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# Datasheet for the decision of 30 January 2009

**Case Number:** T 1260/05 - 3.3.07

**Application Number:** 00974739.5

**Publication Number:** 1237652

**IPC:** B01J 23/75

**Language of the proceedings:** EN

**Title of invention:**
Cobalt catalysts

**Applicant:**
Sasol Technology (Proprietary) Limited

**Opponent:**
-

**Headword:**
-

**Relevant legal provisions:**
EPC Art. 83, 84, 123(2)

**Relevant legal provisions (EPC 1973):**
-

**Keyword:**
"Sufficiency (no) - all requests"
"Clarity (no) - all requests"

**Decisions cited:**
-

**Catchword:**
-

**Case Number:** T 1260/05 - 3.3.07

# DECISION
## of the Technical Board of Appeal 3.3.07 of 30 January 2009

**Appellant:**

Sasol Technology (Proprietary) Limited
1 Sturdee Avenue
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Transvaal (ZA)

**Representative:**

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**Decision under appeal:**

**Decision of the Examining Division of the European Patent Office posted 26 April 2005 refusing European application No. 00974739.5 pursuant to Article 97(1) EPC.**

**Composition of the Board:**

**Chairman:** S. Perryman
**Members:** B. ter Laan
F. Rousseau

## Summary of Facts and Submissions

I. European patent application No. 00 974 739.5, originating from international application PCT/IB00/01745 (published as WO-A-01/39882) and having the international filing date of 24 November 2000, was refused by a decision of the Examining Division of the European Patent Office issued in writing on 26 April 2005. The application as filed comprised 16 claims, the independent claims reading as follows:

"1. A cobalt catalyst precursor which includes a catalyst support impregnated with cobalt, with all reducible cobalt being present in the support as supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b \geq 0$."

"5. A process for preparing a cobalt catalyst precursor, which process includes

in a support impregnation stage, impregnating a particulate porous catalyst support with a cobalt salt, and partially drying the impregnated support; and

in a calcination stage, calcining the partially dried impregnated support to obtain the cobalt catalyst precursor, with the calcination being effected at calcination conditions selected so that all reducible cobalt is present in the support as a supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b \geq 0$."

"6. A process for preparing a cobalt catalyst, which process includes

in a support impregnation stage, impregnating a particulate porous catalyst support with a cobalt salt, and partially drying the impregnated support;

in a calcination stage, calcining the partially dried impregnated support to obtain a cobalt catalyst precursor, with the calcination being effected at calcination conditions selected so that all reducible cobalt is present in the support as a supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b \geq 0$; and

in a reduction stage, reducing the cobalt catalyst precursor, to obtain the cobalt catalyst."

II. The decision under appeal was based on three sets of 15 claims as the main and two auxiliary requests filed on 12 April 2005 during the oral proceedings before the examining division.

The independent claims of the (then) main request read as follows:

"1. A Fischer-Tropsch cobalt catalyst precursor which includes a catalyst support impregnated with cobalt nitrate $Co(NO_3)_2.6H_2O$, wherein the catalyst support is selected form alumina ($Al_2O_3$), silica ($SiO_2$), titania ($TiO_2$), magnesia (MgO) and silica-alumina, and with all reducible cobalt after calcination being present in the support as supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b > 0$."

"5. A process for preparing a Fischer-Tropsch cobalt catalyst precursor, which process includes

in a support impregnation stage, impregnating a particulate porous catalyst support with a cobalt salt, and partially drying the impregnated support, with the resultant partially dried impregnated support containing residual moisture; and

in a calcination stage, calcining the partially dried impregnated support to obtain a cobalt catalyst precursor, with the calcination being effected in air between 95°C and 400°C, and by using an air space velocity and a support heating rate such that all reducible cobalt is present in the support of the precursor as a supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1{,}7$ and $b>0$."

Claim 1 of the (then) first auxiliary request read:

"1. A Fischer-Tropsch cobalt catalyst precursor which includes a catalyst support impregnated with cobalt nitrate $Co(NO_3)_2.6H_2O$, wherein the catalyst support is selected form alumina ($Al_2O_3$), silica ($SiO_2$), titania ($TiO_2$), magnesia (MgO) and silica-alumina, and with all reducible cobalt after calcination in air between 95°C and 400°C, and by using an air space velocity and a support heating rate is present in the support of the precursor as a supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1{,}7$ and $b>0$." (sic)

Claim 1 of the (then) second auxiliary request read:

"1. A Fischer-Tropsch cobalt catalyst obtainable by reduction of a cobalt catalyst precursor which includes a catalyst support impregnated with cobalt nitrate $Co(NO_3)_2.6H_2O$, wherein the catalyst support is selected from alumina ($Al_2O_3$), silica ($SiO_2$), titania ($TiO_2$), magnesia (MgO) and silica-alumina, and wherein in air between 95°C and 400°C, and by using an air space velocity and a heating rate such that all reducible cobalt is present in the support of the precursor as a supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b>0$."

III. The following documents were cited in the decision:

| | |
|---|---|
| D1 | WO-A-99 422214, |
| D2 | EP-A-0 736 326, |
| D3 | WO-A-99 34917, |
| D4 | Appl. Catal. A: General, 233 (2002) 263-281 |
| D5 | Handbook of Heterogeneous Catalysis, Ertl et al., chapter 3.2.2.5 |
| D6 | US-A-4 729 981 and |
| D7 | EP-A-0 494 528. |

According to the decision, the claimed subject-matter lacked clarity (main request and auxiliary request I) as well as novelty over D2 and D6 (all requests).

Both D2 and D6 disclosed Fischer-Tropsch cobalt catalyst precursors without indicating the formula-unit. According to the general teaching of the application the formula-unit would only depend on the calcination conditions used, which should be mild in order not to obtain the undesired $Co_3O_4$ spinel structure. In both D2 (examples 60 and 63) and D6 (example 6 and column 5, lines 34 to 63), calcination was carried out in a mild way under conditions very similar to those in the application. In particular, the same calcination temperature was disclosed and the air space velocity could be calculated. There was no evidence that at the air space velocities calculated from D2 and D6 a precursor of any other structure than the claimed one would be produced. Therefore, the cobalt catalyst precursor of both D2 and D6 would have a structure falling within the required formula-unit.

The examining division pointed out that no prior art examples had been repeated and the structure of the precursor assessed. Also, there was not sufficient evidence to prove that the "undesired" area indicated in Figure 6 of the application was directly linked to a formula-unit outside the one required. Furthermore, in view of the calcination conditions, a limited amount of residual nitrates could also not distinguish the claimed precursor from those of D2 and D6. Since the formula-unit constituted an unusual parameter, the burden of proof to establish that the formula-unit distinguished the claimed subject-matter from the prior art, was with the applicant.

As to clarity, contrary to the wording of claim 1 according to which all cobalt of the precursor should fall within the formula-unit, the description made it clear that the precursor did allow for the presence of a spinel structure as well.

IV. On 23 June 2005 a Notice of Appeal was lodged against that decision, together with payment of the prescribed fee. The statement setting out the grounds of the appeal was filed on 2 September 2005, together with a new set of 15 claims as the main request and further experimental evidence.

After a communication from the Board in preparation of the oral proceedings, in which several problems under Articles 123(2), 84 and 54 EPC were addressed, the appellants, with a letter dated 23 January 2009, filed six new sets of 16, 16, 12, 12, 11 and 6 claims resp. as the main and five auxiliary requests.

V. At the oral proceedings before the Board, held on 30 January 2009, after extensive consideration of the requests and of several objections raised under Articles 123(2), 84 and 54 EPC, further requests replacing the ones then on file were successively submitted. This decision is based on the three last requests filed during the oral proceedings as main request and auxiliary requests I and II.

The independent claims of the main request read as follows:

"1. A Fischer-Tropsch cobalt catalyst precursor which includes a catalyst support impregnated with cobalt, wherein the catalyst support is selected from alumina ($Al_2O_3$), silica ($SiO_2$), titania ($TiO_2$), magnesia (MgO) and silica-alumina, with all reducible cobalt being present in the support as supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b > 0$."

"5. A process for preparing a Fischer-Tropsch cobalt catalyst precursor, which process includes

in a support impregnation stage, impregnating a particulate porous catalyst support with a cobalt nitrate, and partially drying the impregnated support; and

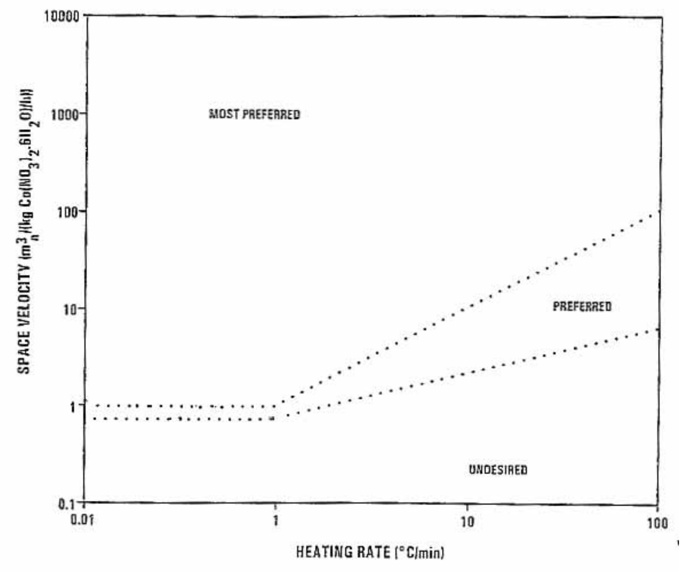
in a calcination stage, calcining the partially dried impregnated support to obtain the cobalt catalyst precursor, with the calcination being effected in air, at a maximum calcination temperatures [sic] between 200°C and 300°C and at calcination conditions selected so that all reducible cobalt is present in the support as a supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b > 0$."

The independent claims of auxiliary request I read:

"1. A Fischer-Tropsch cobalt catalyst precursor which includes a catalyst support impregnated with cobalt, wherein the catalyst support is selected from alumina ($Al_2O_3$), silica ($SiO_2$), titania ($TiO_2$), magnesia (MgO) and silica-alumina, with all reducible cobalt being present in the support as supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b > O$ and wherein all the reducible cobalt is present as $Co_20_3.H_20$ or CoO(OH) or wherein all the reducible cobalt is present as a mixture of $Co_3O_4$ and CoO(OH) or $Co_2O_3.H_2O$."

"5. A process for preparing a Fischer-Tropsch cobalt catalyst precursor, which process includes in a support impregnation stage, impregnating a particulate porous catalyst support with a cobalt nitrate, and partially drying the impregnated support; and

in a calcination stage, calcining the partially dried impregnated support to obtain the cobalt catalyst precursor, with the calcination being effected in air, at a maximum calcination temperatures [sic] between 200°C and 300°C and at calcination conditions of air space velocity and heating rate during fluidized bed calcination in the preferred or most preferred regions of the following diagram



so that all reducible cobalt is present in the support as a supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b > 0$."

Claim 1 of auxiliary request II reads as follows:

"1. A Fischer-Tropsch cobalt catalyst precursor consisting of a catalyst support impregnated with cobalt, wherein the catalyst support is selected from alumina ($Al_2O_3$), silica ($SiO_2$), titania ($TiO_2$), magnesia (MgO) and silica-alumina, optionally palladium, platinum, ruthenium or mixtures thereof, the mass proportion of the palladium, platinum, or ruthenium metal, or the combined mixtures of such metals when such a mixture is used, to the cobalt metal is between 0,01 : 100 to 0,3 :100, with all reducible cobalt being present in the support as supported cobalt oxide of formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b > O$ and wherein all the reducible cobalt is present as $Co_2O_3.H_2O$ or CoO(OH) or wherein all the reducible cobalt is present as a mixture of $Co_3O_4$ and CoO(OH) or $Co_2O_3.H_2O$."

VI. The appellants' arguments submitted in writing and during the oral proceedings can be summarised as follows:

(a) The inventors had found that the kind of cobalt oxide produced during calcination influenced the activity of the catalyst, which was surprising because in a later stage of catalyst preparation the cobalt was reduced to the metal. The formula-unit reflecting the desired form of cobalt oxide did not mean that all the cobalt should fulfil it but rather indicated the average of the cobalt structures present in the precursor. The gist of the invention was, that on average some hydrogen should be present in the cobalt compound; however, by the use of the term "cobalt oxide", $Co(OH)_2$ (cobalt hydroxide) was excluded from the claim.

(b) In order to arrive at the desired structure, special calcination conditions had to be applied, in particular the heating rate and the air space velocity, as reflected in Figure 6. As could be seen from the further diagram submitted during the oral proceedings, it meant that the heating rate should be reduced as soon as the nitrate started to decompose and it could be increased again after the nitrate had decomposed. That procedure could be deduced from the original application, page 7, and Figures 7 and 8 with their description on page 9.

(c) The appellants had submitted sufficient evidence (such as Annexes A and B to the statement of grounds for the appeal) to show that the calcination conditions in the regions indicated as "preferred" and "most preferred" in Figure 6 inevitably resulted in a cobalt precursor having the required structure, whereas calcination conditions outside those regions gave a precursor structure that did not comply with the formula-unit.

(d) Whether the precursor complied with the formula-unit of claim 1 could be established by using TPR (temperature programmed reduction), which gave a reduction pattern of the precursor as a function of the temperature. The different components of the precursor showed peaks at different temperatures, according to their relative hydrogen consumption per reduction step, which could be calculated from the areas under the peaks, as indicated in table 7 of the application in suit.

TPR could distinguish between pure $Co_3O_4$ and cobalt oxide mixtures as claimed and would also reveal the presence of $Co(OH)_2$. The relation between the surface area of the various peaks would remain the same whichever reaction conditions were used. Therefore, the distinction made on the basis of TPR remained valid independent of the reaction conditions used.

(e) The feature distinguishing the claimed subject-matter from D2 was the formula-unit of the cobalt oxide. The appellants had provided evidence of the fact that if the calcination procedure was performed in the range indicated as "undesired" in Figure 6 of the application, no cobalt oxide of the required formula was formed. Such was the case in D2, which therefore did not disclose the cobalt oxide of the claimed structure. The examining division had stated that the burden of proof that D2 did not disclose the required structure lay with the (then) applicant. However, the formula-unit was neither a parameter, nor was the formula unusual. It was a normal chemical formula and there was no other way to characterise the claimed cobalt oxide mixtures. Since on the one hand there was no evidence that the calcination procedure of D2 resulted inevitably in the claimed cobalt oxide, and on the other the appellants had provided evidence that the calcination conditions of D2 resulted in a structure outside the claimed one, D2 did not damage the novelty of the claimed subject-matter.

(f) D6 disclosed a mixed cerium-cobalt oxide, not a cobalt oxide. The other documents were even further away, so that the claimed subject-matter was novel.

VII. The appellants requested that the decision under appeal be set aside and that a patent be granted on the basis of the main request filed on 30 January 2009 during the oral proceedings or, alternatively, on any one of the two auxiliary requests filed at the same time.

## Reasons for the Decision

1. The appeal is admissible.

*Main request*

Clarity and support

2. According to claim 1 of all the requests, all reducible cobalt should be present in the support as supported cobalt oxide of the formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b > 0$. According to the appellants, the formula is the feature that makes the difference between the claimed subject-matter and the prior art, in particular D2. In order to enable the skilled person to decide whether a catalyst belongs to the prior art or falls under the present claims, the distinguishing feature itself as well as the method(s) to measure it should be clear.

2.1 According to the description (page 2, lines 4 to 18) as well as the appellants' explanations during the oral proceedings the requirement that all reducible cobalt should comply with the formula-unit should be interpreted so as to mean that the cobalt oxide may be present in more than one form, as long as the total falls, **on average**, within the formula-unit. As part of the reducible cobalt may therefore be present also in another form than CoO(OH), the question arises how it can be established whether the cobalt oxide of a catalyst fulfils the requirement of falling, on average, within the formula-unit.

It appears that in fact an important part of the cobalt oxide may be outside the required formula, as long as for the cobalt oxide as a whole the average composition falls within the formula. Specific mixtures of reducible cobalt in the form of 45% $Co_3O_4$ with 55% CoO(OH) or $Co_2O_3.H_2O$, which would result in a=1,7 and b=0,55, and 60% $Co_2O_3$ with 40% CoO(OH) or $Co_2O_3.H_2O$, resulting in a=1,7 and b=0,4, are mentioned.

2.2 According to the description, the presence of the supported cobalt oxide phase with the formula-unit $CoO_aH_b$, where $a \geq 1,7$ and $b > 0$, may be determined by using Temperature Programmed Reduction (TPR) as a fingerprint technique (page 5, lines 12 to 15). Four TPR patterns are given: Figures 2 and 9 representing catalysts H and Q, respectively, both according to the formula-unit, and Figures 3 and 10 representing comparative catalysts P and R (description page 16, lines 11 to 14, and page 21, lines 6 to 13).

2.2.1 The TPR patterns of Figures 2 and 3 show multiple peaks, the difference between the two figures being ***inter alia*** the ratio of the peak heights of peak 2 and peak 4, the width of peak 2 at half height and the presence/absence of peak 3, which is situated at a temperature on the X-axis of about 280°C. Figure 2 is said to be typical for a catalyst within the formula-unit, whereas Figure 3 displays the presence of the undesired $Co_3O_4$ spinel phase (description, page 16, lines 18 to 28). From the figures it can also be seen that the peak indicated as "peak 4" in Figure 3 has shifted towards a higher temperature as compared to Figure 2.

2.2.2 Figures 9 and 10 are referred to as TPR patterns in example 7 (description page 21). However as regards both figures, the horizontal axis is not labelled at all, whereas the right hand vertical axis is labelled as T[°C]. The left-hand vertical axis is labelled "TCD signal" but no units are indicated. No meaning is given to the dotted lines shown. It is not clear how to relate any temperature to the various peaks. In the absence of any indication of units for the "TCD signal" the two patterns cannot be compared quantitatively. A qualitative comparison shows a peak 1 in Figure 9 that is not present in Figure 10.

2.2.3 There is no specific indication which peak refers to which form of the cobalt oxide. In particular it is not elucidated which role any differences in the TPR patterns play in establishing the presence or absence of distinguishing features over the prior art, which elucidation would be necessary for an understanding of what the claims require. According to the differences said to exist between a catalyst according to the invention as illustrated in Figure 2, and a catalyst having the undesired $Co_3O_4$ spinel phase said to be illustrated in Figure 3 (page 16, lines 19 to 28), it seems to be suggested that the presence of any $Co_3O_4$ is undesired and that the catalyst according to the formula-unit has no $Co_3O_4$ present. However, that is in clear contradiction to the examples given on page 2 (see point 2.1.1 above). In example 7, Table 7 shows three reactions without any indication which reaction is related to which of the peaks described in Figures 9 and 10. The appellants, in writing (letter dated 23 March 2005) as well as during the oral proceedings, explained that peak 1 was related to the reduction of CoO(OH), peak 2 to the reduction of $Co_3O_4$ and peak 4 to the reduction of CoO. The meaning of peak 3, present in Figure 2, is however not given.

2.2.4 The description does not contain any indication how the values of a and b should be calculated from those TPR patterns. First, the patterns of Figures 2 and 3 have a different look from those of Figures 9 and 10 and it is not clear if the patterns of the latter have perhaps been smoothed by some mathematical method, thus introducing changes in the peaks actually used for the calculation. Secondly, no method is indicated how the surface areas under the peaks are to be calculated. During the oral proceedings the appellants stated that that is, as a standard procedure, done with the help of computer programs. However, no such programs have been specified. Methods to calculate the surface area under peaks are by their nature inexact, certainly in case of small shoulders and vague peaks such as peak 1 in Figures 2, 3 and 9 and peak 3 in Figure 2. In such cases, if the result of that calculation defines the difference between the claimed subject-matter and the prior art (as the appellants argued having regard to D2), a precise indication of the method used and the margins of error that are to be tolerated would be essential.

2.2.5 In addition, the question asked by the Board regarding the measurement of the mixture of CoO(OH) with $Co_2O_3$, also indicated on description page 2 (lines 4 to 18), remained unanswered; in fact, it was stated that that mixture did not exist and was based on an error, which comment is however not in accordance with Example 10, where the presence of $Co_2O_3$ is again mentioned. Furthermore, when asked how a possible presence of $Co(OH)_2$ was taken into account, it was stated that that compound was irrelevant for the formula-unit as cobalt hydroxide was not a cobalt oxide and the claims referred to cobalt oxide only. However, if the formula-unit indicates the average of a mixture of compounds and if compounds not falling within the formula-unit are allowed to be present, it is not clear how and on what basis some compounds falling within the formula-unit should be excluded.

2.2.6 In the present case, not only is any precise information as regards measurement methods and error margins lacking, but also there are numerous unclarities regarding the meaning of the various peaks in the TPR patterns given (peak 3 in Figure 2) and how to deal with the presence or absence of $Co(OH)_2$) and other compounds mentioned in the description. In particular, it appears impossible to establish whether a catalyst fulfils the requirements of present claim 1 so that the clarity requirement of Article 84 EPC cannot be regarded as complied with.

3. Regarding Article 83 EPC, the appellant referred to the description page 7, lines 22 to 25, which reads: "...it is believed that enhanced activity may be obtainable if the heating rate is non-linear in order to tailor the release profiles of the nitrogen oxide(s) and water". The appellant explained that that passage meant that the rate of temperature increase during calcination should be reduced as soon as the nitrate started to decompose and the rate of temperature increase could be increased again after the nitrate decomposition had ended. During the oral proceedings a drawing illustrating that process was submitted. However, the vague and general terms of the relevant passage of the description provide no basis for such an interpretation. As there are discrepancies between Figures 7 and 8 and the explanations relating thereto on page 9 of the description, those figures cannot be taken into account for support of the appellant's arguments. Figure 6 shows the catalyst productivity as a function of the air space velocity and the heating rate during fluidized bed calcination and has no bearing on the time effect. Therefore, there is no guidance for the skilled person how to produce a catalyst complying with the formula-unit and how to change the process if a catalyst should be obtained not fulfilling the formula-unit. Thus, the requirements of Article 83 EPC are not complied with.

### *Auxiliary Request I*

4. In Claim 1 of auxiliary request I the cobalt oxide has been restricted to a mixture of $Co_3O_4$ and CoO(OH) or $Co_2O_3.H_2O$. However, the objections under Article 84 EPC set out in points 2.1 and 2.2 (except point 2.2.5)

above, as well as the objections under Article 83 EPC still apply.

5. In claim 5 the calcination conditions are now specified by incorporation of the diagram of Figure 6. The process for preparing a cobalt catalyst should now be carried out under conditions such that the heating rate and air space velocity during calcination fall within the preferred or most preferred regions indicated in that diagram. The borders of those regions are indicated by dotted lines the exact value of which cannot however be established since the values on the y-axis are not precisely given and the diagram is too small to carry out any measurements with an exact outcome. In the borderline regions it is therefore not possible to know if one works inside or outside the claimed process. In order to comply with Article 84 EPC, the limits of a claim should be clearly indicated. The uncertainty introduced into claim 5 by the reference to Figure 6 is too great for the requirement of Article 84 EPC that the claims be clear to be regarded as met.

6. For those reasons, auxiliary request I cannot be allowed.

*Auxiliary Request II*

7. Claim 1 of auxiliary request II now requires a catalyst precursor "consisting of" a support impregnated with cobalt, while referring to other metals which may optionally be present. The amendments made leave this claim open to the same objections under Articles 84 and 83 EPC as arise for the subject-matter of the claims 1 of the previous requests. In addition the simultaneous

introduction of the words "consisting of a support impregnated with cobalt" and the reference to optional further metal components seems self contradictory and thus objectionable under Article 84 EPC. Moreover, as there is no disclosure in the application as originally filed of a catalyst precursor only consisting of the defined substances and nothing else, claim 1 is objectionable under Article 123(2) EPC as well. For instance, an amount of nitrogen left from the cobalt nitrate with which the support has been impregnated, may still be present (original page 6, lines 4 to 6).

8. For the above reasons, none of the requests fulfils the requirements of the EPC, so that none of the requests is allowable.

## Order

## For these reasons it is decided that:

The appeal is dismissed.

Registrar Chairman

C. Eickhoff S. Perryman