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**Datasheet for the decision
of 30 March 2009**

Case Number: T 1141/06 - 3.3.05

Application Number: 99202908.2

Publication Number: 0988885

IPC: B01D 53/86

Language of the proceedings: EN

Title of invention:

Method of denitrating exhaust gas

Applicant:

mitsubishi heavy industries, ltd.

Opponent:

-

Headword:

Denitrating of exhaust gases/MITSUBISHI

Relevant legal provisions:

EPC Art. 54(1)(2), 56, 84, 123(2)

Relevant legal provisions (EPC 1973):

-

Keyword:

"Main and first auxiliary request: inventive step (no):
obvious implementation in a process of a known catalyst type"
"Second auxiliary request: inventive step (yes): non obvious
implementation in a process of a specific catalyst type not
yet known for efficiently removing ammonia"

Decisions cited:

T 0409/91

Catchword:

-



Case Number: T 1141/06 - 3.3.05

D E C I S I O N
of the Technical Board of Appeal 3.3.05
of 30 March 2009

Appellant:

MITSUBISHI HEAVY INDUSTRIES, LTD.
5-1, Marunouchi 2-chome
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Tokyo (JP)

Representative:

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Decision under appeal:

**Decision of the Examining Division of the
European Patent Office posted 1 February 2006
refusing European patent application
No. 99202908.2 pursuant to Article 97(1) EPC
1973.**

Composition of the Board:

Chairman: G. Raths
Members: J.-M. Schwaller
S. Hoffmann

Summary of Facts and Submissions

- I. This appeal lies from the decision of the examining division refusing European patent application No. 99 202 908.2.
- II. In the contested decision - based on the sets of claims submitted as main and 1st auxiliary requests on 07 September 2005 and 17 January 2006, respectively - the examining division held that claim 1 of both requests lacked an inventive step over the process disclosed in document

D1: EP 0 694 329 A2

when taken in combination with common general knowledge, or alternatively when taken in combination with the teaching of either document

D2: EP 0 261 610 A1 or

D3: DE 42 14 183 A1.

- III. In the grounds of appeal dated 30 May 2006, the appellant requested that a patent be granted on the set of claims as filed on 14 September 2004, claim 1 of which reads:

"1. A method of denitrating exhaust gas for removing nitrogen oxides by catalytic reduction, in which a nitrogen-oxide-containing exhaust gas is introduced into a catalyst filled reaction chamber and ammonia is added as reducing agent which exceeds a theoretical

amount required for reducing the nitrogen oxides (NO_x), the method comprising the steps of: introducing exhaust gas and ammonia into a first denitrating catalyst layer disposed on an upstream side in the gas stream to remove nitrogen oxides; supplying the gas into a first ammonia decomposing catalyst layer disposed on a downstream side from the first denitrating catalyst layer to control the ammonia content to be suitable for the subsequent denitrating process, wherein the first ammonia decomposing catalyst layer having ability to oxidatively decompose unreacted ammonia into nitrogen and nitrogen oxides; then supplying the gas into a second denitrating catalyst layer disposed on a downstream side of the first ammonia decomposing catalyst layer; and supplying the gas into a second ammonia decomposing catalyst layer disposed on a downstream side of the second denitrating catalyst layer to remove the residual ammonia, the second ammonia decomposing catalyst layer being provided with a catalyst which can oxidatively decompose the residual ammonia into nitrogen and nitrogen oxides or a catalyst which can oxidatively decompose the residual ammonia into nitrogen oxides."

- IV. In a communication annexed to the summons to oral proceedings, the board in particular questioned the extent of generalisation of the catalysts defined in above claim 1.
- V. In response to the board's communication, the appellant submitted under cover of the letter dated 16 March 2009 two amended set of claims as a first and second auxiliary request, respectively.

Claim 1 of the 1st auxiliary request reads as follows:

"1. A method of denitrating exhaust gas for removing nitrogen oxides by catalytic reduction, in which a nitrogen-oxide-containing exhaust gas is introduced into a catalyst-filled reaction chamber and ammonia is added as reducing agent, which exceed a theoretical amount required for reducing the nitrogen oxides (NO_x), the method comprising the steps of:
introducing exhaust gas and ammonia into a first denitrating catalyst layer disposed on an upstream side in the gas stream to remove nitrogen oxides;
supplying the gas into a first ammonia decomposing catalyst layer disposed on a downstream side from the first denitrating catalyst layer to control the ammonia content to be suitable for the subsequent denitrating process, wherein the first ammonia decomposing catalyst layer having ability to oxidatively decompose unreacted ammonia into nitrogen and nitrogen oxides;
then supplying the gas into a second denitrating catalyst layer disposed on a downstream side of the first ammonia decomposing catalyst layer;
and supplying the gas into a second ammonia decomposing catalyst layer disposed on a downstream side of the second denitrating catalyst layer to remove the residual ammonia, the second ammonia decomposing catalyst layer being provided with a catalyst which can oxidatively decompose the residual ammonia into nitrogen and nitrogen oxides or a catalyst which can oxidatively decompose the residual ammonia into nitrogen oxides, wherein the catalysts used in the first and second denitrating catalyst layers contain conventional TiO_2 -type catalyst supports and oxides

consisting of vanadium (V), tungsten (W) and/or molybdenum (Mo), wherein the nitrogen selectivity of the catalyst used in the first and second ammonia decomposing catalyst layers provides nitrogen selectivity exceeding 70%, the nitrogen selectivity being defined by the following equation:

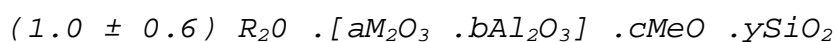
Nitrogen selectivity (%) = $[1 - \{C_{NO_x}^{OUT} - C_{NO_x}^{IN}\} / \{C_{NH_3}^{IN} - C_{NH_3}^{OUT}\}] \times 100$, wherein

$C_{NO_x}^{OUT}$: NO_x (ppm) at the outlet of the ammonia decomposing catalyst layer,

$C_{NO_x}^{IN}$: NO_x (ppm) at the inlet of the ammonia decomposing catalyst layer,

$C_{NH_3}^{OUT}$: NH_3 (ppm) at the outlet of the ammonia decomposing catalyst layer, and

$C_{NH_3}^{IN}$: NH_3 (ppm) at the inlet of the ammonia decomposing catalyst layer, and has, in its dehydrated form, a chemical formula expressed by



(R: alkali metal ion and/or hydrogen ion; M: one or more elements selected from the group consisting of Group VIII element in the periodic table, rare earth element, titanium, vanadium, chromium, niobium, antimony and gallium; Me: alkaline earth metal element; and

$a + b = 1$, $a \geq 0$, $b \geq 0$, $c \geq 0$, $y/c > 12$ and $y > 12$), and contains a crystalline silicate as catalyst support, the crystalline silicate having an X-ray diffraction pattern shown in Table 1 which is described herein, and one or more metals selected from the group consisting of platinum, palladium, ruthenium and iridium as active metal."

The subject-matter of claim 1 of the second auxiliary request reads as follows:

"1. A method of denitrating exhaust gas for removing nitrogen oxides by catalytic reduction, in which a nitrogen-oxide-containing exhaust gas is introduced into a catalyst-filled reaction chamber and ammonia is added as reducing agent, which exceed a theoretical amount required for reducing the nitrogen oxides (NO_x), the method comprising the steps of:

introducing exhaust gas and ammonia into a first denitrating catalyst layer disposed on an upstream side in the gas stream to remove nitrogen oxides;

supplying the gas into a first ammonia decomposing catalyst layer disposed on a downstream side from the first denitrating catalyst layer to control the ammonia content to be suitable for the subsequent denitrating process, wherein the first ammonia decomposing catalyst layer having ability to oxidatively decompose unreacted ammonia into nitrogen and nitrogen oxides;

then supplying the gas into a second denitrating catalyst layer disposed on a downstream side of the first ammonia decomposing catalyst layer;

and supplying the gas into a second ammonia decomposing catalyst layer disposed on a downstream side of the second denitrating catalyst layer to remove the residual ammonia, the second ammonia decomposing catalyst layer being provided with a catalyst which can oxidatively decompose the residual ammonia into nitrogen and nitrogen oxides or a catalyst which can oxidatively decompose the residual ammonia into nitrogen oxides, wherein the catalysts used in the first and second denitrating catalyst layers contain conventional TiO₂-type catalyst supports and oxides

consisting of vanadium (V), tungsten (W) and/or molybdenum (Mo), wherein the nitrogen selectivity of the catalyst used in the first ammonia decomposing catalyst layers provides nitrogen selectivity exceeding 70%, the nitrogen selectivity being defined by the following equation:

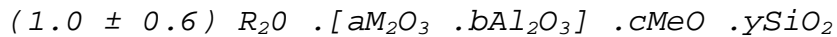
Nitrogen selectivity (%) = $[1 - \{C_{NOx}^{OUT} - C_{NOx}^{IN}\} / \{C_{NH3}^{IN} - C_{NH3}^{OUT}\}] \times 100$, wherein

C_{NOx}^{OUT} : NO_x (ppm) at the outlet of the ammonia decomposing catalyst layer,

C_{NOx}^{IN} : NO_x (ppm) at the inlet of the ammonia decomposing catalyst layer,

C_{NH3}^{OUT} : NH_3 (ppm) at the outlet of the ammonia decomposing catalyst layer, and

C_{NH3}^{IN} : NH_3 (ppm) at the inlet of the ammonia decomposing catalyst layer, and has, in its dehydrated form, a chemical formula expressed by



(R: alkali metal ion and/or hydrogen ion; M: one or more elements selected from the group consisting of Group VIII element in the periodic table, rare earth element, titanium, vanadium, chromium, niobium, antimony and gallium; Me: alkaline earth metal element; and

$a + b = 1, a \geq 0, b \geq 0, c \geq 0, y/c > 12$ and $y > 12$),

and contains a crystalline silicate as catalyst support, the crystalline silicate having an X-ray diffraction pattern shown in Table 1 which is described herein, and one or more metals selected from the group consisting of platinum, palladium, ruthenium and iridium as active metal,

and the catalyst used in the second ammonia decomposing catalyst layer contains one or more oxides selected from the group of silica, alumina, titania and zirconia

as catalyst support, and one or more metals selected from the group consisting of platinum, rhodium, palladium, ruthenium and iridium as active metal".

VI. At the oral proceedings, which took place on 30 March 2009, the discussion focused on inventive step.

VII. The appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the main request filed on 4 September 2004, or alternatively on the basis of the first or second auxiliary request, both filed with letter of 16 March 2009.

Reasons for the Decision

1. Main request - Inventive step

1.1 The present application relates to a method for catalytically removing nitrogen oxides (NO_x) from exhaust gases using ammonia as a reducing agent.

1.2 D1 relates to the same technical field and was acknowledged - as JP 8-38856 - in the application in suit as representing the closest state of the art. This document thus represents a reasonable starting point for assessing inventive step.

In its independent claim 3, D1 discloses a method for removing NO_x from an exhaust gas comprising adding ammonia thereto in an amount not less than the reaction equivalence for the nitrogen oxides, then introducing the gas mixture into a reaction vessel comprising in

sequence a denitration catalyst layer, a catalyst layer capable of decomposing oxidatively ammonia into nitrogen and nitrogen oxides and a denitration catalyst layer capable of decomposing ammonia, thereby removing the nitrogen oxides contained in said exhaust gas.

D1 further discloses (claim 4) that the ammonia decomposition catalyst has as a carrier a crystalline silicate represented, in its dehydrated form, by the formula $(1.0 \pm 0.8) R_2O \cdot [aM_2O_3 \cdot bAl_2O_3] \cdot cMeO \cdot ySiO_2$, wherein R denotes an alkaline metal ion and/or hydrogen ion, M denotes at least one element selected from the group consisting of VIII group elements, rare earth elements, titanium, vanadium, chromium, niobium, antimony and gallium, Me denotes an alkaline earth metal, $a + b = 1$, $a \geq 0$, $b \geq 0$, $c \geq 0$, $y/c > 12$ and $y > 12$, and which has an X-ray diffraction pattern as shown in Table 1 of D1; and at least one metal selected from the group consisting of platinum, palladium, ruthenium, iridium, and rhodium as an active metal.

In the denitration tests 2-1 to 2-3 summarized in Table 8 of D1, NO_x and ammonia concentrations of from 0.02 to 0.04 ppm and 0.7 to 2.3 ppm, respectively, were achieved at the outlet of the third catalyst layer.

1.3 As argued by the appellant, the problem to be solved in the light of D1 is to be seen in the provision of a further reduction of the ammonia concentration in the treated exhaust gas.

1.4 As a solution to this problem, the application in suit proposes the process according to claim 1, characterized in that the exhaust gas is supplied into

a second ammonia decomposing catalyst layer disposed on a downstream side of the second denitrating catalyst layer to remove the residual ammonia, the second ammonia decomposing catalyst layer being provided with a catalyst which can oxidatively decompose the residual ammonia into nitrogen and nitrogen oxides or a catalyst which can oxidatively decompose the residual ammonia into nitrogen oxides.

- 1.5 To the question whether the problem defined under item 1.3 has been solved, evidence in this sense is given in the application in suit (see Table 4), however only with the very specific catalyst "DASH-20M G4S-21 D4 (catalyst support: γ -Al₂O₃; 2 ± 0.2 g Pt/liter)" as the second ammonia decomposing catalyst.

The board has however no doubt that the decomposition of the "residual" ammonia may similarly be achieved with any other type of catalyst "which can oxidatively decompose ammonia into nitrogen oxides" or "which can oxidatively decompose ammonia into nitrogen and nitrogen oxides". It is therefore satisfied that the technical problem underlying the application might be successfully solved on the whole breadth of present claim 1.

- 1.6 It remains to be decided whether the proposed solution to the above problem is obvious or not in view of the state of the art.

In this respect, the appellant argued that D1 was teaching that lowering the ammonia input to the denitration process would decrease the ammonia exhaust emissions without affecting the denitration yields, and

therefore the skilled person confronted with the problem identified under item 1.3 would try this solution without looking for another one.

The board considers that even if the skilled person would have been inclined to try the above solution, it is above all presumed to be aware of everything which is common general knowledge in the art at the relevant filing date of the application in suit. In particular, it is supposed to be aware of any common general knowledge technology suitable for eliminating ammonia from a gas mixture, in particular of that technology according to which ammonia can be catalytically decomposed oxidatively into nitrogen and nitrogen oxides.

Such technology has been used precisely in the denitration process of D1 (see item 1.2 above), in which unreacted ammonia coming out of the first denitration catalyst layer is decomposed in an "ammonia decomposition catalyst layer containing a catalyst capable of decomposing oxidatively ammonia into nitrogen and nitrogen oxides".

The skilled person faced with the problem of reducing the ammonia concentration in the exhaust gas coming out of the second denitration catalyst layer of the same process cannot ignore this previous conversion step wherein the ammonia concentration had already been successfully reduced. As it is trivial that a catalyst decomposing ammonia successfully in a first step would work similarly in a further decomposition step, the board is of the opinion that the skilled person faced with the problem identified under item 1.3 would

obviously try to apply the successful ammonia decomposing catalyst also for decomposing the unreacted ammonia coming out of the second denitration catalyst, and so it would arrive at the subject-matter of claim 1 of the main request, which therefore lacks an inventive step within the meaning of Article 56 EPC.

2. *First auxiliary request - Inventive step*

2.1 The subject-matter of claim 1 of this request differs from that of the main request in that the catalysts have been further defined as follows:

- the catalyst used in the first and second denitrating layers contains conventional TiO₂-type catalyst supports and oxides consisting of vanadium (V), tungsten (W) and/or molybdenum (Mo);

- the catalyst used in the first and second ammonia decomposing layers provides nitrogen selectivity exceeding 70% and has, in its dehydrated form, a chemical formula expressed by
(1.0 ± 0.6) R₂O . [aM₂O₃ . bAl₂O₃] . cMeO . ySiO₂
(R: alkali metal ion and/or hydrogen ion; M: one or more elements selected from the group consisting of Group VIII element in the periodic table, rare earth element, titanium, vanadium, chromium, niobium, antimony and gallium; Me: alkaline earth metal element; a + b = 1, a ≥ 0, b ≥ 0, c ≥ 0, y/c > 12 and y > 12);
and contains a crystalline silicate as catalyst support, the crystalline silicate having an X-ray diffraction pattern shown in Table 1 of the application in suit, and one or more metals selected from the group

consisting of platinum, palladium, ruthenium and iridium as active metal.

- 2.2 The board observes that in D1, the ammonia decomposition catalyst is also defined as having a nitrogen selectivity of 70% or higher (D1, claim 2). Furthermore, the denitration reaction tests 2-1 and 2-2 exemplified in D1 (see also Table 8), in particular the systems 301 to 312, make use as well in their first and second denitration layers as in their ammonia decomposing layer, of catalysts which coincide with the definitions of the catalysts recited in the subject-matter of claim 1 of this request.

In this context, and for the same reasons as for the subject-matter of claim 1 of the previous request (see item 1.6), in particular because it is trivial for the skilled person that the ammonia decomposing catalyst used in the ammonia decomposing layer of D1 is supposed to work similarly in a further ammonia decomposing step, the subject-matter of claim 1 of this request also lacks an inventive step within the meaning of Article 56 EPC.

3. *Second auxiliary request*

3.1 *Allowability of the amendments under Article 123(2) EPC*

Amended claim 1 of this request finds its support in claims 1, 2, 3 and 4 of the application as filed and so meets the requirements of Article 123(2) EPC.

3.2 *Support by the description*

In accordance with Article 84 EPC, a claim must be supported by the description. This means that its scope must not be broader than is justified by the extent of the description and its contribution to the state of the art; it is the definition of the invention in the claims that needs support (T 409/91, OJ 9/1994, 653, reasons 3.3).

In the present case, each kind of catalyst used in the process according to claim 1 having been further specified by insertion of concrete catalyst features, the scope of protection of said claim has been considerably reduced, and the board is satisfied that the definition of the catalysts now in claim 1 is justified, as it reflects the catalyst type used in the example. Its concerns raised in the summons to oral proceedings relating to unjustified generalisation and to a lack of support of claim 1 by the description are therefore considered overcome.

3.3 *Novelty*

The subject-matter of claim 1 of this request is distinguished from the content of the documents cited in the search report, in particular D1, D2 and D3, at least in that the second ammonia decomposing catalyst contains one or more oxides selected from the group of silica, alumina, titania and zirconia as catalyst support, and one or more metals selected from the group consisting of platinum, rhodium, palladium, ruthenium and iridium as active metal. Claim 1 thus satisfies the requirements of Article 54 (1)(2) EPC.

3.4 *Inventive step*

3.4.1 As indicated under item 1.3, the problem to be solved in the light of D1 is to provide a further reduction of the ammonia concentration in the treated exhaust gases.

3.4.2 As a solution to this problem, the application in suit now proposes the process according to claim 1 of the present request, which differs from that of the previous request by the composition of the second ammonia decomposing catalyst, which contains one or more oxides selected from the group of silica, alumina, titania and zirconia as catalyst support, and one or more metals selected from the group consisting of platinum, rhodium, palladium, ruthenium and iridium as active metal.

3.4.3 To the question whether the above-mentioned problem has been successfully solved, Table 4 of the application in suit shows that by providing the second ammonia decomposing layer with a catalyst containing platinum as the active metal and γ -alumina as the supporting material (see also item 1.5 *supra*), the ammonia concentration at the outlet of the denitration process claimed can be reduced to 0.01 to 0.03 ppm, i.e. values well below the concentration ranges (0.7 to 2.3 ppm) achieved with the process of D1.

It is furthermore credible that the above-mentioned problem is solved over the whole breadth of claim 1, because in the present context the other precious metals claimed (palladium, ruthenium, rhodium, iridium) are considered as technically equivalent to platinum as the active metal. Similarly, the other oxides claimed

(silica, titania and zirconia) are considered as technically equivalent to alumina as the supporting material.

3.4.4 It remains to be decided whether the proposed solution is obvious or not in view of the state of the art. In this respect, it is observed that, in contrast to the second ammonia decomposing catalyst defined in claim 1 of the first auxiliary request, which is already known from document D1 as an efficient catalyst for oxidatively decomposing ammonia, neither D1 nor the other state of the art documents cited in the search report, disclose or suggest that a catalyst containing one or more oxides selected from the group of silica, alumina, titania and zirconia as the catalyst support, and one or more metals selected from the group consisting of platinum, rhodium, palladium, ruthenium and iridium as the active metal, might be useful for further removing ammonia from an exhaust gas. Accordingly, the skilled person faced with the problem indicated in item 1.3 *supra* had no hint to arrive at the subject-matter of claim 1 of the present request in the light of the above state of the art documents.

3.5 For the reasons indicated above, it is concluded that having regard to the state of the art, the subject-matter of claim 1 of this request is not obvious to a person skilled in the art and, therefore, it involves an inventive step within the meaning of Article 56 EPC.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to grant a patent on the basis of the claim of the second auxiliary request filed with letter of 16 March 2009, Figure 1 as originally filed, and a description to be adapted.

The Registrar:

The Chairman:

C. Vodz

G. Rath