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Datasheet for the decision of 10 September 2008

Case Number: T 1105/02 - 3.3.05

Application Number: 95119403.4

Publication Number: 0715879

IPC: B01D 53/94

Language of the proceedings: EN

Title of invention:
Catalyst for purifying exhaust gases and process for producing the same

Applicant:
KABUSHIKI KAISHA TOYOTA CHUO KENKYUSHO

Opponent:
-

Headword:
Exhaust gas catalyst/TOYOTA

Relevant legal provisions:
EPC Art. 54(1)(2), 83, 111(1), 123(2)

Relevant legal provisions (EPC 1973):

Keyword:
"Remittal (yes) - substantial amendments of claims"

Decisions cited:
G 10/93

Catchword:
-

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Case Number: T 1105/02 **-** 3.3.05

D E C I S I O N
of the Technical Board of Appeal 3.3.05
of 10 September 2008

Appellant: KABUSHIKI KAISHA TOYOTA CHUO KENKYUSHO
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 12 March 2002 refusing European application No. 95119403.4 pursuant to Article 97(1) EPC 1973.**

Composition of the Board:

Chairman: G. Raths
Members: J.-M. Schwaller
H. Preglau

Summary of Facts and Submissions

I. This appeal lies from the decision of the examining division to refuse the European patent application No 95119403.4.

II. The contested decision was based on three sets of claims submitted on 8 February 1999, 8 June 2000 and 24 January 2002 as main, 1st and 2nd auxiliary request, respectively. Each set of claims enclosed one independent product claim and one independent process claim.

III. In the decision, the examining division held that the subject-matter of the claims 1 of the main request and of the 1st auxiliary request did not satisfy the requirements of Article 123(2) EPC.

Claim 1 of the 2nd auxiliary request read:

"1. A catalyst for purifying exhaust gases, comprising a porous support, cerium oxide or a solid solution including cerium oxide loaded on the porous support, and a nobel metal element loaded on the porous support, characterized in that the cerium oxide or the solid solution including cerium oxide has an average particle diameter of 5 to 100 nm.".

The examining division concluded that it lacked novelty in its subject-matter in the light of the document:

D1 = EP-A-0507590.

In this respect, it argued that the parameter "average particle diameter" defined in claim 1 of the 2nd auxiliary request embraced primary particles having an average particle diameter of e.g. 6 nm (reference was made to page 44, second paragraph of the description). As D1 disclosed a catalyst having primary particles of cerium oxide of less than 25 nm, claim 1 therefore was not novel.

IV. Under cover of the grounds of appeal dated 5 July 2002, the appellant filed *inter alia* a report of comparative results and four amended set of claims as a main and 1st to 3rd auxiliary requests, respectively.

V. In response to a communication annexed to the summons to oral proceedings, in which the board questioned the allowability of the amended claims then on file under Articles 123(2) and 84 EPC, the appellant submitted by fax on 22 November 2007 nine amended sets of claims, respectively as main and 1st to 8th auxiliary requests.

Claim 1 of the main request read as follows:

"A catalyst for purifying exhaust gases, comprising
a porous support,
cerium oxide loaded on the porous support, and
a noble metal element loaded on the porous support,
characterised in that the cerium oxide has an average particle diameter of 5 to 100 nm, said average particle diameter being large enough to prevent most of the cerium oxide from entering pores of said porous support, wherein said average particle diameter is measured by scanning electron microscopy."

VI. At the oral proceedings, which took place on 5 December 2007, the discussion focused on the parameter "average particle diameter" and in this respect the board handed over the document:

D6 = Surface & Coatings Technology, vol. 200 (2005), pages 1751-1754.

The appellant observing that this issue concerned Article 83 EPC, i.e. a ground raised for the first time at this stage of the proceedings, requested that the procedure be continued in writing in order "to allow the applicant to file evidence proving that the sizes of secondary particles in the claimed catalysts can be measured by SEM".

VII. In a communication dated 14 December 2007, the board requested the appellant to provide the proposed evidence. It also raised several objections under Articles 82, 84 and Rule 86(4) EPC [1973].

VIII. Under cover of a letter dated 23 April 2008, the appellant submitted a copy of three (one SEM and two TEM) images - called Reference Fig. 1 to 3 - of a specific γ-alumina (MI-386 by W.R. Grace and Co.).

In replacement of the pending requests, it furthermore filed eight new sets of claims as main and 1st to 7th auxiliary requests, respectively. Each set of claims enclosed two independent product claims and two independent process claims.

Independent product claims 1 and 2 of the main request read as follows:

"1. A catalyst for purifying exhaust gases, comprising a porous support, cerium oxide loaded on the porous support, and a noble metal element loaded on the porous support, characterized in that
the cerium oxide has an average particle diameter of 5 to 100 nm after calcining at 650°C for 1 hour, said average particle diameter being large enough to prevent most of the cerium oxide from entering pores of said porous support, wherein said average particle diameter is measured by scanning electron microscopy."

"2. A catalyst for purifying exhaust gases, comprising a porous support, cerium oxide in a state of a solid solution with a zirconium oxide loaded on the porous support, said solid solution containing cerium oxide and zirconium oxide in a molar ratio of 0.2 : to 1 to 4.0 : 1, and a noble metal element loaded on the porous support, characterized in that
said solid solution has an average particle diameter of 5 to 100 nm after calcining at 750°C for 1 hour, said average particle diameter being large enough to prevent most of the cerium oxide from entering pores of said porous support, wherein said average particle diameter is measured by scanning electron microscopy."

IX. In its letter of 23 April 2008, the appellant in summary pleaded its case as follows:

(a) The values in Table 4 of D1 represent a crystal particle diameter (particle diameter of primary particles) measured by XRD, which does not

represent the real particle diameter. The report of comparative experimental results of July 2002 shows that the average particle diameter of cerium oxide in D1 is outside the range presently claimed.

(b) In order to show that SEM does not allow the measurement of the size of primary particles measured in D1, SEM- and TEM-analyses have been conducted with a commercial γ-alumina. The reference Figure 1 shows a SEM image of γ-alumina particles which appear to be composed of coarse secondary particles, in which primary particles with a diameter of 30 to 50 nm are agglomerated. As shown in reference Figure 2, when using TEM of the same power as that of reference Figure 1, it is obvious that said particles are composed of an aggregation of fibrous fine crystals. By a TEM image of higher power (reference Figure 3) it is observed that some of the fibrous fine crystals are a single crystal (primary particle). Thus, as is clear from a comparison with reference Figure 1, porous particles composed of fibrous fine crystals according to a TEM image would be observed to be dense primary particles in a SEM image. Therefore, it is necessary to confirm that the particle is a single crystal, a dense polycrystal or a porous polycrystal (secondary particle), in which finer primary particles are agglomerated having a slight bond, in order to judge whether it is a primary particle or not. However, this judgement is impossible using solely SEM and also it is necessary to use, for example, TEM. Accordingly, even if the particles of Fig. 2 as shown in document D6 appear to be primary particles in the

SEM image, this would have to be checked by other means like TEM, such that said Fig. 2 is not useful in showing that SEM can provide images of primary particles. As to the practical measurement of an average particle diameter, the measurement of this parameter by SEM is made by an analysis of images. As to all particulate matters found in a SEM image, the form of particulate matter can be obtained by following its outline, wherein the average of its upper diameter value and its lower diameter value is considered to be a particle diameter, wherein an average of each particle diameter thus obtained is taken as the average particle diameter.

(c) In the present technical field, particles existing as a primary particle will not be present, since it is common knowledge of the skilled person that nanometer-scale primary particles agglomerate to form secondary particles, such that it is obvious that the feature "average particle diameter of 5 to 100 nm" included in the claims of the present application indicates a secondary particle diameter.

X. The appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the claims according to the main request filed with the letter of 23 April 2008 or, alternatively, on the basis of one of the sets of claims according to the auxiliary requests 1 to 7, also filed on 23 April 2008. As a further auxiliary request, in case the board considered none of the above requests to be allowable

and did not intend to issue a further communication, oral proceedings were requested.

Reasons for the Decision

1. *Sufficiency of disclosure*

At the oral proceedings before the board, the discussion focused on the issue of whether it was clear for the skilled person from the application that the "average particle diameter", when measured on the finished catalyst by SEM, meant "average particle diameter of secondary particles and, if present at all, of primary particles which are not part of secondary particles and do not adhere thereto", as put forward by the appellant, or whether it could also mean for instance "average particle diameter of primary particles", as suggested by the content of document D6 (in particular Figure 2).

1.1 This issue having been raised for the first time at the oral proceedings, the proceedings were continued in writing in order to allow the appellant to file evidence that the size of secondary particles in the claimed catalysts could be measured by SEM.

In its communication dated 14 December 2007, the board furthermore drew the appellant's attention to the fact that if such evidence were filed, it should be proven up to the hilt that in the present case, SEM would **not** allow measurement of the average particle size of primary particles in the claimed catalyst, but would

unambiguously allow measurement of the particle size of **secondary** particles **only**.

1.2 The evidence submitted by the appellant on 23 April 2008 consisted of one SEM image and two TEM images of a commercial γ-alumina accompanied by the argumentation summarized in item IX. supra.

Apart from the fact that the copy with the three black and white images was of bad quality, the board notes that the evidence submitted by the appellant neither corresponded to that requested by the board in its communication of 14 December 2007, nor to that proposed by the appellant at the oral proceedings. Furthermore, the material - a commercial γ-alumina - on which were conducted the experiments supposed to support said evidence, can neither be equated with the claimed catalyst, nor with the key ingredient of the catalyst, namely cerium oxide.

Furthermore, as the appellant did not give any explanation as to why the submitted evidence should be considered to be of an equivalent standard to that requested, it is the board's opinion that the evidence now at disposal is not conducive to considering the Article 83 EPC issue as overcome.

2. *Remittal*

2.1 Under such circumstances, as the decision under appeal dealt exclusively with amendments which contravened Article 123(2) EPC as well as with lack of novelty, and as the claims have furthermore been substantially amended with respect to those on which the decision was

based, the board considers that present claim 1 generates a fresh case not yet addressed in examination proceedings and requiring re-examination. While Article 111(1) EPC gives the Board of Appeal the power to raise fresh issue in *ex-parte* proceedings where the application has been refused on other issues, proceedings before the Boards of Appeal in *ex-parte* cases are primarily concerned with examining the contested decision (see decisions G 10/93, OJ EPO 1995, 172, points 4 and 5 of the reasons), fresh issues normally being left to the Examining Division to consider after a referral back, so that the Appellant has the opportunity for these to be considered without loss of an instance. The examination not having been concluded, the Board considers it appropriate to exercise the power conferred on it by Article 111(1) EPC to remit the case to the examining division for further prosecution.

2.2 The issues below merit consideration when resuming examination proceedings.

2.2.1 Main request - Allowability of the amendments under Article 123(2) EPC

Although the independent product claims 1 and 2 and the independent process claim 10 have a basis in the application as filed, namely :

- claim 1: in claim 1; in the passage at page 16, lines 4 to 6 and in the 1st preferred embodiment (pages 30 and 31),

– claim 2: in claim 1; in the passage at page 16, lines 4 to 6 and in the 2nd and 3rd preferred embodiments (pages 31 to 34),

– claim 10: in claims 10 and 1; in the passage at page 16, lines 4 to 6 and in the 1st preferred embodiment (pages 30 and 31),

the question arises whether the requirements of Article 123(2) EPC are met in particular as regards the amended process claims 11, 12, 13 and their dependent claims.

2.2.2 Main request - Novelty

D1 discloses in its claim 1 an exhaust gas purifying catalyst […] having, carried on a monolithic carrier, a mixture comprising a catalytically active component comprising 0.5 to 30 g of an alkaline earth metal oxide, 10 to 150 g of cerium oxide, 0.1 to 50 g of zirconium oxide and 10 to 300 g of an activated alumina, and optionally […].

D1 (page 4, lines 7 to 15) requires that the cerium oxide forms crystals not greater than 250 Å (25 nm) in diameter after calcination in air at 900°C for 10 hours. The crystal diameters are determined by preparing a powder at least partially in the form of a composite or solid solution composed of cerium oxide and zirconium oxide, optionally preparing a catalyst, then firing the powder or the catalyst in air at 900°C for 10 hours, subjecting the calcined product to X-ray diffraction analysis, and performing a calculation using the half-value widths found in the X-ray diffraction chart.

Table 4 of D1 indicates the CeO_2 crystal diameters of six catalyst samples measured by XRD; those of samples c) and d) - which have a CeO_2/ZrO_2 molar ratio falling within the range defined in claim 2 of the present request - have the values 135 and 138 Å, respectively.

The appellant argued on the basis of the experimental results of July 2002 that the actual average particle diameter of CeO_2 in the samples c) and d) lay within a range of from 300 nm to several decades of µm, i.e. outside the range defined in present claim 1.

The board observes that in order to assess the above argument and to decide on novelty, it is necessary to know whether, for the skilled person, the meaning of the parameter "average particle diameter" is sufficiently clear from the content of the application. But insofar as the answer to the question of novelty is dependent upon the pending Article 83 EPC issue, no conclusion can be taken in this respect.

It should nevertheless be noted that in the experiments of July 2002, the CeO_2 average particle diameter in the catalyst samples c) and d) was determined according to a photon correlation method - i.e. a method which measures the particle diameter of secondary particles and if present at all, of primary particles which do not adhere to the secondary particles (see in particular the "Fourth Preferred Embodiment" (pages 43 to 45; Figures 1 and 2 of the present application) - whereas in present claim 1 the average particle diameter is measured by SEM. The question thus arises whether any conclusion as to the novelty of the

subject-matter presently claimed can be drawn from the results obtained with the photon correlation method.

2.2.3 The objections raised in items 2.1, 2.2, 2.3, 2.4 (first paragraph), 2.6, 2.7 and 2.8 in the board's communication have been overcome and no longer apply to the present claims.

The objection under Article 84 EPC regarding the dependency of claims 11 to 13 on claim 2 (see item 2.4 (second paragraph) of said communication) is still pending.

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 for further prosecution on the basis of the main request submitted under cover of the letter dated 23 April 2008.

The Registrar: The Chairman:

C. Vodz G. Raths