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Datasheet for the decision
of 14 March 2008

Case Number: T 0081/04 - 3.3.07
Application Number: 96931288.3
Publication Number: 0801978
IPC: B01J 21/18
Language of the proceedings: EN

Title of invention:
Heat treated activated carbon for denitration, process for preparing the same, method of denitration using the same, and system of denitration using the same

Applicant:
OSAKA GAS CO., LTD., et al

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54

Relevant legal provisions (EPC 1973):
-

Keyword:
"Novelty - (yes)"

Decisions cited:
-

Catchword:
-
**Case Number:** T 0081/04 - 3.3.07

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**DECISION of the Technical Board of Appeal 3.3.07 of 14 March 2008**

**Appellants:**

(Applicants)

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**Decision under appeal:**

Decision of the Examining Division of the European Patent Office posted 14 August 2003 refusing European application No. 96931288.3 pursuant to Article 97(1) EPC.

**Composition of the Board:**

Chairman:  S. Perryman
Members:  B. ter Laan
G. Santavicca
Summary of Facts and Submissions

I. European patent application No. 96 931 288.3, originating from international application PCT/JP96/02737 (published as WO-A-97/12671) and having the international filing date of 24 September 1996, was refused by a decision of the Examining Division of the European Patent Office issued on 14 August 2003.

II. That decision was based on a set of nine claims filed on 15 May 2003 as the sole request, claim 1 reading:

"A denitration method which consists essentially of bringing exhaust gas containing nitrogen oxides and not more than 80% of water as water vapour, and ammonia gas having the same concentration as the nitrogen oxides, into contact with a heat-treated active carbon at a temperature ranging from ordinary temperature (about 5 to 40°C) to 100°C, in order to reduce the nitrogen oxides selectively and thereby decompose them to nitrogen and water, wherein said heat-treated active carbon is obtained by heat-treating raw active carbon fibres at 600 to 1200°C in a non-oxidising atmosphere so as to remove oxygen-containing functional groups present at the surfaces thereof and thereby reduce the atomic surface oxygen/surface carbon ratio to 0.05 or less."

The other independent claim referred to the use of such process (claim 8).

III. The Examining Division held that the subject-matter claimed according to the main request did not satisfy the requirements of Articles 54 and 56 EPC in view of
In particular, it was found that the subject-matter of claim 1 also included the use of a carbon treated by more process steps than actually indicated in the claim, i.e. a heat-treatment. D2 disclosed the contacting of exhaust gases with a carbon fibre catalyst prepared by heating active carbon and then treating it with acid. Although the atomic surface oxygen/surface carbon ratio was not mentioned in D2, in view of the similarity of the process with that of the application in suit, the carbon produced in D2 would also have an atomic surface oxygen/surface carbon ratio below 0.05, so that no difference could be seen between the carbon used in both exhaust gas treatment processes. Therefore, claim 1 was not novel.

The examining division also commented that in case of a limitation of claim 1 to include only the heat treatment of the carbon, the only difference with the process of D2 would be the absence of an acid treatment step. The problem to be solved would be to provide a simpler, cheaper and more environmentally friendly process for the production of active carbon. Although the omission of the acid treatment obviously reduced the preparation costs and provided a better environmental profile to the process, the examples showed that the omission of the acid treatment led to a reduction in the degree of denitration, hence a disadvantage, which was not compensated by any technical (unexpected) advantage. Therefore, the claimed subject-matter also lacked an inventive step.
IV. On 13 October 2003 a Notice of Appeal was lodged against that decision, together with an order for payment of the prescribed fee. The statement setting out the grounds of the appeal was filed on 15 December 2003.

After a communication from the Board dated 1 February 2008 in which several problems under Articles 123(2), 84 and 54 EPC were addressed, the appellant, with a letter dated 28 February 2008, filed a set of five claims as the sole request as well experimental results.

V. At the oral proceedings before the Board, held on 14 March 2008, after discussion of the claimed subject-matter, a new claim was filed as the sole request, that reads as follows:

"A selective catalytic reduction method for the denitration of exhaust gases which consists of the steps of:
(a) heat treating raw active carbon fibres having a pore diameter of 10 to 30 Å, a pore volume of 0.3 to 1.2 ml/g and a specific surface area of 500 to 2000 m²/g at 600 to 1200°C in a non-oxidising atmosphere such that the heat-treated active carbon has micropores with a size of 20 Å or less and an atomic surface oxygen/surface carbon ratio to 0.05 or less; and
(b) bringing exhaust gas containing 500 ppm or less of nitrogen oxides, 3% or more of oxygen and not more than 80% of water as water vapour and ammonia gas having the same concentration as the nitrogen oxides into contact, at a temperature of 100°C or below, with said heat-treated active carbon from step (a) as such."
VI. The Appellant's arguments can be summarised as follows:

(a) The subject-matter of the claim found its basis in the application as filed.

(b) The claim had been formulated in such a way as to make clear that the active carbon fibres used for the denitration had only been heat-treated and that no additional step for its preparation took place. D2 disclosed the use of active carbon fibres that had undergone an acid treatment after the heat treatment. Although the key distinguishing feature in the claimed method was the atomic surface oxygen/surface carbon ratio of 0.05 or less of the active carbon fibres, which was nowhere disclosed in the prior art, the use of the heat-treated carbon fibres as such provided a further feature distinguishing the claimed subject-matter from D2. Therefore, the claimed subject-matter was novel.

VII. The Appellant requested that the decision under appeal be set aside and that the case be remitted to the first instance for further prosecution on the basis of the claim submitted at the oral proceedings on 14 March 2008.

Reasons for the Decision

1. The appeal is admissible.
Amendments

2. The claim refers to a selective catalytic reduction method containing two steps: step (a) concerning the production of active carbon fibres that are directly used in step (b) according to which exhaust gases are denitrated.

2.1 That the claimed method is a selective catalytic reduction process can be seen from original claim 11 as well as original page 6, lines 16 to 25, in particular line 23, and original page 14, lines 14 to 16.

2.2 The basis for step (a) can be found in original claim 1 as well as original page 12, lines 21 to 25, for the properties of the raw material; original page 6, lines 16 to 25, and page 15, lines 18 to 22, for the pore size of the heat-treated product; and page 14, lines 5 to 16 for the properties of the exhaust gas.

2.3 The use of the active carbon fibres prepared directly by the method of step (a) for the denitration of the exhaust gases defined in step (b) at a temperature of 100°C or below can be found on original page 13, lines 1 to 10 in conjunction with page 14, lines 5 to 19.

2.4 Therefore, the subject-matter now being claimed finds a proper basis in the application as filed, so that the requirements of Article 123(2) EPC are fulfilled.

3. The Board also has no objections regarding clarity (Article 84 EPC).
Novelty

4. D2 discloses an activated carbon fibre obtained by calcining an activated carbon fibre at 600 to 1200°C in a non-oxidizing atmosphere and subsequently carrying out a sulphuric acid treatment (claim 1). The carbon fibre so obtained is then used in the catalytic reduction with ammonia of nitrogen oxides contained in exhaust gases (claims 2 and 3). In the working example a carbon fibre identified as "OG-5A" made by Osaka Gas Co. Ltd. was heated to 1000°C for 3 hours in a nitrogen gas atmosphere. Subsequently, the carbon fibre was soaked in sulphuric acid (37%) in an amount three times that of the carbon in weight and heated at 400°C for 4 hours.

4.1 Although the atomic surface oxygen/surface carbon ratio of the carbon fibre is not mentioned in D2, the application in suit contains examples describing treatment of the carbon fibre "OG-5A", which is also used in the process of D2. In example 3 "OG-5A" is only heat-treated, resulting in an atomic surface oxygen/surface carbon ratio of 0.025 (Table 1). In example 12 the treatment of "OG-5A" at 1000°C for one hour under nitrogen is followed by adding sulphuric acid (98%) to the carbon fibres, soaking them fully and heating them at 400°C until the sulphuric acid was evaporated completely. The resulting product has an atomic surface oxygen/surface carbon ratio of 0.040 (Table 2). Example 12 differs from the working example of D2 in the duration of the heat treatment (D2: 3 hours, example 12: 1 hour), in the concentration of the sulphuric acid (D2: 37%; Example 12: 98%) and possibly in the duration of the acid treatment (D2:
4 hours; Example 12: till evaporation). The conditions of the treatment in D2 being less aggressive than in example 12 of the application in suit, they would, if anything, lead to a lower atomic surface oxygen/surface carbon ratio than the 0.040 of example 12. Therefore, it has to be concluded that the active carbon produced in the working example of D2 will have an atomic surface oxygen/surface carbon ratio below 0.050.

4.2 In its letter dated 28 February 2008, the appellant compared the atomic surface oxygen/surface carbon ratio of the carbon fibre obtained in example 26 of the application in suit to an experiment indicated as example 2 of D2. In example 26 of the application in suit, the active carbon was "OG-7A" by Osaka Gas Co. Ltd., which was heated at 850°C during one hour under nitrogen. The resulting product has an atomic surface oxygen/surface carbon ratio of 0.030 (Table 3). According to the appellant's letter, in example 2 of D2, carbon fibre "OG-5A" was heated to 1000°C for 3 hours under nitrogen and then treated with sulphuric acid, resulting in an atomic surface oxygen/surface carbon ratio of 0.250. However, no details of the preparation method, in particular of the sulphuric acid treatment (such as concentration, temperature, duration), are given. Also, in D2 there is only one working example; example 2 is a comparative example. Therefore, it is not clear to which example and which experimental conditions exactly the appellant refers.

Furthermore, the atomic surface oxygen/surface carbon ratio of the comparative example in the appellant's letter of 28 February 2008 is much higher than that mentioned in the comparative examples of the
application in suit. In comparative example 1 an untreated "OG-5A" is described that has a value of 0.122, about twice as low as that of the appellant's additional experiment. For lack of experimental detail regarding the additional comparative example, it is not clear how such a difference with the untreated "OG-5A" could occur. Therefore, the additional experiment cannot be accepted as a valid reproduction of the preparation of a carbon fibre according to D2.

4.3 Consequently, the conclusion drawn above (cf. point 4.1) stands and the claimed upper limit for the atomic surface oxygen/surface carbon ratio of 0.050 cannot serve to distinguish the claimed method from the method described in D2.

5. D2 discloses a process in which raw active carbon is first heat-treated and subsequently treated with sulphuric acid before it is used to denitrate exhaust gases. According to the present claim, the carbon is heat-treated and then directly used as such for the denitration of exhaust gases. An additional treatment of the heat-treated carbon fibres is, by the present formulation of the claimed method, explicitly excluded. For that reason, the method now being claimed is novel over D2.

As no documents are on file that are more relevant for novelty than D2, it can be accepted that the requirements of Article 54 EPC are fulfilled.
Inventive step

6. In the decision to refuse the application, the examining division indicated that it did not consider a claim in which the preparation of the active carbon fibres had been limited to a heat-treatment only, to be inventive over D2. It would have been obvious for the skilled person, desiring to simplify the process, to omit the acid treatment, which omission led to a lower denitration without any compensation by another technical advantage.

6.1 The Board cannot share that point of view. Nowhere in D2 could the skilled person find the suggestion that the omission of the acid treatment might result in a process that still gives acceptable results. On the contrary, D2, for comparative purposes, describes the omission of the heat-treatment, but there is no hint that the acid treatment could be left out. There is no requirement in the EPC for any advantage, nor for a shortcoming brought about as the consequence of a certain measure to be compensated by another technical advantage. Therefore, D2 by itself cannot render the claimed subject-matter obvious.

6.2 In view of its opinion regarding D2, the examining division has not taken into consideration the other documents on file. In order to have a full examination carried out and to give the appellant the opportunity to be heard by two instances if necessary, the board remits the case to the first instance for further prosecution.
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 for further prosecution on the basis of the claim submitted at the oral proceedings on 14 March 2008.

Registrar     Chairman

C. Eickhoff     S. Perryman