using a simple analytical test, as now specified in claim 1. By that test the skilled person is able to measure the leachable copper ions in the catalyst without undue burden.

3.3 The catalyst is furthermore defined by a specified amount of alumina and CuO and includes a crystal phase CuO and CuAl_2O_4 . From these features, the skilled person obtains sufficient technical information about the

composition of the catalyst and its crystalline structure. In addition, according to the application as filed, the spinel structure of copper oxide-aluminium oxide is formed when the catalysts are calcined above 700°C (see page 7, lines 8 to 12) as further illustrated in example 9. Under those calcination conditions, the leachable copper ion content can be kept below 5% as also shown in table 1, page 12.

- 3.4 Although the leachable copper ion content may be influenced by the conditions under which the catalyst is prepared, such as the calcination temperature (page 12, example 8, table 1) and the washing conditions (page 18, example 16, table 6), such process conditions would not contribute to a clearer definition of the catalyst. In that respect, a catalyst defined by its leachable copper content can be directly identified and compared with catalysts of the prior art; by applying the specified test it can be seen whether or not the tested catalyst is within the scope of the claim. In contrast to this, defining a catalyst merely by way of process features might be considered not to contribute to the definition of a different catalyst (compare Case Law of the Boards of Appeal of the European Patent Office, 4th Edition 2001, II.B.6.2).
- 3.5 From the above it follows that the skilled person is able to reduce the instructions in claim 1 to practice without undue burden and can decide whether or not a catalyst meets the requirement of claim 1. Consequently, the functional feature as claimed fulfils the prerequisites of the established case law and overcomes the clarity objection raised.

Novelty

4. According to the decision under appeal, D3 was novelty destroying for the claims underlying that decision, since the claimed catalysts did not differ from the known catalysts and since the same reactions as now claimed were involved.

4.1 D3 discloses a process for hydrogenating a bound oxygen-containing organic compound, said process comprising:
contacting said organic compound as a vapour with hydrogen in the presence of an activated catalyst composition consisting of copper and aluminum under hydrogenation conditions, said catalyst having been made by a process comprising:
reducing a homogeneous mixture of copper and aluminum oxides by heating in the presence of a reducing gas under activation conditions which comprise a gradually increasing temperature from a starting temperature of about 40° to 75°C to a final reduction temperature of about 150°C to 250°C (claim 1).

Said organic compound preferably comprises a mono-ester, a di-ester, or a mixture thereof (claim 2).

4.1.1 D3 furthermore discloses a method for preparing a copper and aluminum catalyst useful for hydrogenating an oxygen-containing hydrocarbon feed, said method comprising:
co-precipitating copper and aluminum from their water soluble salts to form a precipitate;
drying and calcining the precipitate to form a calcined catalyst; and

- activating the calcined catalyst by heating said calcined catalyst in the presence of a reducing gas under activation conditions which comprise a reducing temperature that gradually increases from an initial reducing temperature of about 40° to 75°C to a final reducing temperature of about 150°C to 250°C (claim 12).
- 4.1.2 According to the description, a Cu(54):Al(46) catalyst precursor may be prepared in the following manner. A first solution (Solution A) comprising $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ and $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ in deionized water and a second solution (Solution B) comprising Na_2CO_3 in deionized water are each heated to 60°C. Solution B is then added with stirring to Solution A resulting in the formation of a precipitate. This mixture is stirred for 3 hr while cooling to 25°C. The precipitate is isolated and washed with deionized water. The precipitate is then dried in air at 100°C for 18 hr and calcined in air at 400°C for 2 hr (page 5, lines 25 to 31).
- 4.2 The catalyst sample as specified in D3, page 5, lines 25 to 31 has been reproduced by the appellant (test report of 12 February 2002). In that test report, reference is made to US-A-5 053 380, column 6, lines 14 to 27, which corresponds to the cited passage of D3 (page 5, lines 25 to 31). It has been shown that the content of leachable copper ions of a catalyst precursor prepared along the lines of D3 is 34.46%, i.e. outside the claimed range of less than 5%. Therefore, it can be accepted that the presently used catalyst differs from the one disclosed in D3.
- 4.3 Furthermore, the calcination temperature for preparing the catalyst according to D3 is at most 550°C (page 5,

line 21). Since, according to the application as filed (page 12, lines 15 to 18), at 600°C only a crystalline CuO phase is detected by X-ray defraction and a spinel crystalline phase only appears at a calcination temperature of 700°C or higher (page 7, lines 7 and 8), the temperature conditions of D3 are too low for the formation of a spinel structure as required by the present claims. Thus, the catalyst used according to present claim 1 is distinguished from D3 in its structural features. Hence, the novelty objection based on D3 has been overcome by amended claim 1.

Remittal

5. The claimed subject-matter compared to the claims underlying the decision under appeal has been substantially amended as regards the composition and physical structure of the catalyst as well as by its specific use. Those amendments amount to a "fresh case" not yet having been examined by the first instance (Case Law, *supra*, VII.D.7.5.2.(d)). Since the objections mentioned in the decision under appeal related to a different version of the claims, and since the appellant agreed to a remittal, the board finds it appropriate to remit the case to the department of first instance (Article 111(1) EPC).

When prosecuting the case further, the examining division should take note of the correction of the term "no more than" to "less than" in claim 1 as indicated under point 2. above. Although the board has considered novelty with respect to document D3, the final decision on novelty as regards the other documents on file is left to the examining division. The Board notes that

when dealing with the inventive step of the use claim, attention should be paid to whether or not any of the claimed features leads to a specific technical effect in view of the closest state of the art. If a specific technical effect can be shown, the question may arise, whether or not that effect may be considered to be present in all claimed uses.

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 for further prosecution.

The Registrar:

The Chairman:

P. Cremona

R. Teschemacher