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**Datasheet for the decision
of 31 January 2012**

Case Number: T 0688/08 - 3.3.07

Application Number: 01918662.6

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Language of the proceedings: EN

Title of invention:

Highly active Fischer-Tropsch catalyst comprising doped,
thermally stable catalyst support

Applicants:

Sasol Technology (UK) Limited

Headword:

-

Relevant legal provisions:

EPC Art. 123(2), 54, 56

Keyword:

"Main and Auxiliary Requests 1 to 11 not allowable - added
subject-matter"

"Auxiliary Requests 12 and 13 - inventive step (no) -
improvement not shown over the whole breadth of claim -
obvious embodiment"

"Auxiliary Request 14 - Amendments allowable - novelty (yes) -
inventive step (yes) - improvement not obvious"

Decisions cited:

-

Catchword:

-



Case Number: T 0688/08 - 3.3.07

DECISION
of the Technical Board of Appeal 3.3.07
of 31 January 2012

Appellants: Sasol Technology (UK) Limited
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 16 November 2007
refusing European patent application
No. 01918662.6 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 16 November 2007 refusing European patent application No. 01 918 662.6, filed as international application No. PCT/US01/08155 on 14 March 2001 and claiming priority from US application 09/528 163 filed on 17 March 2000.
- II. The decision of the Examining Division was based on the sets of claims according to the then pending Main and Auxiliary Requests submitted on 16 August 2007 and during the oral proceedings on 11 October 2007, respectively. The respective independent claims 1 of those requests read as follows:

Main Request

"1. A catalyst comprising: a γ -alumina support having an internal structure including a dopant selected from the group consisting of a lanthanum oxide dopant, a barium oxide dopant, and combinations thereof and an amount of cobalt, on said γ -alumina support, effective for hydrocarbon synthesis in a slurry bubble column-type reaction system, wherein said dopant is present in said internal structure of said γ -alumina support in an amount effective for increasing the thermal stability of said catalyst for use in said slurry bubble column-type reaction system without reducing the activity of said catalyst for said hydrocarbon synthesis wherein said amount of said dopant present in said support is in the range of from about 1% to about

5% by weight based on the total weight of said γ -alumina support".

Auxiliary Request

Claim 1 of the Auxiliary Request corresponded to that of the Main Request in which the support was defined to include at least 500 ppm of titania, expressed as elemental titanium, the catalyst was defined to contain at least one promoter and the term "about" had been deleted at each occurrence.

III. According to the impugned decision, the subject-matter of the Main and Auxiliary Requests was novel but lacked an inventive step. D1 (WO-A-99/61550) constituted the closest prior art, as it also related to cobalt catalysts on γ -alumina having activity for Fischer-Tropsch synthesis conducted in a slurry bubble column and other three-phase type reactors. D1 also taught to use lanthanum or barium oxide as dopant by a process of co-hydrolyzing an aluminium alkoxide with a lanthanum or barium alkoxide. Concerning the Main Request, the Applicants' argument that the inventive effect was due to lanthanum or barium providing high thermal stability without sacrificing other desirable properties, whether titania was present or not, was not accepted. It was held in particular that no effect had been established for the stabilizing effect of dopants on γ -alumina at very high temperatures generally. Moreover, the teaching in D1 was sufficient incentive for the skilled person to add lanthanum or barium to the γ -alumina support in any case. Having regard to the choice of a level of lanthanum or barium oxide of 1 to 5% by weight, the selection of this range was not associated with a

particular technical effect and therefore did not involve any inventive step. The Main Request was therefore not allowable. Concerning the Auxiliary Request, the starting point for assessing inventive step was the ruthenium-promoted cobalt catalyst on γ -alumina described in example 4 of D1. Said ruthenium-promoted cobalt catalyst, the γ -alumina support of which also contained titanium in an amount of 1000 ppm, was disclosed to exhibit improved activity compared to corresponding non-promoted catalysts. Hence, the skilled person would readily consider the further step of including lanthanum or barium in the support of said ruthenium-promoted cobalt catalyst. Moreover, no effect had been established for the use of barium oxide or for the choice of a level of lanthanum or barium oxide in the range of 1 to 5% by weight, which was considered to be arbitrary. Hence, claim 1 of the Auxiliary Request also lacked an inventive step. The decision provided the indication in an obiter dictum that the claimed subject-matter according to the Main Request also appeared to lack an inventive step over D2 (US-A-5 939 350).

- IV. With their statement setting out the grounds of appeal dated 26 March 2008, the Applicants (hereinafter the Appellants) submitted sixteen revised sets of claims.

- V. In a communication dated 3 August 2011 sent in preparation of the oral proceedings the Board addressed the Appellants' argument that the expression "internal structure" indicated that the dopant lanthanum or barium oxide was "integral to the molecular structure of the support", but not merely impregnated into it. The Board pointed out that the intended meaning for

this expression appeared to be reflected only in the then pending Auxiliary Requests 1, 5, 9 and 13, where the support was defined to be obtainable by adding a dopant alkoxide to, and co-hydrolyzing said dopant alkoxide with, aluminium alkoxide. Those requests, however, lacked either novelty over D6 (US-A-6 100 304) or an inventive step over D1. D6 was a prior application originating from the same applicants, which deprived the claimed priority document of the present application from being the first application within the meaning of Article 87(4) EPC to disclose the catalysts according to claim 1 of Auxiliary Request 1 or 9. As it had been published before the present date of filing, D6 was therefore novelty destroying for claim 1 of Auxiliary Requests 1 and 9. Furthermore, the expression "internal structure" alone was not appropriate to provide any distinguishing feature over catalysts obtained by incipient wetness of a γ -alumina support with solution comprising salts of lanthanum. The Board's communication also referred to D7 (WO-A-00/71253) published before the filing date of the present application.

- VI. The Appellants submitted on 24 October 2011 in response to the Board's objections new sets of claims as their Main and Auxiliary Requests 1 to 15 replacing those then on file. They also submitted on 4 November 2011 a copy of the document acknowledged on page 5 of the present application. They disputed that said document provided any teaching in respect of increasing thermal stability of γ -alumina by incorporation of lanthanum or barium, as might be suggested by the text of the application as filed addressing this prior art.

VII. With a Board's communication of 18 November 2011, attention of the Appellants was drawn to the fact that the claims according to Auxiliary Requests 12 to 15 might not enjoy priority from US patent application 09/528 163. In addition, the Appellants were invited to indicate which preparation method had been used for the synthesis of the doped alumina supports of catalysts 4 and 5 described in the experimental section of the present application. The following documents dealing with the thermal stabilization of γ -alumina with barium or lanthanum oxide were also communicated to the Appellants:

D8: US-A-5 134 107

D9: C. Narula *et al*, J. Mater. Chem., 1997, 7(9), pages 1821-1829 and

D10: H. Arai and M. Machida, Catalysis Today, 10 (1991), pages 81-94.

VIII. The Appellants with letter of 13 January 2012 submitted arguments in support of an inventive step over D7 of the subject-matter according to Auxiliary Requests 12 to 15. They also indicated that the doped alumina supports of catalysts 4 and 5 described in the experimental section of the application had been prepared by adding a dopant alkoxide to, and co-hydrolyzing the dopant alkoxide with, aluminium oxide. A corrected version of Auxiliary Requests 6 and 14 was also submitted.

IX. Oral proceedings took place on 31 January 2012 in the course of which Auxiliary Request 14 was amended in order to overcome objections under Article 84 EPC.

X. The respective independent claims 1 of the present requests are indicated below:

Main Request

Claim 1 of the Main Request corresponds to that of the Main Request underlying the impugned decision in which the catalyst support has been specified to be obtainable by adding a dopant alkoxide to, and co-hydrolyzing said dopant alkoxide with, aluminium alkoxide, the catalyst is defined to contain at least one noble metal promoter and the two occurrences of the term "about" have been deleted. It reads now:

"1. A catalyst comprising: a γ -alumina support having an internal structure including a dopant selected from the group consisting of a lanthanum oxide dopant, a barium oxide dopant, and combinations thereof and an amount of cobalt, on said γ -alumina support, effective for hydrocarbon synthesis in a slurry bubble column-type reaction system, wherein said dopant is present in said internal structure of said γ -alumina support in an amount effective for increasing the thermal stability of said catalyst for use in said slurry bubble column-type reaction system without reducing the activity of said catalyst for said hydrocarbon synthesis wherein said amount of said dopant present in said support is in the range of from 1% to 5% by weight based on the total weight of said γ -alumina support, wherein said γ -alumina support having an internal structure including a dopant is obtainable by adding a dopant alkoxide to and co-hydrolyzing said dopant alkoxide with aluminum alkoxide, and wherein

said catalyst contains at least one noble metal promoter."

Auxiliary Requests 1 to 3

Claim 1 according to Auxiliary Request 1 differs from that of the Main request in that the amount of dopant has been restricted to 2% to 3% by weight. Claim 1 according to Auxiliary Request 2 differs from that of the Main request in that the dopant is now required to be a lanthanum oxide dopant. Claim 1 of Auxiliary Request 3 corresponds to claim 1 of the Main Request in which both limitations defined above in respect of Auxiliary Requests 1 and 2 have been introduced.

Auxiliary Request 4

Claim 1 of Auxiliary Request 4 reads:

"1. A catalyst comprising: a γ -alumina support; an amount of cobalt, supported on said γ -alumina support, effective for said Fischer-Tropsch hydrocarbon synthesis in said Fischer-Tropsch reaction system; and at least one noble metal promoter on said γ -alumina support with said cobalt, said γ -alumina support having an internal structure comprising γ -alumina, at least 500 ppm of titania, expressed as elemental titanium and based on the total weight of the γ -alumina support, and a controlled amount of a dopant selected from the group consisting of a lanthanum oxide dopant, a barium oxide dopant, and combinations thereof, wherein said controlled amount of said dopant is an amount effective for increasing both the activity

and the thermal stability of said catalyst for said Fischer-Tropsch hydrocarbon synthesis in said Fischer-Tropsch reaction system, wherein said controlled amount of said dopant present in said internal structure of said γ -alumina support is in the range of from 1% to 5% by weight based on the total weight of said γ -alumina support, and wherein said γ -alumina support is produced from aluminum alkoxide which is hydrolyzed to produce an alumina product and wherein said dopant is incorporated in said γ -alumina support by adding a dopant alkoxide to, and co-hydrolyzing said dopant alkoxide with, said aluminum alkoxide in an amount effective to yield said controlled amount of said dopant in said internal structure of said γ -alumina support."

Auxiliary Requests 5 to 7

Claim 1 according to Auxiliary Request 5 differs from that of Auxiliary Request 4 in that the amount of dopant has been restricted to 2 to 3% by weight. Claim 1 according to Auxiliary Request 6 differs from that of Auxiliary Request 4 in that the dopant is now required to be a lanthanum oxide dopant. Claim 1 of Auxiliary Request 7 corresponds to claim 1 of Auxiliary Request 4 restricted by both limitations defined above in respect of Auxiliary Requests 5 and 6.

Auxiliary Requests 8 to 11

Claim 1 according to any of Auxiliary Requests 8 to 11 corresponds to claim 1 according to any of the Main and Auxiliary Request 1 to 3, respectively, in which the noble metal has been defined to be ruthenium.

Auxiliary Requests 12 and 13

Claim 1 according to Auxiliary Requests 12 and 13 corresponds to claim 1 according to Auxiliary Requests 4 and 5, respectively, in which the noble metal has been defined to be ruthenium.

Auxiliary Request 14

Claim 1 of Auxiliary Request 14 corresponds to claim 1 of Auxiliary Request 4 in which in addition to minor editorial amendments the catalyst is defined to contain a ruthenium noble metal promoter, a lanthanum oxide dopant and the expression "wherein said controlled amount of said dopant is an amount effective for increasing both the activity and the thermal stability of said catalyst for said Fischer-Tropsch hydrocarbon synthesis in said Fischer-Tropsch reaction system" has been deleted. Claim 1 of Auxiliary Request 14 reads therefore:

"1. A catalyst comprising: a γ -alumina support; an amount of cobalt, supported on said γ -alumina support, effective for Fischer-Tropsch hydrocarbon synthesis in a Fischer-Tropsch reaction system; and at least one ruthenium promoter on said γ -alumina support with said cobalt, said γ -alumina support having an internal structure comprising γ -alumina, at least 500 ppm of titania, expressed as elemental titanium and based on the total weight of the γ -alumina support, and a controlled amount of a lanthanum oxide dopant, wherein said controlled amount of said dopant present in said internal

structure of said γ -alumina support is in the range of from 1% to 5% by weight based on the total weight of the γ -alumina support, and wherein said γ -alumina support is obtainable from aluminium alkoxide which is hydrolyzed to produce an alumina product and wherein said dopant is incorporated in said γ -alumina support by adding a dopant alkoxide to, and co-hydrolyzing said dopant alkoxide with, said aluminium alkoxide in an amount effective to yield said controlled amount of said dopant in said internal structure of said γ -alumina support."

Claims 2 and 3 of Auxiliary Request 14 read as follows:

- "2. The catalyst of claim 1 wherein said controlled amount of said dopant present in said internal structure of said γ -alumina support is in the range from 2% to 3% by weight based on the total weight of said γ -alumina support.
3. A method of Fischer-Tropsch hydrocarbon synthesis comprising the step of reacting a synthesis gas in a Fischer-Tropsch reaction system in the presence of a catalyst according to any of the preceding claims."

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

- (a) Claim 1 of the Main Request was based on claim 1 of the application as filed, claim 2 and the first paragraph of page 9 defining the amount of dopant, claim 6 defining the co-hydrolysis of alkoxide,

claims 7 and 8 specifying that the dopant was barium oxide or lanthanum oxide, claim 11 defining the use of one promoter and page 7, lines 5-6 specifying that the promoter could be a noble metal. In addition, claim 13, which disclosed the use of ruthenium, made it clear that noble metals could be used as promoter in line with the description of embodiment b) on page 7, lines 5-6 from which claim 1 of the Main Request derived. In addition, the catalysts according to examples 4 and 5 of the application as filed where preferred embodiments which concerned catalysts with a noble metal promoter and a γ -alumina support doped with lanthanum or barium oxide. Present claim 1, which did not require the presence of titanium in the γ -alumina support, was also based on the ruthenium-promoted cobalt catalyst 5 described in Table 1, which support contained only traces of titanium. The language "internal structure" was used in claims 14 and 15, which also defined the same catalysts as claims 1 and 2. It also followed from original 32 that a controlled amount of dopant in the internal structure of the γ -alumina support could be obtained by adding a dopant alkoxide to, and co-hydrolyzing said dopant alkoxide with, aluminium alkoxide. It was also indicated that any method of introducing the dopant before crystallization of the boehmite would allow the dopant to be part of the internal structure, i.e. to be part of the material of the alumina or in other words to be part of the molecular structure of the support. This method led to a different structure than that obtained through impregnation of a γ -alumina support with the dopant, which

dopant through impregnation would be only present on the surface of the support or on the surface of its open pores. In addition, the structure of the doped γ -alumina did not depend on the method used for introducing the dopant before crystallization of the boehmite. The use of a ruthenium promoter was disclosed for example in claims 26 and 29 as originally filed. It was therefore concluded that the subject-matter of claim 1 according to any of the Main and Auxiliary Requests 1 to 15 was based on the application as filed.

- (b) Concerning novelty, the co-hydrolysis of aluminium alkoxide and lanthanum or barium alkoxide resulted in a lanthanum or barium oxide dopant being incorporated in the material of the γ -alumina and not only, as obtained by impregnation, of a lanthanum or barium oxide dopant being deposited on the surface of the γ -alumina and on the surfaces of its open pores.

- (c) As regards inventive step, the successive regeneration at high temperature of γ -alumina supported catalysts suitable for Fischer-Tropsch synthesis resulted in a slow and continuous loss of surface area and a slow conversion of the alumina from its γ -alumina phase to other forms (δ -alumina the θ -alumina) with much lower surface areas. It was therefore desired, without compromising the catalysts' activity, to provide catalysts with a high thermal stability. Furthermore, the alumina support of such catalysts could contain an amount of titania impurity, which had a detrimental effect on the catalysts' activity as for example

demonstrated in example 1 and Figure 1 of D1. The Applicants had found that the problem of providing a higher thermal stability without compromising the activity of the catalyst could be achieved by incorporation of a lanthanum oxide or barium oxide dopant in the alumina support, which incorporation could be obtained for example by adding a lanthanum or barium alkoxide to, and co-hydrolyzing said lanthanum or barium alkoxide with, aluminium alkoxide. Reference was made to catalysts 4 and 5 in Table 1 of the application, which had a doped support alumina prepared by co-hydrolyzing a lanthanum or barium alkoxide with aluminium alkoxide. The support used for catalyst 5 had a support containing 2,7% of barium oxide and a very low amount of titanium impurity (40 ppm TiO_2 , corresponding to about 24 ppm titanium as calculated by the Board), i.e. substantially no titanium. Despite this low amount of titanium the activity of catalyst 5 was substantially the same as the activity of catalyst 4 which contained 2,8% of lanthanum oxide and 1865 ppm TiO_2 (corresponding to about 1097 ppm titanium as calculated by the Board). If the activity of catalyst 5 was compared to that of catalyst 3, which contained also substantially no titanium, it had to be concluded that the addition of barium oxide resulted in the activity staying almost the same. It meant that for practical purposes the activity did not decrease. None of the documents on file provided a teaching towards a lanthanum oxide dopant or barium oxide dopant being effective to increase the thermal stability of a noble-metal promoted catalyst without reducing its activity. On this effect alone,

the claimed subject-matter involved an inventive step over D1. Moreover, the measures applied in D8 to D10 which dealt only with automotive combustion catalysts would not be applied by the skilled person in the field of Fischer-Tropsch catalysts. Hence, D8 to D10 could not suggest the presently claimed solution. An inventive step was therefore to be acknowledged.

XII. 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 Auxiliary Requests 1 to 15, Main Request and Auxiliary Requests 1 to 5, 7 to 13 and 15 as submitted on 24 October 2011, Auxiliary Request 6 as submitted on 13 January 2012 and Auxiliary Request 14 as submitted at the oral proceedings.

XIII. The decision was announced at the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

Main Request

Article 123(2) EPC

2. In accordance with the established Case Law of the Boards of Appeal of the EPO, the relevant question to be decided in assessing whether the subject-matter of an amended claim extends beyond the content of the application as filed is whether a skilled person would

have directly and unambiguously derived the proposed subject-matter as amended from the application as filed (Case Law of the Boards of Appeal of the EPO, 6th edition 2010, III.A.7.). In the present case, the amendments contained in claim 1 of the Main Request result in the definition of a cobalt catalyst which among others is defined to have a noble metal-promoter and a γ -alumina support with an internal structure including a lanthanum oxide dopant, a barium oxide dopant or a combination thereof, said γ -alumina support being specified to be obtainable by co-hydrolyzing an aluminium alkoxide with a dopant alkoxide (i.e. a lanthanum or barium alkoxide).

3. As far as the disclosure of the application as filed in relation to the structure of the γ -alumina support comprising a lanthanum oxide dopant, a barium oxide dopant or a combination thereof, is concerned, the application fails to provide a meaning for the wording "*internal structure*" as used in the expressions "*a γ -alumina support having an internal structure comprising γ -alumina and a controlled amount of a dopant*" and "*a γ -alumina support having an internal structure comprising γ -alumina, at least 500 ppm titania, expressed as elemental titanium and based on the total weight of the γ -alumina support and a controlled amount of a dopant*" contained in original independent claims 14 and 28, respectively. In the Board's judgement, those expressions do not necessarily mean that the dopant is part of the molecular structure of the support, as could be achieved by co-hydrolyzing an aluminium alkoxide with a dopant alkoxide, but only that the dopant is present within the support, e.g. on the internal surface of the γ -alumina support, which is

accessible to barium or lanthanum by impregnation of an aqueous solution of their appropriate salts. Hence, the use of the wording "internal structure" in original independent claims 14 or 28 does not necessarily imply that the support structure containing the dopant is one which is obtainable by co-hydrolysis of an aluminium alkoxide with a dopant alkoxide.

4. Furthermore, it is stated on page 9, lines 8-9 of the application as filed, that "*The dopant can be added at substantially any time, but will most preferably be added prior to crystallization of the boehmite*". Hence, the application as filed conveys the teaching that the method used for introducing the dopant (i.e. barium oxide or lanthanum oxide) in the γ -alumina is not essential for the purpose of the claimed invention. Moreover, no evidence has been provided that addition of the dopant to boehmite before transformation of the boehmite to γ -alumina, which also is covered by the general teaching provided in the application as filed, would necessarily lead to the same structure as that obtainable by a method comprising co-hydrolysis of an aluminium alkoxide and a lanthanum or barium alkoxide. It therefore follows from the above that the definition of a specific method by which lanthanum oxide or barium oxide can be incorporated into the γ -alumina support also implies that a particular structure of the doped support has been selected among those originally encompassed by the original independent claims. The question therefore arises whether the application as filed provides a direct and unambiguous disclosure for catalysts having a promoter selected from noble metals and a support obtainable by a method comprising the co-

hydrolysis of an aluminium alkoxide with a lanthanum or barium alkoxide.

5. Concerning the teaching of the application as filed in respect of promoters, it follows from the passage at page 7, lines 3-12, and in agreement with independent claims 1 and 14, that the use of promoters for the claimed catalysts, including a noble metal promoter, which is one of the classes of promoters proposed by the application, is optional. The only passage of the application as filed disclosing the combined use of a promoter and a support obtainable by adding a dopant alkoxide (i.e. lanthanum or barium alkoxide) to, and co-hydrolyzing said dopant alkoxide with, aluminium alkoxide is claim 32 by virtue of its dependence on claim 28, which, however, requires a γ -alumina support comprising at least 500 ppm of titania expressed as elemental titanium and based on the total weight of the γ -alumina support. Claim 28, however, only defines the use of an unspecified promoter, without pointing in the direction of noble metals in general. The indication in dependent claim 29 that "said one promoter" is ruthenium only confirms in line with the passage from page 11, line 5 to page 6, line 2 that the preferred promoter is ruthenium. It does not, however, allow the inference that noble metal promoters are generally preferred.

6. The Appellants also referred to the ruthenium-promoted cobalt catalysts 4 and 5 used in the experimental section of the application, which are supported on a γ -alumina support containing barium oxide or lanthanum oxide. The application as filed, however, does not provide any indication on the structure of the barium

oxide or lanthanum oxide doped γ -alumina support used for catalysts 4 and 5. Neither does it indicate any method by which the dopants were introduced into the γ -alumina for those embodiments. Hence, the disclosure of exemplified catalysts 4 and 5 does not fill the gap between parts of the original application dealing with the incorporation in the support of lanthanum or barium oxide by co-hydrolysis of alkoxides and parts dealing with the use of promoters, let alone noble metals in general.

7. Hence, the combination of features defined in claim 1 of the Main Request amounts to technical information which does not emerge directly and unambiguously from the content of the application as filed. Claim 1 of the Main Request is therefore not allowable having regard to the requirements of Article 123(2) EPC. The Main Request is therefore rejected.

Auxiliary Requests 1 to 3

8. The additional requirements in Auxiliary Requests 1 to 3 concern only the definition of a preferred amount of dopant from 2% to 3% by weight or the definition that lanthanum oxide must be contained as dopant. As these requests thus still define the combined use of a noble metal promoter with a support obtainable by adding a dopant alkoxide to, and co-hydrolyzing said dopant alkoxide with, aluminium alkoxide, the reasoning given in connection with the Main Request, also applies to present Auxiliary Requests 1 to 3. Therefore, Auxiliary Requests 1 to 3 are also not allowable.

Auxiliary Requests 4-7

9. Claim 1 of Auxiliary Requests 4 to 7 has been obtained by specifying in original claim 28, among others, that the "at least one promoter" is a noble metal. It follows, however, from the reasoning given in point 5 above that the application as filed does not disclose the combined use of noble metal promoters in general in the context of original claim 28. Hence, claim 1 according to any of Auxiliary Requests 4 to 7 does not fulfil the requirements of Article 123(2) EPC either.

Auxiliary Requests 8-11

10. Claim 1 according to Auxiliary Requests 8 to 11 corresponds to claim 1 of the Main Request or of Auxiliary Requests 1 to 3, respectively, in which the noble metal promoter has been defined to be ruthenium. It follows, however, from the reasoning given in point 5 above with respect to the Main Request that the disclosure of using a support obtainable by adding a dopant alkoxide to, and co-hydrolyzing said dopant alkoxide with, aluminium alkoxide, as well as a promoter, which preferably is ruthenium as follows from claim 29, is only given in the context of γ -alumina support comprising at least 500 ppm of titania. Claim 1 according to any of Auxiliary Requests 8 to 11, however, does not specify the feature that the γ -alumina support comprises at least 500 ppm of titanium. As the application as filed, however, does not contain any pointer towards the generalization of the above teaching to any γ -alumina support obtainable by adding a dopant alkoxide to, and co-hydrolyzing said dopant alkoxide with, aluminium alkoxide, irrespective of the

amount of titanium to be achieved in the γ -alumina support, claim 1 according to any of Auxiliary Requests 8 to 11 is not considered to have a basis in the application as filed, as required by Article 123(2) EPC. Thus, Auxiliary Requests 8 to 11 are rejected.

Auxiliary Request 12

Amendments

11. Claim 1 according to Auxiliary Request 12 corresponds to that of Auxiliary Request 4, wherein the "at least one promoter" is defined to be a ruthenium promoter, which in fact is the preferred promoter, as can be understood from claim 29, in line with the passage from page 11, line 5 to page 6, line 2 of the original disclosure. Claim 32 as filed discloses a promoted cobalt on γ -alumina catalyst, wherein the γ -alumina support comprises at least 500 ppm of titania and a controlled amount of a dopant selected from the group consisting of a lanthanum oxide dopant, a barium oxide dopant, and combinations thereof and wherein the γ -alumina support is obtainable from aluminium alkoxide which is hydrolyzed to produce an alumina and wherein the dopant is incorporated by adding a dopant alkoxide to, and co-hydrolyzing said dopant alkoxide with, the aluminium alkoxide. Original claim 32, when read in the light of the passages of the application as filed specifying the preferred amount of dopant (claim 30 and page 9, lines 3-5) and the preferred promoter, namely ruthenium (claim 29, in line with the passage from page 11, line 5 to page 6, line 2), provides a direct and unambiguous disclosure for claim 1 according to Auxiliary Request 12. The amount of dopant as defined

in claim 2 is disclosed in original claim 31 and on page 9, lines 5-6 as the most preferred amount of dopant. The catalysts disclosed in claim 32 are indirectly disclosed by reference to claim 28 to be used in a method of Fischer-Tropsch hydrocarbon synthesis, as is now defined in claim 3 of Auxiliary Request 12. Consequently, claims 1 to 3 of Auxiliary Request 12 meet the requirements of Article 123(2) EPC.

Meaning of claim 1

12. Claim 1 of Auxiliary Request 12 indicates that the amount of dopant is an amount effective for increasing both the activity and the thermal stability of the catalyst for Fischer-Tropsch hydrocarbon synthesis, said controlled amount being further defined to be within the range of 1% to 5% by weight based on the total weight of the γ -alumina support. The question therefore arises whether the first condition defining in functional terms the amount of dopant imposes a further restriction on the second condition defining an amount of 1% to 5% by weight for said dopant, or whether claim 1 is meant to define any amount of dopant between 1% and 5% by weight, this range being considered in the application as filed to bring about the above defined alleged advantage with respect to activity and thermal stability. In the Board's judgment, in line with the Appellants' submissions for inventive step, the first condition must be understood from the perspective of the Appellants to be redundant in view of the second condition imposing a concrete amount of dopant. Otherwise in the absence in present claim of any definition for conditions of the Fischer-Tropsch hydrocarbon synthesis under which stability and

activity must be tested, claim 1 would have to be rejected for lack of clarity and also for lack of support as the application does not provide any guidance as to what the skilled person must do, should the preferred range of 1% to 5% by weight not provide the desired result in terms of activity and stability. For the avoidance of doubt, it is pointed out that the Appellants' belief that the above range of 1% to 5% by weight provides the technical effect defined in present claim 1 serves only to attribute to that claim its intended meaning with regard to the amount of dopant. It does not preclude any assessment of the test results presented in the application for the purpose of defining the technical problem solved by the claimed catalysts over the closest prior art.

Novelty

13. As argued by the Appellants and indicated in above point 3, co-hydrolysis of an aluminium alkoxide with a lanthanum or barium alkoxide is considered to result, after the necessary treatment at high temperature of the gel obtained for producing an alumina product, in the lanthanum or barium oxide dopant being incorporated in the material of the γ -alumina support and not only, as obtained by impregnation of a pre-formed γ -alumina support, of the lanthanum or barium oxide being deposited on the surface of the γ -alumina, in particular its open pores. The Board is therefore satisfied that none of the prior art documents on file discloses a cobalt ruthenium-promoted catalyst with a γ -alumina support as specified in present claim 1, i.e. which contains at least 500 ppm titanium based on the total weight of the γ -alumina support and which are

obtainable by using the above described method in order to provide a γ -alumina support containing between 1% and 5% by weight of lanthanum or barium oxide based on the total weight of the support. Hence, claim 1 according to Auxiliary Request 12 meets the requirements of Article 54 EPC.

Inventive step

Closest prior art

14. The present invention aims at providing highly stable and highly active alumina-supported cobalt catalysts for use in Fischer-Tropsch synthesis process, especially in slurry bubble column reactor systems. Document D1 discloses in example 1 on page 16 a ruthenium-promoted cobalt catalyst on γ -alumina with 20% by weight cobalt and 0.5% by weight ruthenium. The γ -alumina support is CATAPAL B, which contains according to the last paragraph on page 16 of D1, 1000 ppm of titanium. This catalyst is employed in slurry bubble column reactor systems for a Fischer-Tropsch synthesis process and provides an activity of about 1400 g-HC/kg-cat/hr (D1, page 17, first full paragraph). Hence, the disclosure of the Fischer-Tropsch synthesis process described in example 1 of D1 which employs a catalyst structurally close to that presently claimed and which meets one of the two objectives set out in the present application, namely highly active catalysts for use in Fischer-Tropsch synthesis process, especially in slurry bubble column reactor systems, is in the Board's judgment and in line with the contested decision, the closest state of the art.

Problem and solution

15. The Appellants argued that the problem solved by the claimed catalysts over that disclosed in example 1 of D1 was the provision of catalysts exhibiting a higher thermal stability without compromising the activity for Fischer-Tropsch synthesis. The solution to this problem is alleged to lie in the incorporation of a lanthanum oxide or barium oxide dopant in an amount ranging from 1% to 5% by weight based on the total weight of the support in the γ -alumina support, said incorporation being obtainable by co-hydrolyzing a lanthanum or barium alkoxide with aluminium alkoxide. In order to show that said problem was successfully solved by the claimed solution, the Appellants referred to the experimental evidence presented in Table 1 of the application.

16. Figure 1 of the present application summarizes thermal stability tests carried out for γ -alumina supports and comparable γ -alumina supports doped with 3% by weight of lanthanum or barium oxide. These tests were carried out at temperatures of 1050°C to 1200°C, i.e. at temperatures higher than those usually employed for catalysts' regeneration of the order of 300-500°C. They serve to demonstrate in an accelerated manner that the claimed catalysts, i.e. their γ -alumina support, will have in comparison with the catalyst disclosed in D1 a higher thermal stability when subjected to repeated regeneration treatments required for reactivation. There no reason for the Board to cast doubt on those test results. It is therefore accepted that the problem of increasing the thermal stability of the catalyst

- disclosed in D1 has been successfully solved by the claimed solution.
17. As concerns the catalysts' activity, test results for three ruthenium-promoted cobalt catalysts on γ -alumina with 20% by weight cobalt and 0.5% by weight ruthenium, which comprise various amounts of titanium dopant in the γ -alumina support, but no lanthanum or barium oxide dopant, have been summarized in Table 1 of the application. These test results correspond to those presented in example 1 of D1. They demonstrate that increasing amounts of titanium in the γ -alumina support result in a decrease of the ruthenium-promoted cobalt catalyst activity for Fischer-Tropsch synthesis, when the γ -alumina support contains neither a barium oxide, nor a lanthanum oxide. Variations of the catalyst activity for Fischer-Tropsch synthesis as a function of the titanium amount in the presence of a constant amount of barium or lanthanum oxide contained in the γ -alumina support, let alone when the dopant is introduced by co-hydrolysis of corresponding alkoxides, are not available.
18. The present application makes it possible to compare catalyst 1 having a γ -alumina support comprising 1000 ppm titanium, i.e. the catalyst described in the closest prior art and catalyst 4, the γ -alumina support of which comprises, based on the total weight of the support, 1097 ppm titanium and 2,8% by weight of lanthanum oxide introduced into the support, according to the Appellants, by co-hydrolysis of lanthanum and aluminium alkoxides. Despite the presence of a slightly higher amount of titanium in the support for catalyst 4, which has a negative influence on the catalyst's

activity as shown in point 17 above, the introduction of lanthanum oxide into the support of the catalyst by the co-hydrolysis method results in an 8% increase of activity in a Fischer-Tropsch synthesis. This result renders credible in the Board's judgement that the introduction of lanthanum oxide in the γ -alumina support of the closest prior art by co-hydrolysis of lanthanum and aluminium alkoxides provides a beneficial effect on the activity of the catalyst for Fischer-Tropsch synthesis.

19. Such an effect, however, has not been shown to exist when barium oxide is introduced in the γ -alumina support, by co-hydrolysis of barium alkoxide and aluminium alkoxide. A comparison between the results for catalyst 3 and catalyst 5 summarized in Table 1, which both are considered to contain only negligible traces of titanium, respectively 7 and 23 ppm compared to the minimum amount of 500 ppm defined in present claim 1, indicates a decrease of activity of 4%. The Appellants argued that the difference of activity between catalyst 3 and catalyst 5 was within the margin of error of 5% associated with the measure of the catalyst's activity. The Appellants who carry the burden of proof for their allegation in support of a technical effect arising from the introduction of barium oxide dopant in the γ -alumina support, however, did not provide statistical data rendering credible that with a difference of activity of 4% between catalysts 3 and 5 and an alleged margin of error of 5% for the measurement, the activity of catalyst 3 which does not contain titanium in its support was not reduced by the introduction of barium oxide in the γ -alumina support. Moreover, even if this effect had

been rendered credible for γ -alumina supports which contain only traces of titanium, it would not have shown that this effect can be extrapolated to γ -alumina supports having a content of titanium of at least 500 pm, which as demonstrated by the application data, also affect the catalysts' activity. Hence, the technical problem of providing catalysts for Fischer-Tropsch synthesis having higher thermal stability without compromising their activity cannot be considered to be solved over the whole breadth of claim 1. It follows therefore that the technical problem solved over the closest prior art can only be seen in the provision of catalysts having higher thermal stability.

Obviousness

20. It remains to be decided whether or not the skilled person starting from the catalyst described in example 1 of D1 and wishing to provide catalysts having higher thermal stability would have been guided by the available prior art to apply the additional measure defined in claim 1 of Auxiliary Request 12, namely the introduction of a lanthanum oxide or barium oxide dopant in the γ -alumina support, said incorporation being obtainable by co-hydrolyzing with an aluminium alkoxide used for the production of the γ -alumina support a lanthanum or barium alkoxide.

21. As acknowledged on page 5, lines 8-18 of the application, alumina, one of common oxides used as support for cobalt-based F-T catalysts, is well known to be sensitive to the pre-treatment temperatures and the amount of time it is subjected to high temperatures. It is in particular known that heating, either during

the pre-treatment step, during the use of the catalyst or during catalyst regeneration may result in a slow and continuous loss of surface area and a slow conversion of the alumina from its γ -alumina phase to other forms which have much lower surface areas, those structural changes resulting in the catalytic metal being less accessible to the reactants and therefore to a loss of catalyst's activity (see application page 5, lines 2-7). The generally known lack of thermal stability of γ -alumina supports used in the field of catalysts is also confirmed in documents D8 to D10, which suggest as solution to this problem the introduction by the sol-gel technique of lanthanum or barium oxide in the aluminium oxide matrix, namely by a process comprising co-hydrolysing an aluminium alkoxide with a lanthanum alkoxide (D8, column 1, lines 22-45; D9, page 1821, third and fourth paragraph) or barium alkoxide (D10, page 84, last paragraph, page 86, first and second paragraph, table 3, paragraph bridging pages 86 and 87). Thus, the skilled person who wanted to improve the thermal stability of the Fischer-Tropsch ruthenium-promoted cobalt catalyst disclosed in example 1 of D1, would have in particular turned to the teaching of document D10 which suggests in Table 3 the incorporation of barium oxide in the aluminium oxide matrix by co-hydrolysing an aluminium alkoxide with a barium alkoxide. The Appellants' argument that documents D8 to D10 do not suggest the claimed solution as those prior art documents relate to other types of catalysts fails to convince, as the thermal stability problem addressed does not originate in the use of one particular catalytic substance present on the γ -alumina support, but in the use of the catalytic inactive γ -alumina support itself. Thus, the skilled person

would have been prompted to apply the solution proposed by documents D8 to D10 also to Fischer-Tropsch catalysts on γ -alumina, which also suffer from lack of thermal stability of γ -alumina. The preferred amount of barium oxide, according to the present application, which is now defined in claim 1 of Auxiliary Request 12, or even the more preferred amount of 2% to 3% by weight also indicated in the first paragraph on page 9 of the present application, are merely the result of routine experimentation work carried out by the skilled person testing various amounts to find suitable thermal stability, the above more preferred amount being suggested in the third paragraph of D9 for thermal stabilization of commercial alumina-based washcoats. Consequently, the skilled person, who starting from the catalyst of example 1 of D1 wanted to provide catalysts having higher thermal stability, would have been motivated to replace the γ -alumina support used for catalyst 1 of D1, by a γ -alumina support comprising 1% to 5% or 2% to 3% barium oxide, based on the total weight of the support, said support being obtained by a process comprising co-hydrolysis of barium and aluminium alkoxides.

22. Hence, claim 1 of Auxiliary requests 12, which encompasses an obvious embodiment, does not meet the requirements of Article 56 EPC.

Auxiliary Request 13

23. It follows from above point 22 that the introduction of 2% to 3% barium oxide, based on the total weight of the support, also lacks an inventive step. Auxiliary Request 13 is therefore not allowable.

Auxiliary Request 14

24. In addition to two editorial amendments in lines 3 and 4, claim 1 according to Auxiliary Request 14 differs in substance from claim 1 of Auxiliary Request 12 in that (i) the γ -alumina support is defined to contain a lanthanum oxide dopant, (ii) the expression "*wherein said controlled amount of dopant is an amount effective for increasing both the activity and the thermal stability of said catalyst for said Fischer-Tropsch hydrocarbon synthesis in said Fischer-Tropsch reaction system*" has been deleted and (iii) the expression "*said γ -alumina support is produced*" has been replaced by "*said γ -alumina support is obtainable*".
25. Claims 1 to 3 of Auxiliary Request 14 find a basis in the application as filed for the same reasons as those given in above point 11. Moreover, in view of amendments (ii) and (iii) indicated in above point 24, the subject-matter according to present claims 1 to 3 is not objectionable any more for lack of clarity or support. In addition, as the subject-matter of present claims 1 to 3 is a restriction of that defined in Auxiliary Request 12, their novelty is acknowledged for the same reasons as those indicated in point 13 above.
26. As regards inventive step, and in view of the analysis of the experimental results given in above points 17 and 18, the problem solved by the subject-matter of present claim 1 over the catalyst disclosed in example 1 of D1, which has a γ -alumina support containing titanium impurities, can be seen in the provision of catalysts exhibiting a higher thermal stability and an

- improved activity for Fischer-Tropsch synthesis. The solution to this problem lay in the incorporation of a lanthanum oxide dopant in an amount ranging from 1% to 5% by weight based on the total weight of the support in the γ -alumina support, said incorporation being obtainable by a process comprising the co-hydrolysis of an aluminium alkoxide with a lanthanum alkoxide.
27. Whereas the incorporation of lanthanum oxide in the γ -alumina support by a process comprising the co-hydrolysis of a lanthanum alkoxide with an aluminium alkoxide would have been obvious for the skilled person in order to improve the thermal stability of the catalyst as shown in above point 21, it is not suggested in the prior art that the same measure would provide an increase of catalytic activity in a Fischer-Tropsch synthesis. Although lanthanum oxide is a known additional promoter for Fischer-Tropsch ruthenium-promoted cobalt catalysts (see D2, Table 3) when it has been incorporated in the catalyst support by impregnation of the pre-formed γ -alumina (see D2, passage from column 11, line 66 to column 12, lines 11 and column 11, lines 12-51), D2 does not suggest a structure of the γ -alumina support obtainable by co-hydrolysis of lanthanum and aluminium alkoxides, let alone any property which would result from it.
28. A structure of the γ -alumina support obtainable by co-hydrolysis of lanthanum and aluminium alkoxides is suggested in D1 for increasing the activity of Fischer-Tropsch catalysts which are not promoted with any noble metal or any near noble metal (paragraph bridging pages 8 and 9 and last paragraph of page 9). Such a teaching is not extended in D1 to noble metal promoted cobalt

catalysts. D1 rather suggests the contrary, as it demonstrates that the incorporation of titanium in the structure of the γ -alumina support results in a reduced activity of the ruthenium-promoted cobalt Fischer-Tropsch catalyst (see point 17 above). Moreover, the amounts of dopant (titanium, lanthanum, barium) used in D1 are more preferably from 0,08 to 0,2% based on the total weight of the γ -alumina support (page 14, third paragraph), i.e. well below the level defined in present claim 1.

29. Should the subject-matter of Auxiliary Request 14 not enjoy priority from US patent application 09/528 163, D6 would become prior art pursuant to Article 54(2) EPC. D6 is directed to palladium-promoted cobalt on alumina catalysts for Fischer-Tropsch synthesis having a γ -alumina support (claim 1). According to column 4, lines 16-19 of that document, the γ -alumina support can be doped with at least titanium, lanthanum, barium, calcium, vanadium, tungsten, or potassium, the dopant being preferably added prior to crystallization or to the boehmite prior to calcination in an amount most preferably in the range of from 0,1 to 5% by weight based on the total weight of the support (column 4, lines 27-32). This document, however, does not suggest that doping the γ -alumina support with lanthanum could counterbalance the negative effect of titanium on the activity of palladium-promoted cobalt-on-alumina catalysts and *a fortiori* for catalysts outside of the teaching of D6 promoted with another noble metal.
30. Finally, as demonstrated by the test results discussed in above point 19, contrary to the incorporation of lanthanum, the incorporation of barium in the structure

of the γ -alumina support by co-hydrolysis of aluminium and barium alkoxides results in a decreased activity. Hence, achieving higher activity cannot be seen as a mere collateral effect obtained when achieving improved thermal stability by incorporation of dopants through co-hydrolysis of suitable alkoxides.

31. Consequently, the claimed solution is not derived in an obvious manner from the prior art. The subject-matter of claim 1 and by the same token that of dependent claim 2 and of independent claim 3, which include all the features of claim 1, meets the requirements of Article 56 EPC.

32. Auxiliary Request 14 is therefore allowable.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the examining division with the order to grant a patent on the basis of Auxiliary Request 14 filed at the oral proceedings and a description to be adapted thereto.

The Registrar

The Chairman

S. Fabiani

J. Riolo