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**Datasheet for the decision
of 26 May 2010**

Case Number: T 1409/08 - 3.3.06

Application Number: 02018807.4

Publication Number: 1287876

IPC: B01D 53/94

Language of the proceedings: EN

Title of invention:

Composite oxide, process for producing the same, and exhaust
gas reducing co-catalyst

Patentee:

KABUSHIKI KAISHA TOYOTA CHUO KENKYUSHO

Opponent:

RHODIA CHIMIE

Headword:

Composite oxide/RHODIA

Relevant legal provisions:

EPC Art. 123(2)

Relevant legal provisions (EPC 1973):

-

Keyword:

"Added subject-matter (all requests): yes"

Decisions cited:

-

Catchword:

-

Summary of Facts and Submissions

- I. The present appeal is from the decision of the Opposition Division to revoke the European patent no. 1 287 876 concerning a composite oxide and a process for producing it.
- II. In its notice of opposition the Opponent sought revocation of the patent on the grounds of Article 100(a) EPC, because of lack of novelty and inventive step of the claimed subject-matter, and of Article 100(c) EPC.
- III. The Opposition Division found in its decision *inter alia* that
- claims 1 and 4 according to the then pending main request complied with the requirements of Articles 84, 123(2) and (3) EPC;
 - however, the subject-matter of claim 4 lacked novelty;
 - furthermore, the two auxiliary requests submitted during oral proceedings were not admitted.
- IV. An appeal was filed against this decision by the Patent Proprietor (Appellant).

The Appellant submitted with letter of 30 September 2008 three sets of amended claims according to the main request and the first and second auxiliary requests, respectively.

Oral proceedings were held before the Board on 26 May 2010.

V. The independent claims 1 and 4 of the set of claims according to the **main request** read as follows:

"1. A process for producing a composite oxide, comprising the steps of:
adding a precipitant to an aqueous solution consisting of a cerium compound, a zirconium compound and an aluminium compound, the remainder being water and inevitable impurities, stirring the resulting solution at a shear rate of 1,000 sec^{-1} or more, thereby generating precipitates by a co-precipitation method; calcining the precipitates; and reducing the precipitates by heating to and holding at a temperature of from 700 to 1,200°C in a reducing atmosphere."

"4. A composite oxide obtainable by the process of any one of claims 1 to 3, consisting of CeO_2 , ZrO_2 and Al_2O_3 , the remainder being inevitable impurities, wherein the composite oxide has a regularly oriented phase in which at least a part of Ce cations and Zr cations is oriented regularly, and wherein the composite oxide has a specific surface area of $20\text{m}^2/\text{g}$ or more."

Both claims 1 and 4 according to the **first auxiliary request** differ from claims 1 and 4 according to the main request insofar as the atomic ratio of Al (aluminium) to the sum of Ce (cerium) and Zr (zirconium) ($\text{Al}/(\text{Ce}+\text{Zr})$) has to fall within the range of 1/5 to 5/1.

The set of claims according to the **second auxiliary request** consists of the process claims 1 to 3 of the main request.

VI. The Appellant submitted in writing and orally *inter alia* that

- the invention disclosed in the documents of the application as originally filed related to a binary or a ternary composite oxide; the binary composite oxide contained CeO₂ (cerium oxide) and ZrO₂ (zirconium oxide) as major components whilst the ternary composite oxide contained an additional metallic oxide being free from reacting with the previously mentioned oxides at 700°C or more, preferably Al₂O₃ (aluminium oxide), and did not require CeO₂ and ZrO₂ to be the major components;

- the description disclosed explicitly that the amount of the additional metallic oxide could be greater than that of CeO₂ and ZrO₂; moreover, all the quantitative limitations for such an additional metallic oxide indicated in the description related to preferred embodiments and were not compulsory;

- therefore, the description of the original application contained a support for claim 4 according to the main request;

- process claim 1 according to the main and the second auxiliary request was supported by the original claim 10 read in combination with the description;

- the additional technical feature of claims 1 and 4 according to the first auxiliary request was supported

by paragraph 36 of the original application as published;

- therefore, also these claims complied with the requirements of Article 123(2) EPC.

VII. The Respondent (Opponent) submitted in writing and orally *inter alia* that

- the application as originally filed did not relate to two distinct embodiments of the invention, one being a binary composite oxide containing CeO_2 and ZrO_2 as major components and the other being a ternary composite oxide not comprising any longer the quantitative requirements of the binary composite oxide;

- the invention originally disclosed concerned only a composite oxide containing CeO_2 and ZrO_2 as major components and additional metallic oxides; this interpretation was supported by claims 1 to 3 and by the examples of the invention, all of them relating to ternary composite oxides containing a major amount of CeO_2 and ZrO_2 , whilst a binary composite oxide of CeO_2 and ZrO_2 without further metallic oxides was only indicated as comparative (comparative example 4);

- the process of original claim 10 not containing any limitation as to the amounts of the various compounds used had to be interpreted in the light of the description wherein the described process was directed to the preparation of the composite oxide disclosed therein, i.e. one containing CeO_2 and ZrO_2 as major components;

- moreover, even though the description related in some other parts to composite oxides containing additional metallic oxides in amounts greater than that of CeO_2 and ZrO_2 , there was no disclosure of a composite oxide wherein Al_2O_3 as the additional oxide could be contained in any possible amount;

 - therefore, claims 1 and 4 according to the main request and claim 1 according to the second auxiliary request did not comply with the requirements of Article 123(2) EPC;

 - furthermore, the additional technical feature of claims 1 and 4 according to the first auxiliary request, disclosed in paragraph 36 of the application as filed, was a preferred feature which could not be selected independently from the other preferred features also disclosed in the same paragraph, such as the atomic ratio of Ce to Zr; in fact, all these technical features were taught to have an effect on both the OSC (oxygen storage-and-release capability) and the specific surface area of the composite oxide;

 - therefore, also the claims according to the first auxiliary request did not comply with the requirements of Article 123(2) EPC.
- VIII. The Appellant requests that the decision under appeal be set aside and that the patent be maintained on the basis of any of the main request or of the first or second auxiliary requests submitted with letter of 30 September 2008.
- IX. The Respondent requests that the appeal be dismissed.

Reasons for the Decision

1. Main request

1.1 *Article 123(2) EPC*

- 1.1.1 It is the established case law of the Boards of Appeal of the EPO that the relevant question to be decided in assessing whether an amendment adds subject-matter extending beyond the content of the application as filed is whether such an amendment was directly and unambiguously derivable from the application as filed (Case Law of the Boards of Appeal of the EPO, 5th edition, 2006, III.A.2 and 2.1).

Claim 4 according to the main request requires that the claimed composite oxide consists of CeO_2 (cerium oxide), ZrO_2 (zirconium oxide) and Al_2O_3 (aluminium oxide), the remainder being inevitable impurities (see point V above). Therefore, according to this claim the claimed composite oxide can comprise any possible concentration of these three oxides, i.e. also a greater amount of Al_2O_3 with respect to the other oxides.

It thus has to be evaluated if a composite oxide containing any possible amount of the above mentioned oxides was directly and unambiguously derivable from the application as filed.

- 1.1.2 Paragraph 16 of the application as filed (reference being made hereinafter to the published version of the application) relates to the summary of the invention and discloses that the composite oxide according to the invention comprises CeO_2 and ZrO_2 as major components

whilst the following paragraph 17 specifies that the composite oxide can desirably be composed of CeO_2 , ZrO_2 and a further metallic oxide being free from reacting with the previously mentioned oxides at 700°C or more, which further oxide can be desirably Al_2O_3 .

Since there is no indication that paragraph 17 refers to an alternative embodiment of the invention which is distinct from that of paragraph 16, the embodiment of paragraph 17 can only be interpreted in the Board's view as a preferred embodiment of the composite oxide disclosed in the preceding paragraph, wherein CeO_2 and ZrO_2 are major components and other oxides may be present. This interpretation is confirmed by claim 1 which concerns a composite oxide wherein CeO_2 and ZrO_2 are the major components, and by claim 3, dependent on claim 1, according to which a further oxide such as Al_2O_3 can be comprised in the composite oxide containing CeO_2 and ZrO_2 as the major components.

Therefore, neither this part of the description nor the product claims can be seen as a support for the composite oxide of claim 4 according to the main request in which Al_2O_3 can be present in largely preponderant amounts with respect to CeO_2 and ZrO_2 .

The fact that the composite oxide according to the invention comprises CeO_2 and ZrO_2 as major components is repeated in paragraphs 26 and 33.

Moreover, paragraph 33 discloses also that when the CeO_2 and ZrO_2 based composite oxide includes further oxides such as Al_2O_3 , these oxides can desirably occupy up to 70 atomic % of the entire composite oxide.

Therefore, it can be derived directly and unambiguously from the description that the composite oxide of the invention does not necessarily contain major amounts of CeO_2 and ZrO_2 but has to contain the amount of these oxides necessary to form a CeO_2 and ZrO_2 based composite oxide, which can have the amount of further oxides indicated in paragraph 33 but not any preponderant amount of them which would not allow the formation of a CeO_2 and ZrO_2 based composite oxide.

This interpretation is confirmed by the following paragraph 34, which clarifies that the CeO_2 and ZrO_2 based composite is a ternary composite oxide of CeO_2 , ZrO_2 and a metallic oxide being free from reacting with the previously mentioned oxides at 700°C or more, e.g. Al_2O_3 . Therefore, the disclosures of paragraphs 33 and 34 do not contradict the previous disclosures of the description relating to composite oxides having as major components CeO_2 and ZrO_2 .

A similar disclosure is contained in paragraph 36, according to which the CeO_2 and ZrO_2 based composite oxide containing further metallic oxides being free from reacting with CeO_2 and ZrO_2 at 700°C or more has a preferred ratio of the additional metals, e.g. Al, to Ce and Zr in the range of 1/5 to 5/1.

Therefore, the part of the original description relating to the product of the invention does not disclose directly and unambiguously that Al_2O_3 can be contained in the composite oxide of the invention in any possible amount but only that, if this oxide is selected as preferred further metallic oxide, then the

relative amounts of the oxides must be compatible with the composition of a CeO_2 and ZrO_2 based composite oxide.

The Board thus cannot agree with the Appellant's submission that this part of the description relates to two distinct embodiments, one relating to a binary composite oxide wherein CeO_2 and ZrO_2 are the major components and one to a ternary composite oxide wherein the oxides contained therein do not require any limitation as to their relative amounts.

- 1.1.3 The method of preparation of the CeO_2 - ZrO_2 based composite oxide or ternary composite oxide disclosed from paragraph 41 onwards as well as the process of preparation disclosed in the summary of the invention in paragraph 21 do not specify Al_2O_3 as the further metallic oxide and can only be understood as being directed to the preparation of the composite oxides described in the respective preceding parts of the description which do not allow any possible amount of the further metallic oxides being free from reacting with CeO_2 and ZrO_2 at 700°C or more as explained hereinbefore.

Similarly, even though the wording of process claim 10 does not contain any limitation as to the amounts of the various compounds used, it also does not relate to the presence of a compound of Al. Therefore, by reading this claim in combination with the description wherein the oxide of Al is considered to be a desirable component of the final composite oxide, it can only be derived that the amount of such a further oxide has to be limited.

1.1.4 This interpretation of the description is corroborated by the examples since all the examples of the invention relate to a composite oxide containing CeO_2 , ZrO_2 and Al_2O_3 wherein the oxides of Ce and Zr constitute the major amount whilst a binary composite oxide of Ce and Zr not containing further metallic oxides is reported as comparison (see comparative example 4 and table 3).

Therefore, the Board concludes that the subject-matter of claim 4 cannot be derived directly and unambiguously from the application as filed.

Claim 4 thus contravenes the requirements of Article 123(2) EPC.

2. First auxiliary request

2.1 *Article 123(2) EPC*

2.1.1 Claim 4 according to the first auxiliary request differs from claim 4 according to the main request insofar as the atomic ratio of Al to the sum of Ce and Zr ($\text{Al}/(\text{Ce}+\text{Zr})$) has to fall within the range of 1/5 to 5/1.

Paragraph 36 of the application as filed relates to particular embodiments of the CeO_2 - ZrO_2 based composite oxides containing further metallic oxides being free from reacting with CeO_2 and ZrO_2 at 700°C or more (page 5, lines 39 to 41) already disclosed in the previous paragraph 34 (page 5, lines 19 to 21), which composite oxides can comprise up to 70 atomic % of further oxides (see paragraph 33; page 5, lines 17 to 18)).

According to paragraph 36, Al_2O_3 is especially desirable as further metallic oxide (page 5, line 41).

Furthermore, it is preferred that Ce and Zr in such CeO_2 - ZrO_2 based composite oxides are contained in an atomic ratio of 1/9 to 9/1 in order not to deteriorate the OSC or the specific surface area of the composite oxide (page 5, lines 41 to 46); moreover, the atomic ratio of the metal of the further metallic oxide with respect to Ce and Zr is preferably within the range of 1/5 to 5/1 for similar reasons (page 5, lines 46 to 50).

Therefore, in the Board's view, it can be derived directly and unambiguously from paragraph 36 that the CeO_2 - ZrO_2 based composite oxide contains as preferred features not only the presence of Al_2O_3 but also both ratios indicated in this paragraph, which bring about beneficial effects as to the OSC and the specific surface area of the composite oxide, i.e. the ratio of Ce to Zr and the ratio of the further metal to Ce and Zr.

In fact, also all the examples of the invention comply with both ratios indicated in said paragraph 36 and the description does not contain any indication that only one of these features would be desirable for obtaining such beneficial effect independently from the other equally preferred feature indicated in the same paragraph.

The Board concludes that the selection from this paragraph 36 of a composite oxide containing Al_2O_3 as further metallic oxide being free from reacting with CeO_2 and ZrO_2 at 700°C or more and having the specified

ratio of Al to Ce and Zr without any limitation as to the ratio of Ce to Zr is not directly and unambiguously derivable from the application as filed.

Therefore, claim 4 according to the first auxiliary request contravenes the requirements of Article 123(2) EPC.

3. Second auxiliary request

3.1 *Article 123(2) EPC*

3.1.1 Claim 1 according to the second auxiliary request is identical to claim 1 according to the main request which relates to a process for preparing a composite oxide from compounds of Ce, Zr and Al not containing any limitation as to the relative amounts of the different compounds.

Claim 10 and paragraph 21 of the application as filed relate to a process for preparing a composite oxide from compounds of Ce, Zr and a compound whose metallic oxide is free from reacting with CeO_2 and ZrO_2 at 700°C or more.

A compound of Al is not specified in this claim or in this paragraph of the description. Therefore, by referring to the description, it is necessarily derived that there exist limitations as to the amounts of the further metallic oxides, in the present case Al_2O_3 , present in the resulting CeO_2 - ZrO_2 based composite oxide of the invention as explained above with respect to claim 4 of the main request (see point 1.1.3). This implies that also the amount of the compound of Al used

in the process must be limited with respect to the other compounds of Ce and Zr.

3.1.2 Since claim 1 according to the second auxiliary request does not contain any limitation as to the amounts of the compounds to be used in the process for preparing the composite oxide of the invention, the Board concludes that the claimed process is not directly and unambiguously derivable from the application as filed.

Therefore, this claim contravenes the requirements of Article 123(2) EPC.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

G. Rauh

P.-P. Bracke