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## Datasheet for the decision of 2 July 2014

Case Number: T 0958/11 - 3.3.05

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B01J37/03, B01D53/94, B01J35/00

Language of the proceedings: ΕN

#### Title of invention:

Porous composite oxide and production method thereof

## Patent Proprietor:

TOYOTA JIDOSHA KABUSHIKI KAISHA

## Opponents:

Rhodia Opérations ExxonMobil Research and Engineering Company

## Headword:

Composite oxide/TOYOTA

#### Relevant legal provisions:

EPC Art. 54, 56, 83

## Keyword:

Novelty - all requests (yes) reworked prior art example (not accepted)
Sufficiency of disclosure main and auxiliary requests 1 to 10 (no) parameter insufficiently disclosed
Inventive step - auxiliary request 11 (yes) alternative processes

## Decisions cited:

T 0409/91, T 0435/91, T 1743/06, T 0641/07, T 1276/08, T 0045/09

#### Catchword:



## Beschwerdekammern Boards of Appeal Chambres de recours

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Case Number: T 0958/11 - 3.3.05

# DECISION of Technical Board of Appeal 3.3.05 of 2 July 2014

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Decision under appeal: Decision of the Opposition Division of the

European Patent Office posted on 8 March 2011 rejecting the opposition filed against European patent No. 1479651 pursuant to Article 101(2)

EPC.

## Composition of the Board:

Chairman G. Raths

Members: J.-M. Schwaller

P. Guntz

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## Summary of Facts and Submissions

- I. The present appeal lies from the decision of the opposition division to reject the two oppositions filed against European patent No. 1 479 651, independent claims 1, 3, 11 and 15 of which read as follows:
  - "1. A porous composite oxide comprising an aggregate of secondary particles in the form of aggregates of primary particles of a composite oxide containing two or more types of metal elements, and having mesopores having a pore diameter of 2-100 nm between the secondary particles; wherein the particle diameter of the primary particles is 3-15 nm and the particle diameter of the secondary particles is 30-100 nm, the percentage of the mesopores between the secondary particles having a diameter of 10 nm or more is 10% or more of the total mesopore volume after firing for 5 hours at 600°C in an oxygen atmosphere."
  - "3. A production method of the porous composite oxide according to claim 1 comprising:
    a solution in which a first metal element compound that forms a hydroxide by hydrolysis is dissolved in organic solvent, and an emulsion containing a second or other metal element ion in an aqueous phase within reverse micelles formed by a surfactant in organic solvent, are mixed, the first metal element compound is hydrolyzed at the interface of the reverse micelles together with incorporating the second or other metal element, primary particles of a precursor of a composite oxide are formed by polycondensation, and in a system containing these primary particles, the primary particles are aggregated to form secondary particles, and

the secondary particles are aggregated;

wherein, the volume of the organic phase outside the reverse micelles is made to be larger than the volume of the aqueous phase inside the reverse micelles and the volume of the surfactant so that mesopores having a pore diameter of 2-100 nm are formed between the secondary particles while maintaining a distance of 20 nm or more between the reverse micelles during hydrolysis."

"11. A production method of the porous composite oxide according to claim 1 comprising:

an aqueous solution containing a first metal element ion and an aqueous solution containing a second or other metal element ion are mixed and allowed to react in an aqueous phase inside reverse micelles formed by a surfactant in organic solvent,

a compound containing the first metal and second or other metal elements is allowed to precipitate, this is then hydrolyzed to form primary particles of a precursor of a composite oxide by polycondensation, and in a system containing these primary particles, the primary particles are aggregated to form secondary particles, and

the secondary particles are aggregated; wherein, the volume of the organic phase outside the reverse micelles is made to be larger than the volume of the aqueous phase inside the reverse micelles and the volume of the surfactant so that mesopores having a pore diameter of 2-100 nm are formed between the secondary particles while maintaining a distance of 20 nm or more between the reverse micelles during hydrolysis."

"15. A production method of the porous composite oxide according to claim 1 comprising:

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an aqueous solution containing a first metal element ion and an aqueous solution containing a second or other metal element ion are mixed and allowed to react, the mixture containing the first metal and the second or other metal element is allowed to precipitate, this is then hydrolyzed to form primary particles of a precursor of composite oxide by polycondensation, and in a system containing these primary particles, the primary particles are aggregated to form secondary particles, and

the secondary particles are aggregated; wherein a compound is precipitated by making the total metal element ion concentration in the mixture of the aqueous solution containing ions of a first metal element and the aqueous solution containing ions of a second or other metal element to be 0.3 mol/L or less, after which a solution containing this precipitate is concentrated to aggregate secondary particles."

II. The following document filed during the opposition proceedings is relevant for the present decision:

D1: WO 95/18068

III. In its decision, the opposition division held the patent to be in conformity with the requirements of Article 83 EPC, in particular for the following reasons:

The determination of primary particle sizes could be performed by methods well known in the art, for instance TEM or XRD. The aggregates of secondary particles, on the one hand, and the secondary particles having a particle diameter of 30 to 100 nm in the form of aggregates of primary particles, on the other hand, were the result of the production method described in

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the description. The patent furthermore disclosed the BJH method for characterising the porosity of the porous composite oxide.

- IV. With its grounds of appeal dated 8 July 2011, opponent I (hereinafter "the appellant") submitted an experiment reporting the reproduction of example 2 of document D1. On the basis of this report, the appellant objected to the novelty of product claims 1 and 2 as granted. The appellant furthermore contested the inventive step and sufficiency of disclosure of the invention as claimed.
- V. With its response to the grounds of appeal dated 22 November 2011, the patentee (hereinafter "the respondent") argued that the evidence for lack of novelty could not be retained, because the reworking of example 2 of D1 had not been done properly. The respondent further argued that the invention was sufficiently disclosed and involved an inventive step, in particular over document D1. As a precautionary measure, it also filed 22 sets of amended claims as auxiliary requests 1 to 22.

Claim 1 of auxiliary request 1 reads as follows:

"1. A porous composite oxide comprising an aggregate of secondary particles in the form of aggregates of primary particles of a composite oxide containing two or more types of metal elements, and having mesopores having a pore diameter of 2-100 nm between the secondary particles; wherein the particle diameter of the primary particles is 3-15 nm and the particle diameter of the secondary particles is 30-100 nm before aggregation of these secondary particles, the percentage of the mesopores between the secondary

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particles having a diameter of 10 nm or more is 10% or more of the total mesopore volume after firing for 5 hours at 600°C in an oxygen atmosphere."

Claim 1 of auxiliary request 2 corresponds to claim 1 as granted, to which the passage "wherein there are hardly any change in peak height of pore diameter or in pore volume even when compared to the resulting porous composite oxide after firing at 600°C and resulting porous composite oxide after firing at 900°C." has been appended.

Claim 1 of auxiliary request 3 corresponds to claim 1 as granted, to which the passage "wherein after firing in air for 5 hours at 900°C, when compared to 600°C, there is no decrease in pore volume and pore distribution is shifted only towards higher peak values." has been appended.

Claim 1 of auxiliary request 4 differs from claim 1 as granted in that the term "consisting" replaces the term "comprising".

Claim 1 of auxiliary request 5 corresponds to claim 1 as granted, to which the passage "wherein the porous composite oxide is obtainable by aggregating the primary particles to obtain the secondary particles, followed by aggregation of the secondary particles." has been appended.

Claims 1 to 13 of auxiliary request 6 relate to production methods and claim 14 reads: "A porous composite oxide obtainable by a method according to any of claims 1 to 13, the porous composite oxide comprising an aggregate of secondary particles in the form of aggregates of primary particles of a composite

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oxide containing two or more types of metal elements, and having mesopores having a pore diameter of 2-100 nm between the secondary particles; wherein the particle diameter of the primary particles is 3-15 nm and the particle diameter of the secondary particles is 30-100 nm, the percentage of the mesopores between the secondary particles having a diameter of 10 nm or more is 10% or more of the total mesopore volume after firing for 5 hours at 600°C in an oxygen atmosphere."

Claim 1 of auxiliary request 7 differs from claim 1 as granted in that the passage "the bonding state between the primary particles of the secondary particles is stronger than the bonding state between the secondary particles and" has been inserted between the features ".. 30-100 nm" and "the percentage of the mesopores..".

Claim 1 of auxiliary request 8 differs from claim 1 as granted in that the features "the porous composite oxide is obtainable by a production method comprising a solution in which a first metal element compound that forms a hydroxide by hydrolysis is dissolved in organic solvent, and an emulsion containing a second or other metal element ion in an aqueous phase within reverse micelles formed by a surfactant in organic solvent, are mixed, the first metal element compound is hydrolyzed at the interface of the reverse micelles together with incorporating the second or other metal element, primary particles of a precursor of a composite oxide are formed by polycondensation, and in a system containing these particles, the primary particles are aggregated to form secondary particles, and the pH is adjusted to approach the isoelectric point which causes the secondary particles to aggregate" have been added at the end of the claim.

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Claim 1 of auxiliary request 9 differs from claim 1 as granted in that the passage "the porous composite oxide is obtainable by a production method comprising a solution in which a first metal element compound that forms a hydroxide by hydrolysis is dissolved in organic solvent, and an emulsion containing a second or other metal element ion in an aqueous phase within reverse micelles formed by a surfactant in organic solvent, are mixed, the first metal element compound is hydrolyzed at the interface of the reverse micelles together with incorporating the second or other metal element, wherein the pH is initially shifted by 1-4 from the isoelectric point during hydrolysis, primary particles of a precursor of a composite oxide are formed by polycondensation, and in a system containing these particles, the primary particles are aggregated to form secondary particles, and the pH is adjusted to approach the isoelectric point which causes the secondary particles to aggregate" has been added at the end of the claim.

Claim 1 of auxiliary request 10 corresponds to claim 1 as granted.

Claims 1, 9 and 13 of auxiliary request 11 read as follows:

"1. A production method of the porous composite oxide comprising an aggregate of secondary particles in the form of aggregates of primary particles of a composite oxide containing two or more types of metal elements, and having mesopores having a pore diameter of 2-100 nm between the secondary particles; wherein the particle diameter of the primary particles is 3-15 nm and the particle diameter of the secondary particles is 30-100 nm, the percentage of the mesopores between the

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secondary particles having a diameter of 10 nm or more is 10% or more of the total mesopore volume after firing for 5 hours at 600°C in an oxygen atmosphere, the production method comprising:

a solution in which a first metal element compound that forms a hydroxide by hydrolysis is dissolved in organic solvent, and an emulsion containing a second or other metal element ion in an aqueous phase within reverse micelles formed by a surfactant in organic solvent, are mixed,

the first metal element compound is hydrolyzed at the interface of the reverse micelles together with incorporating the second or other metal element, primary particles of a precursor of a composite oxide are formed by polycondensation, and in a system containing these primary particles, the primary particles are aggregated to form secondary particles, and

the secondary particles are aggregated; wherein the volume of the organic phase outside the reverse micelles is made to be larger than the volume of the aqueous phase inside the reverse micelles and the volume of the surfactant so that mesopores having a pore diameter of 2-100 nm are formed between the secondary particles while maintaining a distance of 20 nm or more between the reverse micelles during hydrolysis."

"9. A production method of the porous composite oxide comprising an aggregate of secondary particles in the form of aggregates of primary particles of a composite oxide containing two or more types of metal elements, and having mesopores having a pore diameter of 2-100 nm between the secondary particles; wherein the particle diameter of the primary particles is 3-15 nm and the particle diameter of the secondary particles is

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30-100 nm, the percentage of the mesopores between the secondary particles having a diameter of 10 nm or more is 10% or more of the total mesopore volume after firing for 5 hours at 600°C in an oxygen atmosphere, the production method comprising:

an aqueous solution containing a first metal element ion and an aqueous solution containing a second or other metal element ion are mixed and allowed to react in an aqueous phase inside reverse micelles formed by a surfactant in organic solvent,

a compound containing the first metal and second or other metal elements is allowed to precipitate, this is then hydrolyzed to form primary particles of a precursor of a composite oxide by polycondensation, and in a system containing these primary particles, the primary particles are aggregated to form secondary particles, and

the secondary particles are aggregated; wherein the volume of the organic phase outside the reverse micelles is made to be larger than the volume of the aqueous phase inside the reverse micelles and the volume of the surfactant so that mesopores having a pore diameter of 2-100 nm are formed between the secondary particles while maintaining a distance of 20 nm or more between the reverse micelles during hydrolysis."

"13. A production method of the porous composite oxide comprising an aggregate of secondary particles in the form of aggregates of primary particles of a composite oxide containing two or more types of metal elements, and having mesopores having a pore diameter of 2-100 nm between the secondary particles; wherein the particle diameter of the primary particles is 3-15 nm and the particle diameter of the secondary particles is

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30--100 nm, the percentage of the mesopores between the secondary particles having a diameter of 10 nm or more is 10% or more of the total mesopore volume after firing for 5 hours at  $600\,^{\circ}\text{C}$  in an oxygen atmosphere, the production method comprising:

an aqueous solution containing a first metal element ion and an aqueous solution containing a second or other metal element ion are mixed and allowed to react, the mixture containing the first metal and the second or other metal element is allowed to precipitate, this is then hydrolyzed to form primary particles of a precursor of composite oxide by polycondensation, and in a system containing these primary particles, the primary particles are aggregated to form secondary particles, and

the secondary particles are aggregated; wherein a compound is precipitated by making the total metal element ion concentration in the mixture of the aqueous solution containing ions of a first metal element and the aqueous solution containing ions of a second or other metal element to be 0.3 mol/L or less, after which a solution containing this precipitate is concentrated to aggregate secondary particles."

- VI. With letter of 21 May 2014, opponent II informed the board that it would not attend the oral proceedings.
- VII. At the oral proceedings, which took place on 2 July 2014, the issues of sufficiency of disclosure, novelty and inventive step were discussed. In particular, as regards sufficiency of disclosure, the debate focused on the secondary particles and the determination of their particle diameter.
- VIII. After closing the debate, the chairman established the parties' requests as follows:

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

The respondent requested that the appeal be dismissed, or, alternatively, that the patent be maintained on the basis of one of the sets of claims according to auxiliary requests 1 to 22 dated 22 November 2011.

#### Reasons for the Decision

- 1. Main request Novelty
- 1.1 The novelty of the product according to claim 1 as granted was contested on the basis of two experiments in which the process disclosed in example 2 of D1 was reproduced.
- 1.2 Regarding the first experiment, the board agrees with the findings of the opposition division that this reworking is manifestly not acceptable, as it does not correspond to the strict reproduction of said example, the mixing having been carried out in an agitated reactor while in D1 it occurred in a reactor having a Y form.
- 1.3 Concerning the second experiment, the respondent expressed severe doubts regarding the proper reworking of example 2 of D1, in particular as regards the concentration of the starting materials before mixing.

The appellant contested these findings and stated that the concentrations of the starting materials cerium nitrate and zirconyl nitrate were conventionally expressed in g/l of  $CeO_2$  and  $ZrO_2$ . The concentrated starting materials were then diluted with water so as

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to exactly obtain the same concentrations before mixing as in example 2 of D1.

The respondent expressed its surprise regarding the appellant's statements and requested an adjournment of the oral proceedings to carry out counter experiments in case the board would conclude to a lack of novelty on the basis of the second experiment.

The board observes that the information provided at the oral proceedings by the appellant is of such an importance that it would have required an adjournment of the oral proceedings, because — as argued by the respondent — the concentration is one of the key features of the invention (see in particular claim 15 as granted which requires a low concentration of the starting materials).

Since the respondent's doubts had already been raised with its response to the grounds of appeal, i.e. almost three years ago, the board objects to the appellant's silence during the written procedure on this important feature. The oral explanations of the appellant revealed that it was perfectly aware of the obscure presentation of the relevant part of its test. Instead of redressing this defect, the appellant chose to leave the respondent in dark until the oral proceedings.

The appellant's behaviour not to comment during the written procedure on this relevant issue could - if done with the intention to surprise the respondent during oral proceedings - amounts to an abuse of procedure.

At least its explanations during the oral proceedings as to the concentrations of the starting materials and

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as to the calculation method used constitute an amendment to a party's case, the admittance of which lies within the board's discretion (Article 13(1) RPBA).

Amendments sought to be made after oral proceedings have been arranged shall not be admitted if they raise issues that the other party cannot reasonably be expected to deal with without adjournment of the oral proceedings (Article 13(3) RPBA). This is the case here. Having uncovered the concentration of the starting materials and the calculation method, the appellant questioned the relevance of the respondent's doubts as to the proper reworking of example 2 of D1. To show that these doubts prevail it would have now been up to the respondent to perform comparative tests. Whereas these could have been easily performed during the last almost three years, had only the appellant reacted directly to the respondent's response to the grounds of appeal, it is obvious that this is not possible on the day of the oral proceedings.

The Board therefore exercises its discretion not to admit the latest amendment of the appellant's case into the proceedings.

In the absence of a clear and unambiguous reproduction of example 2 of D1, and thus, of a direct and unambiguous disclosure of the claimed subject-matter, the board therefore decides to reject the evidence submitted for lack of novelty.

1.4 The objection of lack of novelty was solely based on the reworked example 2 of D1. None of the documents cited during the opposition proceedings disclose the subject-matter of the four independent claims of the

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main request at issue. Therefore, the board is satisfied that these claims, and those which depend thereon, meet the requirements of Article 54(1) and (2) EPC.

- 2. Main request Sufficiency of disclosure of the invention
- It is established jurisprudence that the requirements for sufficiency of disclosure are met if the invention as defined in the claims could be performed at the filing date of the application by a person skilled in the art in the whole area claimed without undue burden, using common general knowledge and having regard to further information given in the patent in suit (see e.g. T 0409/91, OJ 1994, 653, point 3.5 of the reasons; T 0435/91, OJ 1995, 188, point 2.2.1 of the reasons; T 1743/06, point 1.1 of the reasons).

When the definition of the claimed invention moreover includes one or more parameters, the skilled person should also be able to check whether the parameters are complied with while the invention is carried out (see e.g. decisions T 0045/09, points 1.1 and 1.3 of the reasons; T 1276/08, point 1.1 of the reasons; T 0641/07, point 1. of the reasons).

- 2.2 In the case at issue, the invention (claim 1) concerns a porous composite oxide comprising:
  - an aggregate of secondary particles having mesopores with a diameter of 2-100 nm between the secondary particles;
  - the secondary particles having a diameter of 30-100 nm and forming aggregates of primary particles of a composite oxide containing two or more metal elements;

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- the primary particles having a diameter of 3 to 15 nm;
- and the percentage of mesopores between the secondary particles having a diameter of 10nm or more being 10% or more of the total mesopore volume after firing for 5 hours at 600°C in an oxygen atmosphere,

and three alternative production processes of said composite (claims 3, 11 and 15).

2.3 Regarding the question as to whether the invention as defined in the claims could be performed at the filing date of the application by a person skilled in the art, the board observes that the patent specification (paragraphs [0025] to [0073]) discloses ample details regarding the production of the claimed composite oxide. Furthermore, in paragraphs [0074] to [0082], the preparation of six specific composite oxides of cerium and zirconium (Examples 1, 3, 4 and 5), strontium and zirconium (Example 2) and lanthanum and zirconium (Example 6) is extensively described.

The burden of proof is upon the opponent (here the appellant) to show that a skilled person is unable to carry out the invention. In the present case none of the preparation processes - in particular those detailed in examples 1 to 6 - have been reworked by the appellant to identify any gap of information. So, in the absence of any evidence to the contrary, it is credible that the above-mentioned production processes can lead to composite oxides according to the invention.

2.4 Since the definition of the composite oxide according to the invention is extremely broad and includes several parameters, the question arises whether the

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patent provides sufficient guidance to the skilled person to perform the invention in the whole claimed area and in particular to check whether the parameters defining the claimed composite oxide are complied with without undue burden, using common general knowledge and having regard to the information in the patent in suit.

- 2.4.1 With respect to the parameters defining the claimed composite oxide, the board observes that the patent specification does not disclose any information as to how the size of the different types of particles, in particular the "particle diameter" of the "secondary particles", is to be determined.
- 2.4.2 The examples in particular do not provide any data as regards the particle diameter of the primary particles in the specific composite oxides prepared in examples 1 to 6. As regards the particle diameter of the secondary particles, it is even questionable from the available data whether the composite oxide according to claim 1 can be prepared at all, since the sole available value (10 nm) for the diameter of secondary particles (see examples 1, 5 and 6) falls outside the claimed range of 30 to 100 nm.

The respondent declared in this respect that said examples contained typographical errors, and that the value of "10 nm" should be read as corresponding to the diameter of the primary particles. The board cannot accept this explanation, because examples 5 and 6 unambiguously describe the occurrence of a "slight aggregation of colloidal particles (about 10 nm) (secondary particles)", that the skilled reader necessarily understands as being the "secondary particles" in the sense of claim 1 at issue.

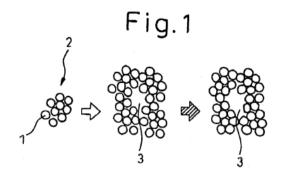
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It follows from this lack in information and from the absence of reliable data in the examples that the missing information regarding in particular the determination of the "particle diameter" of the "secondary particles" is not recoverable by a mere reworking of the examples, as in the case underlying e.g. decision T 0641/07.

- 2.4.3 Regarding the determination of the "particle diameter" of the "secondary particles", the respondent argued that Figure 1 of the patent and the corresponding passage in paragraph [0019] gave the necessary information for identifying the "secondary particles" in an "aggregate of secondary particles" and that the skilled person would necessarily use the most precise method at the priority date, namely TEM (transmission electronic microscopy). TEM furthermore allowed the identification of the boundaries between the individual secondary particles since the bonding state between two secondary particles was weaker than the boundary between primary particles. Concerning the "particle diameter" of a secondary particle which was not round, the skilled person knew that it had to measure the greatest dimension of said particle.

There is no indication in the patent, nor is it common general knowledge, that the "particle diameter" of a secondary particle would correspond to its greatest dimension. In paragraph [0019] of the patent, the porous composite oxide of figure 1 (see below) is described as being "composed by aggregating secondary particles 2 having a particle diameter of about 100 nm, which are aggregates of primary particles 1 of a

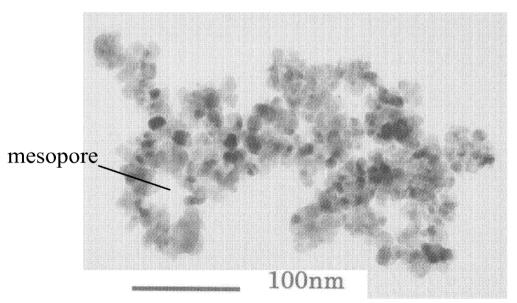
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composite oxide having a particle diameter of 5-15 nm, and not only has pores between primary particles 1, but also has mesopores 3 having a diameter of 10-100 nm between secondary particles 2.".

It is however not apparent from figure 1 that for determining the "particle diameter" the skilled person had to measure its greatest dimension, as suggested by the respondent. It is also not apparent from the above figure where the boundaries of the secondary particles constituting the above aggregate are located.

From the following photograph - that the respondent



[Fig.2] <u>Microemulsion Method</u> TEM Picture (A Porous Composite Oxide)

EP1479651B1 (Toyota Jidosha K.K.) Example 1 [0074][0075]

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submitted in opposition proceedings - which does not represent a theoretical composite as in Figure 1, but the composite oxide of example 1 of the patent as examined by TEM, it can be seen that it is quite impossible in the absence of any guidance in the patent to identify the presence of secondary particles and the boundaries between the said particles, and a fortiori, it is impossible from the photograph to determine the "particle diameter" of the "secondary particles".

The board notes that there is also no indication in the patent, nor does this information appear to be common general knowledge, that the bonding between two secondary particles was weaker than the bonding between primary particles and that this weaker bonding could be identified by TEM, as alleged by the respondent, which explained that the boundaries between two secondary particles were identifiable by TEM when the above composite oxide was examined from another angle. The board cannot retain this argument, for which there is no basis in the patent, and no evidence has been provided that this information was common general knowledge at the priority date of the patent.

Furthermore, this argument was contested by the appellant's technical expert at the oral proceedings.

2.5 It follows from the above considerations that the skilled person seeking to assess whether or not the claimed invention is complied with is left without guidance as regards the method for determining the "particle diameter" of the "secondary particles".

In this context and in the absence of any specific information and guidance concerning the ability of the skilled person to check whether said parameter is complied with when the invention is carried out, the

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board holds the disclosure of the contested patent to be insufficiently clear and complete for the invention to be carried out in the whole claimed area by a person skilled in the art, contrary to Article 83 EPC, insofar as product claims are concerned.

3. Auxiliary requests 1 to 10 - sufficiency of disclosure of the invention

The sets of claims according to auxiliary requests 1 to 10 each comprise an independent product claim (see item V above) including in its definition the parameter "secondary particles" having a "particle diameter" of 30 to 100 nm, which in points 2.1 and 2.5 above has been found to be insufficiently disclosed for a skilled person to check whether the claimed parameters are complied with when the invention is carried out. Hence, for the same reasons, these product claims are also held not to meet the requirements of Article 83 EPC.

- 4. Auxiliary request 11
- 4.1 Sufficiency of disclosure

The claims of this request, which concern three different production methods of the composite oxide defined inter alia in claim 1 as granted, cannot be held to infringe the requirements of Article 83 EPC. The reason is that the burden of proof is upon the opponent, and in the absence of any evidence in this respect, it is to be admitted that the production methods thus claimed can be carried out without undue burden by a skilled person.

Furthermore, in the absence of evidence to the contrary, it is credible - as indicated in point 2.3

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above - that the claimed production steps lead directly to the composite oxide according to the invention.

## 4.2 Novelty

The three production methods according to claims 1 to 13 do not infringe the requirements of Article 54(1)(2) EPC, since none of the cited documents discloses the sequence of process steps defined in said claims.

## 4.3 Inventive step

Claims 1 to 13 of this request meet the requirements of Article 56 EPC for the following reasons:

#### 4.3.1 Invention

The invention concerns three different methods for producing a porous composite oxide.

## 4.3.2 Closest prior art

Document D1, which the parties have acknowledged as representing the most appropriate starting point for assessing inventive step, discloses (claim 7) a process for the preparation of a cerium-zirconium composite oxide comprising:

- preparing a liquid mixture containing trivalent cerium and zirconium compounds;
- placing said mixture in contact with carbonate or bicarbonate, thus forming a reactive medium exhibiting a neutral or basic pH during the reaction;
- collecting a precipitate comprising a compound comprising cerium carbonate and a zirconium oxyhydroxide; and

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calcining said precipitate.

## 4.3.3 Problem

According to the contested patent (paragraph [0009]), the problem was to provide a method of producing a composite oxide with a uniform atomic distribution, with no decrease in pore volume and minimal thermal degradation after high temperature treatment.

## 4.3.4 Solution

As a solution to this problem, the patent proposes the following three methods:

- (1) The solution as proposed in claim 1, characterised in that:
- a solution in which a first metal element compound that forms a hydroxide by hydrolysis is dissolved in organic solvent, and an emulsion containing a second or other metal element ion in an aqueous phase within reverse micelles formed by a surfactant in organic solvent, are mixed, the first metal element compound being hydrolysed at the interface of the reverse micelles together with incorporating the second or other metal element,
- primary particles of a precursor of a composite oxide are formed by polycondensation and aggregated to form secondary particles, and
- the secondary particles are aggregated,
- with the volume of the organic phase outside the reverse micelles being made to be larger than the volume of the aqueous phase inside the reverse micelles and the volume of the surfactant so that mesopores having a pore diameter of 2-100 nm are formed between

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the secondary particles while maintaining an adequate distance between the reverse micelles during hydrolysis.

- (2) The solution as proposed in claim 9, characterised in that:
- an aqueous solution containing a first metal element ion and an aqueous solution containing a second or other metal element ion are mixed and allowed to react in an aqueous phase inside reverse micelles formed by a surfactant in organic solvent,
- a compound containing the first metal and second or other metal elements is allowed to precipitate,
- this system is hydrolysed to form primary particles of a precursor of a composite oxide by polycondensation,
- the primary particles are aggregated to form secondary particles, and
- the secondary particles are aggregated,
- with the volume of the organic phase outside the reverse micelles being made to be larger than the volume of the aqueous phase inside the reverse micelles and the volume of the surfactant so that mesopores having a pore diameter of 2-100 nm are formed between the secondary particles while maintaining an adequate distance between the reverse micelles during hydrolysis.
- (3) The solution as proposed in claim 13, characterised in that an aqueous solution containing a first metal element ion and an aqueous solution containing a second or other metal element ion are mixed and allowed to precipitate by making the total metal element ion concentration in the mixture of the aqueous solution containing ions of a first metal element and the

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aqueous solution containing ions of a second or other metal element to be 0.3 mol/L or less, after which a solution containing this precipitate is concentrated to aggregate secondary particles, and the secondary particles are aggregated.

#### 4.3.5 Success of the solution

Figures 6 and 7 of the patent show that for the composite oxides prepared with the processes claimed there were "hardly any changes in peak height or pore volume, even when fired at high temperatures" (see paragraph [0088], last sentence). Furthermore, as shown by figures 8 and 10, a minimal thermal degradation and low decrease in surface area (see paragraph [0095]) as well as a high cerium and zirconium dispersity (see paragraph [0091]) were achieved. So, it is credible that the problem underlying the patent was solved.

The board is aware that the above mentioned characteristics concern the product obtained. However, at stake is the process.

The board observes that the production process of the composite oxides of document D1 is based on coprecipitation of Ce and Zr ions (see claim 1 and examples). It is commonly known that such a production process leads to a uniform metallic distribution. D1 moreover relates to the production of porous composite oxides having high thermal stability (page 1, first lines), and so this document deals with the same problem as the patent in suit, which means that the problem underlying the invention is to be reformulated as the provision of alternative production methods of thermally-stable porous composite oxides.

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## 4.3.6 Obviousness

As none of the three solutions proposed in independent claims 1, 9 and 13, is disclosed or suggested in the documents on file, the skilled person faced with the problem of providing alternative methods for producing a thermally-stable porous composite oxide cannot arrive in an obvious manner at the different process steps defined in the respective claims 1, 9 and 13, which therefore are held to involve an inventive step.

- 4.3.7 Claims 2 to 7 and 10 to 12, which are dependent on claims 1 and 9, respectively, derive their patentability from that of the independent claim on which they depend.
- 4.4 It follows from the above considerations that the auxiliary request 11 is allowable.

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## Order

## For these reasons it is decided that:

- 1. The contested decision is set aside.
- 2. The case is remitted to the department of first instance with the order to maintain the patent in amended form on the basis of claims 1 to 13 of the auxiliary request 11 submitted by letter of 22 November 2011 (former auxiliary request 4 dated 15 October 2010), and a description to be adapted.

The Registrar:

The Chairman:



C. Vodz G. Raths

Decision electronically authenticated