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**Datasheet for the decision
of 3 February 2011**

Case Number: T 1835/07 - 3.3.07

Application Number: 99944639.6

Publication Number: WO 00/15333

IPC: B01J 23/50

Language of the proceedings: EN

Title of invention:

Process for removing ionizable species from catalyst surface
to improve catalytic properties

Applicant:

Shell Internationale Research Maatschappij B.V.

Opponent:

-

Headword:

-

Relevant legal provisions:

EPC Art. 56, 111(1), 123(2)

Relevant legal provisions (EPC 1973):

EPC Art. 84

Keyword:

"Amendments - clear (yes) - allowable (yes) - Main Request,
First and Second Auxiliary Requests"

"Inventive step (no) - Main and First Auxiliary Requests"

"Remittal (yes) - Second Auxiliary Request"

Decisions cited:

T 0197/86, T 1188/00

Catchword:

-



Case Number: T 1835/07 - 3.3.07

DECISION
of the Technical Board of Appeal 3.3.07
of 3 February 2011

Appellants: Shell Internationale Research Maatschappij B.V.
Carel van Bylandtlaan 30
NL-2596 HR The Hague (NL)

Representative: -

Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 18 April 2007
refusing European application No. 99944639.6
pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: J. Riolo
Members: G. Santavicca
P. Schmitz

Summary of Facts and Submissions

- I. The appeal by the applicants lies from a decision of the Examining Division refusing European patent application 99 944 639.6, concerning a "process for removing ionizable species from catalyst surface to improve catalytic properties".
- II. The decision under appeal was based on five sets of claims submitted at the oral proceedings held on 27 March 2007 as Main and Auxiliary Requests 1 to 4. Independent Claim 11 of the Main and First Auxiliary Requests as well as Claim 1 of the First to Fourth Auxiliary Requests read as follows.

Main Request

"11. A process for the catalytic epoxidation of an alkene with an oxygen containing gas, wherein a catalyst as prepared according to any one of claims 1 to 10 is used."

First Auxiliary Request

"1. A process for the preparation of a catalyst which comprises depositing a catalytically effective amount of one or more catalytically reactive metals comprising silver, and depositing one or more promoters prior to, coincidentally with, or subsequent to, the deposition of said one or more catalytically reactive metals, on one or more materials from which the carrier is formed and which treatment is effective in lowering the

concentration of one or more ionizable species present on the surface of the complete carrier, the lowering being expressed as the lowering of the relative concentration of Si atoms, as measured by X-ray Photoelectron Spectroscopy (XPS) performed on the untreated and treated carrier."

"11. A process for the catalytic epoxidation of an alkene with an oxygen containing gas, which comprises the steps of

- preparing a catalyst by a process as claimed in any one of claims 1 to 10, and,
- contacting the alkene with an oxygen containing gas in the presence of a catalyst."

Second Auxiliary Request

Compared with Claim 1 of the First Auxiliary Request, Claim 1 of the Second Auxiliary Request comprises the limitation "onto a carrier which comprises α -alumina wherein a treatment has been performed on the α -alumina from which the carrier is formed" as a replacement of the feature "on one or more materials from which the carrier is formed".

Third Auxiliary Request

Compared with Claim 1 of the First Auxiliary Request, Claim 1 of the Third Auxiliary Request comprises the limitation "onto a carrier which comprises α -alumina wherein a treatment has been performed on the α -alumina from which the carrier is formed and on the complete carrier" as a replacement of the feature "on one or more materials from which the carrier is formed".

Fourth Auxiliary Request

Compared with Claim 1 of to the First Auxiliary Request, Claim 1 of the Second Auxiliary Request comprises the limitation "onto a carrier which comprises α -alumina wherein a treatment has been performed on the α -alumina from which the carrier is formed" [as a replacement of the feature "on one or more materials from which the carrier is formed"] as well as the further feature "which treatment is washing performed with an aqueous and/or organic solvent-based solution, selected from water, tetraethylammonium hydroxide, ammonium acetate, lithium carbonate, barium acetate, strontium acetate, crown ether, methanol, ethanol, dimethylformamide, and mixtures thereof."

III. According to the decision under appeal:

- (a) Claim 11 of the Main Request concerned a catalytic epoxidation of an alkene with an oxygen-containing gas, the catalyst of which was defined by its process of preparation. There was no evidence that the process of preparation imparted distinguishing properties to the defined catalyst over the known catalysts, and the other features of Claim 11 were known. Thus, the process of Claim 11 was not novel.
- (b) Claim 11 of the First Auxiliary Request comprised a step for the preparation of the catalyst, so that it was novel. Claim 1 of the First Auxiliary Request encompassed any carrier but referred only to an Al-containing carrier, so that it was not

clear (Article 84 EPC). The process of Claim 1 was in any case obvious over D7 (DE-A-2 933 950).

- (c) Claim 1 of the Second Auxiliary Request defining a carrier comprising α -alumina was clear. The closest prior art was still described by D7, which addressed the problem of providing a method for preparing an epoxidation catalyst having good activity and selectivity being stable over time. Although D7 did not mention any treatment of the α -alumina material before forming the carrier, the effect achieved by washing the α -alumina before forming the carrier, compared to the washing of the formed carrier, was very small, if any. Hence, the problem was to provide a further process of preparation for epoxidation catalysts and the defined process was obvious over D7, which taught that alkali soluble silica should be avoided, by using Si-free α -alumina or by treating commercial α -alumina with NaOH.

- (d) Claim 1 of the Third Auxiliary Request concerned a method of preparation of the catalyst, in which the source α -alumina and the formed carrier were both treated. The examples submitted did not prove that any unexpected effect over D7 was achieved, so that the problem solved was still the provision of a further method of preparation of the catalyst. The process defined in Claim 1 was obvious over D7.

- (e) Claim 1 of the Fourth Auxiliary Request no longer contained the treatment of the complete carrier and the washing was performed with an aqueous or organic solvent-based solution selected from the

defined media, *inter alia* tetraethylammonium hydroxide (TEAH) and water, which made an alkaline solution, i.e. a further alkaline solution capable of removing the alkali soluble silica encompassed by the definition "ionizable silicates" of the application and disclosed by D7. These limitations did not change the problem solved over D7, i.e. the provision of a further process. The further washing media as defined, e.g. the further alkaline solutions such as TEAH, were arbitrary and obvious choices for the skilled person.

(f) Therefore, none of the requests was allowable.

- IV. In their statement setting out the grounds of appeal, the appellants enclosed a new Main Request, First, Second and Third Auxiliary Requests corresponding to the Second, Third and Fourth Auxiliary Requests underlying the decision under appeal as well as comparative test results over D7.
- V. In a communication in preparation for oral proceedings, annexed to the summons to oral proceedings dated 23 November 2010, as well as in a letter faxed on 1 February 2011, the Board drew attention to the points that needed to be discussed.
- VI. Oral proceedings took place on 3 February 2011. The appellants submitted 6 sets of claims as the Main and First to Fifth Auxiliary Requests replacing all of the previous requests on file. After the closure of the debate and the deliberation by the Board the decision was announced orally.

VII. Claim 1 of each of the Main, First and Second Auxiliary Requests read as follows:

Main Request

"1. A process for the preparation of a catalyst for the vapour phase epoxidation of an alkene which process comprises performing a treatment on one or more of the materials from which an α -alumina-based carrier is formed, which treatment is effective in lowering the concentration of one or more ionizable species present on the surface of the complete carrier, the lowering being expressed as the lowering of the relative concentration of Si atoms by at least 5%, as measured by X-ray Photoelectron Spectroscopy (XPS) performed on carrier and on a corresponding carrier for which no treatment has been performed, depositing a catalytically effective amount of one or more catalytically reactive metals comprising silver as the main catalytically reactive metals, and depositing one or more promoters prior to, coincidentally with, or subsequent to, the deposition of said one or more catalytically reactive metals, onto said α -alumina-based carrier , wherein said treatment is washing performed with an aqueous and/or organic solvent-based solution, selected from water, ammonium acetate, lithium carbonate, barium acetate, strontium acetate, crown ether, methanol, ethanol, dimethylformamide, and mixtures thereof."

First Auxiliary Request

Compared with Claim 1 of the Main Request, Claim 1 according to the First Auxiliary Request contained the further limitations "depositing from 1 to 40 wt% of silver, basis the weight of the total catalyst, and depositing one or more promoter selected from Group IA metals in an amount in the range of from 10 ppm to 1500 ppm, and Group VIIb metals in an amount less than 3600 ppm, each amount expressed as the metal and by weight of the total catalyst, prior to, coincidentally with, or subsequent to, the deposition of the silver, onto said α -alumina-based carrier".

Second Auxiliary Request

"1. A process for the preparation of a catalyst for the vapour phase epoxidation of an alkene which process comprises performing a treatment on one or more of the materials from which an α -alumina-based carrier is formed, which treatment is effective in lowering the concentration of one or more ionizable species present on the surface of the complete carrier, the lowering being expressed as the lowering of the relative concentration of Si atoms by at least 5%, as measured by X-ray Photoelectron Spectroscopy (XPS) performed on carrier and on a corresponding carrier for which no treatment has been performed, submerging the carrier in a silver impregnation solution containing a metal hydroxide, depositing from 1 to 40 wt% of silver, basis the weight of the total catalyst, and

depositing one or more promoter selected from Group IA metals in an amount in the range of from 10 ppm to 1500 ppm, and Group VIIb metals in an amount less than 3600 ppm, each amount expressed as the metal and by weight of the total catalyst, prior to, coincidentally with, or subsequent to, the deposition of the silver, onto said α -alumina-based carrier , wherein said treatment is washing performed with an aqueous and/or organic solvent-based solution, selected from water, ammonium acetate, lithium carbonate, barium acetate, strontium acetate, crown ether, methanol, ethanol, dimethylformamide, and mixtures thereof."

VIII. The appellants essentially argued as follows:

- (a) The claims of the Main and First to Fifth Auxiliary Requests were clear and based on the original application, thus admissible.
- (b) None of the prior art cited in the proceedings anticipated the process of Claim 1 of the Main Request nor that of Claim 1 of each of the auxiliary requests. In particular, the washing treatment performed on the material from which the carrier was formed was not disclosed by D7.
- (c) The closest prior art was described by D7, which taught to reduce the alkali soluble silica to very low amounts by using NaOH or HF. However, NaOH and HF were aggressive media capable of extracting alumina materials and of generating basic or acidic sites.

- (d) Having regard to the results proven, e.g. the examples in the application as filed and the further examples provided with letter of 25 March 2007 as well as the crush tests carried out after treatment of a commercial carrier with the method of D7, as mentioned in the statement setting out the grounds of appeal, the effectively solved problem was an improvement of the properties of a promoted silver-on-alumina catalyst for the production of ethylene oxide without compromising the physical integrity of the catalyst, i.e. its physical form, hence whilst maintaining crush strength, which if below 5 kg was commercially unacceptable. If the maintenance of the crush strength could not be considered as part of the problem, the latter should be formulated as a process for preparing a catalyst with useful activity and selectivity properties as one that is prepared with aggressive media but without the use of aggressive media. The latter formulation was based on the deletion of the tetraethylammonium hydroxide, objected to by the Examining Division.
- (e) As to obviousness, D7 disclosed the treatment of the formed carrier, not that of the materials from which the carrier was formed. Furthermore, D7 taught that only a minimal level of alkali-soluble Si should be present after treatment of the carrier, i.e. a virtually total removal of it was necessary, or that Si-free carriers should be used, to attain useful catalytic properties. However, silicates were not an inevitable component of alumina source material. Hence, D7 gave no suggestion that washing the source material of the

carrier was beneficial to attaining a catalyst with useful activity and selectivity properties while safeguarding the physical properties of the catalyst. Therefore, the skilled person starting from D7 could not obviously arrive at the solution of Claim 1 of Main and First Auxiliary Requests.

- (f) Claim 1 of the Second Auxiliary Request defined the use of a metal hydroxide in the silver impregnation solution, to bring the claim closer to examples 10 and 11 and to catalyst G of Annex II to the letter dated 25 March 2007.

IX. The appellants requested that the decision under appeal be set aside and that the case be remitted to the examining division for continuation of the examination procedure based on the Main Request or one of Auxiliary Requests 1 to 5 filed during the oral proceedings before the Board.

Reasons for the Decision

- 1. The appeal is admissible.

Main Request

- 2. *Amendments*

- 2.1.1 Claim 1 of the Main Request is based on Claim 7 (process for the preparation of a catalyst including the deposition steps for reactive metals and promoters) of the application as filed and includes the further features of Claim 1 (treatment performed on one or more

of the materials from which the carrier is formed), Claim 4 (washing step and relevant media as defined), Claim 5 (measure of the lowering of the concentration on the surface by at least 5% by XPS), Claim 9 (catalyst for the vapor phase epoxidation of an alkene) and Claim 10 (containing silver as the main catalytically reactive metal on alumina-based carrier). The further restriction to α -alumina-based carrier is based on page 8, line 10, of the original description.

2.1.2 The clarity objections raised in the communications by the Board have been addressed by the amendments made.

2.1.3 Therefore, the Board is satisfied that Claim 1 of the Main request fulfils the requirements of Articles 84 and 123(2) EPC.

3. *Inventive step*

3.1 The present application concerns a process for removing ionizable species from catalyst surface to improve catalytic properties.

3.2 According to the application as filed:

- (a) An "ionizable" species is a species which is capable of being rendered ionic, where the term "ionic" or "ion" refers to an electrically charged chemical moiety (page 4, line 22-25).
- (b) "Ionizable species" typically present on the inorganic type carriers include "soluble silicate" (page 5, lines 9-11).
- (c) Lowering the undesirable ionizable species concentration can be accomplished by any means which is effective in rendering the ionizable

species ionic and removing that species (page 5, lines 14-18).

- (d) The "surface" of the carrier is the area which may be measured by the standard method of Brunauer, Emmett and Teller (BET), i.e. the site at which reaction takes place (page 4, lines 18-22).
- (e) "Improvement in catalytic properties" means that the properties of the catalyst are improved as compared with a catalyst made from the same carrier which has not been treated to lower surface ionizable species, whereby catalytic properties include catalyst activity, selectivity, activity and/or selectivity performance over time, operability (resistance to runaway), conversion and work rate (page 4, lines 8-15).

4. *Closest prior art*

4.1 D7 has been considered as the document describing the closest prior art by the Examining Division and by the appellants. The Board has no reason to take a different position, for the following reasons:

4.1.1 D7 (page 2, lines 3-5) relates to improved supported catalysts for the production of ethylene oxide from ethylene and oxygen, as does the present application.

4.1.2 D7 addresses the problem that for catalysts of this kind the activity and selectivity achievable cannot be maintained at their original level over long periods of operation (page 3, lines 11-15).

4.1.3 The problem of D7 corresponds to the problem mentioned in the application as filed of improving activity

and/or selectivity performance over time (page 4, lines 13-14).

4.1.4 Hence, D7 and the present application relate to the same technical field and address the same problem.

The disclosure of D7

4.2 D7 discloses catalysts for the production of ethylene oxide from ethylene and oxygen based on α -Al₂O₃ as the support material and silver as the active mass, characterised by the fact that the catalysts' content of alkali-soluble Si compounds is below 0.001 wt-% calculated as SiO₂ and relative to the total weight of the catalyst (Claim).

4.2.1 According to D7 (page 4, lines 1-6 and 21-22), alkali-soluble Si compounds - measured as SiO₂ and determined as Si compounds dissolving in an excess of 0.1 N NaOH within 10 minutes at 100°C - have an adverse effect on the catalytic properties, in contrast to poorly soluble silicates, which are harmless.

4.2.2 To achieve particularly long operating periods without large activity or selectivity penalties, D7 teaches to reduce the content of alkali-soluble Si compounds to below 0.001 wt.-%, calculated as SiO₂ and relative to the total weight of the catalysts (page 3, lines 20-27; page 4, lines 8-19).

4.2.3 Thus, D7 is about lowering the silicates that are alkali soluble and that are present in the α -Al₂O₃, material, in the bulk, hence on the surface thereof.

- 4.2.4 According to D7 (page 4, lines 8-26), the catalysts are prepared by using from the beginning Si-free chemicals for the active mass and pure $\alpha\text{-Al}_2\text{O}_3$ as the carrier material. If $\alpha\text{-Al}_2\text{O}_3$ with the required very low content of alkali soluble Si compounds is not commercially available, normal commercial $\alpha\text{-Al}_2\text{O}_3$ is boiled for 30 minutes with an approximately equal quantity of 1 wt.% NaOH and then is washed with water until a pH of 8, or normal commercial $\alpha\text{-Al}_2\text{O}_3$ is boiled for 10 minutes with an approximately equal quantity of 1 wt.% HF acid and then is washed with water until a pH of greater than 5.
- 4.2.5 D7 (page 8) also illustrates the treatment carried out on carriers T₂ and T₃ to reduce the alkali soluble silica below 1 ppm, e.g. 30 minutes with boiling 1 wt.% NaOH followed by washing with demineralised water until neutral pH, or 10 minutes with 1 wt.% HF followed by washing with demineralised water until neutral pH.
- 4.2.6 The process of Claim 1 of the present application is novel over that of D7, because the washing media defined in Claim 1 are not disclosed by D7.

5. *Problem and solution*

- 5.1 As regards the effects of the claimed subject-matter, the applicants have primarily drawn attention to Examples 8 and 10 of the application as filed, concerning carriers A and E, whereby E is formed from the same materials as carrier A but with water washing of the alumina source material, to show that washing the alumina source material provides a benefit. Since they consider that D7 does not disclose washing of the source material, this benefit is also attained over D7.

- 5.2 Further comparative test results were provided by the applicants with their letters dated 8 June 2004 and 25 March 2007. The comparative results of the former letter concern carrier washing, not washing of the source material. As regards Annexes I and II to the latter letter: Annex I dealt again with Examples 8 and 10 of the application as filed but the caesium target for both catalysts was the same. The washing of the raw material improved the selectivity by 1% but increased the temperature (i.e. reduced the activity) by 4°C. The comparative tests of Annex II dealt with two carriers F and G, made of the same materials, but the raw material for carrier G had been washed before the carrier was formed. According to Table V, the washing of both the raw material and the formed carrier reduced the loss of selectivity and the increase in temperature, i.e. the loss of activity.
- 5.3 The application as filed contains no comparative results over D7. Also the further tests submitted with the mentioned letters are not comparative over D7.
- 5.4 Only in their statement setting out the grounds of appeal, the applicants have provided results based on D7, as follows: the extraction process of D7 was applied to commercial catalyst support pellets made of normal commercial α -alumina, boiled for 30 minutes with an equal quantity of 1 wt.% NaOH solution, and then washed with deionised water. Finally the crush strength (test according to "Pfizer Hardness Tester: Crush Strength of Catalyst Pellets", issued 1969 (measured perpendicular to the pellet extrusion axis)) and the attrition index (according to ASTM method No. D4058-92)

of the resulting treated pellets were measured. The same crush strength and attrition tests were also run on the untreated pellets for comparison. The untreated pellets had a crush strength of 13.4 lbs and an attrition index of 13.9% weight loss, while the pellets treated in accordance with the D7 process had a crush strength of 8.2 lbs and an attrition index of 20.2% weight loss. The change in crush strength was held to be significant - from 13.4 to 8.2 lbs, a 38.8% relative decrease; the change in attrition index was held to be dramatic - from 13.9% to 20.2% weight loss, a 45.3 % relative increase. Both extracted physicals were held to reflect "failing grades" for commercial carriers' specifications, meaning that the procedures of D7 were too aggressive to be practical for commercial use.

5.5 As established in the Case Law of the Boards of Appeal of the EPO (6th edition 2010, I.D.4.4, particularly T 1188/00), this finding can be used to reformulate the problem to be solved only if

- (a) the results mentioned are comparative, i.e. the alleged effects are convincingly shown to have their origin in the feature distinguishing the claimed process from that of D7 (T 197/86, OJ EPO 1989, 371, Point 6.1.3 of the Reasons); and, in the affirmative, that
- (b) it is plausible that this effect is attained over the whole breadth of the claims; and, that
- (c) the alleged effects are derivable from the problem mentioned in the application as filed.

5.6 As regards the comparative nature of the test, a feature distinguishing the claimed subject-matter from the process disclosed by D7 (closest prior art) is the

use of washing media other than NaOH and HF, *inter alia* solutions of lithium carbonate, which are alkaline and can be considered as the closest alkaline solution to that of D7, for washing the material from which the carrier is made. However, in the test mentioned in the statement setting out the grounds of appeal the carriers as such have been treated with NaOH, and no comparison has been made with carriers or materials thereof treated with alkaline solutions such as lithium carbonate, so that no alleged maintenance of the physical integrity of the carrier after washing its source material with e.g. lithium carbonate can be gathered. Thus, these tests are not comparative over D7.

- 5.7 Furthermore, the alleged maintenance of crush strength and attrition index is not derivable from the problem mentioned in application as filed. Neither from the list of the "improvement in catalytic properties" given on page 4, lines 4-15, which does not mention them. Nor from Table I that only mentions the initial crush strength of the carrier.
- 5.8 Therefore, the problem to be solved cannot be reformulated to include the maintenance of the physical integrity measured by crush strength and attrition index as invoked by the applicants.
- 5.9 As regards the second formulation of the problem to be solved as proposed by the applicants (a process of preparation of catalyst with useful properties without using aggressive media), the application as filed does not describe what aggressive media dissolve the carrier, extract too much material from the bulk and generate acidic or basic sites in the pores of the carrier.

Moreover, the expression "without aggressive media" anticipates or hints at the solution, as the media defined in Claim 1 are considered to be less aggressive than those of D7 (Case Law, *supra*, I.D.4.3.1).

5.10 It follows from the foregoing analysis of the examples submitted that the only problem effectively solved by the claimed features over D7 is to provide a further process for the preparation of a catalyst for the epoxidation of an alkene with an oxygen gas.

6. *Character of the solution*

6.1 Although D7 illustrates in its examples a treatment carried out on the formed carrier, further passages of D7 specifically mention the α -Al₂O₃ material as such. It is referred to in the following passages: "auf der Basis von α -Al₂O₃ als Trägermaterial" ("based on α -Al₂O₃ as the support material") (Claim; page 3, lines 9 and 22) and "ein entsprechend reines α -Al₂O₃ einsetzt" ("a correspondingly pure α -Al₂O₃"), "derartige α -Al₂O₃-Qualität" (" α -Al₂O₃ of this quality"), "geht man von normalen handelsüblichem α -Al₂O₃ aus, kocht es 30 min ... " ("one starts with normal commercial α -Al₂O₃, boils it for 30 minutes ...") (page 4, lines 8-26). This disclosure makes it at least implicit that the treatment disclosed by D7 is similarly applicable to the source α -Al₂O₃ material.

6.2 This interpretation is in line with the teaching of D7 that the harmful material generally identified as "alkali soluble silica" present in the α -Al₂O₃ can be reduced or removed either by solubilisation in alkaline solution (this property is implicit in the first part

of the definition, "alkali soluble") or by attack with HF (because of its being "silica", second part of the definition). Therefore, the general teaching of D7 cannot be reduced to the treatment of the formed carrier with NaOH as illustrated.

6.3 Since the disclosure "alkali soluble" of D7 definitely implies a solubility in alkaline media of the harmful silica present on $\alpha\text{-Al}_2\text{O}_3$, and since Claim 1 of the Main Request encompasses a washing treatment of the material from which the carrier is made, i.e. $\alpha\text{-Al}_2\text{O}_3$, with an aqueous solution of lithium carbonate, which is an alkaline medium, the claimed process is nothing more than the possibility of using another alkaline solution within a further process of preparation over D7.

6.4 The fact that lithium carbonate is less alkaline than NaOH, hence that it can be disadvantageous, is not decisive, since the skilled person is merely looking for a further process of preparation of the catalyst.

6.5 The process of Claim 1 was obvious and lacks an inventive step over D7 (Article 56 EPC).

6.6 Therefore, the Main Request is not acceptable.

First Auxiliary Request

7. *Amendments*

8. Compared with Claim 1 of the Main Request, Claim 1 of the First Auxiliary Request comprises the further limitations "from 1 to 40 wt% of silver, basis the weight of the total catalyst" and "one or more

promoters selected from Group IA metals in an amount in the range of from 10 ppm to 1500 ppm, and Group VIIb metals in an amount less than 3600 ppm, each amount expressed as the metal and by weight of the total catalyst".

- 8.1 The amendments are clear (Article 84 EPC) and based on the application as filed (page 9, lines 22-25; page 9, line 30 to page 10, line 28) (Article 123(2) EPC), so that also the First Auxiliary Request is admissible.

Inventive step

9. D7 (page 5, lines 23-25) discloses a process for preparing a catalyst having from 2 to 12 wt.% Ag and containing up to 0.035 wt.% of Li, Na and/or K, as well as up to 0.025 wt.% of Cs, as promoters, all of the promoters belonging to Group IA.
- 9.1 Since D7 also discloses the further features of Claim 1 of the First Auxiliary Request, the conclusion reached on the Main Request applies mutatis mutandis to the claimed subject-matter of the First Auxiliary Request, i.e. there is still a lack of inventive step over D7.
- 9.2 Therefore, the First Auxiliary Request is not acceptable either.

Second Auxiliary Request

10. *Amendments*

10.1 Claim 1 of the Second Auxiliary Request (Point VIII, *supra*), compared with Claim 1 of the First Auxiliary Request, includes the further feature of "submerging the carrier in a silver impregnation solution containing a metal hydroxide".

10.2 This further feature is based on Claim 8 of the application as filed (i.e. "submerging the carrier in an impregnation solution of lowered hydrogen ion activity) and on its limitation as given on page 7, lines 7-9, of the application as filed (the use of a metal hydroxide for lowering the hydrogen ion activity).

10.3 Hence, the Second Auxiliary Request also is clearly based on the application as filed (Article 123(2) EPC), and is consequently admissible.

11. *Remittal*

The applicants have requested from the beginning of the appeal to remit the case to the Examining Division for continuation of the examination proceedings.

The ground for refusal of the Fourth Auxiliary Request underlying the decision under appeal was the lack of inventive step having regard to D7. Claims 1 to 16 of Auxiliary Request 2 filed at the oral proceedings before the Board contain further limiting features over the Fourth Auxiliary Request underlying the decision under appeal, *inter alia* the use of metal hydroxide in

the impregnation solution, which has been taken from the description and is not disclosed by D7. Hence, the amended claims of the Second Auxiliary Request define a new combination of features, not disclosed by D7, and not yet examined by the examining division, which thus lies outside any review of the decision under appeal.

Since the limiting feature relating to the use of metal hydroxide in the impregnation solution has been taken from the description, it is also not apparent whether a further search is necessary.

It follows from the above that an examination and possibly also a search of the new subject-matter have to be carried out by the Examining Division.

Therefore, the Board, in the exercise of its discretion under Article 111(1) EPC, considers it appropriate to remit the case to the Examining Division.

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 on the basis of Auxiliary Request 2 filed during the oral proceedings before the Board.

The Registrar:

The Chairman:

S. Fabiani

J. Riolo