BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

Internal distribution code:
(A) [ ] Publication in OJ
(B) [ ] To Chairmen and Members
(C) [X] To Chairmen
(D) [ ] No distribution

Datasheet for the decision of 19 October 2012

Case Number: T 0478/10 - 3.3.05

Application Number: 03024667.2

Publication Number: 1415956

IPC: C01G 25/00, B01J 23/10, B01D 53/94, B01J 23/63

Language of the proceedings: EN

Title of invention:
A method for preparing metal oxide particles and an exhaust gas purifying catalyst

Patentee:
Toyota Jidosha Kabushiki Kaisha

Opponent:
Rhodia Opérations

Headword:
CeO_2/ZrO_2 composite/TOYOTA

Relevant legal provisions:
EPC Art. 54(1)(2), 56

Keyword:
"Novelty (main request): yes"
"Inventive step (main request): yes - improvement (no evidence) - reformulation of the problem - non obvious alternative"

Decisions cited:
-

Catchword:
-

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0478/10 - 3.3.05

D E C I S I O N
of Technical Board of Appeal 3.3.05
of 19 October 2012

Appellant I: (Patent Proprietor)
Toyota Jidosha Kabushiki Kaisha
1, Toyota-cho
Toyota-shi, Aichi-ken, 471-8571 (JP)

Representative:
TBK
Bavariaring 4-6
D-80336 München (DE)

Appellant II: (Opponent)
Rhodia Opérations
40 Rue de la Haie-Coq
F-93306 Aubervilliers (FR)

Representative:
Dubruc, Philippe
Rhodia Services
Direction de la Propriété Industrielle
40 Rue de la Haie-Coq
F-93306 Aubervilliers (FR)

Decision under appeal:
Interlocutory decision of the Opposition Division of the European Patent Office posted 30 December 2009 concerning maintenance of the European patent No. 1415956 in amended form.

Composition of the Board:

Chairman: G. Raths
Members: J.-M. Schwaller
D. Prietzel-Funk

Summary of Facts and Submissions

I. The present appeals lie from the decision of the opposition division maintaining European patent No. 1 415 956 on the basis of the second auxiliary request filed on 13 November 2009, independent claims 1 and 2 of which read as follows:

"*1. The use of metal oxide particles having cores comprising larger molar amounts of zirconia than of ceria, and surface layers comprising larger molar amounts of ceria than of zirconia for manufacturing an exhaust gas purifying catalyst.*

2. Metal oxide particles having cores comprising larger molar amounts of zirconia than of ceria, and surface layers comprising larger molar amounts of ceria than of zirconia, wherein the metal oxide particles carry a noble metal.

II. Independent claims 1 and 12 of the patent as granted read as follows:

"*1. Metal oxide particles having cores comprising larger molar amounts of zirconia than of ceria, and surface layers comprising larger molar amounts of ceria than of zirconia.*"

12. A method for preparing metal oxide particles as defined in claim 1 wherein the method comprises:
preparing a solution comprising zirconia sol and ceria sol;
adjusting the pH of the solution within ± 0.5 on the basis of the isoelectric point of zirconia; and

aggregating zirconia and then aggregating ceria around the aggregated zirconia from said solution to make metal oxide particles."

III. The following documents cited during the opposition proceedings are relevant for the present decision:

D2: Li Wei-Bin, Yang Xu-Fei, Chemical Journal of Chinese Universities, Vol. 23, 6, pages 1140 to 1142

D2': Translation into English of D2

D3: EP 1 035 074 A1.

IV. In the contested decision, the opposition division held claim 1 as granted to lack novelty over document D2/D2'. The opposition division argued in particular that the process according to D2/D2' - whereby ZrO_2 particles were impregnated with a $Ce(NO_3)_3$ solution, dried and calcined - led to ZrO_2 particles with an outer layer comprising larger amounts of ceria than zirconia.

V. With its statement of grounds of appeal, the patent proprietor (hereinafter "appellant I") submitted an experimental report and requested that the patent be upheld as granted.

VI. With its statement of grounds of appeal, the opponent (hereinafter "appellant II") filed a new document:

D4: "*Removal of nitrogen oxides from the exhaust of a lean-burn gasoline engine*", W. Bögner et al.,

Applied catalysis B: Environmental, 7, pages 153 to 171 (1995).

Appellant II argued that D4 in association with document D2/D2' anticipated the subject-matter of claim 1 as maintained by the opposition division in terms of novelty and inventive step. Appellant II also contested the inventive step of claim 2 as maintained on the basis of the combined disclosure of documents D2/D2' and D4, optionally in combination with the teaching of document D3.

VII. With a further letter dated 17 September 2010, appellant II contested the experimental report of appellant I, arguing in particular that in contrast to the specific embodiments disclosed in document D2/D2', those proposed in the experimental report had not been calcined.

VIII. With a letter dated 28 September 2010, appellant I requested the board not to admit document D4 into the proceedings.

IX. With a further letter dated 22 March 2011, appellant I filed a second experimental report, which this time included samples which were calcined.

X. With a letter dated 20 July 2012, appellant II declared that it would not be attending the oral proceedings to be held on 19 October 2012.

XI. With a letter dated 7 August 2012, appellant I submitted 9 auxiliary requests.

XII. At the oral proceedings, which took place on 19 October 2012 in the absence of appellant II, the discussion focused on novelty and inventive step of the claims as granted.

XIII. The parties' requests were established as follows:

Appellant II requested in writing that the decision under appeal be set aside and that the patent be revoked. Alternatively, it requested that claims 1 to 11 as granted be not allowed.

Appellant I requested that the decision under appeal be set aside and that the patent be maintained as granted (main request) or, alternatively, that the patent be maintained on the basis of the claims according to one of the auxiliary requests 1 to 9 filed on 7 August 2012.

Reasons for the Decision

1. *Admissibility of document D4*

Document D4 was filed with appellant II's grounds of appeal; appellant I thus had sufficient time to take it into consideration. In the board's view, D4 is relevant to the case at issue, since it is referred to in document D2/D2'. It follows that D4 satisfies the requirements of Article 12(2) and (4) RPBA. It is thus admitted into the appeal proceedings.

2. *Main request - novelty*

2.1.1 Document D2/D2' - that the opposition division held to anticipate the subject-matter of claim 1 as granted -

discloses (page 2, item 1.2) the impregnation of a ZrO_2 carrier having a specific surface area of 120 m^3/g with 0.1 mol/L of a $Ce(NO_3)_3$ aqueous solution at a pH controlled to be between 6.0 and 7.0 under stirring for 2 hours, the impregnated carrier being then dried for several hours at 120°C to get a powdery sample and calcined for 6 hours at 450°C in an oxygen atmosphere to get a 5% Ce-ZrO_2 sample.

2.1.2 In the second experimental report annexed to its letter dated 22 March 2011, appellant I prepared different samples of Ce (5 mass%)/ZrO_2 by impregnating 53.2 g ZrO_2 having a specific surface area of 107 m^2/g with 8.68 g $Ce(NO_3)_3/6H_2O$ dissolved in 200 cc ion-exchanged water, adjusting the pH to 6.9, stirring for 2 hours, with the pH after stirring being 6.4, drying for 12 hours at 120°C and calcining at different temperatures between 450°C and 900°C.

The measurement by XPS (point 3-1 of the report) of the Ce surface concentration of the different calcined samples revealed a Ce atomic ratio varying from 6.30 to 7.12 and a Zr atomic ratio between 92.88 to 93.70.

The measurements by TEM/EDX (point 3-2 of the report) revealed Ce surface concentrations varying between 3.71 and 4.80 atomic% and Zr surface concentrations varying between 95.20 and 96.29 atomic%.

2.1.3 For the board, the above experiments show that Ce/ZrO_2 metal oxide particles prepared by impregnating ZrO_2 particles according to the impregnation process disclosed in document D2/D2' do not have a surface layer comprising larger molar amounts of ceria than of

zirconia, and thus do not fall under the wording of claim 1 as granted. The metal oxide particles prepared in document D2/D2' thus do not anticipate the subject-matter of claim 1 as granted.

The same conclusion is drawn for the subject-matter of claim 12 as well as for claims 2 to 11 and 13 to 15, which depend on claim 1 and claim 12, respectively.

2.1.4 Document D4 (point 2.1. Catalyst preparation) discloses a refractory oxide composed primarily of Al, Ce and Zr impregnated with a solution containing precious metal and a proprietary adsorbing material. Metal oxide particles having cores comprising larger molar amounts of zirconia than of ceria, and surface layers comprising larger amounts of ceria than of zirconia, are not disclosed in D4.

2.1.5 The board is satisfied – and nor was it in dispute – that the other documents cited in these proceedings do not anticipate the subject-matter of the claims as granted.

2.1.6 It follows from the above that the claims 1 to 15 as granted meet the requirements of Article 54(1) and (2) EPC.

2.2 Inventive step

2.2.1 The contested patent (paragraph [0001]) relates to a catalyst for purifying exhaust gases from an internal combustion engine, and metal oxide particles suitable as a carrier for said catalyst.

2.2.2 The starting point for assessing inventive step is represented - as acknowledged by appellant I - by document D2/D2', which concerns cerium/zirconium oxide particles as a carrier for catalysts. Its disclosure is detailed in point 2.1.1 above.

2.2.3 According to the patent in suit (paragraph [0008]) the exhaust gas purifying catalyst may have a superior heat resistivity and a superior ability to purify exhaust gas after a high-temperature endurance test.

2.2.4 As a solution to this problem, the contested patent proposes the metal oxide according to claim 1, which is characterised in particular by a surface layer comprising larger molar amounts of ceria than of zirconia.

2.2.5 As to the success of that solution, the board observes that there is no evidence for any improvement with respect to the closest state of the art. Therefore the technical problem had to be reformulated - as acknowledged during the oral proceedings - as the provision of a metal oxide carrier alternative to the one disclosed in D2/D2'.

2.2.6 The board is satisfied that the above technical problem is effectively solved. Examples 1 and 2 and Table 2 of the contested patent show that metal oxide particles according to claim 1 as granted are plainly suitable as a carrier for platinum particles. Table 2 further shows that the exhaust gas purifying catalysts thus prepared have a smaller platinum particle size, higher specific surface area and lower HC-T50 after an endurance test - and thus have higher heat resistivity and exhaust gas

purification ability - than comparative samples (catalysts 4 and 5 in Table 2) prepared from homogeneously mixed Ce/Zr metal oxides.

2.2.7 As to whether the above solution is obvious in view of the cited prior art, in particular documents D3 or D4, the board observes the following.

Document D3 (paragraphs [0001]) discloses the preparation of zirconium- and cerium-based mixed oxides having good thermal stability and oxidation-reduction performance, and their use in catalysts for exhaust gas purification. As indicated in paragraphs [0044] to [0046], the above mixed oxides are obtained from "a precipitate with zirconium and cerium uniformly brought together" and have "high chemical homogeneity" and a structure "high in chemical uniformity".

In the board's view, D3 leads away from the solution proposed in claim 1 since the above mixed oxides are homogeneously dispersed throughout the particles while those defined in claim 1 as granted have the claimed core-shell structure. It follows that the skilled person looking for an alternative to the mixed oxide disclosed in D2/D2' would not arrive at the subject-matter of claim 1 from the disclosure in D3.

2.2.8 The same conclusion can be drawn from the disclosure in document D4, which as explained in point 2.1.4 above, does not disclose metal oxide particles having cores comprising larger molar amounts of zirconia than of ceria, and surface layers comprising larger amounts of ceria than of zirconia.

2.2.9 The remaining documents cited during the opposition and appeal proceedings do not contain further information pointing towards the claimed solution.

2.2.10 It follows from the above that the subject-matter of claims 1 and 12 as granted (and that of claims 2 to 11 and 13 to 15, which depend thereon respectively) is not obvious for the skilled person in the light of the disclosure of document D2/D2' taken in combination with the teachings of the other documents cited in these proceedings. Therefore, the subject-matter of claims 1 to 15 as granted involves an inventive step within the meaning of Articles 52(1) and 56 EPC.

3. The board is satisfied that the claims as granted also satisfy the other requirements of the EPC.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the department of first instance with the order to maintain the patent as granted.

The Registrar: The Chairman:

C. Vodz G. Raths