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**Datasheet for the decision  
of 5 February 2009**

**Case Number:** T 0839/07 - 3.3.06

**Application Number:** 99916674.7

**Publication Number:** 1086505

**IPC:** H01M 8/18

**Language of the proceedings:** EN

**Title of invention:**  
Catalyzed wall fuel gas reformer

**Applicant:**  
UTC Fuel Cells, LLC

**Opponent:**  
-

**Headword:**  
Catalyzed alumina washcoat/UTC

**Relevant legal provisions:**  
EPC Art. 123(2), 56

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

**Keyword:**  
"Added subject-matter (no)"  
"Inventive step (yes)"

**Decisions cited:**  
-

**Catchword:**  
-



Case Number: T 0839/07 - 3.3.06

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.06  
of 5 February 2009

**Appellant:**

UTC Fuel Cells, LLC  
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South Windsor  
CT 06074 (US)

**Representative:**

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**Decision under appeal:**

Decision of the Examining Division of the  
European Patent Office posted 29 December 2006  
refusing European application No. 99916674.7  
pursuant to Article 97(1) EPC.

**Composition of the Board:**

**Chairman:** P.-P. Bracke  
**Members:** L. Li Voti  
J. Van Moer

## Summary of Facts and Submissions

I. This appeal lies from the decision of the Examining Division to refuse European patent application no. 99 916 674.7, relating to a hydrocarbon fuel gas steam reformer assembly.

II. As regards claim 1 according to then pending main request the Examining Division, referring to documents

(1): US-A-5733347;

(2): US-A-4522894; and

(3): US-A-4900712,

found in its decision, *inter alia*, that

- the claimed subject-matter was novel over the cited prior art;

- the technical problem underlying the invention concerned the stabilization of the used washcoat against high temperature recrystallization of alumina and the hindrance of carbon deposition on the surface of the walls of the fuel gas passages;

- the closest prior art was represented by document (1), disclosing a hydrocarbon fuel gas steam reformer assembly having a portion of the walls of the fuel gas passages coated with a catalyzed alumina washcoat;

- the subject-matter of claim 1 according to the main request differed from that disclosed in document (1) insofar as the alumina washcoat comprised a lanthanum oxide compound;

- however, it was known from document (2), relating to an autothermal reformer operating at a temperature slightly higher than that used in a fuel gas steam reformer according to the present application, and from document (3), relating to automobile coated catalytic converters, that the addition of lanthanum oxide to a catalyzed alumina washcoat would stabilize it against the recrystallization of alumina at high temperature;

- therefore, the skilled person would have expected that the same effect would occur by adding lanthanum oxide to the catalyzed alumina washcoat of the hydrocarbon fuel gas steam reformer assembly of document (1);

- the subject-matter of claim 1 according to the main request thus lacked an inventive step.

As regards the then pending first auxiliary request the Examining Division found additionally that

- document (1) already disclosed the use of nickel as a catalyst for the washcoat in the fuel gas passages of the fuel gas steam reformer assembly and taught also that the burner gas passages could be washcoated and catalyzed;

- therefore, the subject-matter of claim 1 according to the first auxiliary request differed from that disclosed in document (1) insofar as the alumina washcoat comprised a lanthanum oxide compound and the washcoat of the burner passages was catalyzed with a noble metal or with a mixture of noble metals;

- there was no evidence that the claimed catalysts combination would provide an unexpected effect over the prior art and that the use of a nickel catalyst would allow to inhibit the deposition of carbon on the surface of the fuel gas passages;
- to the contrary, it was known from document (2) that nickel was inferior to noble metals as a reformer catalyst for inhibiting carbon deposition;
- therefore, the technical problem underlying the invention of stabilizing the washcoat against high temperature recrystallization of alumina and inhibiting carbon deposition on the surface of the walls of the fuel gas passages in order to optimize the working of the reformer assembly could not be considered to have been successfully solved by means of the combination of catalysts used according to claim 1;
- the use of noble metals as catalysts for oxidation/burner processes was known from document (2);
- since all the catalysts used according to claim 1 were already known from the prior art for their use and performance in reforming processes, it would have been obvious for the skilled person to use these catalysts in the washcoats used according to the teaching of document (1);
- therefore, the claimed subject-matter did not involve an inventive step.

III. An appeal was filed against this decision by the Applicant (Appellant).

The Appellant submitted with letter of 22 January 2009 an amended set of claims, claim 1 of which reads as follows:

"1. A hydrocarbon fuel gas steam reformer assembly comprising:

a) a plurality of fuel gas passages (24) for receiving a mixture of fuel gas and steam;

b) a plurality of burner gas passages (14) for receiving a burner gas stream, said burner gas passages (14) and said fuel gas passages (24) being disposed in heat exchange relationship with each other whereby heat from said burner passages (14) is transferred to said fuel gas passages(24);

c) a plurality of process gas regenerator passages (26) for receiving gas flowing from said fuel gas passages (24), said process gas regenerator passages (26) and said fuel gas passages (24) being disposed in heat exchange relationship with each other;

d) a washcoat (28) being provided on the walls of said fuel gas passages (24), said washcoat (28) being formed of 2 to 5% by weight lanthanum oxide, 5 to 20% by weight calcium oxide, 0 to 20% by weight cerium oxide, and the remainder alumina, which washcoat composition stabilizes alumina against recrystallization at elevated temperatures and also inhibits carbon deposition on the walls of said fuel gas passages (24); and

e) said washcoat (28) on the walls of said fuel gas passages (24) being catalyzed."

Dependent claims 2 to 9 relate to particular embodiments of the assembly of claim 1.

IV. The Appellant argued in writing *inter alia* that

- the amended claims did not contravene the requirements of Article 123(2) EPC;

- starting from document (1), the technical problem underlying the invention had to be seen in the provision of a hydrocarbon fuel gas steam reformer similar to that disclosed in document (1) but having a longer failure-free useful life;

- this technical problem had been solved by adding specific amounts of lanthanum oxide, calcium oxide and, possibly, cerium oxide to the alumina washcoat, which combination of oxides acts synergistically in stabilizing the alumina washcoat against recrystallization at high temperature and in inhibiting the formation of carbon deposits on the washcoated walls;

- the selected combination of oxides was much more effective in the lower temperature environment of the fuel gas reformer used according to the invention than it would have been in the higher temperature environment of the autothermal reformer disclosed in document (2);

- since the prior art did not suggest the use of the selected combination of oxides for obtaining the above mentioned combination of effects, the claimed subject-matter involved an inventive step.

- V. The Appellant requests that the decision under appeal be set aside and that a patent be granted on the basis of claims 1 to 9, submitted with letter of 22 January 2009.

### **Reasons for the Decision**

1. Article 123(2) EPC

The Board is satisfied that claims 1 to 9 are supported by the original documents of the application, reference being made hereinafter to the published version WO 99/53561.

In particular, claim 1 finds support in claims 1 and 3 read in combination with page 2, lines 12 to 16 and 21 to 22; page 2, line 29 to page 3, line 6; page 3, lines 24 to 29; and in figures 2 to 4 in combination with page 5, lines 5 to 25.

Claim 2 finds support in claim 4.

Claim 3 finds support in page 5, lines 16 to 18 in combination with the passage from page 5, line 33 to page 6, line 2.

Claim 4 finds support in page 3, lines 7 to 10 read in combination with the passage from page 2, line 33 to page 3, line 6 and that on page 5, lines 33 to 34.

Claims 5 to 9 find support in claims 5 to 9.



Therefore, claims 1 to 9 comply with the requirements of Article 123(2) EPC.

2. Novelty

The Board has no reason to depart from the finding of the department of first instance that the claimed subject-matter is novel over the cited prior art.

3. Inventive step

- 3.1 The present invention regards a hydrocarbon fuel gas steam reformer assembly, wherein the reformer fuel gas passages are washcoated with a catalyzed alumina composite, which composite is formed of 2 to 5% by weight lanthanum oxide, 5 to 20% by weight calcium oxide, 0 to 20% by weight cerium oxide, the remainder being alumina.

As explained in the application, the fuel cell power plants normally used in the prior art included fuel gas steam reformers which catalytically convert a fuel gas, such as natural gas, into hydrogen, carbon monoxide and carbon dioxide. This conversion involves passing a mixture of the fuel gas and steam through a catalytic bed which is heated to a reforming temperature of about 1250 to 1600°F (677 to 871°C). Catalysts typically used were nickel catalysts deposited on alumina pellets. A typical reformer consisted of a plurality of reaction tubes contained in a housing that is insulated for heat retention. In such a reformer the reaction tubes were heated by burning excess fuel gas in the housing and passing the burner gases over the reaction tubes (see page 1, lines 12 to 20).

However, such steam reformers required a large catalyst bed surface area in order to provide a high degree of catalyst-fuel mixture interaction as well as a large heat transfer surface area to produce the amount of hydrogen required to operate the fuel cells at peak efficiency. This resulted in undesirably large and heavy reformer assemblies (page 1, line 30 to page 2, line 6).

Therefore, an improved steam reformer structure had been disclosed in document WO 97/24176, corresponding to document (1). This steam reformer structure provides the necessary catalyzed and heat transfer surface areas though being substantially smaller and lighter than the previously available steam reformers, and can be operated at lower service temperatures. The improved steam reformer structure is formed from a series of essentially flat plate reformer components. Each of the reformer components includes reformer passages which are adjacent to a plurality of central regenerator/heat exchanger passages and each of the reformer passage plate units is disposed directly adjacent to a burner passage plate unit so that the adjacent reformer and burner passages share a common wall. All of the metal sheets which make up the flat plate reformer and some of the burner components of the assemblage may have surfaces covered with a catalyzed alumina coating, wherein the surfaces to be catalyzed can be primed by means of a conventional washcoating process (page 2, lines 7 to 21).

The technical problem underlying the invention thus is defined in the present application as the provision of

an improved catalyzed washcoat to be used in a compacter steam reformer assembly as disclosed in WO 97/24176, which washcoat is stable against high temperature recrystallization of alumina and inhibits carbon deposition on the washcoated surfaces (see page 2, lines 28 to 30; page 3, line 30 to page 4, line 10).

- 3.2 Since document (1), the U.S. patent application corresponding to the above cited international application WO 97/24176 discussed hereinabove, is the only prior art document cited relating to an improved compacter fuel gas steam reformer assembly having surface covered with a catalyzed washcoat, the Board finds that document (1) represents the most suitable starting point for the evaluation of inventive step.

Starting from document (1), the technical problem underlying the invention has to be defined as reported in the present application as the provision of an improved catalyzed washcoat which is stable against high temperature recrystallization of alumina and inhibits carbon deposition on the washcoated surfaces.

- 3.3 As taught in the present application, the inclusion of lanthanum oxide in the catalyzed alumina washcoat acts synergistically against high temperature recrystallization of alumina whilst the selected combination of lanthanum, calcium and cerium oxide acts synergistically in suppressing carbon deposition thereby prolonging the operational life of the assembly (see page 3, lines 24 to 29; page 6, lines 19 to 23).

The Board has no reason to contest that the use of the selected combination of oxides in the catalyzed alumina washcoat provides the above mentioned technical effect.

Moreover, even though document (2) shows that the use of nickel on calcium/alumina as a steam reforming catalyst in an autothermal reformer does not appear to prevent completely carbon deposition (column 16, lines 31 to 44 and column 17, lines 16 to 20), this teaching concerns a reformer catalyst which is different from that used according to claim 1 of the present application as it does not contain a combination of lanthanum oxide, calcium oxide and, possibly, cerium oxide. Moreover, such a catalyst is used at a reforming temperature of 942°C, higher than the maximum temperature used in a compacter steam reformer assembly as claimed, which temperature is **lower than 871°C** (see page 1, lines 12 to 16 in combination with page 2, lines 7 to 11 of the present application).

Therefore, there is no reason to believe that by using nickel as a catalyst on an alumina composite containing the combination of oxides of claim 1, the suppression of carbon deposition taught in the present application would not be achieved.

The Board thus finds that the above mentioned technical problem has been convincingly solved by means of a washcoat having a composition according to claim 1.

- 3.4 The compact fuel gas reformer assembly disclosed in document (1) differs from that according to claim 1 of the present application insofar as the catalyzed

alumina washcoat does not comprise the selected combination of oxides of claim 1 (see document (1), claim 1; column 2, lines 15 to 20; column 3, lines 38 to 65; figure 2).

Moreover, this document does not contain any suggestion that the selection of a particular combination of oxides could be helpful for stabilizing the washcoat against high temperature recrystallization of alumina and for inhibiting carbon deposition on the washcoated surfaces.

- 3.5 Document (2) relates to an autothermal reformer. At variance with the compact fuel gas reformer assembly of document (1), which is only operable to convert a fuel gas into hydrogen and carbon dioxide, the autothermal reformer of document (2) includes a first catalyst zone wherein a heavy hydrocarbon feedstock is cracked to lighter hydrocarbons by means of a catalytic partial oxidation and a second catalyst zone wherein the lighter hydrocarbons are subjected to steam reforming thereby producing hydrogen and carbon dioxide (see document (1), column 1, lines 14 to 16; and document (2), claim 1; column 1, line 62 to column 2, line 18; column 3, lines 48 to 54; column 5, lines 14 to 21).

Both the first and second catalyst zones of the autothermal reformer of document (2) are run at a temperature between 871 and 1316°C (see column 4, lines 18 to 22), i.e. at a temperature higher than that used in the fuel gas reformer according to document (1), which is lower than 871°C (see column 1, lines 16 to 21 in combination with lines 54 to 58). Moreover, the catalysts used for oxidation and for steam reforming

are preferably platinum group metals on a stabilized alumina (see document (2), column 4, lines 47 to 57).

Document (2) teaches that alumina can be stabilized against the undesirable high temperature phase transition to alpha alumina by adding one or more rare earth metal oxides and/or alkaline earth metal oxides, such as oxides of one or more of lanthanum, cerium, praseodymium, calcium, barium, strontium, magnesium, the preferred combination being a combination of lanthanum oxide and barium oxide (see column 7, line 59 to column 8, line 2; column 13, lines 29 to 34).

Furthermore, document (2) teaches that the platinum group metal catalysts are effective in preventing carbon deposition (column 8, lines 17 to 18 and 26 to 30; column 9, lines 33 to 38; column 17, lines 23 to 27).

Therefore, even though the skilled person, by following the teaching of document (2) could have tried one or more of the oxides mentioned above for stabilizing alumina against high temperature recrystallization, he would not have found in this document any suggestion that a combination, by weight, of 2 to 5% lanthanum oxide, 5 to 20% calcium oxide and 0 to 20% cerium oxide as selected in claim 1 of the patent in suit, could have any inhibitory effect against carbon deposition at the lower temperatures used in the steam reformer of document (1).

- 3.6 Document (3), relating to automobile coated catalytic converters, teaches that an alumina washcoat used in an automotive exhaust can be stabilized against high

temperature recrystallization of alumina by using oxide dopants like titanium, zirconium, cerium, lanthanum, iron, nickel or chromium oxide (column 1, lines 5 to 21; column 2, lines 27 to 40); the examples show the excellent properties of a combination of cerium and lanthanum oxide (column 9, lines 22 to 33).

However, this document, though suggesting the use of cerium and lanthanum oxides for stabilizing alumina against high temperature recrystallization, does not contain any hint that a combination, by weight, of 2 to 5% lanthanum oxide, 5 to 20% calcium oxide and 0 to 20% cerium oxide as selected in claim 1 of the present application, which combination necessarily comprises calcium oxide, not mentioned in document (3), could have any inhibitory effect against carbon deposition at the temperatures used in the steam reformer of document (1).

- 3.7 Therefore, the skilled person would not have found in the prior art any hint to use the selected combination of oxides in the catalyzed alumina washcoat of document (1) in order **both** to stabilize the washcoat against high temperature recrystallization of alumina and to inhibit carbon deposition on the washcoated surfaces.

Therefore, the subject-matter of claim 1 involves an inventive step.

The same arguments apply *mutatis mutandis* to the dependent claims.

**Order**

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

The decision under appeal is set aside.

The case is remitted to the department of first instance with the order to grant a patent on the basis of claims 1 to 9 submitted with letter of 22 January 2009 and a description to be adapted thereto.

The Registrar:

The Chairman:

G. Rauh

P.-P. Bracke