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Datasheet for the decision of 30 January 2025

Case Number: T 2174/22 - 3.3.06

Application Number: 10833263.6

Publication Number: 2505263

IPC: B01J23/10, B01J33/00, F01N3/10

Language of the proceedings: EN

Title of invention:
COMPLEX OXIDE, METHOD FOR PRODUCING SAME AND EXHAUST GAS PURIFYING CATALYST

Patent Proprietor:
Solvay Special Chem Japan, Ltd.

Opponent:
Neo Chemicals & Oxides (Europe) Ltd.

Headword:
Solvay/Doped Ceria

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

Keyword:
Amendments - added subject-matter (yes) - allowable (no)
Inventive step - (no)

Decisions cited:

Catchword:

Boards of Appeal of the
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Case Number: T 2174/22 - 3.3.06

D E C I S I O N
of Technical Board of Appeal 3.3.06
of 30 January 2025

Appellant: Neo Chemicals & Oxides (Europe) Ltd.
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Decision under appeal: **Interlocutory decision of the Opposition Division of the European Patent Office posted on 19 July 2022 concerning maintenance of the European Patent No. 2505263 in amended form.**

Composition of the Board:

Chairman J.-M. Schwaller
Members: S. Arrojo
C. Heath

Summary of Facts and Submissions

I. An appeal was filed by the opponent against the decision of the opposition division to maintain European patent No. 2 505 263 in amended form on the basis of the claims according to the main request filed on 30 March 2021.

II. In its grounds of appeal, the appellant argued that the set of claims upheld by the opposition division extended beyond the content of the application as filed, was insufficiently disclosed and was not inventive over **D3** (WO 2008/156219 A1) in combination with **D4** (Rocchini et al., "*Relationships between Structural/Morphological Modifications and Oxygen Storage-Redox Behaviour of Silica-Doped Ceria*"), **D5** (Wang et al. "*Structure, Thermal Stability, and Reducibility of Si-Doped Ce-Zr-O Solid Solution*"), **D6** (US 5,529,969) or **D7** (US 6,350,421 Bl); or over D4 alone or in combination with D3, D7 or **D10** (EP 0 444 470 A1). Additionally, none of the auxiliary requests 1 to 17 then on file overcame the objections raised under Articles 123(2), 83 and 56 EPC. Documents D9 and D10 were submitted as annexes.

III. In its reply, the patent proprietor and respondent refuted the above objections and requested that document D10 not be admitted into the appeal proceedings.

IV. In its preliminary opinion, the board concluded that claims 3 and 4 of the main request did not meet the requirements of Article 123(2) EPC, and indicated that this conclusion also applied to auxiliary requests 2 to 5 and 11 to 17. Further, the invention defined in claim

1 of auxiliary request 1 was considered insufficiently disclosed, and thus did not meet the requirements of Article 83 EPC, which objection likewise applied to auxiliary requests 1 to 4, 6, 7, 11, and 14. Furthermore, the subject-matter of claim 1 of auxiliary request 8 was deemed not to involve an inventive step starting from D4 as the closest prior art.

V. In a submission dated 27 November 2024, the proprietor filed new arguments in response to the preliminary opinion of the board.

VI. In a submission dated 23 December 2024, the opponent requested that the new arguments filed by the proprietor be disregarded under Article 13(2) RPBA.

VII. At the oral proceedings, that took place on 30 January 2025, the proprietor withdrew the main request and auxiliary requests 1 to 4, 6 to 7 and 9 to 17.

VIII. Claims 2 and 3 of auxiliary request 5 respectively correspond to claims 3 and 4 of the main request, and read as follows:

"2. A method for producing a composite oxide according to claim 1 comprising the steps of:
(a) providing a cerium solution in which not less than 90 mol% of the cerium ions are tetravalent, wherein said cerium solution is a ceric nitrate solution or an ammonium ceric nitrate solution in water,
(b) heating to and maintaining said cerium solution obtained from step (a) at not lower than 60 °C for a duration of 10 minutes to 48 hours to obtain a cerium suspension,

(c) adding an oxide precursor of at least one element selected from yttrium and rare earth metal elements other than cerium, to said cerium suspension obtained through said heating and maintaining in step (b),

(d) heating said cerium suspension containing said oxide precursor and holding it at 100°C or greater for a duration of 10 minutes to 6 hours,

(e) adding a precipitant to the suspension obtained from step (d) to obtain a precipitate,

(f) calcining said precipitate at 250 to 500°C for a duration of 30 minutes to 36 hours,

(g) impregnating an oxide obtained through said calcining, with a solution of a silicon oxide precursor, and

(h) calcining said oxide impregnated with the solution of a silicon oxide precursor at 300 to 700°C for a duration of 1 to 10 hours."

"3. A method for producing a composite oxide according to claim 1 comprising the steps of:

(A) providing a cerium solution in which not less than 90 mol% of the cerium ions are tetravalent, wherein said cerium solution is a ceric nitrate solution or an ammonium ceric nitrate solution in water,

(B) heating to and maintaining said cerium solution obtained from step (A) at a temperature of not less than 60°C for a duration of 10 minutes to 48 hours to obtain a cerium suspension,

(C) adding a silicon oxide precursor and an oxide precursor of at least one element selected from yttrium and rare earth metal elements other than cerium to said cerium suspension obtained through the heating and maintaining in step (B),

(D) heating said cerium suspension containing said silicon oxide precursor and said oxide precursor, and

holding it at 100°C or greater for a duration of 10 minutes to 6 hours,
(E) adding a precipitant to the suspension obtained from step (D) to obtain a precipitate, and
(F) calcining said precipitate at 300 to 700°C for a duration of 1 to 10 hours."

Claim 1 according to **auxiliary requests 5 and 8** (now main and first auxiliary request, respectively) reads as follows:

"1. A composite oxide consisting of cerium, silicon, oxygen, and at least one element selected from yttrium and rare earth metal elements other than cerium, wherein a mass ratio of said cerium to said at least one element is 85:15 to 99:1 in terms of oxides, wherein a content of said silicon is 5 to 20 parts by mass in terms of SiO_2 with respect to 100 parts by mass of a total of said cerium and said at least one element in terms of oxides, wherein said composite oxide has properties of exhibiting a specific surface area of not less than 40 m^2/g as measured by BET method using nitrogen gas adsorption after calcination at 900°C for 5 hours, and a reducibility represented by a percent of trivalent cerium in the composite oxide reduced from tetravalent cerium of not lower than 40% as calculated from measurement of temperature-programmed reduction from 50°C to 900°C after calcination at 1000°C for 5 hours."

IX. The parties' final requests were as follows:

The appellant requested that the decision of the opposition division be set aside and the patent be revoked in its entirety.

The respondent requested that the patent be maintained on the basis of auxiliary request 5 filed with the reply to the appeal or, as an auxiliary measure, on the basis of auxiliary request 8 also filed with the reply to the appeal.

Reasons for the Decision

1. Auxiliary request 5 - Article 123(2) EPC

1.1 The subject-matters of claims 2 and 3 of this request correspond to those of claims 6 and 7 as filed with the following amendments to steps (d) and (D) (highlighted by the board):

Claim 2: "*(d) heating ~~and maintaining~~ said cerium suspension containing said oxide precursor ~~of at least one of rare earth metal elements other than cerium and including yttrium, up to and at not less than~~ <u>and holding it at</u> 100°C <u>or greater for a duration of 10 minutes to 6 hours,</u>...*"

Claim 3: "*(D) heating ~~and maintaining~~ said cerium suspension containing said silicon oxide precursor and said oxide precursor, ~~of at least one of rare earth metal elements other than cerium and including yttrium, up to and at not less than~~ <u>and holding it at</u> 100°C <u>or greater for a duration of 10 minutes to 6 hours,</u>...*"

1.2 The alleged basis for defining a time period of 10 minutes to 6 hours in connection with the step of holding the temperature is found on page 14, lines 7-8 of the description as filed, which reads: "*In step (d), the duration of the heating and maintaining is usually 10 minutes to 6 hours ...*".

1.3 The discussion primarily relates to the question of whether linking the time period to the step of "holding" (i.e. maintaining) the temperature, rather than to the originally described steps of "heating and maintaining", results in an unallowable extension of the subject-matter of the claims.

1.4 The opposition division and the respondent contended that the wording in the original application should not be interpreted as encompassing the period during which the temperature was ramping up. In this respect, they argued that a skilled person would understand the relevant time period to be that in which the solution or suspension was maintained at a high temperature, as this was when the chemical transformation occurred. This interpretation would be supported by the standard practice of avoiding artificial semantic constructions of the language in the claims and the specification. In support of this, they referred to the passage on page 14, lines 10-12 of the original description, which states that "*heating and maintaining, at lower than 100°C, the crystallinity of the precipitate ... is not sufficiently high*". In view of this indication, the skilled person would understand that the critical time period for carrying out the invention was the one in which the solution was held at a high temperature.

1.5 The opposition division also noted that the term "heating" did not necessarily imply an increase in temperature, as it also encompassed cases where energy was required in order to maintain an elevated temperature. Using this interpretation, the apparent contradiction in the wording of the claim would be resolved.

1.6 At the oral proceedings, the respondent further argued that when considering the original application as a whole (particularly paragraph 39 of the A1 publication, corresponding to page 14, lines 7-16 of the description as filed), it was directly and unambiguously clear to a person skilled in the art that the disclosed time periods could only refer to those during which the temperature was held at the required elevated values. This interpretation was further reinforced by the examples in the patent, which explicitly specified the duration for maintaining the high temperature but made no mention of the heating period. The proprietor also contended that a literal reading of the claims would lead to contradictions with the teaching of the patent as a whole. Specifically, under a literal interpretation, the original claims would encompass embodiments that could not achieve the intended purpose of the invention, such as processes where the temperature ramp-up phase consumes nearly the entire claimed period, failing to provide the necessary conditions for the chemical transformation to occur effectively.

1.7 The board disagrees with the above arguments for the following reasons:

The original disclosure explicitly describes the specified time periods as applying to the combined steps of "heating and maintaining". Interpreting this expression to include the time required for ramping up the temperature is not an artificial semantic construction, as it is standard practice in heating processes to account for both the temperature increase and its duration.

Moreover, the board notes that the only relevant criterion for assessing compliance with Article 123(2) EPC is whether the amendments introduce information that is not directly and unambiguously derivable from the original application. Therefore, the argument that a certain interpretation would encompass embodiments not achieving the desired effect is, in principle, irrelevant. It is also not apparent why the reference in the examples to the period of maintaining and/or the omission of the period of heating should somehow imply that the unambiguous concept of "heating and maintaining" should be reinterpreted.

While the opposition division correctly noted that energy input is required both to increase the temperature and to maintain it at an elevated value, this does not represent the most straightforward reading of the phrase "*the duration of the heating and maintaining...*" found on page 14 of the original application. The reason is that "heating" and "maintaining" are described as distinct, sequential steps rather than as interrelated, simultaneous processes (such as "heating for maintaining", or "heating in order to maintain"). Furthermore, the following wording is used both in original claim 6 and in the description (see page 5): "*heating and maintaining ... up to and at not lower than 100°C*" (emphasis added by the board), which clearly establishes a distinction between two separate steps: "heating up to" and "maintaining at", thus confirming that "heating" refers to the process of increasing the temperature "up to" at least 100°C, while "maintaining" refers to holding the temperature "at" that elevated level.

By limiting the restriction of the time periods to the step of maintaining the temperature at elevated values, claims 2 and 3 clearly extend beyond the disclosure of the application as filed, as they encompass embodiments not covered by the original wording. For instance, these claims cover processes in which step (d) or (D) is carried out slowly by heating the suspension to a temperature above 100°C over an extended period, followed by maintaining it at that temperature for the duration defined in claim 1, which could result in a total duration for the "heating and maintaining" steps exceeding the periods originally disclosed.

1.8 The board thus concludes that the subject-matter of claims 2 and 3 according to auxiliary request 5 extends beyond the content of the application as filed, and thus infringes Article 123(2) EPC.

2. Auxiliary request 8 - Inventive step

The requirements of Article 56 EPC are not met for the following reasons:

2.1 Closest prior art

Document D3 (see page 2, 3rd par.; claim 1) discloses a high specific surface area composite oxide consisting of cerium oxide doped with an oxide of another rare earth metal. According to this document, the composite oxide has a specific surface area after calcination at 900°C for 5 hours up to 33 m^2/g. Example 2 of D3, which appears to correspond to comparative example 3 of the contested patent, achieves a reducibility of 35% after calcination at 1000°C for 5h.

2.1.1 Document D4 (see abstract; page 462, left col.; Table 1; figures 1-2 and passage bridging pages 463 and 464) discloses a high specific surface area composite oxide consisting of silica-doped ceria, and indicates that the doping of ceria with silica in amounts ranging from 0.4 to 12.8% increases both the surface area and the reducibility of the product after calcination. The reducibility of the fresh material after calcination at 923 K is reported to increase from 40% for undoped ceria to 80% when ceria is doped with 12.8% silica.

2.1.2 Even though both documents represent a suitable starting point, the board will formulate the inventive step argumentation starting from D4.

2.1.3 Although D4 discloses the specific surface area and the reducibility after calcination, it does not explicitly anticipate the values obtained after the calcination steps defined in claim 1 at issue, i.e. a specific surface area after calcination at 900°C for 5 hours of not less than 40 m^2/g and a reducibility (measured as a percent of trivalent cerium in the composite oxide reduced from tetravalent cerium) not lower than 40% as calculated from measurement of temperature-programmed reduction from 50°C to 900°C after calcination at 1000°C for 5 hours.

2.1.4 The subject-matter of claim 1 at issue thus differs from D4 in that the composite oxide is doped with yttrium or rare earth metals other than cerium in a mass ratio of cerium to the other element of 85:15 to 99:1 and in the achievement of the above mentioned parametric values.

2.2 Problem solved by the invention

2.2.1 In view of the values of the parameters defined in the claim, the alleged invention aims to provide a composite oxide that achieves both a high specific surface area and high reducibility after calcination.

2.2.2 To support these technical effects, the opposed patent compares (see Table 1) ceria doped with yttrium or rare earth metals alone (comparative examples 1 to 4) with ceria doped by a combination of silica and rare earth metals or yttrium (examples 1 to 14). The proprietor submitted document D8, which compares the examples in the patent with five further comparative examples of silica-doped ceria (samples A to D) allegedly representing the closest prior art D4.

2.2.3 The proprietor argued that the combination of rare earth metals and silica led to the highest values in terms of the specific surface area and reducibility after calcination when compared to ceria doped with only silica or rare earth metals or yttrium. This was apparent from the results in the patent and in D8. Moreover, when these data were graphically plotted and the different basis used to calculate the proportions were taken into account (see pages 6 and 7 of the submission dated 27 November 2024), it was readily recognisable that the materials according to the invention provided significant advantages both in terms of reducibility and specific surface area after calcination when compared to the silica-doped ceria in D4. The problem solved by the invention was thus to provide a doped ceria with an improved specific surface area and reducibility after calcination.

2.2.4 The board is not convinced that the experiments in the patent and in D8 provide sufficient evidence that doping ceria with a combination of silica and yttrium or rare earth metals would in general give rise to an improvement with respect to the silica-doped ceria in the closest prior art D4.

As noted by the appellant, incorporating rare earth metals (Examples 2 and 6) into the 2% silica-doped ceria (Sample A) leads to significantly lower specific surface areas after calcination. Similarly, adding 15% La (Example 11), 10% Nd (Example 13) or 10% Y (Example 14) to the 5% silica-doped ceria (Sample B) also reduces the specific surface area after calcination. These observations also remain valid once the recalculation of the amount of silica proposed in the respondent's submission dated 27 November 2024 is taken into account, as the calculated silica proportions in samples A and B remain the same. Moreover, the board notes that Table 1 and the graphs concerning the specific surface area after calcination in said submission omit some of the exemplary embodiments in which the specific surface area after calcination is lower than in the comparative examples (examples 11, 13 and 14). It is thus apparent that the effect of increasing the specific surface area after calcination is not achieved over the whole scope of the claim.

Although in view of the comparisons shown in the table of D8, the materials according to the invention appear to consistently provide a higher reducibility than the comparative examples, the board has concluded that this cannot be considered to be unexpected or surprising, because it is already known from D4 that both silica and rare earth metals individually provide benefits in terms of reducibility. The technical effect of the alleged invention can therefore only be associated either with a synergistic or with an additive effect.

To support the achievement of a synergistic effect, it should be demonstrated that when ceria is doped with silica and rare earth metals or yttrium, a benefit is observed with respect to the alternative of using silica (or rare earth metals) alone as known from D4. This assessment should be conducted by comparing the results obtained with ceria materials treated with the same or similar overall amounts of dopants. In fact, when the comparison is made in this way, the following is observed: sample D doped only with 20% silica provides a higher reducibility (68%) than the ceria doped with 10% silica and 5% + 5% rare earth metals in examples 4 and 8 (61% and 66%), despite the fact that both contain a similar total amount of dopants. An analogous trend is observed when comparing the other examples in this way: Samples C and E, in which the ceria is doped with 10% silica, achieve a higher reducibility (53% and 59%) than any one of examples 7 to 14 (42% to 51%), despite the fact that the latter contain an overall higher amount of dopants (5% silica plus 10 to 15% of rare earth metals). It is thus apparent in view of these comparisons that combining silica and rare earth metals or yttrium does not give rise to a synergistic effect with respect to the alternative of doping the ceria with silica alone as known from D4.

The above conclusions also remain valid when the proportion of silica of samples C, E and D is recalculated taking into account the different basis as proposed in respondent's submission dated 27 November 2024. In particular, samples C and E would still achieve a higher reducibility than examples 7 to

14 with a lower overall amount of dopants, and sample D would achieve an 11% higher reducibility than examples 4 and 8, despite including only a 3% higher overall amount of dopants.

Finally, the board notes that even if it were accepted that the results shown in D8 demonstrate that incorporating rare earth metals or yttrium into a silica-doped ceria increases the reducibility, this would still not suffice to conclude that the invention achieves an improvement with respect to the teachings in D4, as this document does not only teach that doping ceria with silica improves the reducibility, but also that the reducibility can be further enhanced by increasing the amount of added silica. In particular, according to D4 (figure 1 and page 464, left col.), the reducibility after calcination of the silica-doped ceria increases from 51% to 61%, 74% and 80% when the amount of SiO_2 dopant is respectively increased from 0.4% to 2.1%, 6.4% and 12.8%. The board sees therefore no basis to conclude that adding rare earth metals or yttrium to silica-doped ceria would achieve better results than increasing the amount of silica as proposed in D8.

In view of the above considerations, the board concludes that the results on file do not demonstrate that doping ceria with a combination of silica and rare earth metals or yttrium leads to an improvement in specific surface area and/or reducibility after calcination when compared to the solutions proposed in D4, i.e. doping ceria with silica to provide a high specific surface area and reducibility after calcination and increasing the silica content to further enhance these properties. The only technical effect that can be acknowledged in relation to D4 is thus the provision of an alternative ceria material with high specific surface area and reducibility after calcination.

The invention therefore solves the problem of providing an alternative doped ceria with high specific surface area and reducibility after calcination.

2.3 Obviousness

2.3.1 The proprietor argued that a skilled person seeking to improve the specific surface area and/or reducibility after calcination would not find it obvious to incorporate an additional dopant into the silica-doped ceria disclosed in D4, as this document taught (see page 468) that ceria and silica reacted to form a distinct phase, $Ce_{9.33}(SiO_4)_6 O_2$, which was chemically different from pure ceria. As a result, a skilled person would have no reason to assume that the known benefits of adding rare earth metals or yttrium to pure silica would also apply to a silica-doped ceria. Therefore, the subject-matter of claim 1 was not obvious in view of the teachings in D4.

2.3.2 The board disagrees with the above argumentation for the following reasons:

As explained by the appellant, document D4 includes various teachings regarding the advantages in terms of reducibility of silica-doped ceria: firstly, it indicates (Figure 1 and passage bridging pages 463 and 464) that doping ceria with silica progressively improves the reducibility of a fresh material exposed to 923K calcination, with the reducibility increasing from 51% to 80% as the silica content rises from 0% (CS0) to 12.8% (CS6). D4 then explores the behaviour of undoped ceria and silica-doped ceria after exposure to redox cycles at different temperatures. The conclusion that doping ceria with silica enhances its reducibility remains valid in this context. However, the authors attempt to explain the behaviour of ceria and silica-doped ceria after exposure to one or more redox cycles. It is within this context that the passage on page 468, cited by the proprietor, assesses the structural modifications occurring during the reduction process. The conclusion is that, while the fresh sample consists of separate domains of crystalline ceria and amorphous silica (see page 467, right col.), exposure to an H_2 reduction cycle causes the silica and ceria phases to react and form $Ce_{9.33}(SiO_4)_6 O_2$. As explained in the conclusions (see page 477, right col.), when this phase is subjected to a re-oxidation step, it decomposes, resulting in the reformation of separate phases of amorphous silica and ceria crystallites. This process explains why silica-doped ceria maintains a high reducibility compared to undoped ceria, even after being exposed to one or more redox cycles.

Since, in view of these explanations, D4 clearly teaches that the silica-doped ceria is formed by separate ceria and silica phases, a skilled person would in principle expect that adding rare earth metals to silica-doped ceria will have a similar effect (at least on the ceria crystallites) as that occurring when rare earth metals are added to undoped ceria. Already from this perspective, it can be concluded that a skilled person seeking an alternative composite with high reducibility and being aware in view of D4 itself of the beneficial effects of rare earth metals on the reducibility of ceria, would at least contemplate incorporating rare earth metals into the silica-doped ceria.

For the sake of completeness, it should be noted that the board does not disagree with the general argument of the respondent that combining different dopants may lead to unexpected results and that, consequently, it would not be correct to assume that adding various dopants will necessarily result in an improved reducibility. This is confirmed by the patent results which, for instance, show (see Examples 10 and 11 or Comparative Examples 1 and 2) that adding more La to ceria or silica-doped ceria actually reduces the reducibility of the material. However, the conclusion on obviousness is not based on the argument that a skilled person would find it obvious to combine different dopants to improve the reducibility. As discussed above, the problem addressed by the invention is not to improve the reducibility, but to find alternative ways of achieving a high reducibility. This distinction is crucial, as in this case the skilled person does not require teachings suggesting an improvement in reducibility compared to D4, but rather information from which it can be reasonably inferred that adding rare earth metals to silica-doped ceria will likely lead to a material with good reducibility. As explained above, the board finds that such information can indeed be derived from the teachings in D4.

The board also notes that the specific ratio between ceria and rare earth metals or yttrium of 85:15 to 99:1 does not provide an inventive contribution as it extends over most of the conventional range used for dopants, so that a skilled person applying common knowledge would arrive at values within the claimed range.

Finally, even though D4 does not specify the specific surface area and reducibility after calcination under the conditions defined in claim 1 at issue, the values disclosed in this document (i.e. a specific surface area of 168 m^2/g (not clear if after calcination or in the fresh material) and 80% reducibility after calcination at 923K), as well as those achieved in D8, clearly show that both the specific surface area and the reducibility after calcination in D4 are within the ranges defined in claim 1 at issue (specific surface area of not less than 40 m^2/g after calcination at 900°C for 5 hours and reducibility not lower than 40% after calcination at 1000°C for 5 hours).

2.4 In view of the above considerations, the board concluded that a skilled person starting from document D4 and seeking to find an alternative ceria composite with high specific surface area and reducibility after calcination would, in view of the teachings in D4 and common knowledge, contemplate incorporating rare earth metals into the silica-doped ceria of this document. The subject-matter of claim 1 is therefore obvious in view of document D4. The requirements of inventive step under Article 56 EPC are therefore not met.

3. Since none of the requests submitted by the proprietor is allowable, the patent shall be revoked.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The patent is revoked.

The Registrar: The Chairman:

A. Wille J.-M. Schwaller

Decision electronically authenticated