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Case Number: T 2080/09 - 3.3.05

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Language of the proceedings: EN

Title of invention:
NOx exhaust gas purifying catalyst

Applicant:
HONDA MOTOR CO., LTD.

Headword:

Relevant legal provisions:
EPC Art. 56, 84, 123(2)

Keyword:
Added-subject-matter: main request (yes); auxiliary request (no)
Clarity: auxiliary request (yes)
inventive step: auxiliary request (yes)-non obvious modification

Decisions cited:

Catchword:

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Case Number: T 2080/09 - 3.3.05

D E C I S I O N
of Technical Board of Appeal 3.3.05
of 3 April 2014

Appellant: (Applicant)	HONDA MOTOR CO., LTD. 1-1, Minami-Aoyama, Minato-ku Tokyo 107-8556 (JP)
Representative:	Böhm, Brigitte Weickmann & Weickmann Patentanwälte Postfach 86 08 20 81635 München (DE)
Decision under appeal:	**Decision of the Examining Division of the European Patent Office posted on 8 April 2009 refusing European patent application No. 07013912.6 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman: G. Raths
Members: H. Engl
P. Guntz

Summary of Facts and Submissions

I. This appeal is from the decision of the examining division, posted on 8 April 2009, refusing European patent application EP 07 013 912.6.

The documents cited during the examination procedure included the following:

D1: WO-A-2005/044 426
&
D1a: EP-A-1 685 891
D2: US-A-2002/187 893
D3: JP-A-02 904 862
&
Accession no. 1990-327378, Derwent Publications Ltd., London, GB; Class E36, Section Ch, Week 199043
D4: JP-A-2004 120 873
&
D4a: WO-A-02/41 991
D5: US-A-5 354 720

II. In the contested decision, the claimed catalyst was considered to lack an inventive step having regard to documents D1 (D1a) and D4a (Article 56 EPC). Starting from D1a, the technical problem was to provide an alternative NO_x reduction catalyst for the purification of exhaust gas from combustion engines with periodic lean/rich cycles.

According to the examining division, the selections which hade to be made within the teaching of D1 were arbitrary and obvious, in particular as it was known from D4a that a zeolite beta exchanged with iron and cerium was effective in the selective catalytic

reduction of NO_x with ammonia at temperatures in the range of 250 to 600°C.

III. The notice of appeal of the applicant (henceforth: the appellant) was filed by letter dated 15 June 2009. The statement of grounds of appeal was received under cover of a letter dated 18 August 2009 and was accompanied by a set of claims constituting a main request and an auxiliary request.

The appellant also filed an Experimental Report (Enclosure 4).

The claims of the main request were later substituted by a fresh set of claims 1 to 9 submitted with letter dated 3 September 2009.

IV. The independent claims 1 and 7 of the main request read as follows:

"1. An NO_x purifying catalyst for purifying NO_x in exhaust gas discharged from an internal combustion engine with air-fuel ratios controlled to be lean and rich, comprising:

a first catalytic layer that includes a β-zeolite having an iron element and a cerium element, but not having a noble metal;

a second catalytic layer that includes a noble metal and cerium-oxide based material; wherein

the second and the first catalytic layers are coated on a support in that order, so that the first catalytic layer is the uppermost layer."

"7. Use of an NO_x purifying catalyst according to claims 1-6 for purifying NO_x in exhaust gas discharged from an internal combustion engine with air-fuel ratios controlled to be lean and rich."

Claims 1 and 7 of the auxiliary request read:

"1. An NO_x purifying catalyst for purifying NO_x in exhaust gas discharged from an internal combustion engine with air-fuel ratios controlled to be lean and rich, comprising:

a first catalytic layer that **contains no noble metal components** and that includes a β-zeolite having an iron element and a cerium element;

a second catalytic layer that includes a noble metal and cerium-oxide based material; wherein

the second and the first catalytic layers are coated on a support in that order, so that the first catalytic layer is the uppermost layer."

"7. Use of an NO_x purifying catalyst according to claims 1-6 for purifying NO_x in exhaust gas discharged from an internal combustion engine with air-fuel ratios controlled to be lean and rich."

(Emphasis added by the board).

Dependent claims 2 to 6 and 8 and 9 define particular embodiments of the catalyst of claim 1 and of the use according to claim 7.

V. The arguments of the appellant may be summarized as follows:

It was absolutely clear to the skilled person that the entire first catalytic layer of the catalyst of claim 1 of the main request should not contain any noble metal element. Reference was made to the description, page 11, third paragraph, to support this interpretation.

D1 (D1a) represented the closest prior art. Starting from the two embodiments of catalysts having two layers (paragraphs [0042] and [0045]) and from example 11 of D1a, a multiple selection would have to be made in order to arrive at the presently claimed catalysts.

A characteristic feature of the catalyst according to the invention was the combination of two specific layers, wherein the first catalytic layer contained no noble metal components and includes a β-zeolite having an iron element and a cerium element. This technical feature had the effect of improving the NO_x purification performance. The technical problem consisted in providing a catalyst showing an improved NO_x conversion at low temperatures. Experimental evidence for the improvement was submitted as Enclosure 4.

The prior art did not hint at the claimed catalyst. In particular, the advantageous two layer approach having two active catalytic species, namely Fe/Ce in the first catalytic layer and a noble metal in the second catalytic layer, was not taught or suggested by the prior art. D4a even led away from it, as it disclosed only single layer catalysts and concentrated on improving stability at high temperatures which were not relevant for the present invention.

VI. Requests:

The appellant requested that the decision under appeal be set aside and a patent be granted on the basis of claims 1 to 9 of the main request filed with letter dated 3 September 2009 or in the alternative, on the basis of the claims 1 to 9 of the auxiliary request, filed with the statement of grounds of appeal.

Reasons for the Decision

1. Amendments

1.1 Main request

Current claim 1 is identical with claim 1 in the version underlying the contested decision.

The examining division argued (point 1 of the Reasons of the contested decision) that said claim 1 could be read in two possible ways, depending on which preceding claim element the expression *"but not having a noble metal"* referred to. Thus, in a first claim construction, the β-zeolite was free from a noble metal, in another construction it would be the entire first catalytic layer which was free from a noble metal. In view of the undisputed fact that the original application documents disclosed only the latter embodiment, the claim was rejected under Article 123(2) EPC as "not being supported by the original application".

The board concurs with the argument of the examining division. In present claim 1, in particular the second

paragraph thereof, the term of "not having a noble metal" may be construed to apply to either one of the preceding elements, namely to the β-zeolite or to the entire first catalytic layer. According to the appellant and in the understanding of the board, however, the claim's wording is meant to define a catalyst having a first catalytic layer free of noble metal.

In fact, the application documents as originally filed do not clearly and unambiguously disclose a catalyst wherein the β-zeolite is free from a noble metal, but the remaining components of the first catalytic layer comprise a noble metal element. The claim covers this second possibility, for which a basis is missing in the original disclosure. Therefore, the subject-matter of the claim contravenes Article 123(2) EPC.

The main request is not allowable.

1.2 Auxiliary request

In claim 1 of the auxiliary request, the definition of the first catalytic layer reads:

"a first catalytic layer that contains no noble metal components and that includes a β-zeolite having an iron element and a cerium element."

A basis for this wording is found in the application as filed on page 11, fourth paragraph. Therefore, claim 1 of auxiliary request 1 fulfils the requirements of Article 123(2) EPC.

The issue discussed under point 1.1 above with respect to claim 1 of the main request has been removed by

placing the expression *"that contains no noble metal components"* to a position immediately after the expression *"first catalytic layer"*. Thus, the problem of an ambiguous interpretation is resolved.

Claims 2 to 6 are based on claims 3 to 7 as originally filed. Use claims 7 to 9 are based on the disclosure of original claims 1 and 2 and on the paragraph of the description bridging pages 3 and 4.

The requirements of Article 123(2) EPC are met.

2. Article 84 EPC (auxiliary request)

Potential issues of lack of clarity of claim 1 have been rendered moot by moving the expression *"that contains no noble metal components"* to a position immediately after the expression *"first catalytic layer"*.

The requirement of Article 84 EPC is therefore met.

3. Novelty (auxiliary request)

The board is satisfied that none of the documents (see points 4.6.4 to 4.6.6) discloses a catalyst having all the features of claim 1 of the auxiliary request.

The requirements of Article 54 EPC are thus met.

4. Inventive step (auxiliary request)

4.1 *The invention*

The present invention is concerned with a catalyst for exhaust gas purification, in particular purification

from nitrogen oxides (NO_x).

4.2 *Closest prior art*

D1 is a document in Japanese language. D1a is a patent family member of D1 published after the priority date of the present application. Its contents were accepted by the appellant as a correct translation of D1. In the following, reference will be made to document D1a.

Said document D1a discloses catalysts for reducing nitrogen oxides (NO, NO_2) in exhaust gases of internal combustion engines subjected to combustion with periodic rich/lean excursions (see paragraphs [0001] and [0014]). For this purpose, D1a discloses three types of catalysts, one type being a single layer catalyst (see page 6, paragraph [0033]) and two types of catalysts having two layers (see paragraphs [0042] and [0045]).

One of the two layer catalysts comprises (see paragraph [0045]):

- an inner catalyst layer wherein the catalyst component (A) comprises
(A) (a) ceria or
(b) praseodymium oxide or
(c) an oxide and/or a composite oxide of at least two elements selected from Ce, Zr, Pr, Nd, Tb, Sm, Gd and La;
and the catalyst component (B) comprises
(B) (d) a noble metal catalyst component selected from platinum, rhodium, palladium and oxides thereof and
(e) a carrier,

- an outer catalyst layer comprising the above-

mentioned catalyst component (A) and the catalyst component (C) comprising

(C) (f) a solid acid, and

(g) a solid acid supporting an oxide of at least one element selected from V, W, Mo, Cu, Fe, Co, Ni and Mn.

D1 (D1a) thus qualifies as the closest prior art document not only in view of the similarity of the underlying technical problem, but also in view of the structural similarity of the catalysts.

4.3 *Problem to be solved*

According to the appellant, the technical problem to be solved by the present application can be seen in improving the NO_x conversion rate at low temperature conditions over D1 (see statement of grounds of appeal, dated 18 August 2009, page 6, paragraphs 4 and 5).

4.4 *Solution*

As a solution to this problem, the invention proposes an NO_x purifying catalyst characterized by a a first and uppermost catalytic layer that contains no noble metal components and includes a β-zeolite having an iron element and a cerium element.

4.5 *Success of the solution*

In order to demonstrate the success of the solution, the appellant submitted as Enclosure 4 to the statement of grounds of appeal fresh experimental evidence including a comparison of the performance of the inventive catalyst with catalysts obtained according to D1a.

For comparison purposes, example 11 of D1a was selected. Said example concerns a catalyst having an inner catalyst layer composed of y-alumina and 1% of platinum and an outer catalyst layer comprising cerium oxide powder and β-zeolite powder supporting 2.5% Fe_2O_3 (see paragraph [0120]). For a meaningful comparison, a modified catalyst having the same second catalytic layer as in the present invention (Pt and cerium oxide) was used. The board is satisfied that the so modified catalyst is still representative of the art disclosed in D1a.

The diagrams presented on pages 2/7 to 5/7 of Enclosure 4 demonstrate that the catalyst of the present invention exhibits a superior NO_x purification rate at low temperatures (200°C to 300°C) and an even significantly superior NO_x purification rate at high temperatures (350°C to 450°C), in comparison with the modified catalyst prepared in accordance with example 11 of D1a.

The reason for the superior performance can be explained by the increased NH_3 adsorption capacity of the upper layer which greatly influences NO_x purification performance. This effect is demonstrated by the data presented on page 7/7 of the said Enclosure 4, showing an increased level of NH_3 adsorption capacity for the inventive Fe, Ce ion exchange zeolite layer at all temperatures (150, 250 and 350°C), compared with a layer of Fe ion exchange zeolite.

The board notes that these comparison experiments have been performed under identical conditions as to the measurement of time, cycle number, lean/rich ratio and

model gas composition (see Enclosure 4, page 1/7). Therefore, the objections of the examining division (page 4, second paragraph, of the contested decision) were effectively taken into account.

In view of the above, the board can accept that the problem posed has been successfully solved.

4.6 *Obviousness*

It remains to be decided whether the claimed solution was obvious having regard to the prior art.

4.6.1 The appellant argued that starting from D1a four separate selections would have been necessary to arrive at the claimed catalyst structure (see point 4.2 above). These are:

- selecting Ce from four alternatives as a component (A) of the outer catalyst layer;
- selecting a β-zeolite from the list of acid-type zeolites for solid acid component (f) of the outer catalyst layer;
- selecting Fe from eight alternatives for the oxide (g) of catalyst component (C) in the outer catalyst layer; and
- selecting cerium oxide from four alternatives for catalyst component (A) of the inner catalyst layer.

In the board's view, there is no motivation in D1a itself to make the above-mentioned four selections which resulted in the catalyst according to the present invention.

4.6.2 The appellant furthermore argued that the characteristic feature according to the invention was the combination of two specific catalyst layers,

wherein the β-zeolite-based first catalytic layer includes both Fe and Ce elements. This technical feature had been shown as having the effect of improving the NO_x removal performance, what appears to be plausible.

4.6.3 Looking now to the remaining documents, D4 (D4a) discloses stable, metal-promoted aluminosilicate β-zeolite catalysts for NO_x reduction ion-exchanged with a lanthanide salt, for instance cerium, or with iron (see claims 8 to 10; claim 78). D4 does not hint at the claimed catalysts because it only discloses single layer catalysts. Although a temperature range of 250 to 600°C is disclosed in claim 78 of D4a for the nitrogen oxide reduction reaction, the gist of D4a on maintaining catalytic activity under harsh hydrothermal conditions (see page 4, lines 16 to 21, page 5, lines 1 to 4; page 7, lines 10 to 15). Accordingly, the catalytic activity of the catalysts was examined experimentally only at temperatures on the higher end of the range and above, namely at temperatures of 425°C, 550°C and 650°C (see page 22, Tables 1 and 2; Figures 3 and 4). Catalytic reactions at these high temperatures are not relevant for the present invention whose object is to improve low temperature NO_x reduction. Therefore, D4a would be of no guidance for the skilled person in solving the problem posed.

4.6.4 Document D2 discloses a hydrocarbon (HC) trapping type exhaust gas purifying catalyst comprising on a cordierite honeycomb substrate an under layer of γ-alumina, a Ce-Pr double oxide and hydrate alumina powder and an upper layer comprising β-zeolite and a hydrate alumina powder. The double layer catalyst formed on the honeycomb substrate was then impregnated with a Pt catalyst.

Such a double layer catalyst differs from those of the instant invention in that both layers contain a noble metal catalyst and in that the β-zeolite does not contain Fe and Ce. Therefore, a combination of D1a and D2 would not lead to the claimed catalysts.

4.6.5 D3 (see abstract) discloses a method of purifying a gas stream from nitrogen oxides by contacting the stream and NH3 at 250 to 600°C with a catalyst composition comprising (i) a zeolite of USY beta or ZSM-20 type having a defined SiO_2/Al_2O_3 ratio and a specific pore structure and (ii) 0.1 to 30 wt.-% of a Fe or Cu promotor. D3 thus neither discloses a double layer catalyst nor Ce as a catalyst component.

4.6.6 D5 disclosed a catalyst for reducing nitrogen oxides in lean exhaust gases of motor vehicle engines. The catalyst comprises, on an inert structure reinforcing monolithic or honeycomb body, a first catalyst coating layer of aluminum oxide and/or cerium oxide optionally stabilized with rare earth metals or silicon dioxide. A second catalytic coating layer of a mordenite-type zeolite containing Cu and/or Fe is formed on the first coating layer and contains Ir and Pt as noble metals (see column 2, line 65 to column 3, line 11; claim 1). D5 does not disclose Ce as a component of the outer catalyst layer. The zeolite differs from the β-zeolite used in the present invention.

In summary, neither D3 nor D5 provide conclusive hints at the claimed subject-matter.

4.6.7 Therefore, the subject-matter of claim 1 of the auxiliary request involves an inventive step (Article 56 EPC), having regard to the prior art. For the same

reasons, the subject-matter of use claim 7, referring back to claim 1, and of the dependent claims is also patentable.

5. Oral proceedings

The appellant requested, as a further auxiliary measure, oral proceedings according to Article 116 EPC in case the board rejected both the main and the auxiliary request (see statement of grounds of appeal, page 2, point 1.3). Thus, in compliance with Article 116(1), first sentence, EPC, the board may reject the main request without an oral hearing, provided it grants the auxiliary request.

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 grant a patent on the basis of claims 1 to 9 of the auxiliary request, filed with letter dated 18 August 2009, and a description and figures to be adapted.

The Registrar: The Chairman:

C. Vodz G. Raths

Decision electronically authenticated