

**Internal distribution code:**

- (A)  Publication in OJ  
(B)  To Chairmen and Members  
(C)  To Chairmen  
(D)  No distribution

**Datasheet for the decision  
of 08 January 2008**

**Case Number:** T 0229/04 - 3.3.07

**Application Number:** 95909862.5

**Publication Number:** 0748255

**IPC:** B01J 33/00

**Language of the proceedings:** EN

**Title of invention:**

Copper Catalysts

**Patent Proprietors:**

Johnson Matthey PLC

**Opponents:**

SÜD-CHEMIE AG

**Headword:**

-

**Relevant legal provisions:**

EPC Art. 54, 56

RPBA Art. 10a(2), 10b(1)

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

-

**Keyword:**

"Novelty (yes)"

"Inventive step (yes)"

"Late filed evidence admitted (no)"

**Decisions cited:**

-

**Catchword:**

-



Case Number: T 0229/04 - 3.3.07

**DECISION**  
of the Technical Board of Appeal 3.3.07  
of 08 January 2008

**Appellant:** SÜD-CHEMIE AG  
(Opponents) Lenbachplatz 6  
D-80333 München (DE)

**Representative:** Splanemann Reitzner Baronetzky Westendorp  
Rumfordstraße 7  
D-80469 München (DE)

**Appellant:** Johnson Matthey PLC  
(Patent Proprietors) 40-42 Hatton Garden  
London EC1N 8EE (GB)

**Representative:** Gibson, Sara Hillary Margaret  
Intellectual Property Department  
Johnson Matthey Catalysts  
P.O. Box 1  
Belasis Avenue  
Billingham, Cleveland TS23 1LB (GB)

**Decision under appeal:** Interlocutory decision of the Opposition  
Division of the European Patent Office posted  
11 December 2003 concerning maintenance of  
European patent No. 0748255 in amended form.

**Composition of the Board:**

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

## Summary of Facts and Submissions

I. These appeal proceedings concern the decision of the Opposition Division maintaining in amended form European patent No. 0 748 255 entitled "Copper catalysts".

II. Independent claims 1 and 8 as granted read:

"1. A process for the stabilisation of a reduced copper catalyst containing copper and optionally compounds of zinc, chromium, zirconium, magnesium, silicon and/or aluminium, comprising a) passivating the catalyst by passing a passivating gas stream that is free of reducing gases and which contains less than 5% by volume of oxygen through a bed of the catalyst under conditions such that the temperature of the catalyst does not exceed a specified value until the catalyst is passivated, and then b) increasing the oxygen content of the gas passing through the bed until the oxygen partial pressure corresponds to that of air at atmospheric pressure, characterised in that said passivating gas stream contains 0.05 to 0.5% by volume of oxygen and an amount of carbon dioxide at least twice the amount of oxygen, and the proportion of oxygen in said passivating gas stream and the temperature at which said passivating gas stream is fed to the bed being such that the temperature of the catalyst does not rise to above 100°C during said passivation step."

"8. A stabilised passivated copper catalyst containing copper and optionally compounds of zinc, chromium, zirconium, magnesium, silicon and/or aluminium,

characterised in that the passivated catalyst has a thin copper carbonate layer on the copper surface and does not exhibit substantial oxidation when heated in air from 20°C at a rate of 10°C per minute until the temperature has reached at least 100°C."

The dependent claims 2 to 7 were directed to preferred embodiments within the ambit of claim 1.

III. The patent had been opposed in its entirety on the grounds of Article 100(a) EPC with respect to lack of novelty and lack of an inventive step. The opposition was supported *inter alia* by the following documents:

(1) GB-A-1 319 622

(3) A.V. Krylova *et al.*, Applied Catalysis, 39, (1988), 325-331 and

(10) A.F. Hollemann, E. Wiberg, Lehrbuch der Anorganischen Chemie, Walter de Gruyter & Co. Berlin, 1960, page 452.

IV. The Opposition Division decided that the Proprietors' main request to reject the opposition could not be allowed, as claim 8 as granted lacked novelty over the disclosure of either document (10) or (3). It was held in particular that the feature "does not exhibit substantial oxidation when heated in air from 20°C at a rate of 10°C per minute until the temperature has reached at least 100°C" corresponded to an unusual parameter which was furthermore unclear and therefore should be disregarded when assessing novelty. Document (10) was found to be novelty destroying for the subject-matter of claim 8 as it explicitly disclosed a metallic copper having a basic copper carbonate layer

thereon. The Opposition Division also held that document (3), which disclosed the passivation of a copper-containing low-temperature water-gas-shift catalyst with a carbon dioxide, steam and oxygen mixture, resulted in the passivated catalyst of claim 8 under dispute. Although not explicitly disclosed, the presence of a carbonate layer on the copper catalyst was assumed to be obtained in view of the similarity of the conditions used in document (3) and in the patent in suit.

The Opposition Division also decided that the subject-matter of claims 1 to 7 of the patent as granted, which formed the basis for the Proprietors' first auxiliary request, met the requirements of the EPC. The Opposition Division came to the conclusion that document (1) did not disclose a passivation step in the presence of carbon dioxide. Novelty over the other documents cited in the opposition proceedings was also acknowledged, as none of them disclosed a passivation process comprising the use of a gas stream containing 0.05 to 0.5 vol.% of oxygen, followed by an increase of the oxygen partial pressure up to that of air at atmospheric pressure. As regards inventive step, the Opposition Division considered document (1) as the closest state of the art. Based on the experimental evidence contained in the patent and those submitted by the Proprietors with their facsimile letter dated 24 October 2003, the technical problem solved over document (1) was formulated as the provision of a more effective passivation process. This, according to the contested decision, was in particular achieved by the use of a passivation gas stream, in which the amount of carbon dioxide was at least twice the amount of oxygen.

As none of the documents cited by the Opponents suggested the importance of the ratio between carbon dioxide and oxygen in the passivation gas stream for solving this problem, the claimed process was considered to represent a non-obvious modification of the process of document (1).

V. Notices of appeal were filed both by the Opponents and the Proprietors.

VI. The Opponents, in addition to their statement setting out the grounds for appeal submitted on 16 April 2004 and a further written submission dated 26 October 2004, requested with a letter dated 25 May 2007, that the following documents be introduced into the proceedings:

(13) Holleman-Wiberg, Lehrbuch der Anorganischen Chemie, 81.-90. revidierte Auflage, 1976, page 795

(14) US-A-2 677 669 and

(15) US-A-4 090 980.

They also requested with letter dated 6 December 2007, that the following additional documents be introduced into the proceedings:

(17) Fluka catalogue, page 1206 and

(18) Sigma-Aldrich catalogue, pages 1560-1561.

VII. The Proprietors, with their statement setting out the grounds for appeal requested as main request maintenance of the patent with the claims as granted and submitted three sets of claims, forming the basis for their first to third auxiliary requests, respectively. The claims as maintained by the

Opposition Division formed the basis for their fourth auxiliary request. Further arguments and three additional sets of claims as their fifth to seventh auxiliary requests were submitted by letter dated 18 August 2004. The Proprietors also submitted with letter dated 6 December 2007 an experimental report, hereinafter referred to document (16). They also submitted an additional set of claims forming the basis for their eighth auxiliary request.

VIII. Oral proceedings were held on 08 January 2008 in the course of which the Opponents submitted a combined TGA and DSC measurement of a commercial sample of basic copper carbonate, hereinafter referred to document (19).

IX. The arguments by the Opponents can be summarized as follows:

(a) Document (1) disclosed a method of stabilising a pyrophoric copper-zinc oxide catalyst obtained by reduction of a composition comprising copper oxide and zinc oxide, wherein the catalyst was initially surrounded with a non-oxidising gas stream. The elemental copper was then partially oxidised by introducing oxygen into the gas stream and gradually increasing the concentration of oxygen in the stream to a maximum of 5 volume % whilst maintaining the temperature of the catalyst at or below 65,6°C. The reduction of the composition comprising copper oxide and zinc oxide was carried out by introducing hydrogen into a stream of inert gas such as natural gas, carbon dioxide or nitrogen. According to page 2, lines 44-54 the hydrogen was purged with inert gas after reduction

had occurred and before oxygen was introduced. Document (1) was not restricted to the use of a preferred inert gas. It did not disclose that the inert gas should be changed for the reduction and oxidation steps. The Opponents also argued that valve 27a which was used to provide a supply of nitrogen or carbon dioxide to the reduction vessel was also used for purging the stabilization vessel with the same inert gas. Thus, the same inert gas as specified page 2, lines 19-20 of document (1) for the reduction of the pellets comprising copper oxide and zinc oxide, which could be carbon dioxide, could be used for purging the hydrogen and introducing oxygen. Hence, document (1) disclosed the combined use of carbon dioxide and oxygen for the passivation step, so that the process as defined in claim 1 of the contested patent lacked novelty.

- (b) Regarding the meaning of the wording "copper carbonate" present in claim 8, the Opponents were of the opinion that copper carbonate as such, i.e.  $\text{CuCO}_3$ , did not exist, as shown by documents (13), (17) and (18). They alleged at the oral proceedings before the Board, that X-ray photoelectron spectroscopy (XPS), contrary to the Patentee's assertion in paragraph [0019] of the patent, was not a method which would allow detection of the presence of a thin layer of copper carbonate on the surface of a copper catalyst. However, no objection for lack of sufficiency of disclosure under article 100b) EPC was raised against claim 8. The Opponents rather took the view that the passivation process



according to claim 1 of the patent in suit, either resulted in a passivation layer of copper oxide on the surface of the catalyst, should the process take place in the absence of water, or that the wording "copper carbonate" as used in the patent in suit, in particular in claim 8, in fact meant basic carbonate, i.e. a mixture comprising  $\text{CuCO}_3$  and  $\text{Cu}(\text{OH})_2$ , which however meant that the passivation process of the patent in suit took place in the presence of water.

- (c) Document (3) disclosed the reaction of a copper catalyst in the presence of oxygen, carbon dioxide and water so that a thin basic copper carbonate layer would build up on the copper catalyst. Hence, document (3) was novelty destroying for claim 8, should the wording "copper carbonate" mean basic copper carbonate. Document (10) which disclosed that a layer of basic copper carbonate was formed on metallic copper when in contact with air and carbon dioxide, also anticipated the subject-matter of claim 8.
- (d) As to inventive step, the closest state of the art was document (3), which, as did the patent in suit, related to the stabilization of pyrophoric copper catalysts with a gas mixture comprising carbon dioxide, water and oxygen. The problem underlying the present invention lay in the further development of the process of document (3) for an industrial application. The Opponents accepted at the oral proceedings before the Board that the experimental evidence on file showed an improvement of the passivation. The selection of

an oxygen atmosphere as defined in the different steps of claim 1 for achieving that purpose was however rendered obvious by the teaching of document (1).

- (e) The Opponents also sought to rely on documents (14) and (15) in support of the argument that claim 1 lacked an inventive step in case the claimed process did not result in a copper carbonate layer, but instead in a copper oxide layer.

X. The arguments by the Proprietors can be summarized as follows:

- (a) According to the patent under dispute and as shown by the experimental evidence provided by document (16), the passivation process according to claim 1 of the patent in suit resulted in the formation of a layer of copper carbonate on the copper catalyst.
- (b) The feature defining in claim 8 that the passivated catalyst "does not exhibit substantial oxidation when heated in air from 20°C at a rate of 10°C per minute until the temperature has reached at least 100°C" could be clearly determined by thermogravimetric analysis as specified in the patent specification. Weight gain indicated that copper oxide was formed when the catalyst was exposed to air. The presence of that feature in claim 8 ensured that the copper carbonate layer was thick enough to protect the underlying copper from undesirable pyrophoric oxidation in the temperature range used to handle copper catalysts.

- (c) Concerning novelty, document (1) could not anticipate the claimed subject-matter as carbon dioxide was not used in document (1) as an inert gas for the stabilization of the copper catalyst. Furthermore, document (1) did not disclose that the quantity of carbon dioxide should be at least twice the quantity of oxygen. The stabilization in document (1) took place by the formation of copper oxide, not copper carbonate. The catalysts disclosed in document (3) did not exhibit the absence of substantial oxidation on heating in air until a temperature of at least 100°C. Document (10), which disclosed that copper surfaces in air slowly form a coating of green basic carbonate, was not relevant to the question of protecting copper catalysts from pyrophoric oxidation. There was no indication in document (10) that the copper should not exhibit any substantial oxidation on heating in air until the temperature had reached at least 100°C.
- (d) As regards inventive step, document (1) formed the closest state of the art. The Patent Proprietors argued that the Opponents had failed to provide experimental evidence that copper oxide was formed instead of copper carbonate, while the examples of the contested patent as well as the experimental evidence presented in document (16) showed that a passivation layer of copper carbonate was formed on the copper. The technical problem solved over document (1) was to provide passivated catalysts that could be more efficiently activated, as less hydrogen was consumed for the activation. It was

not be obvious to combine the teaching of document (1) with that of document (3) as the carbon dioxide used in document (1) was not an oxidant but an inert gas. In addition, none of the documents cited suggested the passivating gas stream as defined in the present claims, in particular the relative amounts of oxygen and carbon dioxide which were required to provide an effective passivation of reduced copper catalysts and also ensure that the temperature of the catalyst would not rise above 100°C during the passivation step.

- (e) Document (13) said nothing about copper catalyst passivation. Documents (14) and (15), which rather concerned the passivation of nickel catalysts, did not disclose in relation to copper catalysts the conditions required by the present invention in order to obtain a layer of copper carbonate. Thus, the late filed documents (13) to (15) were not relevant and should not be admitted into the proceedings.

XI. The Opponents requested that the decision under appeal be set aside and the patent be revoked.

XII. The Proprietors requested that the decision under appeal be set aside and the patent be maintained as granted, or alternatively on the basis of any one of the sets of claims as set out in auxiliary requests 1 to 8.

XIII. At the end of the oral proceedings the decision of the Board was announced.

## Reasons for the Decision

1. The appeals are admissible.

### *Process claims 1 to 7 of the main request - Novelty*

2. The Opponents objected to the novelty of the subject-matter of claim 1 in view of the disclosure of document (1).

- 2.1 Document (1) discloses in claim 1 a method of stabilising a pyrophoric copper-zinc oxide catalyst obtained by reduction of a composition comprising copper oxide and zinc oxide, wherein the catalyst is initially surrounded with a non-oxidising gas stream, i.e. an inert gas, and the elemental copper is partially oxidised in a further step by introducing oxygen into the gas stream. The concentration of oxygen in the gas stream is gradually increased to a maximum of 5 volume % whilst maintaining the temperature of the catalyst at or below 150°F (65,6°C).

- 2.2 The Opponents' objection is based on the assertion that the oxidation step of the copper catalyst is carried out in the presence of carbon dioxide, either as a residue of the inert gas used for the reduction step or as an inert gas injected for the oxidation step.

- 2.3 The details of the oxidation step, as well as those of the preceding reduction steps are disclosed on page 2, line 62 to page 3, line 53 with reference to the drawing. The process is carried out in two different vessels, one for reduction, i.e. the furnace vessel 10

(page 2, lines 67-89) and a second one for stabilization of the catalyst, i.e. the stabilization vessel 26 (page 2, lines 88-89 and 92-102). After reduction of the copper in the furnace vessel, the pellets are cooled down and discharged into the stabilization vessel, which has been previously purged with nitrogen or another (unspecified) inert gas via a line 44 (page 3, lines 27-34). When the temperature of the pellets has fallen to an appropriate level for controlled oxidation of the copper (page 3, lines 36-39), air (or oxygen) is admitted into the inert gas stream of line 44. This means that nitrogen or another unspecified inert gas is injected through line 44 into the stabilization vessel containing the reduced pellets before introduction of any oxygen. Hence, contrary to the Opponents' opinion, it cannot be taken that carbon dioxide, even if should it have been present in the furnace vessel during the reduction step and discharged together with the reduced catalyst, would still be present in the stabilization vessel, when oxygen is introduced via line 44.

- 2.4 Document (1) discloses on page 2, lines 89-92, that conduit 25 between the bottom of the furnace vessel and the top of the stabilization vessel is connected to a gas line 27, controlled by a valve 27a, to provide a supply of nitrogen or carbon dioxide to the conduit 25, and therefore implicitly to the stabilization vessel 26. It does not, however, necessarily mean that carbon dioxide is present during the stabilization step, when oxygen is allowed to flow in the stabilization vessel, because, as shown above, nitrogen or another (unspecified) inert gas is injected through line 44 before any oxygen is introduced into the stabilization

vessel. The Opponents' argument, that valve 27a between the furnace vessel and the stabilization vessel, which is used to provide a supply of nitrogen or carbon dioxide (page 2, lines 89-92) in the furnace vessel, is also used for purging the stabilization vessel with the inert gas, is purely speculative and not based on any evidence.

2.5 The Board is also not able to find any indication in document (1) for the Opponents' contention that carbon dioxide can be used as an inert gas within the meaning of document (1) for the stabilization step, as the only inert gas mentioned in document (1) for the stabilization step is nitrogen and document (1) fails to disclose that any inert gas used for the reduction step should be also considered as an inert gas for the passivation step.

2.6 Thus, the presence of carbon dioxide and oxygen during the stabilization step has not been shown to be clearly and unambiguously disclosed in document (1).

2.7 Consequently, the subject-matter of present claim 1 and claims 2 to 7 dependent thereon is novel over document (1) and therefore meets the requirements of article 54 EPC.

*Meaning of product claim 8 of the main request*

3. Product claim 8 relates to a stabilised passivated copper catalyst, characterized among other things by having a thin copper carbonate layer on the copper surface.

- 3.1 The Opponents, based on document (13), alleged that a thin layer of copper carbonate could not be obtained on the copper surface of the catalyst, as "normal" copper (II) carbonate, i.e. the compound of formula  $\text{CuCO}_3$ , did not exist. They concluded therefore that the only possible copper carbonate which could be present on the surface of the copper should be basic copper carbonate, i.e. a mixture comprising  $\text{CuCO}_3$  and  $\text{Cu}(\text{OH})_2$ , which however necessitated the use of water during the passivation step.
- 3.2 The presence of a thin copper carbonate layer on the copper surface of the claimed passivated catalyst is discussed in paragraphs [0017], [0019] and [0031] of the patent in suit. According to paragraph [0017], *"it is believed that the passivation is effected as a result of the formation of a very thin copper carbonate, possibly cuprous carbonate, layer on the copper surface of the catalyst: calculations based upon the amount of oxygen employed and the copper surface area of the catalyst in a typical passivation, show that it is probable that only the first few, e.g. 2, layers of copper atoms at the catalyst surface react to form the copper carbonate"*. According to example 1 (paragraph [0031]) and to paragraph [0019], the presence of a layer of copper carbonate is demonstrated by X-ray photoelectron spectroscopy (XPS). It can also be observed by infra-red spectroscopy (paragraph [0019]). Furthermore, the patent in suit does not describe, in addition to carbon dioxide and oxygen, the use of water, which would be necessary in order to obtain basic copper carbonate. It follows therefore that the thin layer of copper carbonate referred to in claim 8 of the patent in suit has to be understood in the light of the



specification as being a material formed of (emphasis added by the Board) carbonate and copper ions, possibly cuprous ions, which copper was before passivation present in the first few layers of copper atoms at the surface of the catalyst.

- 3.3 The Board does not dispute that pure copper (II) carbonate, i.e. the compound per se, is not known, as it has not been reported to have been isolated. This fact is part of the general knowledge of the skilled person as for example evidenced by document (13). The teaching of document (13), however, only relates to the impossibility of obtaining a pure copper (II) carbonate, i.e. a compound made solely of copper (II) carbonate. It does not indicate that the surface of a copper bulk material such as a copper catalyst, could not be covered by carbonates only, linked to copper ions, possibly cuprous ions, present in the first few layers of copper atoms at the surface of the catalyst surface.
- 3.4 The Opponents did not provide any experimental evidence that following the passivation method disclosed in the patent in suit, one would not obtain a passivation layer as defined in above point 3.2. According to the established jurisprudence of the Boards of Appeal, each of the parties to the proceedings carries the burden of proof for the facts it alleges. In the present case, where the Opponents rely on the impossibility to obtain a thin layer of copper carbonate on the surface of the copper catalyst, contrary to the experimental evidence presented in the patent in suit, the burden of proof for those facts lies on their side. The Opponents' allegation at the oral proceedings before the Board that X-ray photoelectron spectroscopy (XPS), contrary

to the Patentee's assertion in paragraph [0019] of the patent, was not a method which would allow to detect the presence of a thin layer of copper carbonate on the surface of a copper catalyst is not corroborated by any evidence. Having not presented any convincing evidence for their contention, the Opponents have not discharged their burden of proof. Thus, the unsubstantiated allegation, that copper carbonate can only mean basic copper carbonate and, consequently, that water must have been used in addition to carbon dioxide and oxygen in the passivation process, must be disregarded.

*Novelty of product claim 8*

4. The Opponents objected to the novelty of the subject-matter of claim 8 in view of either of document (3) or (10).

- 4.1 Novelty over document (3)

Document (3) presents the results of a study of the oxidative interaction and stabilization of a wide variety of pyrophoric catalysts with carbon dioxide, water vapour or a mixture of two or three oxidants. One of the catalysts tested is a copper-containing low-temperature shift catalyst (HTK-4) (page 326, lines 7-9 and 18-24, page 329, figure 4) which is treated with a mixture of carbon dioxide, water vapour and oxygen. Document (3) does not describe the nature of the protective layer obtained on that copper catalyst. It also fails to give any detail on the proportions of carbon dioxide and oxygen in the gas flow used for passivation. It follows from paragraph [0020] of the patent in suit that a mixture comprising carbon dioxide

and oxygen does not necessarily lead to the formation of a carbonate layer on the copper surface, as it is critical that the ratio of carbon dioxide to oxygen exceeds a certain value, in line with the requirement in claim 1 according to which the amount of carbon dioxide must be at least twice the amount of oxygen. Thus, in the absence of any disclosure for the ratio of carbon dioxide to oxygen used in document (3), it cannot be assumed that the process employed in document (3) leads to the existence of a carbonate layer on the copper catalyst. Summing up, the formation of thin layer of copper carbonate on the copper catalyst used in document (3) is neither explicitly disclosed, nor can it be deduced from the described treatment conditions, nor has any experimental evidence in the form of an attempted reworking of the process of document (3) been provided by the Opponents showing that such a carbonate layer could be determined to exist by the measurement methods suggested in the patent in suit. The Opponents' line of argument that the subject-matter of claim 8 was anticipated by the stabilized copper catalyst of document (3) has not been made out and the novelty of the subject-matter of claim 8 over document (3) therefore has to be acknowledged.

#### 4.2 Novelty over document (10)

Document (10) is an excerpt of an inorganic chemistry textbook describing chemical properties of copper. It discloses that a copper surface oxidizes in the presence of air so as to slowly form copper (I) oxide and that in the presence of carbon dioxide a plating of green basic copper carbonate referred to as "patina" is built up, which protects the copper from further

destruction. To illustrate this phenomenon, the Opponents referred to the green coating on copper roofs. However, a layer of basic copper carbonate does not fall under the definition of a thin copper carbonate layer the meaning of which is explained in above point 3.2. Furthermore, document (10) fails to disclose that the copper below the plating of green basic carbonate is suitable as a catalyst, which would require that the copper is explicitly or implicitly defined as to be in a form allowing catalytic activities. Hence, document (10) cannot be considered to directly and unambiguously disclose the subject-matter of claim 8 of the patent under dispute and the novelty of claim 8 of the patent in suit over document (10) also has to be accepted.

- 4.3 Thus, the objection to lack of novelty raised by the Opponents are not founded.
- 4.4 Although the novelty of claim 8 over document (1) was not objected to by the Opponents, the Board notes for the sake of completeness, that document (1) does not disclose the use of a mixture of carbon dioxide and oxygen for the stabilization step (see points 2.3 to 2.6 above), which is essential to obtain a thin layer of copper carbonate. Thus, novelty of the subject-matter of claim 8 of the patent in suit over document (1) is also acknowledged.

*Inventive step of process claims 1 to 7 of the main request*

5. *Closest state of the art*

5.1 The closest prior art for assessing inventive step is normally a prior art document disclosing subject-matter conceived for the same purpose or aiming at the same objective as the claimed invention and having the most relevant technical features in common, i.e. requiring the minimum of structural and functional modifications (Case Law of the Boards of Appeal of the European Patent Office, 5<sup>th</sup> edition, 2006, I.D.3.1 and 3.2.).

5.2 The patent in suit relates to copper catalysts and to a process for their stabilization (paragraph [0001], claims 1 and 8). The copper catalysts are often supplied as oxidic precursors which require a lengthy reduction step to become active (paragraph [0002]). They are normally not supplied in the reduced state as they tend to be pyrophoric and so present handling difficulties. Their discharging from a reactor is also on the same ground problematic (paragraph [0003]). According to paragraph [0004], the problems of both the lengthy reduction of fresh oxidic copper catalyst precursors and the discharge of spent copper catalysts can be overcome by effecting passivation of the reduced copper. It follows from paragraphs [0003], [0004] and [0008] and example 2, where a charge of about 40 m<sup>3</sup> of a spent catalyst from the synthesis reactor of a commercial methanol plant is treated, that the patent in suit is directed to a process which allows stabilization of very large batches of copper catalyst. Stabilization of copper catalysts by passivation is according to the application as filed already known

from document (1) (paragraph bridging pages 1 and 2, corresponding to paragraph [0005] of the patent in suit), which the patent in suit implicitly describes as the starting point for the present invention (paragraphs [0005] and [0008]).

- 5.3 The Opponents nevertheless argued that document (3) represented the closest state of the art, because it disclosed the passivation of a copper-containing low-temperature shift catalyst (HTK-4) using a mixture of gas comprising carbon dioxide, steam and oxygen.
- 5.4 Document (3) is a study on the oxidative interaction of a wide variety of catalysts with carbon dioxide, water vapour or a mixture of two or three oxidants. It is apparent from the abstract of this document and the part describing the experimental conditions (page 326, third paragraph), that the passivation process disclosed in document (3) is not a process under industrial conditions, but rather a model serving to study the oxidative interaction and stabilisation of some pyrophoric catalysts. As the patent in suit relates to industrial scale processes, as does document (1), the Board considers that the process disclosed in document (1) represents a more realistic starting point for the skilled person, who wishes to provide an industrial process for the stabilization of copper catalysts.

## 6. *Problem solved and solution*

- 6.1 Starting from document (1), the patent in suit aims at a method for passivating copper catalysts which is faster and which results in a stable passivated

catalyst wherein, for catalysts of similar copper crystallite size, a greater proportion of the copper is still in the elemental form (see paragraph [0008]). A greater proportion of the copper in the elemental form after passivation, means that less reduction will be necessary to put the catalyst in its active state. Both passivation step and the activation step are therefore shortened, which provides significant advantages for the customer (see paragraph [0008]).

- 6.2 According to the patent in suit, the solution to the above problem lies in the simultaneous use of carbon dioxide and oxygen as passivating gas, wherein the amount of carbon dioxide is at least twice the amount of oxygen (see paragraph [0009]).
- 6.3 According to example 1 of the patent in suit (paragraph [0031]), X-ray diffraction spectroscopy fails to detect any copper phases except copper metal for a catalyst passivated according to the present method, indicating that no significant bulk oxidation had occurred during the passivation. In addition, the passivated catalyst of example 1 was found to be stable in air below 125°C. From the total weight gain which was observed at temperatures above 125°C in air, which was only slightly less than the weight gain exhibited by a fully reduced sample of the catalyst, it could be deduced that bulk oxidation of the catalyst had not occurred during passivation (paragraph [0032]).
- 6.4 In addition, a comparison of the method described in example 3 with that described in example 5, which is outside of the scope of present claim 1 as no carbon dioxide is used, shows that passivation in the absence

of carbon dioxide leads to bulk oxidation of the catalyst, which is less stable, as can be deduced from the fact that it is prone to self heating in air.

6.5 These results are also in line with the results of oxidation experiments (DSC/TGA) that had been submitted by the Proprietors before the Opposition Division and presented again before the Board with their letter dated 18 August 2004. In those experiments a copper catalyst was passivated with either a mixture of carbon dioxide and oxygen in a ratio of 10 to 1 or with a gas mixture comprising equal amounts of carbon dioxide and oxygen. The oxidation experiments show that the copper catalyst passivated with the higher ratio of carbon dioxide to oxygen exhibited a higher oxidation enthalpy and a larger mass gain, indicating that passivation with the higher ratio of carbon dioxide to oxygen, provided passivated copper catalysts which contained a larger amount of copper in the reduced form.

6.6 The Opponents did not provide any evidence, such as counter-experiments, which demonstrate that the method according to the patent in suit encompasses embodiments which do not result in passivated copper catalysts which are more stable in air below 125°C and contain a larger amount of copper in the reduced form, than those obtained according to passivation method of document (1). They also did not contest, apart from the explanation of the phenomenon leading to the results observed, that the experiments presented by the Proprietors in the patent and in their submissions dated 18 August 2004 provide evidence for more stable passivated copper catalysts containing a larger amount of copper in the reduced form.



6.7 In view of the above, the Board is therefore satisfied that the problem underlying the present invention as defined in above point 6.1 has been successfully solved by process defined in claim 1 of the patent in suit.

7. *Obviousness*

7.1 It remains to be decided whether or not the proposed solution to the problem underlying the patent in suit is obvious in view of the cited state of the art.

7.2 As shown in points 2.3 to 2.6 above, the closest state of the art, document (1), does not disclose the use of carbon dioxide in the passivation step and therefore cannot itself suggest the solution proposed by the patent in suit.

7.3 According to the introductory part of document (3), on page 325, the optimal conditions using oxygen to form an oxide protective layer on catalysts are specific to each type of catalyst. The authors of document (3) also reported that they had already developed an efficient and rapid method for the air stabilization of low-temperature shift catalysts, but said method failing to be universal, they had sought to provide a method which could be universally applied to a broad range of catalysts (page 325, introduction). For this reason, document (3) presents a study of the oxidative interaction of a wide variety of catalysts with carbon dioxide, water vapour or a mixture of two or three oxidants. One of the catalysts studied is a copper containing low-temperature shift catalyst (HTK-4). The results show that carbon dioxide and the mixture of

carbon dioxide, water vapour and oxygen, the proportions of which are not defined in document (3), provide an effective stabilization of that low temperature shift catalyst (see figures 2 and 4, page 329 and page 331, last paragraph). A comparison between figures 2 and 4 show that the copper catalyst passivated with carbon dioxide and the same catalyst passivated with the mixture of carbon dioxide, water vapour and oxygen, exhibit over the entire range of temperature about the same residual pyrophoricity. Therefore, those results do not suggest that an improvement of the stability for the passivated copper catalyst could be achieved using a mixture of carbon dioxide and oxygen. Furthermore, document (3) does not suggest that the passivation using carbon dioxide or a mixture of carbon dioxide, water vapour and oxygen would be an improvement compared to that carried out in previous studies with air or oxygen. The teaching of document (3) is not that the passivation of copper-containing low-temperature shift catalysts with oxygen or air could be improved by the used of carbon dioxide, but that the method using carbon dioxide can be universally applied to a broad range of catalysts. Document (3) is also silent on the ratio between carbon dioxide and oxygen. It is *a fortiori* silent on the importance of the ratio of carbon dioxide to oxygen in order to obtain sufficient protection, while increasing the amount of copper in the reduced form. It therefore follows that the solution proposed by the patent in suit was not obvious in view of document (3) either.

8. It is therefore concluded that the subject-matter of process claim 1 of the main request and dependent

claims 2 to 7 thereon fulfil the requirements of Article 56 EPC.

*Inventive step of product claim 8 of the main request*

9. Document (1) is also considered for the same reasons as given in above point 5. to represent the closest state of the art for analysing inventive step of present claim 8.
  
10. The passivated catalyst of claim 8 is not defined as being obtainable by the process of present claim 1, but is merely characterized in that it has "a thin copper carbonate layer on the copper surface" and that it "does not exhibit substantial oxidation when heated in air from 20°C at a rate of 10°C per minute until the temperature has reached at least 100°C". The term "thin" can only be interpreted consistent with the specification as a thickness which results from the bonding of carbonate with the first few layers of copper atoms present at the surface of the catalyst (see above point 3.2). The number of layers concerned however cannot be more precisely determined in the light of the specification. The second feature concerned is also defined by a substantial absence of oxidation, whereby the specification is also silent on how much oxidation is not considered to be substantial. Even though no precise meaning in the context of the present patent can be attributed to the features "thin" and "does not exhibit substantial oxidation when heated in air from 20°C at a rate of 10°C per minute until the temperature has reached at least 100°C", the subject-matter of claim 8 is distinguished over the passivated catalysts obtained in document (1) by the presence on

the copper surface of a layer formed of copper carbonate.

11. In the absence of any evidence that the mere replacement of copper oxide by copper carbonate on the surface of the copper provides, other things being equal, any technical advantage to the passivated catalyst, the problem solved by the product of present claim 8 over the closest state of the art must be seen in the provision of further passivated copper catalysts.
  
12. As already mentioned in above points 4.1, 4.2 and 4.4 documents (3), (10) and (1) are silent on the possibility of obtaining copper carbonate on the copper surface of the catalyst. Document (10), in particular, does not relate to a layer of copper carbonate in the meaning of the patent in suit, i.e. a layer formed of (i) copper ions, possibly cuprous ions, present at the first few layers of copper atoms at the surface of the catalyst and (ii) carbonate, but to a layer of basic copper carbonate (see point 4.2 above), which contains copper (II) hydroxide. As the Opponents have not argued that the skilled person was aware of the possibility to obtain a thin layer of copper carbonate on the copper surface of the catalyst in the meaning of the patent in suit and the Board is not aware of such prior art teaching, it is concluded that it would have not been obvious for a skilled person to arrive at the subject-matter of present claim 8, even if he merely wanted to provide further passivated copper catalysts.
  
13. The subject-matter of product claim 8 of the main request is therefore inventive within the meaning of Article 56 EPC.

14. The main request being thus allowable, it is not necessary to consider either of the auxiliary requests.

*Procedural issues - late filed evidence (Article 114(2) EPC)*

15. According to the Rules of Procedures of the Boards of Appeal (RPBA), the statement of grounds of appeal and the reply thereto shall contain a party's complete case including all the facts, arguments and evidence relied on (see Article 10a, paragraph (2), of the RPBA in the version of 12 December 2002, published in OJ EPO 2003, 62). Any amendment to a party's case after it has filed its grounds of appeal or reply may be admitted and considered at the Board's discretion (see Article 10b, paragraph (1), of the RPBA, above).
16. The Opponents submitted documents (13), (14) and (15) with their letter dated 25 May 2007, i.e. more than three years after filing of their statement setting the grounds for appeal. The Patent Proprietors submitted in turn document (16) with their facsimile letter dated 6 December 2007. The Opponents submitted in addition documents (17) and (18) with a further letter dated 6 December 2007 and document (19) during the oral proceedings.
17. Documents (13), (17) and (18) have been submitted in order to show that pure copper (II) carbonate does not exist. Documents (14) and (15) have been submitted as closest prior art for the analysis of inventive step, should the claimed process not form a copper carbonate layer, but a copper oxide layer. It follows from the present decision that the impossibility of forming pure

copper (II) carbonate or the formation in the claimed process of a copper oxide layer are not relevant issues for the present decision. In addition, documents (14) and (15) are not more relevant than document (1) for the analysis of inventive step of the claimed subject-matter, as documents (14) and (15), contrary to document (1) does not focus on a method for passivating copper catalysts, but on a method for passivating nickel catalyst.

18. Document (16) are experimental data in support of the formation of a layer of copper carbonate on the copper surface of the catalyst when applying the method of claim 1 under dispute. As the Board has found that the experimental evidence on file already supports the formation of a layer of copper carbonate on the copper surface of the catalyst (see above points 3.2 to 3.4), document (16) can also be disregarded (Article 114(2) EPC). Document (19) is a combined TGA and Differential Scanning Calorimetry (DSC) measurement of a commercial sample of basic copper carbonate. It was submitted by the Opponents in order to show that the material tested in document (16) by TGA could not be basic copper carbonate. Document (19) is also of no relevance to the present decision, because document (16) is itself not relevant to the present decision and the passivation layer obtained with the method of the patent in suit has not been shown to be formed of basic copper carbonate as alleged by the Opponents.
19. Consequently, the Board, exercising its discretion under Article 114(2) EPC, decides to disregard documents (13) to (19).

**Order**

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

1. The decision under appeal is set aside.

2. The patent is maintained as granted.

The Registrar

The Chairman

C. Eickhoff

S. Perryman