

Veröffentlichung im Amtsblatt Publication in the Official Journal Publication au Journal Officiel	Ja/Nein Yes/No Oui/Non
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Aktenzeichen / Case Number / N° du recours : T 286/87 - 3.4.1

Anmeldenummer / Filing No / N° de la demande : 82 305 702.1

Veröffentlichungs-Nr. / Publication No / N° de la publication : 0 079 171

Bezeichnung der Erfindung: Non-catalytic process for reducing NO emissions
Title of invention: of combustible effluents
Titre de l'invention :

Klassifikation / Classification / Classement : B 01 D 53/34

ENTSCHEIDUNG / DECISION

vom / of / du 12 January 1989

Anmelder / Applicant / Demandeur : Exxon Research and Engineering Company

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 123(2) EPC - Article 56 EPC

Schlagwort / Keyword / Mot clé: "Extension beyond the content of the application
as filed (auxiliary requests, yes)" "Inventive
step (main and auxiliary requests, no)"

Leitsatz / Headnote / Sommaire

Europäisches
Patentamt

Beschwerdekammern

European Patent
Office

Boards of Appeal

Office européen
des brevets

Chambres de recours



Case Number : T 286/87 - 3.4.1

D E C I S I O N
of the Technical Board of Appeal 3.4.1
of 12 January 1989

Appellant : Exxon Research and Engineering Company
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Decision under appeal : Decision of Examining Division 031
of the European Patent Office
dated December 2nd 1986 refusing
European patent application
No. 82 305 702.1 pursuant to
Article 97(1) EPC

Composition of the Board :

Chairman : K. Lederer
Members : E. Turrini
C. Payraudeau

Summary of Facts and Submissions

- I. European patent application 82 305 702.1 (publication number 0 079 171) was refused by decision of the Examining Division. The decision was based on a set of six claims as submitted on 12 July 1986 of which Claim 1, the sole independent claim reads as follows:

"1. A non-catalytic process for reducing the amount of NO emissions in combustion effluent wherein a reducing gas comprising ammonia, either alone or in combination with one or more additional reducing gases, is injected into a flowing combustion effluent containing NO and oxygen at an elevated temperature whereby the resulting effluent gas has a reduced concentration of NO, and passing the resulting effluent gas in contact with a catalyst to reduce the content of ammonia therein characterized in that the said reducing gas is injected into the flowing combustion effluent when at least a portion of said combustion effluent is in a reduction zone at a temperature within the range of from greater than 700°C to no more than 1100°C to produce a treated effluent containing unconverted NO and residual ammonia, and in that the catalyst is a metallic material placed or located substantially at the end of the reduction zone and which is contacted by the treated effluent at a temperature in the range of from greater than 700°C to no more than 1100°C to produce a final effluent downstream of the catalyst which has a reduced content of ammonia compared with its ammonia content upstream of the catalyst and an NO content which is not significantly or substantially greater than its NO content upstream of the catalyst."

- II. The reason given for the refusal was that the subject-matter of independent Claim 1 lacked novelty in the sense of Article 54 EPC with respect to the disclosure of document JP-A-54 4698 (D1).
- III. The Appellant lodged an appeal against the decision.
- IV. The Appellant requests that the decision of the Examining Division be set aside and a European patent be granted on the basis of the application documents as filed on 12 July 1986, on which the refusal was based (main request).

As respective first to fifth auxiliary requests, the Appellant further requests a patent to be granted on the basis of either one of five sets of amended claims as submitted on 8 August 1988.

Claim 1 according to the first auxiliary request differs from Claim 1 of the main request in that the full stop of the end of that claim is replaced by ", and wherein the residence time of the effluent gas in the reduction zone is less than one second."

Claim 1 according to the second auxiliary request reads:

"1. A non-catalytic process for reducing the amount of NO emissions in combustion effluent wherein a gas comprising ammonia is injected into a flowing combustion effluent containing NO and oxygen at an elevated temperature whereby the resulting treated effluent gas has a reduced concentration of NO, and passing the resulting treated effluent gas in contact with a catalyst to reduce the content of ammonia therein, characterized in that the said treated effluent gas contains NO, the ammonia-comprising gas is injected into the flowing combustion effluent when at least a portion of the combustion effluent is in a reduction zone at a temperature in the range of from 700 to

1100°C and the said treated effluent is contacted with a catalyst of heat-resistant alloy selected from the group consisting of iron-based alloys, nickel-based alloys, chromium-based alloys, and oxide dispersion strengthened alloys, at a temperature in the range (a) between 900 and 1100°C when the reducing gas consists only of ammonia, or (b) 700 to 850°C when the reducing gas comprises ammonia and another reducing gas selected from hydrogen, carbon monoxide, paraffinic, olefinic and aromatic hydrocarbons and oxygenated hydrocarbons to produce a final effluent downstream of the catalyst which has a reduced content of ammonia downstream of the catalyst compared with the ammonia content upstream thereof and an NO content which is not significantly or substantially greater than its NO content upstream of the catalyst."

Claim 1 according to the third auxiliary request differs from Claim 1 of the second auxiliary request in that the full stop at the end of that claim is replaced by "and wherein the residence time of the effluent gas in the reduction zone is less than one second."

Claim 1 according to the fourth auxiliary request reads:

"1. A non-catalytic process for reducing the amount of NO emissions in combustion effluent wherein a gas comprising ammonia is injected into a flowing combustion effluent containing NO and oxygen at an elevated temperature whereby the resulting treated effluent gas has a reduced concentration of NO, and passing the resulting treated effluent gas in contact with a catalyst to reduce the content of ammonia therein, characterized in that the said treated effluent gas contains NO, the ammonia-comprising gas is injected into the flowing combustion effluent when at least a portion of the combustion effluent is in a reduction zone at a temperature in the range of from 700 to

1100°C and the said reducing gas comprises ammonia and another reducing gas selected from hydrogen, carbon monoxide, paraffinic, olefinic and aromatic hydrocarbons and oxygenated hydrocarbons, the temperature of contact with the catalyst is in the range 700 to 850°C to produce a final effluent downstream of the catalyst whose NH₃ and NO contents are respectively less than and not significantly or substantially greater than the NH₃ and NO contents upstream of the catalyst, and the catalyst is a heat-resistant alloy selected from the group consisting of high temperature iron-based alloys, nickel-based alloys, chromium-based alloys, and oxide dispersion strengthened alloys."

Claim 1 according to the fifth auxiliary request differs from Claim 1 of the fourth auxiliary request in that the full stop at the end of that claim is replaced by ",and wherein the residence time of the effluent gas in the reduction zone is less than one second."

The Appellant further offers to disclaim from the scope of the various Claims 1 the feature that the reduction zone contains metallic heat transfer surfaces, if need be.

- V. In support of his requests, the Appellant essentially argues that document D1 fails to disclose that the metal net used in the known process does in fact catalyse the reaction of residual ammonia and oxygen in the combustion exhaust gas to form harmless nitrogen and steam and that it describes a process for removing residual ammonia from a gas which is substantially free of NO. In particular, Example 1 and Figure 2 of D1 relate to a SO_x, not an NO_x removal process. The remaining Examples 2 and 3 disclose treating temperatures (550°C and 590°C, respectively) which are far below those employed in the process of the present invention (700°C - 1100°C).

The Appellant further alleges the existence of a technical prejudice against contacting an ammonia-treated gas containing NO, oxygen and residual ammonia with a transition metal or compound thereof in order to remove ammonia without significantly increasing the NO-content of the resulting gas, since the fact that such metals tend to promote the conversion of ammonia to NO_x in the presence of oxygen was not only well-known in the art, but also used on a very large scale for the industrial production of nitrogen oxides from ammonia as evidenced for instance by the document "Riegel's Industrial Chemistry, edited by J.A. Kent, Reinhold Book Corporation, 4th Printing, 1968, pages 105 to 111 (in particular page 106).

With respect to the feature specifying a residence time of less than one second of the effluent gas in the reduction zone as set out in Claims 1 in accordance with first, third and fifth auxiliary request, he submits that the disclosure of such residence time in the document US-A-4 129 651 (D4) is of no relevance to the claimed process. For D4 addresses the problem of maintaining the effectiveness of ammonia to eliminate NO_x from combustion gas in the presence of metallic heat transfer surfaces, which are absent from the zone where NO_x reduction is achieved in accordance with the process of the present invention.

Reasons for the Decision

1. The appeal is admissible
2. Main request
 - 2.1 Novelty

2.1.1 Document D1, an English translation of which (D'1) was submitted by the Appellant, discloses a non-catalytic process for reducing the amount of NO_x emissions in combustion effluent wherein a reducing gas comprising ammonia (alone) is injected into a flowing combustion effluent containing NO_x at an elevated temperature whereby the resulting effluent gas has a reduced concentration of NO_x (NO_x is reduced to nitrogen but the NO_x removal rate may "not reach 100%" or be "less than 90%"), and passing the resulting effluent gas in contact with a catalyst to reduce the content of ammonia therein. The said reducing gas is injected into the flowing combustion effluent when at least a portion of said combustion effluent is in a reduction zone at high temperature to produce a treated effluent containing residual ammonia. The catalyst is a metallic material (3) placed or located substantially at the end of the reduction zone and which is contacted by the treated effluent at a temperature in the range of from 300°C to 900°C (which partially overlaps the contact temperature range of from greater than 700°C to no more than 1100°C specified in Claim 1) to produce a final effluent downstream of the catalyst which has a reduced content of ammonia compared with its ammonia content upstream of the catalyst (since ammonia is decomposed to harmless gas) (D'1, page 2, lines 2 to 9 and 16 to 23; Figure 1).

Since the process disclosed in D1 can be used for reducing NO_x in exhaust gases of boilers or diesel engines (D'1, page 4, 3rd paragraph) which are well-known to contain NO and oxygen (present description page 1, lines 17 to 22 and page 4 lines 5 to 6, or US-A-3 900 544 (D2), column 2, lines 20 to 32), this process is implicitly disclosed as being suitable for reducing the concentration of NO in combustion effluent gases containing NO and oxygen.

Document D1 does not explicitly disclose that the final effluent downstream of the catalyst has a NO content which is not significantly or substantially greater than its NO content upstream of the catalyst as defined at the end of present Claim 1. This statement, however, defines a result to be achieved by the process rather than a feature of the process. Moreover, when interpreted in the light of the description, it fails to introduce any clear limitation to the scope of the claim for the increase in NO resulting from the catalytic ammonia decomposition reaction according to the claimed process may amount to 75% (see comparative Example C and Example 3), and even to more than 400% (see comparative Example H and Example 12). The above statement, therefore, cannot be regarded as clearly distinguishing the subject-matter of Claim 1 over the process known from document D1.

Having regard to Appellant's different interpretation of the content of this document, it is to be noticed that Document D1 discloses the use of a metallic catalyst for the decomposition of residual ammonia and oxygen in the treated combustion effluent to form harmless nitrogen and steam (D'1, page 3, lines 6 to 13 and page 4, 2nd paragraph), and that it also discloses a temperature range for the latter reaction (300°C - 900°C, page 2, Claim (1)) which partially overlaps the corresponding range as claimed. The mere fact that Examples 2 and 3 (D'1, page 5) refer to temperatures in the lower portion of the disclosed range (550°C and 590°C) does not render the disclosure of its upper portion ineffective, the more so since Example 1 (D'1, page 5) and Figure 2 clearly teach that NH₃ decomposition is in fact more effective at the higher limit of the disclosed temperature range than at its lower limit.

It is also acknowledged that the specific examples cited in D1 (D'1, page 5) all relate to the treatment of gases which are not specified to contain substantial amounts of NO_x as further submitted by the Appellant, but these specific examples, obviously, are meant only to illustrate the effect of the metallic catalyst on the residual NH₃ in the effluent combustion after NO_x-reduction (page 5, line 14). The process disclosed and claimed in D1 however, clearly comprises a prior NO_x-reduction step, wherein the NO_x removing rate may be less than 90% (page 2, lines 21 to 23) and therefore this document is fully relevant.

Thus the subject-matter of Claim 1 distinguishes over the process known from document D1 essentially in that the temperature in the NO reduction zone is specified to be within the range of from greater than 700°C to no more than 1100°C. In this respect, it should be noticed that the temperature data given throughout document D1 relate only to the temperature at which the residual ammonia containing gas is contacted by the metallic catalyst (page 2 Claim 1; page 3, 1st paragraph; page 4, 1st paragraph; page 5 Examples 1 to 3; Figure 2). The temperature in the reduction zone is merely stated to be "high" (page 2, lines 11 to 13; page 3, lines 5 to 6), and the fact that the temperature in the zone where catalytic ammonia decomposition occurs may be as low as 300°C or 450°C (page 4, 1st paragraph), which is far below the usual temperatures for NO reduction (D2, Claim 1), suggests that the temperatures in both zones may well be different.

- 2.1.2 Document (D2) discloses a non-catalytic process for reducing the amount of NO emissions in combustion effluent wherein a reducing gas comprising ammonia, either alone (Claim 19) or in combination with one or more additional reducing gases (Claim 1), is injected into a flowing combustion effluent containing NO and oxygen at an elevated

temperature whereby the resulting effluent gas has a reduced concentration of NO, in which the said reducing gas is injected into the flowing combustion effluent when at least a portion of said combustion effluent is in a reduction zone at temperature within the range of from 704°C to 1093°C (1300°F to 2000°F, Claim 1), which is substantially the range of from greater than 700°C to no more than 1100°C as defined in Claim 1, to produce a treated effluent containing unconverted NO (Table I).

Since the amount of ammonia injected in the NO containing combustion effluent gas may be up to 10 moles of ammonia per mole of NO (column 5, lines 28 to 33), it is further implicit from the disclosure of document D2 that the treated effluent may also contain residual ammonia as is well known in the art (D'1, page 2, last paragraph) and pointed out also in the description of the present application (page 4, lines 18 to 24).

The decomposition of such residual ammonia is not addressed in D2 and the subject-matter of Claim 1 thus distinguishes over the process known from document D2 in that it comprises further steps for catalytically reducing the content of residual ammonia in the treated effluent gas.

2.1.3 Document D4 relates to an improvement to the process of document D2, which aims at avoiding that, in industrial apparatus comprising heat exchanging means, ammonia injected in the flowing combustion effluent for non-catalytically reducing its NO content be itself converted into NO_x under catalytic activity of the metallic heat exchange surfaces (column 3, lines 4 to 29).

This document does not address the decomposition of residual ammonia after completion of the non-catalytic NO reduction reaction.

2.1.4 The remaining documents do not come closer to the claimed subject-matter.

2.1.5 Thus the subject-matter of Claim 1 is novel within the meaning of Article 54 EPC.

2.2 Inventive step

2.2.1 The Board considers document D1 as being the nearest prior art. This document clearly lacks sufficient information as regards the temperature range for the NO-reduction. Therefore, the objectively assessed technical problem to which the subject-matter of Claim 1 affords a solution is to determine a suitable temperature range for the reduction reaction of NO by ammonia in a known process.

2.2.2 However, putting into practice any technical process necessarily requires that the different operating parameters involved in the process be set properly.

2.2.3 Document D2 relates to a process for reducing the amount of NO emissions in combustion effluent which is obviously of the type used in the first stage of the process disclosed in D1, since it also involves the reduction without catalyst of NO by ammonia injected into a flowing combustion effluent containing NO and oxygen at an elevated temperature (Claim 1). The temperature at which NO-reduction occurs being specified to be in the range from 1300°F to 2000°F (Claim 1, line 49) i.e. from 704°C to 1093°C, the skilled person would contemplate a similar range for the reduction reaction in the process disclosed in D1 and thus arrive to the process defined in Claim 1, in which the corresponding temperature range (from greater than 700°C to no more than 1100°C) is substantially identical.

2.2.4 The Appellant's arguments in support of the patentability of the subject-matter of Claim 1 are not convincing.

In particular, since document D1, discloses a process wherein removing of the residual ammonia is achieved by contacting the residual ammonia containing gas with a metal surface in a temperature range overlapping the range claimed for that purpose (see point 2.1.1 above), there is no room for a general technical prejudice against the capability of such surface to decompose residual ammonia within the claimed temperature range.

The Appellant did not submit any argument in support of the non-obviousness of the claimed temperature range from greater than 700°C to no more than 1100°C for the temperature in the NO reduction zone, which is the sole distinguishing feature of the subject-matter of Claim 1 in accordance with the main request as compared with the process disclosed in document D1.

2.2.5 For these reasons, the subject-matter of Claim 1 lacks an inventive step in the sense of Article 56 EPC.

2.2.6 The subject-matter of the remaining Claims 2 to 6 of the set of claims in accordance with the main request does not imply an inventive step in the sense of Article 56 EPC either.

In particular, document D1 already discloses the use of iron-based alloys and stainless steel as a metallic catalyst (D'1, page 3, lines 14 and 15) as defined respectively in dependent Claims 2 and 3.

Document D2 discloses temperature ranges in the reduction zone of from 871°C to 1093°C (1600°F to 2000°F, Claim 19) and from 704°C to 871°C (1300°F to 1600°F, Claim 34) which

are substantially those defined in dependent Claims 4 and 6, respectively, and the use of hydrogen as an additional reducing gas (Claim 1, line 54) as defined in dependent Claim 5.

- 2.3 Thus, the claims in accordance with the main request fail to define patentable subject-matter in the sense of Article 52(1) EPC.

Applicant's main request therefore cannot be allowed.

3. Auxiliary requests

- 3.1 Claim 1 in accordance with Appellant's first auxiliary request is distinguished from Claim 1 in accordance with his main request only by the additional feature that the residence time of the effluent gas in the reduction zone is less than one second, as set out at the end of the claim.

This feature, however, is known already from document D4 which teaches that in a method for reducing the amount of NO_x in an exhaust combustion gas, wherein said combustion gas is preferably maintained at a temperature between 800°C and 1100°C in the presence of ammonia and oxygen, which is substantially the method used in the NO reduction step of the present process, "the length of time the combustion gas is maintained at said temperature is dependent upon the degree of distribution of the ammonia source in the combustion gas" and that "when ammonia is uniformly mixed, the time may be on the order of 0,01 seconds, possibly even less" (column 4, lines 22 to 38).

- 3.1.1 It is not denied in this respect that as submitted by the Appellant document D4 addresses the specific problem of maintaining the effectiveness of the NO_x reduction reaction in an industrial combustion apparatus such as a boiler for electric power generation, which comprises metallic heat

exchange surfaces in the high temperature zone in order to attain high heat efficiency (column 1, lines 61 to 66), and that it also discloses a preferred residence time of the combustion gas in the high temperature zone of "about one second" to achieve both effective ammonia mixing by diffusion and high heat efficiency in the boiler (column 4, lines 39 to 46). The indication in D4 that the latter value is preferred when the process is performed in industrial combustion apparatus, comprising metallic heat exchange surfaces in the reduction zone, would not in the Board's view dissuade the skilled person from contemplating using lower values such as those disclosed also in the document (0,01 seconds or less), in particular when the process is not intended to be implemented in an industrial apparatus, involving heat transfer through metallic surfaces located in the reduction zone, as is the case both for the process disclosed in D1 and for that of the present application.

- 3.1.2 For these reasons, the subject-matter of Claim 1 in accordance with Appellant's first auxiliary request is not considered to involve an inventive step within the meaning of Article 56 EPC, and the claim is not allowable under Article 52 EPC, accordingly.

Appellant's first auxiliary request cannot therefore be allowed either.

- 3.2 The introduction in Claims 1 in accordance with the second, third, fourth and fifth auxiliary requests of an upper limit of 850°C for the temperature range at which the treated effluent is contacted with the catalyst is not supported by the description as originally filed, which discloses such value in connection only with the temperature range at which NO-reduction occurs (page 2 lines 20 to 25, page 3, lines 15 to 25, claims 4 and 6). Since there is no suggestion in the original description that the temperatures should be constant throughout the

reduction zone, the wording "at least a portion of said combustion effluent is in a reduction zone at a temperature within the range of from ..." (Claim 1) suggesting instead that further portions of the combustion effluent (e.g. at the end of the reduction zone where catalytic decomposition of residual ammonia occurs) may be at a different temperature, specific temperature values originally disclosed only in relation with the NO-reduction process cannot be considered to apply also to the ammonia-decomposition reaction.

This objection was not contested by the Appellant, nor did he submit any argument in favour of the admissibility of the value 850°C as a new limit of the claimed temperature range.

Therefore, Claims 1 in accordance with the second, third, fourth and fifth auxiliary requests are all considered to define subject-matter extending beyond the content of the application documents as filed and, for that reason, to contravene the provisions of Article 123(2) EPC.

Appellant's second, third, fourth and fifth auxiliary requests cannot be allowed, accordingly.

- 3.3 While documents D1 and D2 do not relate to an NO reduction process involving heat transfer through metallic surfaces in the reduction zone the skilled person would not, in the Board's view, for the reasons set out above in paragraph 3.1.1, disregard the disclosure in document D4 of residence times of the effluent gas in the reduction zone of less than one second on the mere ground that this document is concerned with a NO_x reduction process involving heat recovery. The cited documents would therefore be equally relevant against the allowability of independent claims amended in such a way as to include a disclaimer specifying

that the reduction zone does not contain metallic heat transfer surfaces, as was further offered by the Appellant.

Introducing such disclaimer could not therefore confer patentability to the subject-matter of any of the various Claims 1 which have been submitted in the proceedings, and the Appellant need not be invited to do so, accordingly.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar

The Chairman

F. Klein

K. Lederer