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**Datasheet for the decision
of 5 April 2017**

Case Number: T 1014/14 - 3.3.05

Application Number: 02026805.8

Publication Number: 1317953

IPC: B01D53/94, B01J21/00,
B01J23/58, B01J23/63

Language of the proceedings: EN

Title of invention:

Use of a catalyst for lowering the amount of nitrogen oxides
in the exhaust gas from lean burn engines

Patent Proprietor:

Umicore AG & Co. KG

Opponent:

Johnson Matthey Public Limited Company

Headword:

Use of NO_x catalyst/UMICORE

Relevant legal provisions:

EPC Art. 56

Keyword:

Inventive step - main request (yes)

Decisions cited:

T 0405/00, T 1646/12, T 0542/13

Catchword:



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Case Number: T 1014/14 - 3.3.05

D E C I S I O N
of Technical Board of Appeal 3.3.05
of 5 April 2017

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Decision under appeal: **Decision of the Opposition Division of the
European Patent Office posted on 24 February
2014 revoking European patent No. 1317953
pursuant to Article 101(3)(b) EPC.**

Composition of the Board:

Chairman J.-M. Schwaller
Members: G. Glod
P. Guntz

Summary of Facts and Submissions

- I. The present appeal lies from the decision of the opposition division to revoke European patent EP-B1-1 317 953.

The following documents were cited in the decision:

D1A: English translation of D1 (JP 8 117 601)

D5: CA 2 280 631

- II. The statement of grounds included Annexes 1 and 2 relating to comparative examples. A further comparative test (Annex 3) was filed on 5 September 2014.

Claim 1 of the sole request - identical to the one underlying the decision - reads as follows:

"1. Use of a catalyst for lowering the amount of nitrogen oxides in the exhaust gas from lean burn engines, wherein the catalyst comprises at least one noble metal of the platinum group of the periodic table of the elements, as well as at least one nitrogen oxides storage material comprising at least one nitrogen oxides storage component on one or more support materials, in combination with a homogeneous Mg/Al mixed oxide of magnesium oxide and aluminum oxide, wherein magnesium oxide is present in a concentration of 1 to 40 wt.-%, based on the total weight of the Mg/Al mixed oxide, wherein the nitrogen oxides storage component is selected from barium oxide and strontium oxide and wherein the support material for the nitrogen oxides storage components consists of one or more high-melting metal oxides which are selected from the group consisting of cerium oxide and

cerium mixed oxides."

- III. The respondent (opponent) filed its reply by letter of 14 November 2014.
- IV. In its preliminary opinion the board endorsed the interpretation of claim 1 as given in T 542/13. Further, it held claim 1 to involve an inventive step.
- V. The respondent filed further arguments by letter of 3 March 2017.
- VI. Oral proceedings were held on 5 April 2017.
- VII. The appellant essentially argued as follows:

The skilled person with a mind willing to understand the claimed subject-matter would immediately recognise that the nitrogen oxides storage component could only be barium oxide and strontium oxide and that the support had to be cerium oxide and/or cerium mixed oxides. Barium oxide and strontium oxide could not be present on a different support such as the homogeneous Mg/Al mixed oxide. If a different interpretation were chosen, the restricted language used in the last part of claim 1 would be meaningless.

The subject-matter of claim 1 differed from example 1 of D5 in that the catalyst used contained a homogeneous Mg/Al mixed oxide.

The problem to be solved was to improve thermal stability and to provide better nitrogen oxides storage capability even after ageing. Figure 7 showed that said problem was solved.

D5 taught to use cerium/zirconium mixed oxide as a support for the nitrogen storage component barium oxide, but did not provide any incentive to change the support for platinum and/or rhodium. The same applied to the older document D1, which proposed the use of a complex oxide MAl_2O_4 (M: alkaline earth metal) as support for barium oxide.

There was no proof that in a catalyst according to example 1 of D5, barium was dissolved and subsequently moved to the aluminium oxide support.

Neither D5 nor D1 taught towards a homogeneous Mg/Al mixed oxide support for the noble metal, and so the solution to the problem was not obvious.

VIII. The respondent essentially argued as follows:

It agreed with the opposition division's interpretation of the claim.

In line with T 405/00, the wording "catalyst comprises" allowed barium oxide to be present as an unspecified material, in addition to the nitrogen oxides storage component that was part of the nitrogen oxides storage material recited in claim 1.

The support material was defined as consisting of "one or more high-melting metal oxides which are selected from the group consisting of cerium oxide and cerium mixed oxides". It was accepted that the language "selected from the group consisting of" defined a closed group for "the support material" specified in the last part of claim 1. However, the term "consists of" defined a closed group of possible support materials for at least one of the "one or more support

materials".

The plural form "nitrogen oxides storage components" in the expression "support material for the nitrogen oxides storage components" did not restrict the support material for all possible nitrogen oxides storage components that might be present in the catalyst of claim 1, including any nitrogen oxides storage components that might be present due to the wording "catalyst comprises".

The nitrogen oxides storage material that was explicitly defined in claim 1 could also include additional, unspecified features in addition to the "at least one nitrogen oxides storage component on one or more support materials".

It was evident that there was nothing technically illogical about the interpretation that claim 1 encompassed the use of a catalyst where barium oxide was supported on the homogeneous Mg/Al mixed oxide. The literal wording of claim 1 related to a catalyst where both lithium and barium oxide were supported on the homogeneous Mg/Al mixed oxide, in addition to barium oxide being supported on a cerium/zirconium mixed oxide. The open wording of "*the catalyst comprises*" in combination with the feature "*a homogeneous Mg/Al mixed oxide*" permitted the presence of other components on the Mg/Al mixed oxide.

The expression "in combination with" was a non-specific definition because it was not clear if it was intended to define the homogeneous Mg/Al mixed oxide as part of the nitrogen oxides storage material or only as part of the overall catalyst. The broadest technically sensible meaning was that the homogeneous Mg/Al mixed oxide was

part of the overall catalyst, but not part of the nitrogen oxides storage material. This was also in line with dependent claims 11 and 12, which referred separately to the Mg/Al mixed oxide and the nitrogen oxide storage material, which was indicative of the Mg/Al mixed oxide not being part of the nitrogen oxides storage material. Further, claim 4 allowed for the presence of the nitrogen storage component cerium oxide on the Mg/Al mixed oxide. The features that defined the components of the nitrogen oxides storage material had no limiting effect on the Mg/Al mixed oxide.

According to the appellant's interpretation of claim 1, the subject-matter of claim 1 lacked an inventive step in view of D5 alone or in view of D5 in combination with D1.

The objective technical problem to be solved in view of Example 1 of D5 was to provide a catalyst for use in treating nitrogen oxides in an exhaust gas from a combustion engine which had improved thermal stability. When seeking to solve the objective technical problem, the person skilled in the art would not go against the fundamental teaching of D5, which was that the Ce/Zr mixed oxide should be used as the support material for the NO_x storage component. The skilled person would therefore focus on the Pt/Al₂O₃ and/or Rh/Al₂O₃ components. Among the limited number of conventional support materials taught in D5 (page 19, lines 24 to 26), magnesium/aluminium spinel would be tried with a reasonable expectation of success, especially since it had a surface area that was the closest to that of aluminium oxide (Table 1).

Due to the melting points of the barium hydroxide and/or carbonate and the presence of water in the exhaust

gas, the skilled person would be concerned that some of the barium species in Example 1 of D5 would dissolve from the Ce/Zr mixed oxide and migrate onto the nearby alumina during use. Therefore, he would consider replacing the alumina support material with a "barium resistant" support material. He would turn to D1 and inevitably arrive at the claimed subject-matter.

- IX. The appellant (patent proprietor) requested that the opposition division's decision be set aside and that the patent be maintained on the basis of the set of claims filed on 5 February 2013 and resubmitted with the statement of grounds of appeal of 7 July 2014.

The respondent (opponent) requested that the appeal be dismissed.

Reasons for the Decision

1. In its reply of 14 November 2014 the respondent indicated that claim 1 contravened Articles 100(a), 100(b) and 100(c) EPC, but only provided substantiation with respect to Article 56 EPC throughout the appeal procedure. Article 100(b) was not substantiated during opposition procedure and so is not part of the proceedings.

The board sees no reason to raise objections under Articles 54 or 123(2) EPC. Therefore the only point of debate is inventive step.

2. Interpretation of claim 1

Prior to deciding on inventive step, the scope of the claim needs to be established. The opposition division followed the interpretation of the opponent and

concluded that barium oxide may also be present on the homogeneous Mg/Al mixed oxide.

The board does not follow this interpretation for the following reasons:

The last part of claim 1 would not provide any real limitation to claim 1 if the opposition division's view were followed. The wording "*wherein the nitrogen oxides storage component is selected from barium oxide and strontium oxide*" limits the mandatory at least one nitrogen oxides storage component to barium oxide, strontium oxide or mixtures thereof. The use of "*is selected from*" means that only barium oxide and strontium oxide are eligible as nitrogen oxides storage components.

Further, the wording "*and wherein the support material for the nitrogen oxides storage components consists of one or more high-melting metal oxides which are selected from the group consisting of cerium oxide and cerium mixed oxides*" limits the support materials for barium oxide, strontium oxide or mixtures thereof to cerium oxide and cerium mixed oxides. The closed wording "*consisting of*" restricts the support material to cerium oxide and cerium mixed oxides. This means that when barium oxide and strontium oxide are present, they can only be supported by cerium oxide and cerium mixed oxides.

The expression "*catalyst comprises [...], as well as at least one nitrogen oxides storage material comprising at least one nitrogen oxides storage component on one or more support materials [...]*" means that several nitrogen oxides storage materials can optionally be present in the catalyst. However, these materials must

be made from components other than barium oxide, strontium oxide or mixtures thereof, such as for example supported or unsupported zeolites or supported magnesium oxide. In addition, the nitrogen oxides storage material that comprises barium oxide and/or strontium oxide as storage component can further comprise other non-oxygen-storing materials such as a noble metal.

This conclusion is also in line with claims 11 and 12, according to which platinum can be deposited on the Mg/Al mixed oxide and on the nitrogen storage component barium oxide and/or strontium oxide. Further, claim 4 is not in contradiction with the present interpretation, since it does not relate to barium and/or strontium oxide. The fact that the cerium oxide deposited on the Mg/Al mixed oxide may have nitrogen oxide storage capacity is also not in contradiction with the present interpretation.

Even accepting that the homogeneous Mg/Al mixed oxide of magnesium oxide and aluminium oxide is not part of the nitrogen oxides storage material, it cannot contain barium and/or strontium oxide, since the support for these components is limited to cerium oxide and cerium mixed oxides (see above).

This interpretation is in line with T 542/13 (Reasons 1.1.3), which concluded that nitrogen storage components other than those explicitly mentioned could also optionally be present in the catalyst.

This seems further to be in agreement with the description and the position taken in T 1646/12 concerning the interpretation of claims with respect to the description (Reasons 2.1). Since the meaning of

claim 1 is not unambiguous, the interpretation of claim 1 should be in line with the description. It is evident from the numerous examples that barium oxide is present only on cerium mixed oxide and not on Mg/Al mixed oxide (Table 3).

The interpretation is also not contradictory with T 405/00 (Reasons 2.2), since it is accepted that other nitrogen oxides storage materials/components other than the mandatory barium and/or strontium oxide can be present in the catalyst.

To summarise, the catalyst according to claim 1 needs to contain a nitrogen oxides storage material that contains barium oxide and/or strontium oxide supported on cerium oxide and/or cerium mixed oxides. It can contain other nitrogen oxides storage materials and/or components, but these cannot be barium oxide and/or strontium oxide. Therefore barium oxide and strontium oxide cannot be supported on the "homogeneous Mg/Al mixed oxide of magnesium oxide and aluminum oxide".

3. Inventive step

3.1 Invention

The invention relates to the use of a catalyst for lowering the amount of nitrogen oxides in the exhaust gas from lean-burn engines.

3.2 Closest prior art

The catalyst of example 1 of D5 is the closest prior art. This was not disputed by the parties. In said catalyst, barium oxide is coated on cerium/zirconium mixed oxide, platinum is deposited on aluminium oxide

and rhodium on aluminium oxide.

3.3 Problem

According to the contested patent, the problem underlying the invention is to provide a catalyst that has an improved thermal stability, a wider temperature window and an improved nitrogen conversion in that window (paragraph [0016]).

3.4 Solution

As a solution to this problem, the use of a catalyst according to claim 1 is proposed, which is characterised in that it comprises a homogeneous Mg/Al mixed oxide of magnesium oxide and aluminium oxide, wherein magnesium oxide is present in a concentration of 1 to 40 wt.-%, based on the total weight of the Mg/Al mixed oxide.

3.5 Success of the solution

It is accepted that the problem is solved. Figure 7 of the patent comparing comparative catalyst CCI to catalysts C4a and C4b gives an indication that the replacement of Al_2O_3 by a homogeneous mixed oxide of magnesium oxide and aluminium leads to improved storage efficiencies, especially for the aged catalysts. Although the comparative example is not completely in line with example 1 of D5 - in which the aluminum oxide contains some lanthanum - there is no reason to believe that the results would differ when choosing the catalyst according to example 1 of D5 as comparative catalyst. The respondent has not provided any evidence that would contradict these findings and has not

contested the results.

3.6 Obviousness

- 3.6.1 D5 also relates to catalysts with storage efficiency and improved resistance to ageing (page 7, lines 33 and 34). In view of the known problem of reaction of nitrogen oxides storage material such as barium oxide with certain support materials (page 3, lines 20 to 38), D5 proposes using for example cerium/zirconium mixed oxide as support material (page 8, lines 5 to 13). The catalytically active component may be present on the storage material itself or on a separate support material such as aluminium oxide (page 17, lines 36 to 38).

Example 1 illustrates such a catalyst containing cerium/zirconium (90/10) mixed oxide as support for barium oxide (page 25, lines 17 and 18) and aluminium oxide as support for the catalytically active components platinum and rhodium. Table 1 shows that conventional support materials exhibit strong mixed oxide production with barium oxide and potassium oxide, while the support used in the catalyst of example 1 has only a weak tendency to produce mixed oxide (page 19, lines 24 to 28, and Table 1). Therefore, the skilled person recognises that the support material used for barium in example 1 of D5 is optimally chosen.

D5 is however completely silent about any effect of the support material on the catalytically active component after ageing of the catalyst. The skilled person finds no incentive in D5 to exchange the aluminium oxide support for platinum and rhodium for a different one in order to solve the posed problem. All the data

presented in Table 1 relates to support for barium oxide.

- 3.6.2 The catalyst of example 1 was coated with barium oxide by impregnating with aqueous barium acetate solution, then drying and calcining at 500°C in air. As a result of calcining, the soluble barium acetate was fixed on the support material in the form of barium oxide and/or barium carbonate. This ensured that the barium acetate did not go into solution again during the subsequent preparation steps (D5, page 25, line 20, to page 26, line 5), which included calcination for 4 hours at 500°C (D5, page 26, lines 35 and 36). During operation, the catalyst was operated at temperatures between 150 and 500°C (D5, page 24, line 24, and figures 2 to 18). The skilled person would not expect barium to dissolve at these temperatures, since the melting temperature for barium carbonate is 811°C.

The ageing of the catalysts was done at a maximum temperature of 750°C at the entrance to the catalyst. This increased to about 830°C due to the exothermic reaction on the particular catalyst (D5, page 22, lines 22 to 27). However, these temperatures do not reflect the normal operating temperatures of the catalyst, but relate to an artificial ageing process.

The assertion that barium would dissolve during use and migrate to the close aluminium oxide support, thereby leading to the formation of mixed oxides, is based entirely on speculation not corroborated by any evidence. Therefore, D5 does not provide any hint for changing the support of the catalytically active compounds in order to solve the posed problem.

3.6.3 D1 (see paragraph [0005] of D1a) discloses that the NO_x clean-up performance of Pt/Ba/Al₂O₃ catalysts deteriorates after prolonged use, because at elevated temperatures barium reacts with alumina, so that Ba is lost and the NO_x cleaning performance declines. The solution proposed by D1 is to support barium on a complex MAl₂O₄ support to which an alkali metal has been added, with M being an alkaline earth (see paragraph [0008] of D1a).

D1 is thus also concerned with the reaction of barium with aluminium oxide and proposes a suitable support to overcome said problem. However, D1 like D5 does not relate to the type of support intended for the sole catalytic material, since in D1 the catalyst is deposited on the same support as barium (see Table 2). Therefore D1 does not provide any incentive to change the aluminium oxide support of example 1 of D5.

It follows that the solution to the posed problem is not obvious in view of the state of the art.

3.7 Independent claim 1, and claims 2 to 19 that directly or indirectly refer to claim 1, therefore involve an inventive step, and so the requirements of Article 56 EPC are fulfilled.

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 the set of claims as submitted with the statement of grounds of appeal dated 7 July 2014 and a description to be adapted thereto.

The Registrar:

The Chairman:



C. Vodz

J.-M. Schwaller

Decision electronically authenticated